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EFFECT OF THE LIFSHITZ TOPOLOGICAL TRANSITIONS INDUCED BY TENSILE DEFORMATION ON THE THERMOPOWER AND RESISTANCE OF BISMUTH WIRES

This paper presents the results of studying the effect of elastic deformation on the resistance and thermopower of bismuth nanowires at 4.2 - 300 K. Glass-coated single-crystal Bi wires of different diameters (70 and 320 nm) were prepared by high-frequency liquid phase casting by the Ulitovsky method. According to X - Ray diffraction, all the wires had the same $(10\underline{1}1)$ orientation along the wire axis. Methods of investigation of Shubnikov-de Haas oscillations in the pure and Sn - doped Bi wires have conclusively proven that the point of the thermopower sign inversion on the deformation dependences $\alpha(\xi)$, which coincides with the position of the maximum resistance on $R(\xi)$, is associated with an electronic topological transition induced by the elastic deformation whereby the T - holes at T - point of the Brillouin zone disappear. The enhancement of the thermoelectric figure of merit of Bi wires under high-temperature deformation is discussed.

Key words: thermoelectricity, bismuth wires, elastic deformation, Shubnikov-de Haas oscillations, quantum size effect, topological transitions.

Introduction

A rising tide of interest in thermoelectricity in recent years is due to development of new concepts, theories and nanotechnologies, opening up new ways for enhancement of thermoelectric figure of merit both in the nanostructures and the bulk nanostructured materials [1 – 4]. Thermoelectric figure of merit is defined by the expression: $Z = \alpha^2\sigma/\chi$, where α is thermopower, σ is electric conductivity, $\chi = \chi_e+\chi_p$, χ_p is lattice thermal conductivity, χ_e is electron thermal conductivity.

Of particular interest are nanowires based on bismuth and its alloys [1 – 3, 5]. Quantum and classical size effects observed in nanowires open up the prospects for electron and phonon transport control by means of structural engineering. The authors of [1 – 3] demonstrated the enhancement of thermoelectric figure of merit in size-restricted structures, specifically in *Bi* nanowires. Increase in *Z* can be due to quantum size effect leading to semimetal - semiconductor transition with an increase in the density of states close to the Fermi level, which, in turn, results in considerable thermopower increase. As a result of this, power factor ($\alpha^2\sigma$) grows as compared to the bulk samples. On the other hand, carrier and phonon scattering on the boundary will lead to reduction of thermal conductivity, which also leads to *Z* increase. In so doing, realization of phonon scattering mechanism (impairing figure of merit) must not impede effective transport of charge carriers [6]. Small diameter of nanowires leads to efficient phonon scattering by the surface and gigantic reduction of thermal conductivity [7, 8].

Only a few papers pursued experimental research of thermal conductivity in bismuth nanowires [5, 9, 10] of different structure and manufactured by different methods. Both in single crystal and polycrystal wires there was a considerable suppression of thermal conductivity as the wire diameter d was reduced. Moreover, in single crystal *Bi* wires there was considerable thermal conductivity anisotropy [10] exceeding that value in the bulk samples of *Bi*. In [5] it was shown that in single crystal *Bi* nanowires grown with (110) orientation (i.e. normal to trigonal axis) the thermal conductivity is a factor of 4 smaller than in the wires with (102) orientation and is considerably reduced with decreasing diameter, which points to strong scattering of hot carriers at the boundaries.

A search for materials with a combination of such properties as high electric conductivity is the task of optimization and thermoelectric figure of merit enhancement. Complex properties control can be realized with the knowledge of interrelation between the structure and properties of semimetals and semiconductors of different composition, relying upon the theoretical concepts of condensed matter physics.

One of possible methods of thermoelectric parameters control is elastic deformation whereby there is an essential change in the Fermi surface topology in *Bi* and its alloys [11, 12, 13].

In [13] it was shown that elastic deformation in glass – coated single crystal *Bi* nanowires can reach 2 – 3 % of relative elongation, which brings about essential changes in the Fermi surface topology.

