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Gigantic magnetoresistive effect in n-Si/SiO_2/Ni nanostructures fabricated by the template-assisted electrochemical deposition

Abstract. *The study of the carrier transport and magnetotransport in n-Si/SiO_2/Ni nanostructures with granular Ni nanorods embedded into the pores in SiO_2 was performed over the temperature range 2 – 300 K and at the magnetic field induction up to 8 T. In n-Si/SiO_2/Ni nanostructures at temperatures of about 25 K a huge positive MR effect is observed. Possible mechanisms of the effect is discussed.*

Streszczenie. *Przeprowadzono badania mechanizmu przenoszenia ładunków i magnetoprzewodzenia w nanostrukturach n-Si/SiO_2/Ni z ziarnistymi nanocząstkami Ni rozmieszczonymi w porach SiO_2 w zakresie temperatur 2 - 300 K oraz przy indukcji magnetycznej do 8 T. W nanostrukturach n-Si/SiO_2/Ni w temperaturze około 25 K zaobserwowano bardzo wyraźny dodatni efekt MR. Omówiono prawdopodobne mechanizmy pojawienia się opisanego efektu. (**Gigantyczny efekt magnetorezystywności w nanostrukturach n-Si/SiO_2/Ni wytworzonych przy użyciu metody nanoszenia elektrochemicznego**).*

Keywords: Ion implantation, granular materials, nanostructures, magnetoresistance.
Słowa kluczowe: implantacja jonowa, ziarniste materiały, nanostruktury, magnetorezystancja.

Introduction

At the present time commercially produced magnetic nanosensors, memory nanocells, etc. are not free from such limitations as too high sensitivity to temperature, fairly high costs, poor properties at high frequencies and relatively large sizes. Therefore, of special interest is the development of the methods to create nanostructures and/or their arrays exploiting the giant (GMR) or tunneling (TMR) magnetoresistive effects [1]. The GMR/TMR effects are created in the artificially layered systems composed of a large number of magnetic/nonmagnetic layers, forming some kind of a superlattice [2]; multilayer structures in spin valve configurations [3]; point contacts between two ferromagnetic nanowires [4]; nanobridges fabricated from ferromagnetic semiconductors [5]; etc.

Another approach to the fabrication of the arrays of magnetic sensors and related nanodevices is based on usage of porous templates (SiO_2, Al_2O_3, etc.), where the ordered or randomly distributed mesa- or nanopores are filled with different nanostructured substances and/or their compositions [6]. As regards the preparation of nanoarrays, it should be noted that currently the template-assisted synthesis is a well-established and reliable technique for the production of numerous metallic and semiconductor nanoarrays (for example, see [1, 3, 7]). One of the template-assisted fabrication techniques is based on the Nuclear Ion Track Etch Method (NITEM). Swift heavy ions irradiating any substrate can significantly transform the material within a narrow (cylindric) regions with a diameter of the order of 10 nm. Selective etching of these latent ion tracks leading to the creation of channels (pores) with a large aspect (length-to-diameter) ratio enables one to form porous templates for further nanorods array growth.

Of particular interest is the use of the NITEM technology for the fabrication of micro- or nanoelectronic devices by the electrodeposition procedure. The advantages of the electrodeposition technique are its effective control of thickness, stoichiometric composition, and grain dimensions of deposited substances. By the adequate selection of the potential for deposition one can realize the low-rate deposition regime, in the process of which subsequent atomic layers are deposited step by step. A growth mechanism during electrodeposition enhances the uniformity of deposited elements, allows for the creation of compounds, solid solutions, and also heterogeneous systems including the nanogranules (clusters) of different composition or their interchanging layers. Electrodeposition allows for achieving the atomic-level control of the growth process, leading to the formation of well-quality deposits of the desired material [8].

Note that such synthesis of the nanoarrays containing nanogranular materials on Si substrate, utilizing the enhanced TMR/GMR effects, shows much promise for the design of Si-compatible electronic devices. Moreover, deposition of nanoarrays set onto semiconductor substrates enables its application with the additional electrode that joins nanoarrays to a system of MOSFET-like electronic elements called the TEMPOS structures [9, 10].

In this work we study the carrier transport and magnetotransport properties in the bundles of Ni nanorods embedded into the n-Si/SiO_2 porous template created by selective etching of swift heavy ion tracks in a SiO_2 layer when the pores are filled with nickel nanoparticles.

