Negative magnetoresistance in indium antimonide whiskers doped with tin

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Negative magnetoresistance of InSb whiskers with different impurity concentrations $4.4 \cdot 10^{16} - 7.16 \cdot 10^{17}$ cm⁻³ was studied in longitudinal magnetic field 0–14 T in the temperature range 4.2–77 K. The negative magnetoresistance reaches about 50% for InSb whiskers with impurity concentration in the vicinity to the metal–insulator transition. The negative magnetoresistance decreases to 35 and 25% for crystals with Sn concentration from the metal and dielectric side of the transition. The longitudinal magnetoresistance twice crosses the axis of the magnetic field induction for the lightly doped crystals. The behavior of the negative magnetoresistance could be explained by the existence of classical size effect, in particular boundary scattering in the subsurface whisker layer.

PACS: **76.60.-k** Nuclear magnetic resonance and relaxation; 72.15.Rn Localization effects (Anderson or weak localization); 73.43.Ot Magnetoresistance.

Keywords: negative magnetoresistance, InSb whiskers, transverse and longitudinal magnetoresistance, metal-insulator transition.

Introduction

Negative magnetoresistance (NMR) was observed in InSb crystals at low temperatures and weak magnetic fields [1-9]. There are various interpretations of this effect due to different mechanisms of scattering of charge carriers. In particular: 1) surface boundary scattering [1,2]; 2) scattering on magnetic impurities [3-5]; 3) scattering on non-magnetic impurities with concentration in the vicinity to metal–insulator transition (MIT) [5-9].

Negative magnetoresistance was found in InSb films [1,2]. Transverse and longitudinal magnetoresistance in InSb thin films grown on GaAs substrates were studied at temperatures 4.2, 80 and 300 K [1]. NMR of undoped InSb film was observed only in magnetic fields parallel to the film at high temperatures. The negative magnetoresistance effect is connected with surface boundary scattering in the plane normal to InSb film. Positive magnetoresistance shows the logarithmic increase with anisotropy between parallel and perpendicular orientation of magnetic field. It's arising from the two-dimensional weak anti-localization that reflects strong spin–orbit interaction caused by the asymmetric potential at the interface. At low tempera-

tures (about 80 K), the transport is dominated by the twodimensional electrons in the accumulation layers at the InSb/GaAs heterointerface [1].

In Sn-doped InSb films, the negative magnetoresistance was found in extremely weak magnetic fields [2]. Its appearance was observed before the Shubnikov–de Haas oscillations. The negative magnetoresistance crossovers to the positive magnetoresistance occurs with the decrease of the film thickness to 0.1 mm. These effects were analyzed and the spin–orbit scattering rate in the intrinsic InSb film due to the bulk inversion asymmetry has been found. The crossover from weak localization to weak anti-localization with decreasing InSb film thickness from 1 to 0.1 mm was found for Sn-doped films in weak magnetic fields before the appearance of the Shubnikov–de Haas oscillations [2].

Magnetic and transport properties of indium antimonide doped with manganese were studied in the temperature range 1.6–300 K and magnetic fields up to 15 T [3,4]. Negative magnetoresistance was revealed in diluted magnetic semiconductor InSb:Mn with nanosize MnSb precipitates [4]. The positive magnetoresistance was observed at temperatures above 10 K [4]. It transforms into negative magnetoresistance with the decrease of the temperature. The temperature dependence of negative magnetoresistance was explained by damping of the spin-dependent scattering of charge carriers in magnetic field.

Magnetoresistance of nonmagnetic InSb single crystal doped with manganese with impurity concentration $1.5 \cdot 10^{17}$ cm⁻³ was investigated in the temperature range 40 mK-300 K and magnetic fields 0–25 T [5–8]. Colossal decrease of resistivity in *p*-type InSb(Mn) crystals was revealed in magnetic fields 0–4 T at superlow temperatures [5]. The Hall constant changes its sign under variable temperature and magnetic field.

