Effect of Weak and High Magnetic Fields in Longitudinal and Transverse Configurations on Magneto-Thermoelectric Properties of Quantum Bi wires

A. Nikolaeva[a,b], L. Konopko[a,b], T. E. Huber[c], Gh. Para[a], and A. Tsurkan[a]

[a]*Ghitu Institute of Electronic Engineering and Nanotechnologies, ASM, Academiei, str. 3/3, MD-2028, Chisinau, Republic of Moldova, e-mail: A.Nikolaeva@nano.asm.md*
[b]*International Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw, Poland*
[c]*Department of Chemistry, Howard University, 500 College St. N.W., DC 20059 Washington, U.S.A*

We report on the magneto- thermopower of single-crystal Bi nanowires with diameters 75 nm and 250 nm in a longitudinal and transverse magnetic fields of 0-14T. The temperature range is 1.5–300K. Bi nanowires in a glass capillary have been prepared by the high frequency liquid phase casting. The temperature dependence $R(T)$ shows the transition from metallic to semiconducting behavior due to quantum size effect, where the Bi-wire diameter is reduced to less than 80 nm. It is for the first time that the effect of the negative magneto-resistance in a transverse magnetic field, due to the quantum size effect on 75 nm Bi wires, has been observed. The thermopower is very sensitive to the wire diameter, up to a change in the sign from negative to positive at low temperatures, and to a significant extent in a weak longitudinal magnetic field. The field dependences of longitudinal and transverse magneto-resistance have features characteristic of the occurrence of the quantum size effect and galvanomagnetic size effect, and provide information on the parameters of the energy spectrum and charge carrier mobility. The enhancement of the thermoelectric figure of merit for Bi nanowires is discussed. We also discuss the power factor $\alpha^2\sigma$ and its dependence on the diameter, magnetic field and temperature.

Keywords: Bi nanowires, thermoelectricity, size quantization effect.

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INTRODUCTION

The electrical transport and magneto-thermoelectric properties of single crystal Bi nanowires have attracted considerable attention because of the quantum size effect (QSE). In a semimetal, the QSE causes the conduction band and the valence band to break up into subbands whose numbers correspond to discrete values of the wave vector along the "quantizing" dimension. By introducing a quantum confinement, a semimetal – semiconductor transition can be achieved [1]. Due to a long electron mean free path (m.f.p.) l_e (~ 100 nm at room temperature) and a very large Fermi wavelength λ (40–60 nm), material Bi is the best candidate to study the classical and quantum size effects for the object size comparable to l_e and λ [1–9].

In [10, 11] a significant increase in thermoelectric efficiency Z was predicted in quantum Bi-wires at the semimetal – semiconductor transition due to the QSE. In the thermoelectric figure of merit $ZT = \frac{\alpha^2\sigma}{\chi}T$, σ is the electrical conductivity, α is the Seebeck coefficient (thermopower), $\chi – = k_e+k_l$ is the thermal conductivity (k_e, k_l are the electron and lattice contributions, T is the absolute temperature).

An approach to increase Z is to increase the density of states near the Fermy level at the size quantized and to decrease thermal conductivity due to an additional strong phonon scattering on the surface of quantum Bi nanowire walls [1, 10, 11]. Single-crystal wires are required so as to observe the QSE.

The galvanomagnetic size effect (GMSE) was studied theoretically and experimentally in bismuth wires with d > 200 nm, prepared by various methods [3, 6–10]; in particular, its occurrence in $\rho(H)$, $(H||I)$ is in fairly good agreement for both individual Bi wires and nanowire arrays.

As for the thermoelectric power, the available experimental results obtained in most cases on Bi nanowire arrays embedded in a porous Al_2O_3 dielectric matrix are very contradictory [12–14] and differ not only quantitatively but also qualitatively. This is probably due to some variation in the diameter of nanowire arrays, filling of pores Al_2O_3 as well as to the presence of uncontrolled structural defects, especially in establishing contacts, because the length of the nanowire arrays is less than 200 μm.