This work presents the results of research on the conductivity and thermopower changes under elastic deformation of glass-coated single crystal *Bi* wires of different diameters in the temperature range 4.2 to 300 K. A change in the Fermi surface topology was controlled by means of Shubnikov - de Haas (ShH) oscillations in the wires of pure and *Sn* – doped bismuth.

Samples, experimental procedure

Glass-coated pure and *Sn* – doped *Bi* wires were prepared by liquid phase casting by the Ulitovsky-Taylor method [13 – 16] and had the form of a strictly cylindrical single crystal with diameters from 75 nm to several microns with (1011) orientation along the wire axis (Fig. 1).

Tests performed with the help of rotation *X* - ray diagrams of submicron *Bi* wires showed that the resulting wires were single crystal, as a rule, of the same orientation: wire axis coincides with ΓL – orientation in reduced Brillouin zone, which is located in bisector – trigonal plane and makes with bisector axis an angle of 19.5˚ (Fig. 1).

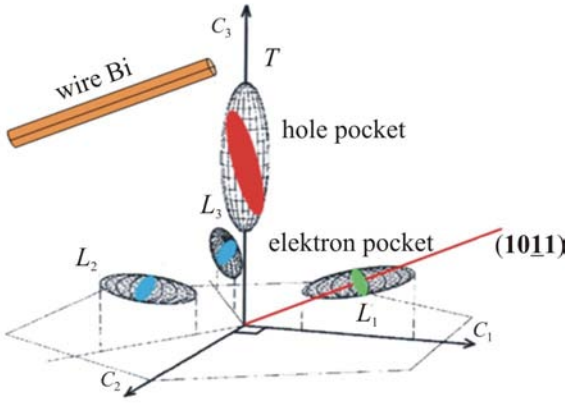
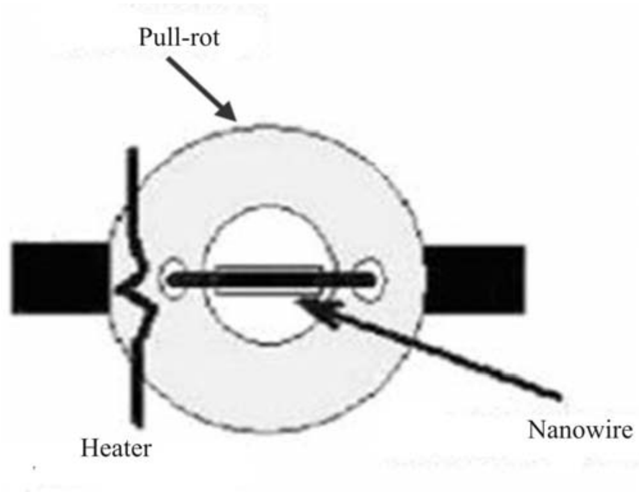


Fig. 1. $(10\underline{1}1)$ *orientation of Bi Fermi surface with respect to wire axis.*

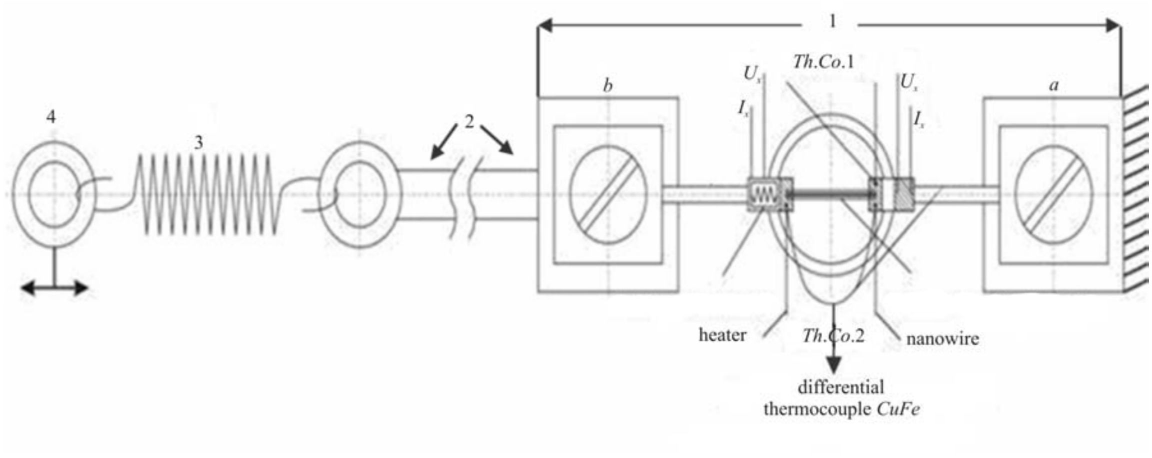
In so doing, trigonal axis C_3 is inclined to the wire axis at an angle of 70°, and one of the binary C_2 axes is normal to that axis.