The behavior of magnetic Mn- and nonmagnetic Geimpurities was compared in InSb. Ge like Mn forms shallow acceptor level and demonstrates exactly the same critical concentration of metal–insulator transition at $N_{\rm cr} = 2 \cdot 10^{17}$ cm⁻³. The magnetotransport characteristics of *p*-InSb(Ge) crystals at low temperatures differ from *p*-InSb(Mn) characteristics [6]. InSb crystals doped by Ge do not demonstrate resistivity dependense on impurity concentration, temperature and magnetic field, but it demonstrates the variable range hopping conductivity, and positive magneotresistance typical for disordered localized nonmagnetic impurities.

The resistivity near the metal–insulator transition in InSb:Mn and InSb:Ge were studied and negative magnetoresistance was observed at temperature 1.6 K [9]. InSb:Mn exhibits a strong enhancement of the resistivity below 10 K. Negative magnetoresistance effect increases by applying hydrostatic pressure. The exchange interaction between the hole spin of the Mn acceptor is the dominant correlation effect leading to the formation of an antiferromagnetic alignment of the Mn spins along the percolation path which inhibits hopping of holes between neighboring Mn sites.

Our previous magnetoresistance studies of InSb whiskers doped with Sn [10,11] revealed negative magnetoresistance at low temperatures in weak magnetic fields, but its behavior wasn't analyzed at different temperatures and doping levels. It is interesting to consider transport mechanisms in InSb whiskers to explain the magnetoresistance behavior.

The aim of this paper is to study conditions of the negative magnetoresistance existence in InSb whiskers with Sn concentration in the vicinity to MIT in the temperature range 4.2–77 K at magnetic fields 0–14 T.

Experimental procedure

The objects of studies were to observe the behavior of InSb whiskers with *n*-type conductivity obtained by the chemical transport reactions method [11]. Investigated whiskers were doped by tin during microcrystals growth. InSb whiskers were selected with length 2–3 mm and lateral dimensions about 30–40 μ m. Electrical contacts to InSb whiskers were created by using Au wires with diameter 10 μ m that form an eutectic with the microcrystal under pulsed welding. This technique was tested and described in previous works [12] for contact creation to solid solution

SiGe whiskers. It allows measuring whisker resistance using four contacts scheme.

InSb whisker conductivity was studied in the temperature range 4.2–300 K. For these studies crystals were cooled to temperature 4.2 K in the helium cryostat. The temperature was measured by Cu–CuFe thermocouple calibrated with CERNOX sensor.

The magnetic field effects of the whiskers was studied using the Bitter magnet with the induction up to 14 T and the time scanning of field 1.75 T/min in the temperature range 4.2–77 K. Stabilized electric current along the whisker was created by the current source Keithley 224 in the range 1–10 mA depending on the crystal resistance. CERNOX sensor was used for magnetic measurement, in particular for its thermostabilization. It is weakly sensitive to magnetic field induction. The change of its output signal in the field with induction B = 15 T is about 1%.

All characteristic such as: electrical voltage of the whisker contacts, output signals from the thermocouple and the sensor's magnetic field were measured using the digital voltmeters type Keithley 199 and Keithley 2000 with precision up to $1 \cdot 10^{-6}$ V and simultaneous automatic registration.

Four groups of *n*-type InSb whiskers with different doping concentration (Sn) and varying degrees of approximation to the critical concentration $N_{\rm cr} = 3 \cdot 10^{17} \text{ cm}^{-3}$, which corresponds to the phase metal–insulator transition, were selected in order to study their magnetoresistance:

— InSb whiskers with the impurity concentration $3.26 \cdot 10^{17} \text{ cm}^{-3}$ which correspond to MIT;

— InSb whiskers with the impurity concentration $2.3 \cdot 10^{17}$ cm⁻³ in the vicinity of MIT at the dielectric side of the transition;

— Heavily doped microcrystals with the impurity concentration $7.16 \cdot 10^{17}$ cm⁻³ in the vicinity of MIT at metal side of the transition;

— Lightly doped InSb whiskers with the impurity concentration $4.4 \cdot 10^{16}$ cm⁻³ removed from MIT at dielectric side of the transition.