The most suitable material to study the QSE and GMSE is strictly cylindrical single-crystal Bi wires, die-cast from the liquid phase in a glass envelope with a length of a few mm [6, 8, 9].

In this paper we report the dimensional features in the magneto-field dependences of resistance and thermopower in longitudinal and transverse magnetic fields up to 14T for single crystal Bi wires with diameters of 250 nm and 75 nm (at the semimetal – semiconductor transition due to the QSE).

In weak magnetic fields, H_{max}, the maximum in the longitudinal magneto-resistance (LMR) $R(H)$, corresponds to the "cutoff" magnetic field of SdH oscillations at 4.2 K. With decreasing diameter of the wires (d < 80 nm) this maximum disappears and magneto thermopower achieves the maximum positive value at 20–30 K.

It is for the first time that the effect of the negative magneto-resistance in a transverse magnetic field (TMR) due to the QSE has been observed. The power factor $\alpha^2\sigma$ and its dependence on the diameter, magnetic field and temperature were calculated from the experimental data.

SAMPLES AND EXPERIMENT

Individual glass-coated Bi wires with the diameter < 100 nm were prepared by the high frequency liquid phase casting (the improved Ulitovsky-Taylor method) [6, 8, 9].

The orientation of the wires was verified by the X-Ray diffraction. Studies on the Diffractometer Xcalibur-E reveal that the investigated wires are single crystals and have the $(10\bar{1}1)$ orientation along the wire axis (Fig. 1, inset). In this orientation the wire axis makes an angle of 19.5^o with the bisector axis C_1 in the bisector-trigonal plane. The trigonal axis C_3 is inclined to the wire axis at the angle of 70^o, and one of the binary axes C_2 is perpendicular to it (Fig. 1, inset).

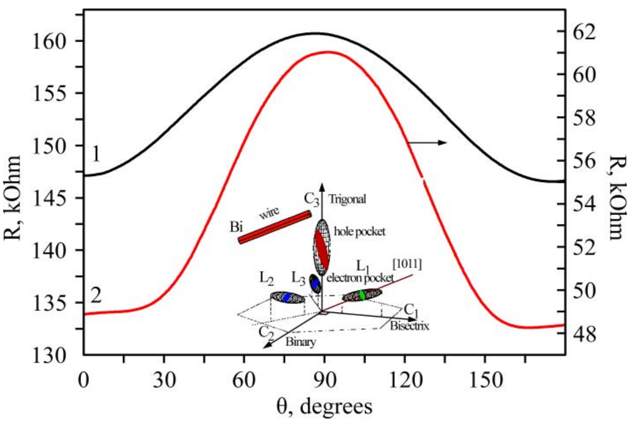

Fig. 1. Angle diagrams of transverse MR $R(\theta)$ ($H\perp T$), of Bi wires at 100K, H = 1 T. 1 – d = 75 nm; 2 – d = 250 nm. Inset: Schematic drawing the three Fermy surface electron pockets L and T hole pocket. The orientation of (1011) and Bi wire is also indicated.

Figure 1 shows the rotation angular diagram of the transverse magneto-resistance (ADTMR) $R(\theta)$ Bi wires with d = 75 nm and d = 250 nm at 100K. The curves qualitatively correspond to the similar ADBMR for bulk single crystal Bi samples for the case where the current is directed along the bisector axis [15]. In weak magnetic fields ADBMR curves have a simple bell-shape with a periodicity of 180^0. The maximum on the $R(\theta)$ ($\theta = 90^0$) corresponds to $H||C_2$ and the minimum ($\theta = 0^0 = 180^0$) corresponds to the situation when the wire axis, the crystallographic C_3 axis and the vector $\vec{H}$ are in one plane, and the angle $\angle HC_3 \approx 20°$.

Electrical contact in the butt-end of the wire to the copper foil was performed either by a fusible solder (58°) or by In or InGa-eutectics (low melting alloys). The type of the contact solder did not influence the results of measurements.