Orientation of crystallographic axes in *Bi* and its alloys and structural perfection were also confirmed by the investigations of angular diagrams of revolution of transverse magnetoresistance, as well as by means of studying SdH oscillations [13 – 16].

To study the deformation curves of resistance $R(\xi)$, thermopower $\alpha(\xi)$ and SdH oscillations, a special holder was designed (Fig. 2) making possible investigations in the temperature range 4.2 to 300 K both pointwise and in the automatic mode with elastic wire tension to 2 – 3% of relative elongation in magnetic fields up to 14*T* [17].



a)



b)

Fig.2. (a) – schematic of sample mounting on a ring of beryllium bronze when measuring kinetic properties of wires under elastic tension. b) – schematic of sample (wire) holder allowing investigations under elastic tension in the temperature range 2 to 300 K. (1 – construction of beryllium bronze ring, Th. Co. 2 – differential thermocouple CuFe, 2 – rod regulating stress from spring to collar, 3 – spring, 4 – screw).

When studying the wires exposed to elastic tension, the wire was mounted on a ring of beryllium bronze, diameter ≈ 5 mm (Fig. 2, *a*). Two contact pads of copper clad fiberglass of size 2 mm × 1 × 0.5 mm fastened on a ring at the distance of 2 – 3 mm from each other served as contact pads for mounting a sample and a heater for creation of temperature gradient when measuring thermopower. Temperature gradient was maintained by a miniature heater and measured by a differential $Cu-Fe$ thermocouple. All the data, namely a signal from the sample, temperature, temperature gradient and the rated value of thermopower were output to computer and observed in the process of measurement on a display. The whole construction – a ring of beryllium bronze with the sample – was placed in the holder (Fig. 2, *b*).

One ring end (*a*) was secured immovably, and the other (*b*) was connected to a stretching moving member. Travel of screw (4) on rotation is passed to the free end of the spring (3), connected by rod (2) with beryllium ring. This leads to a change in force action on a ring of thermally processed beryllium bronze and, hence, to a change in the deformation of this ring, and to a change in the elongation *Bi* microwire section rigidly attached to the opposite points of beryllium ring. Screw axis by means of gear mechanism is connected to DC motor, as well as to a multiturn potentiometer. By the change in potentiometer resistance one can determine the turn angle of the screw and, accordingly, determine a change in sample elongation. Such a schematic allowed operating the experiment automatically. In this case, on switching the motor, program commands digital multimeters to measure potentiometer resistance and to measure sample resistance or thermopower, displays onscreen curve $R(\xi)$ or $\alpha(\xi)$ and saves the measured data in a file.

Preliminary calibration of ring stretched due to applied force was performed with the microscope at $T = 300$ K. The above construction allowed gradual stretching of sample to 2 – 3% of relative elongation $\xi = \frac{\Delta l}{l_0} = \frac{l-l_0}{l_0} \cdot 100\%$, where l_0 is sample length prior to stretching. The accuracy of measuring tensile force was ~ 2 %.

