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Quantum magnetoresistance in Si <B, Ni> whiskers

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It was studied the electrical magnetoresistance of nickel- and boron-doped filamentary silicon crystals in which a metal-insulator transition is observed. A giant magnetoresistance reaches up to 280 % in the Si whiskers with doping concentration of boron $p_{300\mathrm{K}} = 5\cdot 10^{18}$ cm^{-3} in the magnetic fields with induction up to 14 T at temperature 4.2 K. Peculiarities of magnetoresistance at low temperatures were shown to be caused by "core-shell" structure of crystals. A giant magnetoresistance nature was considered within quantum magnetoresistance model. The analysis was performed to determine the critical field of transition from classical parabolic magnetoresistance to quantum magnetoresistance, realized in the near-surface region of the crystal. The silicon whiskers were used for design of magnetic field sensors.

Keywords: silicon, whiskers, hopping conductivity, weak localization, spin-orbit interaction.

1. Introduction

State-of-art sensor electronics devices based on the giant magnetoresistive effect are created on the basis of metal films with ferromagnetic or antiferromagnetic interactions [1, 2], while most modern technologies are focused on semiconductors. Giant magnetoresistance (GMR) material has been developed into various applications of sensor based on magnetic field sensing, such as magnetic field sensor, a current sensor, linear and rotary position sensors, data storage, head recording, nonvolatile magnetic random access memory [1, 3]. The operation of such devices is based on the strong dependence of resistance on the magnetic field, caused by the exchange interaction of charge carriers with each other or with magnetic impurities. For example, the operation principle of magnetic field sensors manufactured by Murata Manufacturing Co., Ltd. is based on the effect of anisotropy of magnetoresistance. Such sensors have a change in magnetoresistance in the environment of about 2 % in the magnetic fields with the induction of 1 T, however. The voltage consumption of such sensors reaches 6 V. Equally important is the development of new technologies for the manufacture of sensors that are operational in harsh operating conditions (for example, ultra-low temperatures, high magnetic fields, etc.). In this case, the study of magnetotransport characteristics of materials is important because in such conditions there are quantum interference effects, such as weak localization [4, 5] or antilocalization of charge carriers, which lead to special behavior of magnetoresistance, in particular negative or abnormally positive magnetoresistance, and in some cases to the emergence of a giant magnetoresistance [6, 7]. Therefore, it is important to develop semiconductor spintronics compatible with modern technology of silicon chips, which is embodied in semiconductors diluted with magnetic impurities [8–10].

As you know [11], semiconductor microcrystal are good model objects for studying the magneto-transport properties of crystals due to the possibility of their doping in the process of growth by vapo-liquid-solid (VLS) mechanism. It is obvious that the study of the magnetic properties of a material doped with a magnetic impurity that has a magnetic moment due to an unfilled outer 3*d*-orbital, such as Ni or Mn, is fundamental and determines the predicted characteristics of the crystals. So, a hysteresis of magnetoresistance was observed in Si whiskers doped with a magnetic impurity (Ni) depending on the concentration of boron impurity, corresponding to the metal–insulator transition ($5\cdot 10^{18}$ cm^{-3}) [11]. At the same time, the occurrence of abnormally positive or negative magnetoresistance [12, 13] caused by the peculiarities of the near-surface transfer of charge carriers in crystals, where the magnetic impurity is mainly localized, was considered within the framework of low-temperature hopping conductivity. In this case, it is clear that the electron-electron interaction at localized impurity levels is significant. Basic electrophysical parameters such as coherence length, spin-orbit interaction energy were evaluated in the work [14]. However, the presence of an abnormally large positive magnetoresistance in silicon microcrystals at low temperatures in magnetic fields up to

14 T has not been sufficiently studied. Positive magnetoconductance maybe is due by the deformation of the wave functions of localized holes due to the magnetic field, which leads to the emergence, depending on the doping, a giant magnetoresistance in the crystals.

Therefore, the aim of the work is to study the magnetoresistance of silicon whiskers doped with a complex of impurities (B, Ni) to the concentration, which corresponds to the transition of metal–insulator, at low temperatures (down to the temperature of liquid helium) in magnetic fields up to 14 T.