For investigations in the transverse magnetic field, a special device was applied, which allows rotating the sample in two directions: $\perp$ and || to the magnetic field. The TMR ($H\perp I$) was measured at θ = 0 (Fig. 1, inset). Measurements of the LMR ($H||I$) and TMR ($H\perp I$) were carried out in a superconducting solenoid field up to 14 T in the temperature range of 1.5–300K in the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland).

RESULTS AND DISCUSSION

The semimetalic Bi wires with a diameter of 250 nm exhibit Shubnikov de Haas (SdH) oscillations. Figure 2 (inset) shows SdH oscillations on the magneto-resistance $R(H)$ (derivative $\delta R/\delta H(H)$) in longitudinal orientations $H||I||C_1$ of Bi wires, with d = 250 nm.

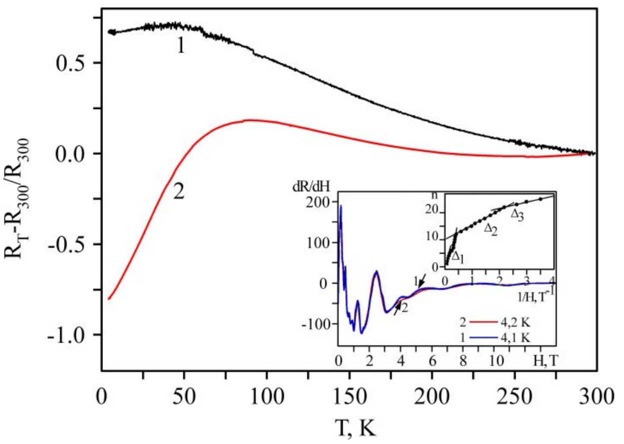

Fig. 2. Temperature dependences residual resistance $\frac{\Delta R}{R}(T) = \frac{R_T - R_{300}}{R_{300}}(T)$ of single Bi nanowires: 1 – d = 75 nm; 2 – d = 250 nm. Inset: Field dependences of LMR derivative $\delta R/\delta H(H)$ of Bi wire (d = 250 nm) at T = 2.1K and 4.2K and dependences of quantum number n of SdH oscillations on reverse field H^{-1}.

The SdH oscillations are periodic in $1/H$, with a period of $\Delta(1/H) = 2\pi e\hbar/cS$, which is inversely proportional to the extreme cross-section S of the Fermy surface in the plane normal to the magnetic field H.

In the longitudinal magnetic field ($H||I$), there are three different extreme cross sections S (cross-hatched in Fig. 1, inset) and three respective SdH oscillation periods: 1 – $\Delta_1 = 7\cdot 10^{-5}$ Oe^{-1} from one

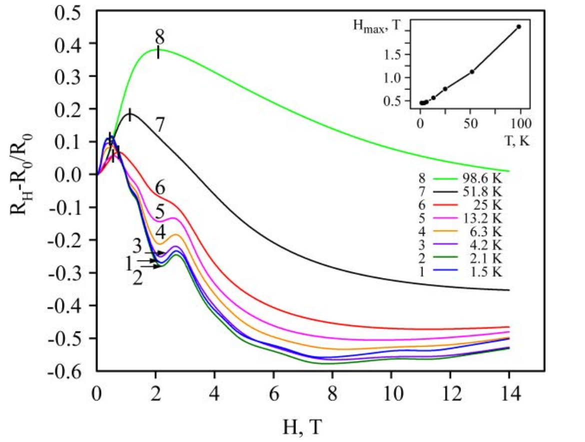

Fig. 3. Magnetic field dependences of longitudinal residual MR $\Delta R/R(H)$ for 250 nm Bi wire at different temperatures (temperatures indicated). Vertical bars indicate maximum position on $\Delta R/R(H)$. Inset: *P* peak position H_{max} as function of *T*.