Special attention was focused on the observance of elastic deformation condition. For this purpose, numerous deformation cycles were performed, and reproducibility of results was estimated. Temperature dependences of resistance and thermopower both with and without elastic deformation were obtained automatically on a computerized setup in the temperature range 77 to 300 K in the Laboratory of Electronics of Size-Restricted Structures, D.Gitsu Institute of Electronic Engineering and Nanotechnologies of the Academy of Sciences of Moldova, and in the range 4.2 to 300 K - in the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland) in magnetic fields up to 14 *T*.

Results and discussion

Complex measurements of temperature dependences of resistance $R(T)$, thermopower $\alpha(T)$, as well as deformation dependences of resistance $R(\xi)$ and $\alpha(\xi)$ of *Bi* wires of different diameters were performed in the temperature range 4.2 to 300 K.

For an unambiguous interpretation and recording of change in the section of the Fermi surface due to T - holes at elastic deformation, use was made of the wires doped with *Sn* acceptor impurity wherein the Fermi level is shifted down the energy scale and the concentration of T - holes increases essentially. This made it possible to study SdH oscillations at elastic deformation on $R(B)$ dependences, rather than on $\partial R/\partial B(B)$ derivatives, which simplified the experiment considerably.

It is known that SdH oscillations from T - holes in the longitudinal configuration $B \parallel I$ of pure *Bi* wires (at $\xi = 0$) are observed only in strong magnetic fields, on $\partial R/\partial B(B)$ derivatives, as a rule on the wires where longitudinal magnetoresistance is not saturated in a strong magnetic field, but starts to increase [18] at $B > 8$ T. Though at precise orientation of magnetic field $B \parallel I$ and parallel to

(10$\underline{1}$1) direction in *Bi* wires with $d < 500$ nm the magnetoresistance is saturated in magnetic fields 6 T < B < 14 T [16, 19].

Note that, as was found earlier, on the wires with $d < 75$ nm there was a semimetal-semiconductor transition caused by quantum size effect that manifested itself in “semiconductor” nature of dependence $R(T)$ and inversion of thermopower sign to $\alpha(T)$ [13 – 15, 19].

The figure represents deformation dependences of resistance R on the tension of *Bi* wires of diameters d at 4.2 K. The elasticity of deformation (tension) process was confirmed by the reproducibility of results with multiple cycles of elastic tension. For *Bi* wires with $d \geq 90$ nm a typical feature is that in low temperature region (4.2 K) on the deformation curve $R(\xi)$ there is a minimum, at $\xi = 0.8 – 1.2$ % in the area of electron topological transition $3e^L+1h^T \rightarrow 2e^L+1h^T$, then the resistance is drastically increased by a factor of 2 – 4, and in the area of maximum tensions an abnormal maximum is formed which was followed by plasticity area. Such a nonmonotonous dependence of resistance on tensile force is typical of wires with $d > 90$ nm.

At wire diameters smaller than 80nm, the shape of deformation curves at 4.2 K is changed, the maximum on $R(\xi)$ is reduced and at wire diameters 60 – 45nm the deformation curve $R(\xi)$ is declining (Fig. 3, curve 1). Resistance $R(\xi)$ is reduced by 20 – 30%. Such a shape of deformation curve of resistance $R(\xi)$ is typical of bismuth wires doped with *Te* (Fig. 3, curve 2), when holes at T - point disappear and conductivity is defined only by L - electrons. Thus, this fact is an indirect proof of semimetal-semiconductor transition in *Bi* wires with $d < 80$ nm due to quantum size effect.