2. Experiment

Si whiskers were grown by chemical transport reactions in a closed bromide system using impurities of boron and gold. Detailed description of obtaining such crystals is described in the [13]. The temperature of the source zone was 1370 K, the temperature of the crystallization zone was 1070–1150 K. The diameter of the microcrystals was 30–40 μm. The whiskers of such diameters are considered as bulk materials. We have investigated crystals with a concentration of acceptor impurity corresponding to the metal–insulator transition (MIT) in silicon ($5 \cdot 10^{18}$ cm^{-3}) on the dielectric and metal sides, respectively. Nickel impurity was introduced by low-temperature diffusion (up to 800 °C), pre-deposited a film of Ni on the grown crystal by electrochemical deposition. Studies on the introduction of magnetic impurities into the crystal were carried out in the work [11]. The main idea of the method is to immerse the ends of the crystal in an aqueous solution of electrolyte, the main component of which are nickel salts. It should be noted, that Ni is not electrically active impurity in silicon [14, 15], but magnetic impurity. Thus, Ni presence in the whiskers should lead to a substantial change in their magnetotransport properties. This method makes it easy to control the total amount of metal, i.e., the doping dose for subsequent low-temperature diffusion processes, taking into account that the entire amount of Ni dissolved in the electrolyte is deposited on the surface of the crystal. Ohmic contacts to the crystals were created by the method of the molten zone with the formation of eutectic Au–Si, which was described in the paper for Pt [16]. A four-probe technique was used for further magneto-transport studies. A study was conducted *I-V* characteristics to assess of the ohmic contacts [13]. In the temperature range 4.2–77 K linear characteristics are obtained, and the resistance of the contacts provided the requirement $R_{\text{contacts}} << R_{\text{volume}}$. The study of electrophysical characteristics was carried out according to [13] in the temperature range 4.2–77 K in strong magnetic fields up to 14 T Bitter magnet. Stabilized electric current in the circuit of the sample was provided by a current source type Keithley 224 in the range 1–100 μA. Scanning in the field took place in two modes. The induction of the magnet was 14 T, sweep speeds were 1.75 and 3.5 T/min at 4.2 K and higher temperature range, respectively. Automatic registration of measurement data via the parallel port of PC with their subsequent visualization and storage into file were performed. Experimental research results were obtained in the framework of the international agreement on scientific cooperation for 2019–2022 between the Lviv Polytechnic National University and the Institute of Low Temperature and Structure Research, Wroclaw, Poland.

3. Experimental results

Investigations of silicon whiskers doped with boron and nickel impurities to concentrations corresponding to the dielectric side of the metal–insulator transition were carried out in the temperature range 4.2–77 K in strong magnetic fields up to 14 T. Thus, Fig. 1 shows the temperature dependence of the magnetoresistance of Si <B, Ni> crystals.

The peculiarities of the behavior of the magnetoresistance of silicon crystals at low temperatures caused by spin-polarization processes are described in the work [11] (Fig. 1, part A). Thus, in the framework of hopping conductivity [11], the transport of charge carriers along localized impurity levels occurs in the presence of twice localized carriers with oppositely or parallel-directed spins. Studies of magnetoresistance [12] showed that if the ratio $H_0^1 / H_0^2 = (T_1 / T_2)^{1/2}$ holds, where H_0 is the magnetic field, at which magnetoresistance is equal to zero, negative magnetoresistance of crystals is caused by localization of charge carriers with parallel directed spins (in contradistinction to anomalous positive magnetoresistance). In addition, studies of magnetization and magnetoresistance [14] showed that the low-temperature transport of charge carriers of Si < B, Ni > whiskers. That is due to the ferromagnetic regime, where with increasing magnetic field induction there is a decrease in the magnitude of calculated values of phase coherence length l_φ and spin-orbit coherence length l_{so} that comprise approximately 45 and 750 nm,

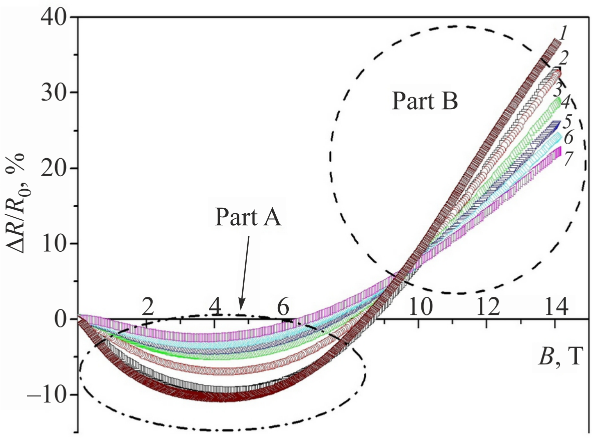

Fig. 1. (Color online) Temperature dependence of the magnetoresistance of Si <B, Ni> whiskers with charge carriers concentration corresponding to the metal–insulator transition on the dielectric side for fixed temperature, K: 2.4 (*1*), 4.2 (*2*), 15 (*3*), 30 (*4*), 40 (*5*), 50 (*6*), 60 (*7*).

respectively, at 4.2 K and exponentially decrease with temperature growth. The important in terms of the numerical experimental works, in which originally grown silicon contains the boron impurity, while the resulting material is introduced the Ni atom impurity using a diffusion method.