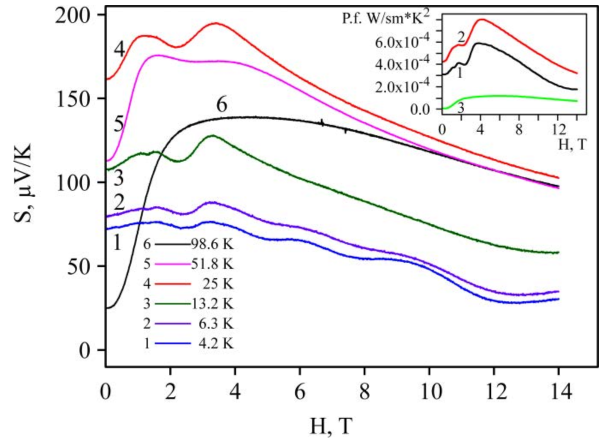

Fig. 4. Longitudinal thermopower ($H \| \Delta T$) as function of magnetic field at various temperatures (temperatures indicated) Bi wire, d = 250 nm. Inset: Magnetic field dependences ($H \| I$) of P.f. = $\alpha^2\sigma(H)$ for various temperatures calculated from Figs. 3, 4 of Bi wire, d = 250 nm. 1 – T = 13K; 2 – T = 25K; 3 – T = 98 K.

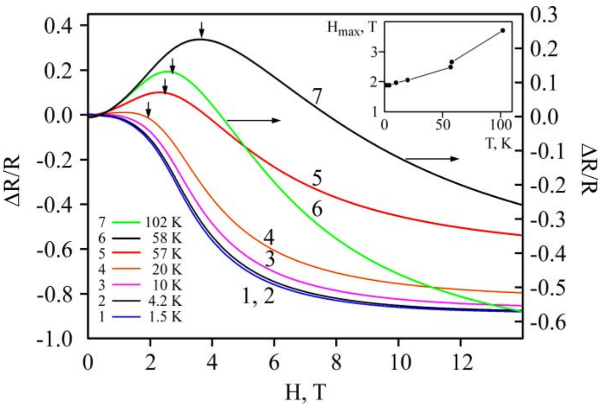

Fig. 5. Magnetic field dependences of longitudinal residual MR $\Delta R/R(H)$ Bi wire d = 75 nm at different temperatures (temperature indicated). Inset: Peak position H_{max} as function of temperature *T*.

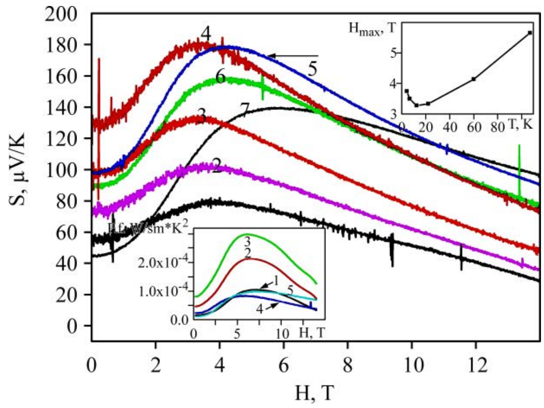

Fig. 6. Longitudinal thermopower ($H \| \Delta T$) as function of magnetic field at various temperatures (temperatures indicated in Fig. 5) Bi wire, d = 75 nm. Inset on the right: Peak position H_{max} as function of temperature *T*. Inset down: Magnetic field dependences ($H \| \Delta T$) of P.f. = $\alpha^2\sigma$ for various temperatures calculated from Figs. 5, 6 of Bi wire d = 75 nm. 1 – T = 5K; 2 – T = 10K; 3 – T = 20K; 4 – T = 57K; 5 – T = 100K.

electron ellipsoid L_1 of a smaller size S_{L1}, 2 – $\Delta_2 = 3.2 \cdot 10^{-5}$ Oe^{-1} from two equivalent electron ellipsoids of a larger size S_{L2}, 3 – $\Delta_3 = 0.5 \cdot 10^{-5}$ Oe^{-1} from the hole ellipsoid S_{T3} (Fig. 2, inset). The oscillations are in good agreement with those determined from bulk single crystals [16].

The Dingle temperature was determined from the dependences of the SdH oscillation amplitude versus the magnetic field at 2.1K. The Dingle temperature $T_D = \hbar/\kappa_B\tau$, where $\tau = l/\upsilon_F = em_*/\hbar\kappa_F$ is the carrier relaxation time. In our 250 nm Bi wire $T_D \approx 1$K [6]. This suggests that the investigated single Bi wires have very high structural perfection.