Experimental results and discussion

Studies of InSb whiskers with different impurity concentration (Sn) in the vicinity of MIT in the temperature range 4.2–77 K at magnetic fields 0–14 T show the occurrence of NMR (see Figs. 1–4). A similar behavior of magnetoresistance, including its reduction in the magnetic field, was observed for whiskers on the base of other materials, such as *p*-SiGe solid solution [13] and germanium with *p*- and *n*-type conductivity [14]. The negative magnetoresistance in the InSb whiskers was observed in magnetic field applied parallel to the crystal axis. NMR reaches about 50% at the impurity concentration $3.26 \cdot 10^{17}$ cm⁻³ and is observed at temperatures 4.2-77 K and in the magnetic field 2–14 T (Fig. 1). Value of the NMR in the InSb whiskers with Sn

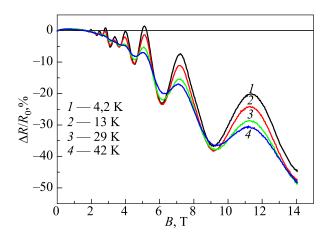


Fig. 1. (Color online) Longitudinal magnetoresistance of InSb whiskers with impurity concentration $3.26 \cdot 10^{17}$ cm⁻³ at different temperatures.

concentration in the vicinity of MIT at the metal and dielectric side of the transition decreases to 35%, 25% for concentrations $2.3 \cdot 10^{17}$ cm⁻³, $7.16 \cdot 10^{17}$ cm⁻³ and begins at magnetic field 3 T, 5 T, accordingly (Figs. 2, 3). The NMR of the lightly doped InSb whiskers with impurity concentration $4.4 \cdot 10^{16}$ cm⁻³ is revealed in all investigated magnetic fields 0–14 T and the longitudinal magnetoresistance twice crosses the field axis (Fig. 4).

Figure 1 shows the longitudinal magnetoresistance (**B** parallel to the wire axis) for InSb whiskers with impurity concentration $3.26 \cdot 10^{17}$ cm⁻³, which corresponds to MIT at temperature 4.2 K. The maximum ratio $\Delta R/R$ increases approaching to MIT and reaches 50% at 4.2 K. As the temperature rise, the maximum value decreases (up to 15% at temperature 42 K).

The observed NMR effect is like that of the reference [15] for NiMn/InSb structure. According to data [15] the NMR cannot be explained by disorder effect [16], and by the scattering of the conduction electrons by localized spins through an s-d exchange interaction [17]. An expla-

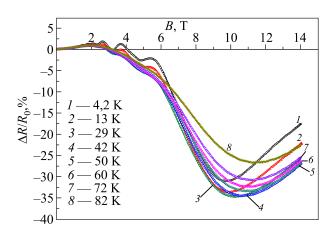


Fig. 2. (Color online) Longitudinal magnetoresistance of InSb whiskers with impurity concentration $2.3 \cdot 10^{17}$ cm⁻³ at different temperatures.

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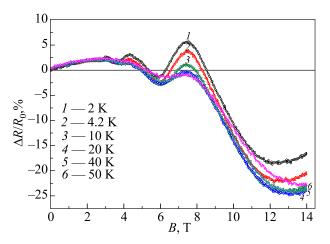


Fig. 3. (Color online) Longitudinal magnetoresistance of InSb whiskers with impurity concentration $7.16 \cdot 10^{17}$ cm⁻³ at different temperatures.

nation of the NMR is the interface containing microscopic magnetic entities (NiMn or Ni precipitates). Upon increasing the magnetic field, these magnetic entities gradually align their magnetic moments with the external magnetic field leading to a decrease in the spin-dependent resistance of the system [15].

Noteworthy explanation revealed negative magnetoresistance in the field dependences of the investigated longitudinal magnetoresistance in the present work. Possible reasons for the demonstrated effects may include: 1) the presence of size quantization in whiskers [18]; 2) the presence of magnetic ordering of electron spins in InSb whiskers with concentration in the vicinity of metal-insulator transition [19]; 3) the presence of magnetic ordering in InSb whiskers by introducing uncontrolled magnetic impurities [20]; 4) quantum interference of electron wave function [21,22]; 5) classical size effect [23].