Special attention was drawn to the study of Si whiskers doped with impurities of B and Ni, to the concentrations of boron, which corresponds directly to the metal–insulator transition. The research results are shown in Fig. 2.

A significant growth in positive magnetoresistance (without a transition to saturation) is observed with an increase in the magnetic field, both in silicon crystals with a boron impurity concentration corresponding to the dielectric side of the transition (Fig. 1, part B) and in crystals with a boron impurity concentration corresponding to MIT at low temperatures (Fig. 2). In the first case, the magnetoresistance increases to values $\Delta R/R \approx 35$ % (curve *1*), in the second case — $\Delta R/R \approx 280$ % (curve *2*). Nature of the magnetoresistance of silicon alloys doped with boron and nickel with a semiconductor conductivity is well explained within the model according to which the positive magnetoresistance is explained by the deformation of the wave functions of localized holes at low temperatures due to the influence of the magnetic field [17]. This model can be used in our case as well. Taking into account the results of work on "core-shell" structures in the samples [13] a more detailed estimation of the giant magnetoresistance at low temperatures in such crystals can be given in the model of quantum magnetoresistance (QMR), which is the subject of a separate discussion.

Discussion

Today, numerous theoretical and experimental works are devoted to the creation of new 1D and 2D functional materials based on semiconductors [18, 19]. In this case, due to the naturally formed heterostructure, the Si <B, Ni> whiskers is a good model object for studying kinetic properties, in particular magnetoresistance. It is known [13] that at cryogenic temperatures, when there is a significant freezing of charge carriers, their number is too small, and the main mechanism of their transfer must take into account the quantum transfer mechanism within the hopping conductivity of twice-localized states. There are several theoretical models for interpreting the magnetic and transport properties of such systems [20–22]. Under the influence of the magnetic field, the tails of density states of localized carriers can polarize the surrounding magnetic impurities. The transfer of holes (in the ferromagnetic mode) occurs with the help of the nearest neighbor by the hopping mechanism of the charge carrier. This model assumes that the average distance between localized holes should be greater than the radius of localization of holes, namely $\alpha_0^3 p << 1$, where α_0^3 represents the characteristic of the exponential decrease of the wave function of the hole in localized states, p is the effective concentration of holes [20].

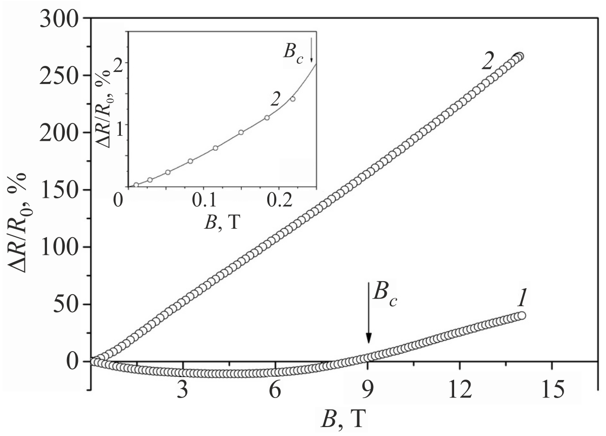

Fig. 2. Magnetoresistance of silicon whiskers with boron impurity concentration corresponding to MIT $N_B \sim 5\cdot10^{18}$ cm^{-3} at temperature $T = 4.2$ K. Inset: Magnetoresistance of silicon whiskers with boron impurity concentration corresponding to MIT in fields up to 0.25 T, where the classical parabolic change of magnetoresistance is performed.

During experimental studies of the magnetization of Si microcrystals deviations from the Curie–Weiss law were observed. The negative Curie temperature is determined, indicating the presence of antiferromagnetic interaction in the crystals [14]. The results of experimental studies do not exclude the fact (Fig. 1, part B), that with increasing magnitude of the magnetic field (in the presence of a local magnetic moment caused by a magnetic impurity) the spins of twice localized carriers are reoriented in opposite directions. This is manifested in the transition from negative to positive magnetoresistance, which increases linearly with the subsequent increase of magnetic field induction. These theoretical assumptions can obviously be interpreted and quantified using quantum magnetoresistance theory.