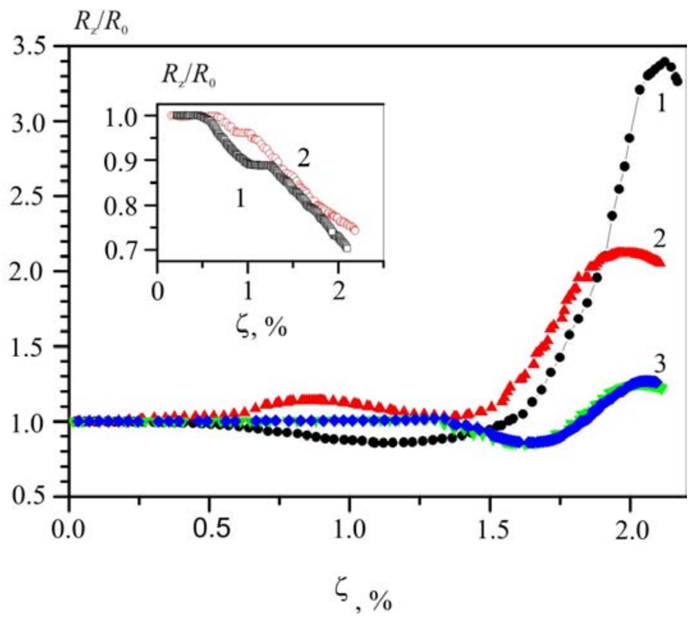


Fig. 3. Deformation curves of reduced resistance $R_\xi/R_0(\xi)$ of Bi wires of different diameters at T = 4.2 K. 1. d = 300 nm, 2. d = 110 nm, 3. d = 80nm. On the insert: deformation dependences of resistance R(ξ) of Bi wire (1), T = 4.2K, d=50nm and (2) Bi - 0.025 at % Te, d = 200nm. The scale for curve 2 is doubled.

Figs. 4, 5 represent transformation of deformation curves of resistance $R(\xi)$ and thermopower $\alpha(\xi)$ of *Bi* wires, $d = 320$ nm, with a rise in temperature from 4.2 K to 300 K. There is a gradual transition from a nonmonotonous dependence $R(\xi)$ at 4.2 K to a linear dependence (with two slopes) of resistance decrease with tensile deformation at temperatures 250 – 300 K (Fig. 4, curve 9).

The smaller wire diameter d, the higher the temperature of transition from a nonmonotonous dependence $R(\xi)$ to a linear resistance decrease with a rise in temperature. The value of resistance maximum $R(\xi)$ and the behaviour of deformation dependence $R(\xi)$ are essentially dependent on temperature T (Fig. 4).

With a rise in temperature, the value of maximum on the deformation curve of resistance was reduced essentially (Fig. 4). The insert in Fig. 4 shows exponential growth of maximum resistance value by $R(\xi)$ with temperature decrease, which points to reduction of charge carrier concentration.

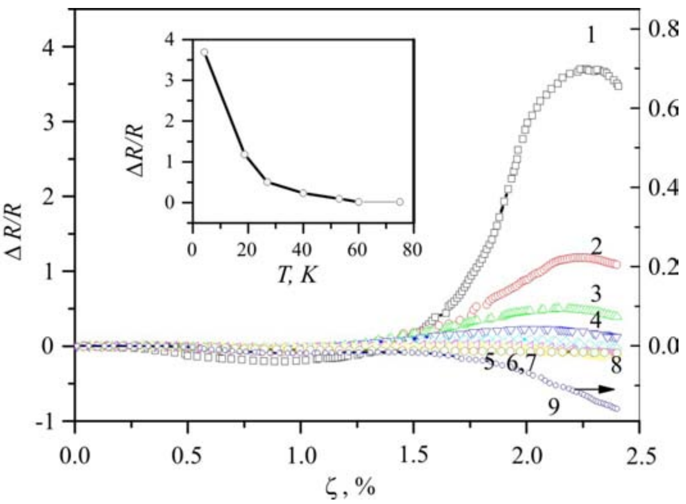


Fig. 4. Deformation dependences of relative resistance $\Delta R/R(\xi)$ of Bi wire with d = 320 nm: 1- T = 4.2, 2 – T = 18,7, 3 – T = 27, 4 – T = 40, 5 – T = 53, 6- T = 60, 7- T = 75, 8- T = 187, 9 – T = 203K. On the insert: maximum resistance value $(\Delta R/R)_{max}$ on the deformation dependences of resistance versus temperature T.