Experimental

In the present work we used underpotential electrodeposition of Ni nanoparticles into the nanopores in a SiO_2 layer as a template created on the n-Si(100) substrate with 4.5 $\Omega\cdot$cm resistivity to produce magnetosensitive nanostructures n-Si/SiO_2/Ni. The regimes and electrochemical properties of the samples studied were described in [11-13]. A SiO_2 layer with the thickness 700 nm was thermally grown on the Si(100) substrate by the standard procedure (1100°C, 10 hours, pure oxygen). To produce a mesaporous SiO_2 layer, scanned beams of 350 MeV $^{197}Au^{26+}$ ions were employed for the bombardment of the oxidized Si wafers with fluences of about $5\cdot10^8$ cm^{-2}. The pores were formed by chemical etching of latent tracks with in a dilute HF acid for a whole depth of the SiO_2 layer down to the Si substrate. Etching resulted in the formation of the nanopores randomly distributed over the sample surface and shaped like truncated cones with heights about 400 - 500 nm and down/upper diameters of 100/250 nm. The nanopores filled with Ni nanoparticles enable one to form a system of nanorods (or NanoRods-In-Pores – NRIPs) randomly distributed within the SiO_2 layer.

The structure of nanoporous templates and Ni-NRIPs was studied using SEM LEO-1455VP and X-Ray diffraction analysis. As follows from [11-14], electrodeposited Ni-NRIPs had granular structure with the Ni particles having the face-centered cubic structure and dimensions of about 30 – 70 nm.

The procedure of electrical measurements carried out at temperatures ranging 2 - 310 K and at magnetic fields with the induction B up to 8 T is described in [12, 14]. The I-V characteristics and DC resistance of n-Si/SiO_2/Ni nanostructures were measured between two electric probes shown in the sketch of the sample shown in insert (a) in Fig. 1. Two electric probes were prepared by ultrasonic soldering of In on the surface of the studied samples. The DC resistance was measured in two regimes **1r** and **2r**. In regime **1r** we estimated equilibrium resistance $R_o = dV/dI$ at $V \to 0$ from I-V characteristics measured at a number of constant temperature values. In regime **2r** we measured temperature dependences of $R = V/I$ at current I = const when cooling from room temperature to 2 K. As is seen in Fig. 1, at such a scheme of electric measurements for the studied n-Si/SiO_2/Ni nanostructures the electric current vector I was always directed along NRIPs bundles (which were embedded into the SiO_2 layer, lied under two In probes and electrically joined through Si substrate) and along Si substrate plane. Magnetoresistance MR(B) = $\Delta R(B)/R(0) = [R(B) - R(0)]/R(0)$ (here $R(B)$ and $R(0)$ are structure resistances with and without a magnetic field, respectively) was measured in three different regimes. In regime **1mr** we estimated MR from I-V characteristics measured at a number of constant temperatures at B = 0 and B = 8 T (see, [12, 13]). In regime **2mr** we measured MR sweeping out of B between 0 and 8 T at some constant temperatures (see, [13]). In regime **3mr** we estimated MR by point-to-point subtracting of the temperature dependencies $R(T)$ measured in regime **2** (at I = const) at B = 8 T from $R(T)$ at B = 0.

Moreover, as is seen from Fig. 1b, for regimes **2a** and **3a** MR was measured in 3 different mutual alignments i of vectors B and I and the substrate plane in n-Si/SiO_2/Ni nanostructures. For i = 1 vector B is parallel to NRIPs axis and normal to vector in them and to Si substrate plane. For i = 2 vector B is parallel to NRIPs axis and vector I in them but parallel to vector I in Si substrate plane.

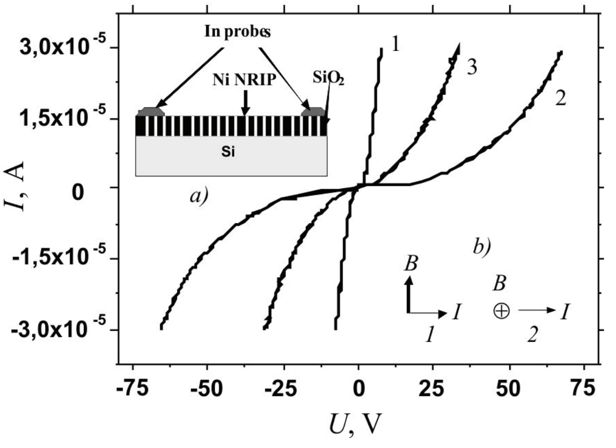

Fig.1. I-V characteristics at 25 K for the magnetic field inductions B = 0 (*1*) and B = 8 T with B-I configurations i = 1 (*2*) or i = 2 (*3*) shown in insert (*b*). Inserts show a schematic view Ni-NRIPs and In probes in n-Si/SiO_2/Ni nanostructures (*a*) and a scheme of mutual alignements of magnetic induction vector B, current vector I and Si substrate plane at MR measurements (configurations i = 1, 2)

Results and Discussion

Here we present in more details the temperature dependence of resistance in B = 0 and MR in n-Si/SiO_2/Ni nanostructures than was reported previously in [12, 13]. Fig. 1 shows that the I-V characteristics of n-Si/SiO_2/Ni nanostructures exhibit non-linear behavior and strong dependence on mutual alignements of magnetic induction vector B, current vector I and Si substrate plane (NRIPs axis). The symmetrisity of I-V branches and the behavior of I-Vs at different temperatures described in oue previous paper [12] shows that two nanorod bundles under the electric probes contacting the Si substrate electrically resemble two Si/Ni Schottky diodes serially joined and switched-on opposite to each other.