It is known that [13] silicon filamentous crystals are considered as a naturally formed heterostructure, in which the transfer processes are interpreted as kinetic effects, which, due to the "core-shell" structure, occur in the near-surface layers. On the other hand, in the works [23–25] the behavior of magnetoresistance for different types of materials, such as narrow-band semiconductors, semimetals, metals, heterostructures or chalcogenides is considered in a sufficiently wide temperature range. According to the classical Parish–Littlewood model (PL model) for disordered systems [23] a significant increase in longitudinal magnetoresistance and its linear temperature dependence was noticed. As can be seen from the experimental results of the magnetoresistance study at low temperatures (Figs. 1, 2) there is a sharp linear increase in magnetoresistance beginning from certain critical magnetic field B_{cr} that are different depending on the concentration of charge carriers in the Si<B, Ni>samples corresponding to MIT. As a rule, the classical behavior of the magnetoresistance is its quadratic field dependence (Fig. 1, part A and the inset, Fig. 2). In addition, the next surprising feature of the magnetoresistance is its rather high value (of about 280 %) (Fig. 2). The observed peculiarities of the magnetoresistance could not be explained in the framework of classic theory. Several prevailing models have been discussed in this work to explain the observed GMR in silicon whiskers, such as: the QMR model and the two-band models that were applied in layered systems [26]. This model indicates that if we have more than two conducting channels with different carrier mobility then the current flows through the channel with high mobility in low magnetic field. After that, as the magnetic field is increasing, the current will go into the low mobility channel as the longitudinal conductivity of both channels drops as $1/[1 + (\mu B)^2]$.

Such "core-shell" structure was created due to the two-stage filamentary crystal growth of leader (core) according to vapor-liquid-solid (VLS) mechanism and shell creation according to vapor-crystal (VC) mechanism. To observe the impact of the interfaces of silicon whisker channel contrast experiments were performed [13]. For example, the transport properties of an identical whisker were studied before and after the surface layer was removed due to etching. The results of the whisker resistance investigation showed that the resistance value grows after the surface layer etching as compared with non-etched one.

Similar assumptions have been discussed for deformed InSb microcrystals. In particular, in [27] it was determined, that the InSb whisker magnetoresistance changes non-monotonically when the temperature drops and reaches 700 % at 70 K and magnetic field induction 10 T. GMR seen in InSb whiskers could be also explained in the framework of the quantum magnetoresistance model. However, the main origin of the effect is deformed by the whisker "core-shell" structure. Due to strong transverse deformation (of about $3\cdot10^{-3}$ relat. units) electron transport change occurs in the whisker shell. The same behavior was found in our previous works [27], where the application of strong compressive strain leads to arising Berry phase in the whiskers, which evidences in the transition to topological insulator of the material at low temperatures. The Landau fan diagrams confirmed Berry phase presence at low temperatures in the InSb whiskers with doping concentration in the vicinity to the MIT [27]. In contrast to the creation of a stress-strain state in InSb crystals, the presence of a magnetic impurity in Si <B, Ni> crystals affects the electronic transport of charge carriers, which greatly simplifies the technology of obtaining silicon crystals. Due to low-temperature diffusion, the concentration of the magnetic impurity of Ni is concentrated in the near-surface region of the crystal [13], where there is mainly low-temperature transport of charge carriers. Given the spin-polarization transport within the hopping conductivity at low temperatures at twice occupied states, the orientation of the spins of localized charge carriers in the field leads to a significant increase in the magnetoresistance of crystals in an antiferromagnetic regime with increasing induction of the magnetic field. The increase in magnetoresistance is characteristic of heterostructures in which there is a, so-called, "barrier layer", charge carriers in antiferromagnetic when a magnetic field is turned on [28].

GMR is a result of material defects or inhomogeneity, which means that the origin of observed GMR in our silicon whiskers is intrinsic. The QMR theory proposed by Abrikosov explains the linear magnetoresistance observed in nonmagnetic semiconductor silver chalcogenides [24–26]. Such GMR could be seen in semimetals and narrow-gap semiconductors that have small electron effectiveness and a low concentration of carriers. Silicon whiskers perfectly fit this characteristic.

The quantum magnetoresistance theory shows that the linear field-dependent magnetoresistance will appear under the "extreme quantum limit" (EQL) $h\omega_c > E_F$, at the time when all carriers stay at the lowest Landau level. Recently, some work released that the quantum GMR could still emerge with a few occupied Landau levels [29]. The results can be expressed the next way:

$$\rho_{xx} = \rho_{yy} = \frac{N_i B}{\pi n_0^2 ec}, \tag{1}$$

where ρ_{xx} and ρ_{yy} are the transverse components of the resistivity tensor, n_0 is the carrier concentration, and N_i is the scattering center concentration. The carrier concentration and the requirements for temperature are:

$$n_0 \le \left(\frac{eB}{hc}\right)^{3/2},$$

$$T \le \frac{eBh}{m^* c}, \tag{2}$$

where m^* is the effective mass.