The presence of size quantization in InSb whiskers is excluded due to large transverse dimensions (20–40 μ m) of whiskers (much larger than de Broglie wavelength).

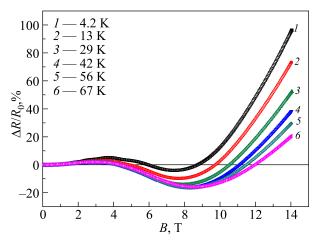


Fig. 4. (Color online) Longitudinal magnetoresistance of InSb whiskers with impurity concentration $4.4 \cdot 10^{16}$ cm⁻³ at different temperatures.

Probable cause of the detected features of InSb whisker magnetoresistance may be the presence of magnetic ordering of electron spins on impurities in heavily doped semiconductors. According to well-known model of Y. Toyozawa an appearance of NMR occurs due to reorientation of spins localized on impurity atoms [19]. As consequence, one of the possible channel of charge carrier scattering is excluded leading to magnetoresistance decrease. For example, NMR effect was found in Ge whiskers [14]. In InSb whiskers strong spin-orbital interaction of charge carriers should occur, which is confirmed by magnetoresistance splitting and the obtained giant g-factor 45-60 [11]. However, this effect should disappear quickly with the increase of the temperature above 50 K, where transition from hopping conductance to classical mechanism of conductance should take place. Nevertheless, in experiments conducted by this study effect of NMR is observed at temperatures near 77 K. That is, the presence of NMR at higher temperatures is so far unclear. Besides at low temperatures Toyozawa model does not explaine so large values of NMR.

The presence of magnetic ordering in InSb whiskers due to introducing magnetic impurities such as Mn, actually leads to the observation of negative magnetoresistance in the field dependences of the magnetoresistance [20]. The authors of [20] believe that presence of NMR is dependent on the orientation of spin of electrons scattering on Mn^{2+} ions. Results of the present study of elemental content of impurities in InSb whiskers by microprobe analysis did not reveal the presence of magnetic impurities in the samples. Besides, the presence of magnetic impurities interacting with charge carriers is rather doubtful in the whiskers due to an absence of magnetoresistance peaksplitting except the one sample corresponding to MIT. Nevertheless, the investigations of the whisker surface will be the content of further research of InSb whiskers.

The observed NMR could be explained by quantum interference of electron wave functions [21,22]. The disappearance of NMR is associated with the destruction of interference of the electron wave functions by the magnetic field. That leads to the effects of weak localization and electron-electron interaction. These effects at low spin-orbit interaction result in increase of the resistance. To determine the presence of quantum interference effects in the whiskers it is necessary to investigate the behavior of resistance in the low magnetic fields (up to 1-2 T) and low temperatures (1.7-4.2 K). Our previous investigation of InSh whiskers have shown an appearance of SdH oscillations at low magnetic fields, which indicates in the presence of quantum interference in the whiskers [10,11]. However, quantum interference effect (small corrections to the conductivity) could not call so much value of negative magnetoresistance (of about 50%) as observed in experiment.

Another explanation of the observed phenomenon was proposed. First of all, it should be noted the prevalence of surface conductance in the specimens as compared with bulk one. This conclusion results from the investigation of longitudinal and transverse resistivity of the whiskers (see Fig. 5). As follows from Fig. 5, transverse resistivity (curve 4) is significantly lesser than longitudinal one (curve 1). This can be explained by the prevalence of surface conductance in transverse specimen geometry. The similar phenomenon was observed in Si whiskers, where increase of dopant impurities approaching the whisker surface was revealed [24].

One can suppose that the same mechanism of the whisker growth by chemical vapour deposition in halogen closed system leads to the similar whisker doping by impurities during the growth process. The second reason of increase of doping impurity near the whisker surface may be diffusion of impurities to the surface during the sample annealing after their growth.