The typical temperature dependences of the resistance $R = V/I$ for n-Si/SiO_2/Ni nanostructures measured in regime **2r** at B = 0 and plotted in logarithmic scale are shown in Fig. 2. We would note three main features of these curves.

Firstly, position of $R(T)$ curves on ordinate scale of Fig. 2 is strongly dependent on the measuring current I although their shape is similar for different I values in the region 10 - 1000 nA. Moreover, experimental points obtained from I-V characteristics at measuring in regime 1 hit directly on curve 3, i.e. curve 3 can be considered as equilibrium.

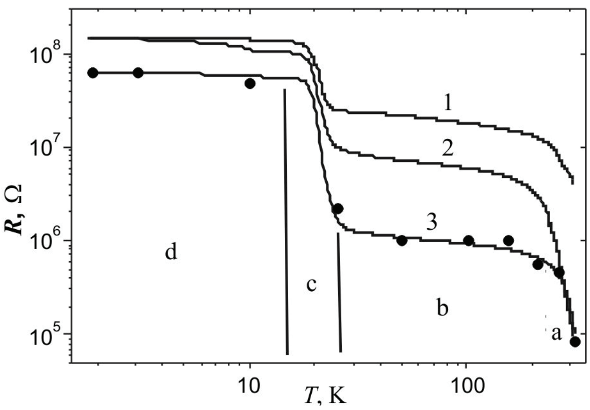

Fig. 2. Temperature dependencies of resistance measured in regime **2** at B = 0 for different currents: 1 – 1000 nA, 2 – 100 nA and 3 – 10 nA. Points in curve 3 present equilibrium values of resistance R_o measured in regime **1r**

The second feature is that the shape of $R(T)$ curves for n-Si/SiO_2/Ni nanostructures are principally different from that for granular Ni films (see, [12, 14]): Ni films observed metallic progress of $R(T)$ with positive sign of dR/dT whereas for n-Si/SiO_2/Ni nanostructures $dR/dT < 0$ that is characteristic for semiconductors or some of granular systems. As was mentioned in [13], the observed behavior of $R_o(T)$ (curve 3) suggests that in the studied nanostructures regions (a) - (c) in Fig. 2 at temperatures ranging 15 – 300 K the activation (exponential) carrier transport by the Si substrate dominates. The ln$Ro(1/T)$ curve 3 gives the activation energy close to a half of the gap width for Si single crystals in region (a) and close to the ionization energy of phosphorus in Si in region (c). In this context the region (b) can be considered as transition one between zone and impurity contribution to conductance.

The third feature is that at the lowest temperatures (less than 15 K), when the impurity carrier transport by Si substrate is freezed-out, the curves ln(R_o) *vs* ($1/T$) are characterized by variable activation energies (region (d) on curve 3 in Fig. 2). Taking into account a metallic behavior of $R_o(T)$ curves for the granular Ni films and large resistance of SiO_2 strata between Ni nanorods, this contribution to the $R_o(T)$ in region (d) seems to be explained by the formation of the electrons-enriched n-Si/SiO_2 interfacial layer due to the band bending where electrons are moving along the interface by hopping mechanism [15].

As is seen from Fig. 1 and 3a, the application of a magnetic field to the n-Si/SiO_2/Ni nanostructure for both configurations i = 1,2 caused strong increase of resistance (MR > 0) approaching a huge values at around 25 K. Note also, that at temperatures higher than 180 K, when measuring currents I decrease, magnetoresistance becames negative. This effect is more pronounced on MR(*B*) curves shown in Fig. 3b which was calculated by point-to-point subtracting of the curves measured in regime **2r** (at different currents) in B = 8 T and B = 0 correspondingly. The MR(B) curves behavior shown in Fig. 3b confirms the results carried out in regimes **1r** and **2r** and given in Fig. 3b in our previous work [12]. We can see that at low temperatures MR strongly increases with measuring current decrease approaching 200 – 600 % at temperatures of about 20 - 27 K for 10 < I < 1000 nA.

a)

b)