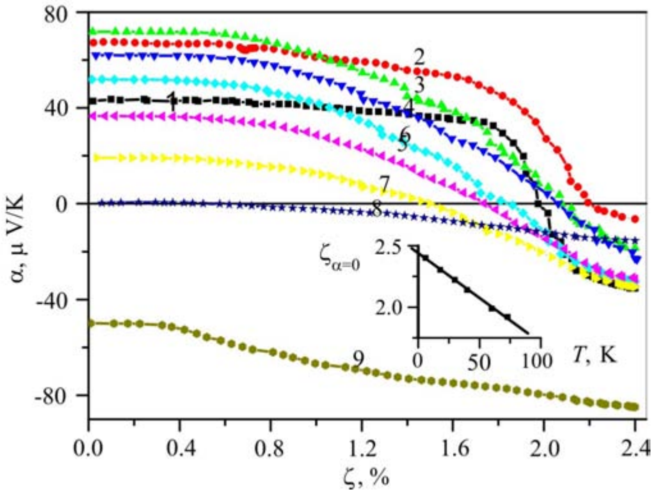


Fig. 5. Deformation dependences of thermopower $\alpha(\xi)$ at different temperatures of Bi wire with d = 320 nm 1- T = 4.2, 2- T = 18.7, 3- T = 27, 4- T = 40, 5- T = 53, 6- T = 60, 7- T = 75, 8- T = 187, 9- T = 203K. On the insert: elastic deformation value ξ, whereby thermopower inverts sign under tension versus temperature T.

Fig. 5 represents deformation dependences of thermopower $\alpha(\xi)$ of *Bi* wire with d = 320nm at different temperatures. It has been established that with a rise in temperature the inversion of thermopower sign is shifted to the area of weaker elastic deformations (insert in Fig. 5).

Dependence of temperature of thermopower sign inversion from (+) to (-) on the deformation curves of thermopower α(ξ) is almost linear (insert in Fig. 5). Different temperature dependences of resistance and thermopower anomalies are proof of the fact that resistance behaviour is due to other mechanism than thermopower, but both facts point to reduction of *T* - holes concentration at tensile deformation of *Bi* wires with $(10\underline{1}1)$ orientation along the axis.