Figure 2 shows temperature dependences of the residual resistance $\frac{\Delta R}{R}(T) = \frac{R_T - R_{300}}{R_{300}}(T)$ of Bi wires with d = 250 nm and d = 75 nm. For wires with d = 75 nm, a semiconducting behavior of $R(T)$ is observed; it indicates the semimetal-semiconductor transition due to the QSE. The temperature dependence $\Delta R/R(T)$ for the wire with d = 250 nm characterizes the transition from bulk bismuth to size-dimensional wires [6, 9].

Figures 3–6 show field dependences of LMR $\Delta R/R(H)$ and longitudinal magneto-thermopower (LMTP) $\alpha(H)$ of Bi wires with d = 250 nm (Fig. 3, 4) and d = 75 nm (Fig. 5, 6) in a temperature range of 1.5–100K. A specific feature of LMR in Bi wires is the presence of a maximum in $R(H)$ in weak magnetic fields, which depends on the wire diameter and negative magneto-resistance in strong magnetic fields. Magnetic fields change the trajectory of the carriers, which leads to a change in the electrical conductivity of metals placed in an external magnetic field. The nature of the electron motion in weak and strong magnetic fields is very different. In a weak magnetic field, the Larmor radius r_L of the electron orbit $r_L = p_\perp c/eH$ is superior to the mean free path $r_L > l$, ($p_\perp$ is the component of the Fermi momentum vector perpendicular to the magnetic field H; m.p.f. evaluated in our paper [9]) and, between the successive acts of scattering, an electron moves along a short arc trajectory. In this case, the electron motion under the influence of an applied electric field is the same as in the absence of a magnetic field. Electrons, due to the curvature of the trajectory in the magnetic field, can reach the surface and be additionally scattered on the surface. In a strong magnetic field (μH >> 1), the electron has time to make several complete cycles of motion without scattering, and in this case $r_L < l$.

As mentioned previously [3, 4, 6] in the wires with 200 nm < d <1 μm, the dependence of $H_{max} \sim d^{-1}$. The Fermi momentum P_F was calculated from the dependence

$$H_{max} = \frac{2P_F c}{ed}, \quad (1)$$

$$P_F = \frac{deH_{max}}{2c} = 1{,}1 \cdot 10^{-21}\,\frac{\text{g} \cdot \text{cm}}{\text{s}}.$$

In the redistribution of the measurement, an error of P_F calculated from (1) coincides with the value of P_F, obtained from SdH oscillations from the two electron ellipsoids $L_{2,\,3}$ symmetrically arranged with respect to the wire axis (inset in Fig. 1). The presence of the maximum H_{max} in weak magnetic fields and negative magneto-resistance in strong magnetic fields testify to the occurrence of the GMSE in Bi wires. Later Dresselhaus [10] observed a similar behavior on Bi nanowire arrays.

In the wires with d = 75 nm, the maximum of the longitudinal magneto-resistance at 4.2K was absent (Fig. 5, curve *1*), indicating that there is a very small contribution to the conductivity of the L carrier, or none at all, due to the absence or reduction of the overlap of L and T bands because of the QSE. However, with increasing the temperature to a certain T value, depending on the wire diameter d (in this case, $T \approx$ 13K) in weak magnetic fields, $R(H)$ exhibits the maximum, the behavior of which with a further rise in temperature T is similar to the behavior of $H_{max}(T)$ in the Bi wires with d = 250 nm. At T > 40K, H_{max} is shifted towards higher temperatures according to the law close to linear (inset in Fig. 5). Apparently, an increase in temperature results in the diminishing of the forbidden band for semiconductors or of a band overlapping for semimetal proportionally κT, promoting the appearance of L carriers and increasing their contribution to the conductivity.