If assumption of the prevalence of surface conductance in the whisker is true, i.e., the main part of charge carriers transport takes place in subsurface region of the whisker, which can be characterized by effective wire radial distance, one can suggest the following explanation of negative magnetoresistance in the InSb whiskers. The MR peaks in Fig. 2 are due to the classical size effect, where the wire boundary scattering is reduced as the cyclotron radius becomes smaller than the effective wire radial distance, resulting in a decrease in the resistivity. The similar behavior is typical for the longitudinal MR of Bi nanowires in the diameter range dW = 45-200 nm, while the peak position B_m varies linearly with 1/dW as the wire diameter increases [23]. The condition for the occurrence of B_m is given approximately by $B_m \approx 2ck_F/EdW$, where k_F is the wavevector at the Fermi energy [23]. Taking into account the obtained Fermi energy E, one can calculate the effective wire radial distance, which for InSb whiskers is about 250 nm.

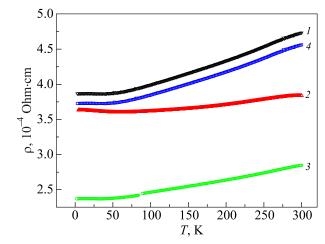


Fig. 5. (Color online) Dependences of longitudinal (*1–3*) and transverse (*4*) resistivity versus temperature for InSb whiskers with various impurity concentrations: $3.26 \cdot 10^{17} \text{ cm}^{-3}$ (*1*, *4*), $2.3 \cdot 10^{17} \text{ cm}^{-3}$ (*2*), $7.16 \cdot 10^{17} \text{ cm}^{-3}$ (*3*).

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The peak position B_m is found to increase linearly with increasing temperature in the range of 4.2–60 K, as shown in Fig. 2. At an increase of temperature phonon scattering becomes more important and leads to shift of the B_m peak position. When the whisker impurity concentration is removed from MIT (as at dielectric and metal side of MIT) a decrease of NMR peak is observed.

The decrease of the magnetoresistance peak may be attributed to a rise of effective radial distance approaching effective wire radial distance, much longer than the carrier mean free path. The negative MR observed for the InSb whiskers above B_m (Figs. 2, 3) shows that wire boundary scattering is a dominant scattering process for the longitudinal magnetoresistance, establishing that the mean free path is larger than the effective wire radial distance.

To check the size effect in the magnetoresistance of InSb whiskers the Larmor radius of the electron trajectories was calculated in the range of magnetic field 0-14 T according to the model described in the work [25]. It decreases with increasing induction in all range. So, the Larmor radius is larger than the effective wire radial distance in magnetic fields up to 2-3 T and it changes from 1 µm to 200 nm. These two parameters are equal to each other at the magnetic induction 4 T. The Larmor radius is less than the radial distance at the higher magnetic fields, and it consists 77 nm at 14 T. As follows, the contribution of size effect on the behavior of magnetoresistance was observed: in the magnetic fields up to 2-4 T strong boundary scatterring occurs leading to rapid growth of magnetoresistance. When Larmor radius becomes less that effective wire radial distance, then magnetoresistance decreases rapidly caused by decreasing in the boundary scattering.

Conclusions

It is established that longitudinal magnetoresistance of all investigated samples changes sign with increase of magnetic field: it is positive in the magnetic fields up to 2–4 T and it becomes negative at higher magnetic fields. The large negative magnetoresistance as well as the change of magnetoresistance sign were discussed due to following possible mechanisms: 1) presence of carrier quantization in the whiskers; 2) presence of magnetic ordering due to superposition of electron spins; 3) presence of magnetic ordering due to introducing uncontrolled magnetic impurities in the whiskers; 4) quantum interference of electron wave functions; 5) classical size effect.

The contribution of all the above terms were discussed and it was proposed that dominant reason of large NMR and change of MR sign could be due to the existence of classical size effect, in particular boundary scattering during their conductance in thin (of about 250 nm) subsurface layer of the whiskers. Presence of quantum interference of electron wave functions as well as magnetic ordering due to superposition of electron spins on impurities leads to nonessential contribution to observed NMR in the whiskers.