Considering that N_i is the scattering center concentration under the conditions of hopping conductivity there are localized carriers on which the charge carriers jump is realized, then for the two presented cases we have the following. For samples with a directly corresponding charge carrier concentration MIT ($n_0 = 5\cdot10^{18}$ cm^{-3}) on the right Eq. (1) ratio $N_i / n_0 \to 1$. We can assume that all charge carriers are ionized and participate in the current transfer. In this case taking as the effective mass in silicon whiskers [30, 31] is small, $m^* \sim 0.57m_0$ (m_0 is the free electron mass), the right-hand side of the inequality in Eq. (2) equals 4.2 K ($B_c \approx 0.2$ T) (see Fig. 2, inset). Then the quantum mechanism of magnetoresistance in the near-surface region of the crystal becomes dominant.

For samples with a concentration of charge carriers corresponding to the dielectric side of the transition in the right part Eq. (1) ratio $N_i / n_0 < 1$. The conditions that are described in Eq. (2) are reached at liquid nitrogen temperature, when $B_c > 6$ T (Fig. 1, Part B). The model indicated that the quantum magnetoresistance is insensitive to the temperature, that is confirmed by our results.

One of the areas of application of the detected QMR effect in crystals with a concentration of charge carriers corresponding to MIT are magnetic field sensors with magnetoresistive principle of operation (Fig. 2). So for a magnetic field sensor based on silicon microcrystals with high sensitivity to a magnetic field of 20 % / T for 4.2 K is proposed.

Conclusions

Experimental investigation have been conducted to study the magnetic resistance of silicon whiskers doped with a complex of impurities (B, Ni), which corresponds to the metal–insulator transition at low temperatures (down to 4.2 K) in magnetic fields up to 14 T. The giant magnetoresistance was detected in such crystals. A significant increase in positive magnetoresistance with increasing magnetic field induction was observed in silicon crystals of both boron concentrations: (i) corresponding to the dielectric side of the transition ($p_{300K} = 2\cdot10^{18}$ cm^{-3}); (ii) corresponding directly to MIT ($p_{300K} = 5\cdot10^{18}$ cm^{-3}). In the first case, the magnetoresistance increases to values $\Delta R/R \approx 35$ %, in the second one to $\Delta R/R \approx 280$ %. Further analysis showed us that the GMR seen in Si whiskers could be explained by the quantum magnetoresistance model. Features of magnetoresistance are realized in the near-surface region of the crystal, thanks to the “core-shell” structure. This is evident in the transition from the classical parabolic magnetoresistance Further analysis showed us a significant linear increase of magnetoresistance with increasing magnetic field corresponding to the transition from the ferromagnetic localization regime of carriers with spins directed parallel to antiferromagnetic with oppositely directed spins. The critical fields in which such a transition occurs are equal 0.25 and 6 T, respectively. Crystals with a concentration of charge carriers corresponding to MIT can be used for design of magnetic field sensors with magnetoresistive principle of operation. Sensitivity of such sensors to a magnetic field induction is of 20 %/ T for 4.2 K, which allows us its measurement with high precision.

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Квантовий магнітоопір у ниткоподібних кристалах Si <B, Ni>

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Досліджено електричний магнітоопір легованих нікелем та бором ниткоподібних кристалів кремнію, в яких спостерігається перехід метал-ізолятор. Гігантський магнітоопір досягає 280 % у ниткоподібних кристалах Si з концентрацією легуючої домішки бору $p_{300\text{K}} = 5 \cdot 10^{18}$ см$^{-3}$ у магнітних полях з індукцією до 14 Тл при температурі 4,2 К. Показано, що особливості магнітоопору при низьких температурах обумовлено структурою кристалів «серцевина–оболонка». Природу гігантського магнітоопору розглянуто в рамках квантової моделі магнітоопору. Аналіз проведено для визначення критичного поля переходу від класичного параболічного магнітоопору до квантового магнітоопору, який реалізований у приповерхневій області кристала. Ниткоподібні кристали кремнію використовували для проєктування датчиків магнітного поля.

Ключові слова: кремній, ниткоподібні кристали, стрибкова провідність, слабка локалізація, спін-орбітальна взаємодія.