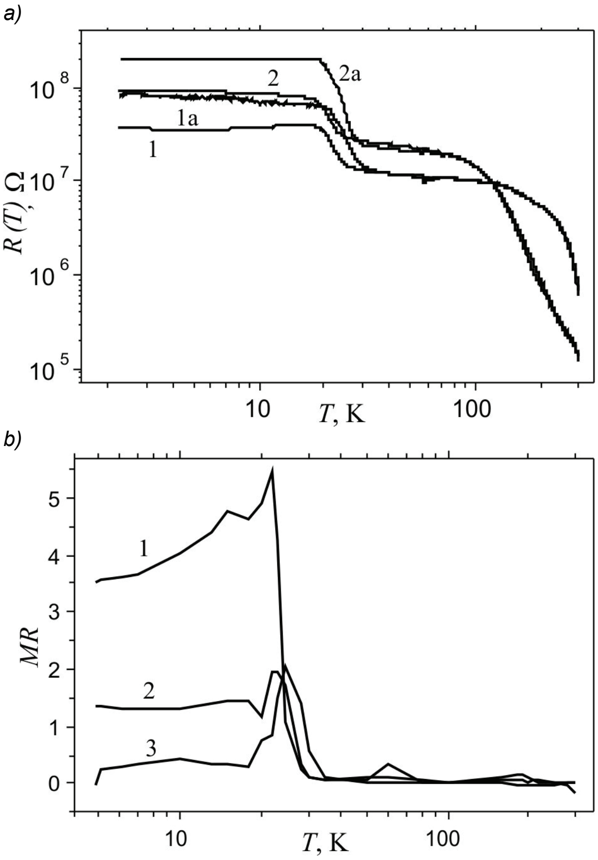

Fig. 3. *a* – Temperature dependencies of resistance measured in regime **2r** at B = 0 (curves 1 and 2) and at B = 8 T (curves 1a and 2a) for different measuring currents: 1, 1a – 1000 nA; 2, 2a – 100 nA. Mutual alignements of magnetic induction vector B, current vector I and Si substrate plane corresponded to configuration i = 1; *b* – Temperature dependence of MR(B = 8 T) measured in regime **3mr** for I = 10 nA (1), 100 nA (2) and 10 nA (3)

As we observed, at T > 180 K, where activational conductance by Si substrate is predominant (region (a) in Fig. 2), the values of MR effects in n-Si/SiO_2/Ni nanostructures for configurations i = 1,2 were negative as for the samples of Ni films. This probably means that magnetoresistance in this temperature region is similar to the anisotropic magnetoresistive effect observed in Ni nanorods [16]. At temperatures lower than 100 K, where impurity and hopping conductance are realized, MR values for n-Si/SiO_2/Ni nanostructures were always positive. Moreover, we observed that enhancement of MR just falls within temperature region (c) in Fig. 2, where impurity conductance is predominant and where a value of the avalanche threshold voltage $V_{av}(T)$ in I-V characteristics begins to grow considerably (see, inset in Fig. 4b in [12]). These results enable one to assume that, in contrast to granular Ni films on the Si substrate, a huge positive magnetoresistive effect at low temperatures in n-Si/SiO_2/Ni nanostructures can be associated with two possible contributions – the influence of Si/Ni Schottky barriers and/or movement of electrons along the electrons-enriched Si/SiO_2 interfacial channel. To separate these two mechanisms we need to make additional experiments.

Resume

The conducted study of the carrier transport in magnetic field in n-Si/SiO_2/Ni nanostructures with granular Ni NRIPs embedded into SiO_2 layer and contacting Si substrate revealed behavior which was considerably differentiated from granular Ni films electrodeposited on n-Si wafers.

As our study have shown, the n-Si/SiO_2/Ni nanostructures, being electrically similar to two Si/Ni Schottky diodes switched-on opposite to each other, display 3 contributions to the temperature dependences of equilibrium DC resistance: the zone-like carrier transport by Si substrate (at T > 250 K); impurity conductance by the phosphorus-doped Si substrate (at 15 K < T < 180 K) when the zone-zone carrier transport by Si wafer is freezed-out; and hopping conductance by the localized states at T < 15 K when electrons become to move along the n-Si/SiO_2 interface over the electrons-enriched layer due to the band bending.

In n-Si/SiO_2/Ni nanostructures at the temperatures ranging 17 - 27 K, where impurity conductance by the phosphorus-doped Si substrate is predominant, a considerable positive contribution to the MR effect is observed, that may be attributed to two possible reasons – the influence of Si/Ni Schottky barriers and/or movement of electrons along the electrons-enriched Si/SiO_2 interfacial channel. To separate these two mechanisms we need to make additional experiments.

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