- S. Ishida, K. Takeda, A. Okamoto, and I. Shibasaki, *Physica E* 20, 255 (2004).
- S. Ishida, K. Takeda, A. Okamoto, and I. Shibasaki, *10th Conf. on Hopping and Related Phenomena*, International Centre for Theoretical Physics, September 1–4, 2003, p. 1–7.
- E. Lahderanta, A.V. Lashkul, A.V. Kochura, S.G. Lisunov, B.A. Aronzon, and M.A. Shakhov, *Phys. Status Solidi A* 211, 991 (2014).
- A. Kochura, Growth, Magnetic and Transport Properties of InSb and II-IV-As₂ Semiconductors Doped with Manganese, Diss. Lappeenranta University of Technology, Lappeenranta (2011).
- S.A. Obukhov, S.W. Tozer, and W.A. Coniglio, *Sci. Rep.* 5, 13451 (2015).
- 6. S.A. Obukhov, Phys. Status Solidi C 9, 247 (2012).
- O.V. Kirichenko and V.G. Peschanskii, *Fiz. Nizk. Temp.* 37, 925 (2011) [Low Temp. Phys. 37, 734 (2011)].
- N.A. Viglin, V.V. Ustinov, V.M. Tsvelikhovskaya, and O.F. Denisov, *JETP Lett.* 84, 79 (2006).
- J. Teubert, S.A. Obukhov, P.J. Klar, and W. Heimbrodt, J. Phys. Rev. Lett. 102, 046404 (2009).
- A. Druzhinin, I. Ostrovskii, Yu. Khoverko, N. Liakh-Kaguy, I. Khytruk, and K. Rogacki, *Mater. Res. Bull.* 72, 324 (2015).
- A. Druzhinin, I. Bolshakova, I. Ostrovskii, Yu. Khoverko, and N. Liakh-Kaguy, *Mater. Sci. Semicond. Proc.* 40, 550 (2015).
- A.A. Druzhinin, I.P. Ostrovskii, N.S. Liakh, and S.M. Matvienko, *Journal of Physical Studies* 9, 71 (2005).
- A.A. Druzhinin, I.P. Ostrovskii, Yu.M. Khoverko, N.S. Liakh-Kaguj, and Iu.R. Kogut, *Mater. Sci. Semicond. Proc.* 14, 18 (2011).
- A.A. Druzhinin, I.P. Ostrovskii, Yu.N. Khoverko, N.S. Liakh-Kaguy, and A.M. Vuytsyk, *Funct. Mater.* 21, 130 (2014).
- S. Gardelis, J. Androulakis, Z. Viskadourakis, E.L. Papadopoulou, J. Giapintzakis, S. Rai, G.S. Lodha, and S.B. Roy, *Phys. Rev. B* 74, 214427 (2006).
- R.G. Mani, L. Ghenim, and J.B. Choi, *Phys. Rev. B* 43, 12630 (1991).
- 17. Y. Katayama and S. Tanaka, *Phys. Rev.* 153, 873 (1967).
- A.A. Nikolaeva, L.A. Konopko, A.K. Tsurkan, E.P. Sinyavskii, and O.V. Botnari, *Surf. Eng. Appl. Electrochem.* 51, 45 (2015).
- 19. Y. Toyozawa, J. Phys. Soc. Jpn. 17, 986 (1962).
- A.V. Kochura, B.A. Aronzon, and M. Alam, J. Nano and Electronic Physics 5, 04015-1 (2013).
- B.L. Altshuler, A.G. Aronov, A.I. Larkin, and D.E. Khmel'nitskii, *JETP* 54, 2, 411 (1981).
- P.A. Lee and T.V. RamaknShnan, *Rev. Mod. Phys.* 53, 287 (1985).
- 23. Z. Zhang, X. Sun, M.S. Dresselhaus, J.Y. Ying, and J. Heremans, *Phys. Rev. B* **61**, 4850 (2000).
- A. Druzhinin, I. Ostrovskii, Y. Khoverko, and R. Koretskii, *Mater. Sci. Semicond. Proc.* 40, 766 (2015).
- 25. Yu.P. Gaidukov and E.M. Golyamina, JETP 48, 719 (1978).