As shown in [2] and subsequently widely used in [17], especially at high temperatures, in order to explain the anomalous peak of the LMR in wires and films at higher temperatures, the following expression should be used:

$$H_{max} = \frac{2P_F}{e\sqrt{dl}} \quad (2)$$

where l is m.p.f., i.e. in the maximum m.f.p. $l = d$. In this case, the temperature dependence of H_{max} at $R(H)$ ($H \,||\, I$) becomes clear. In fact, it represents the temperature of a carrier mobility μ in Bi wires, and the maximum of $R(H)$ separates strong and weak magnetic fields (μH < 1 and μH > 1).

In the longitudinal configuration, the magneto-thermopower $\alpha(H)$ also exhibits its maximum in low magnetic fields (Fig. 6) and generally shows the same trends as the H_{max} on $R(H)$.

In the wires with d < 100 nm, the dependence of the maximum $H_{max}(T)$ on the longitudinal thermopower $\alpha(H)$ ($H \parallel \Delta T$) is nonmonotonic (inset in Fig. 7). At T > 20K, the maximum is shifted to the area of strong magnetic fields under the law close to linear, as in the wires with d = 250 nm.

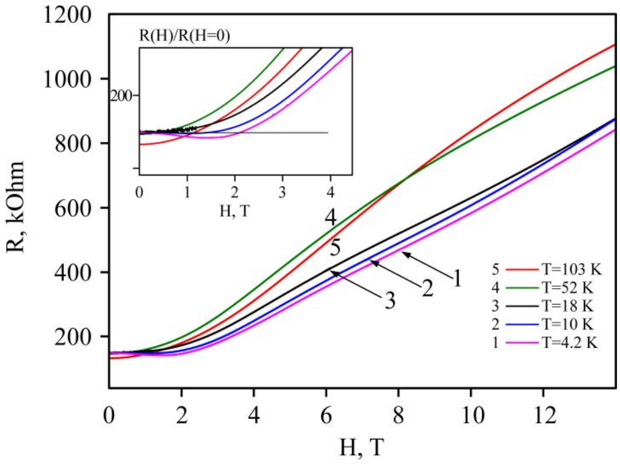

Fig. 7. Magnetic field dependences of transverse MR ($H \perp I$, $H \| C_3$) of Bi wire, d = 75 nm for various temperatures (temperatures indicated). Inset: Magnetic field dependences MR $R_H/R_0(H)$ in initial magnetic field.

In a transverse magnetic field $H \perp I$, ΔT, $H \| C_3$ (θ = 0 point on curve *1* in Fig. 1) in Bi wires with d = 75 nm, we have been first to observe the effect of a negative magneto-resistance at T < 5K associated with the quantum size effect [18, 19], which occurs only in Bi wires with d < 80 nm. At the same time, the field dependence of the thermoelectric power $\alpha(H)$ exhibits a maximum positive polarity, which decreases and shifts to the low magnetic field with increasing temperature (inset in Fig. 8). In strong magnetic fields, the thermoelectric power changes its sign from positive to negative and the point of the sign change is shifted in the region of weak magnetic fields according to the conclusions of the theory, taking into account the QSE [19].

Complex experimental studies of the resistance $R(H)$ and thermopower $\alpha(H)$ of Bi wires with d = 250 nm and d = 75 nm at different temperatures made it possible to calculate the Power factor P.f. = $\alpha^2\sigma$ and its dependence on the value and direction of the magnetic field at various temperatures.

Figures 4, 6 and 8 (insert) show the P.f. as a function of a magnetic field at various temperatures for wires with d = 250 nm and d = 75 nm. At the temperature of 100K, the maximum value of P.f. = $1 \cdot 10^{-4}$ W/cm·K^2 at H = 2 T is achieved for Bi wires with d = 250 nm. At T = 25K, the maximum value P.f. = $8.0 \cdot 10^{-4}$ W/cm·K^2 at H = 5 T is observed. In the transverse field $H \perp \Delta T$, $H \| C_3$ P.f. is almost of an order of a magnitude smaller due to a sharp rise in the positive thermopower in a transverse magnetic field.

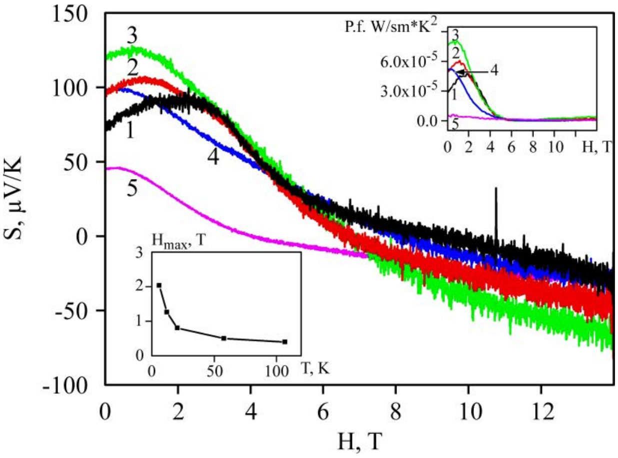

Fig. 8. Transverse magneto thermopower ($H \perp \Delta T$, $H \| C_3$) as function of magnetic field at various temperatures Bi wire, d = 75 nm. Inset left: Peak position H_{max} as function of temperature T. Inset right: Field dependences ($H \perp I$) P.f. = $\alpha^2\sigma(H)$ calculated from Figs. 7, 8 for various temperatures (temperatures indicated in Fig. 7).

It is known that in bulk Bi samples of trigonal orientation, in a temperature range of 100–150K, the thermopower has a negative value and increases 2-fold in magnetic fields < 1T. The effect was used in the magneto-thermoelectric power converters.

It should be noted that in Bi wires (d < 300 nm), the thermopower is positive at T < 100K, which is an important factor for thermoelectric applications because for the n-branches of the thermoelectric energy converter alloys, $Bi_{1-x}Sb_x$ are usually used, and the creation of p branches at low temperatures is problematic.

CONCLUSIONS

Single-crystal wires in glass cover with diameters of 250 and 75 nm have been prepared and their magneto thermoelectric properties investigated. As a result, a semimetal-semiconductor transition has been observed due to size quantization of the energy spectrum. It is shown that field dependences of the longitudinal and transverse magneto-resistance and thermopower contain singular points characterizing the expression of the GMSE and the QSE, and contain information on the parameters of the energy spectrum and on the charge carrier mobility. It is for the first time effect of the negative magnetoresistance in a transverse magnetic field, due to QSE on 75 nm Bi wires has been observed. It is also demonstrated that in quantum Bi wires the thermopower is positive, significantly increases at low temperatures and heavily depends on the wire diameter d. In addition, a significant increase in the positive thermoelectric power in the presence of a longitudinal magnetic field has been observed. The studies carried out by the authors are important not only for fundamental physics of one-dimensional structures, but also for device applications in thermoelectricity.

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Реферат

В работе приведены результаты измерений магнито-термоэлектрических свойств монокристаллических нанонитей Bi с диаметрами 75 нм и 250 нм в продольном и поперечном магнитном полях до 14 Т в интервале температур 1,5–300К. Цилиндрические нити Bi в стеклянной оболочке изготавливались литьем из жидкой фазы. Температурные зависимости сопротивления $R(T)$ показывают переход от «металлической» к «полупроводниковой» зависимости благодаря проявлению квантового размерного эффекта (QSE) при уменьшении диаметра нитей Bi менее 80 нм. Впервые обнаружен эффект отрицательного магнитосопротивления в поперечном магнитном поле, связанный с проявлением квантового размерного эффекта в нитях с $d < 75$ нм. Термоэдс чувствительна к диаметру нитей d и значительно возрастает в слабом продольном магнитном поле. Полевые зависимости продольного и поперечного магнитосопротивления имеют особенности характеризующие проявление квантового и гальваномагнитного размерных эффектов, которые содержат информацию о параметрах энергетического спектра и подвижности носителей заряда. Обсуждается вопрос повышения термоэлектрической эффективности в нанонитях Bi. Из экспериментальных данных рассчитывался силовой фактор $\alpha^2\sigma$ в зависимости от диаметра, нитей, магнитного поля и температуры.

Ключевые слова: нанонити висмута, термоэлектричество, квантовый размерный эффект.