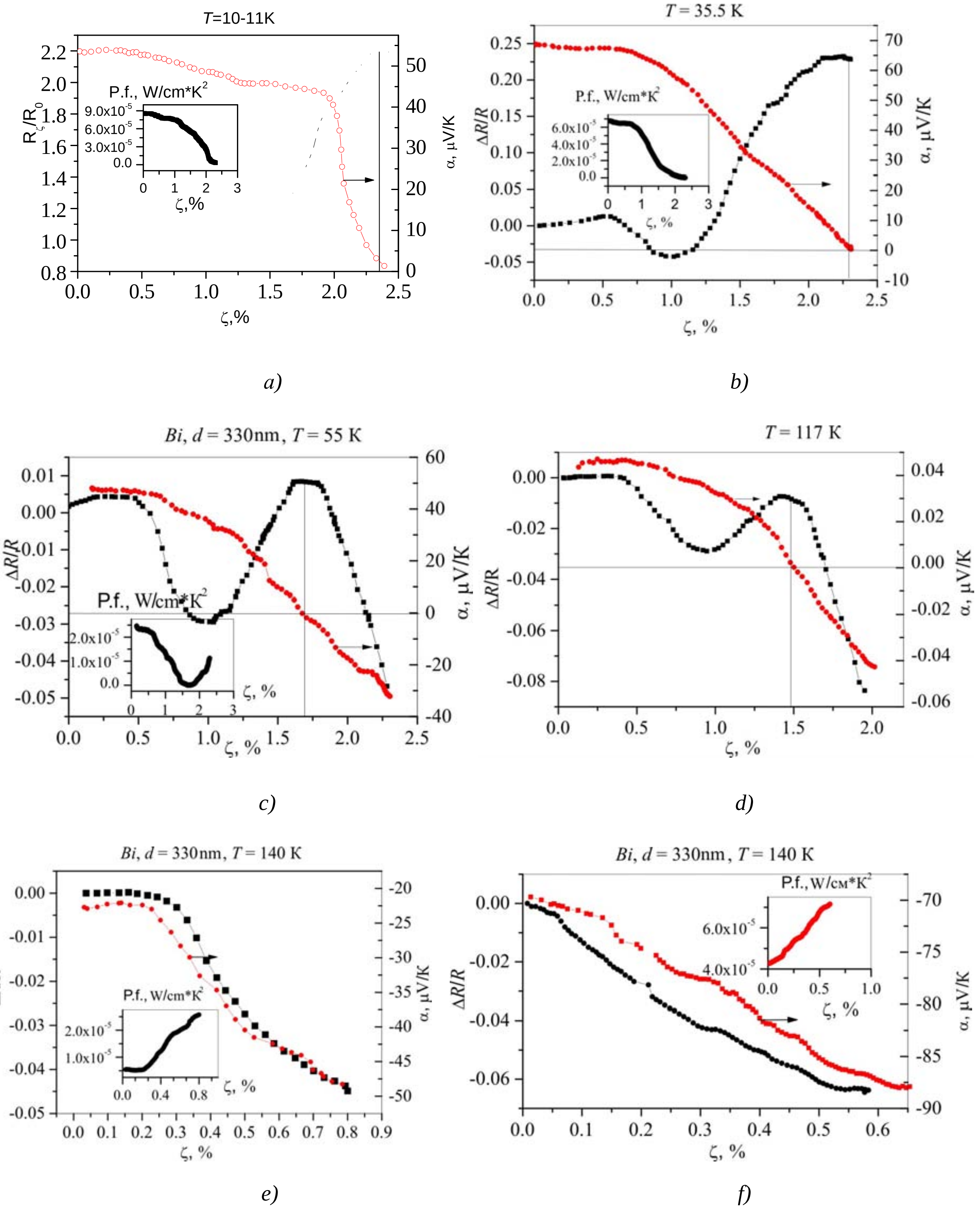
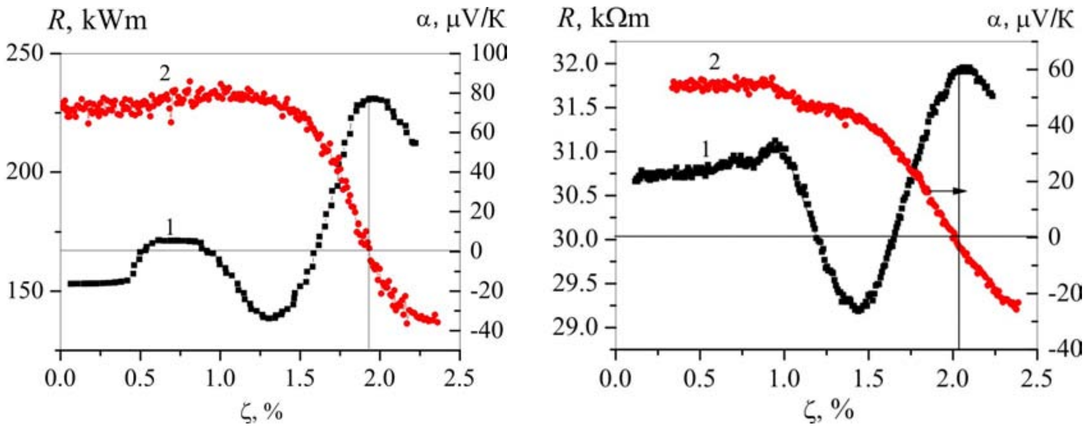


Fig. 6. Deformation dependences of relative resistance ΔR/R(ξ) and thermopower α(ξ) of Bi wire, d = 330nm at different temperatures: a) T = 11, b) T = 35.5, c) T = 55, d) T = 117, e) T = 140 K, f) T = 300K. On the inserts – deformation dependences of power factor P.f. = $\alpha^2\sigma$.

Figs. 6, 7 represent deformation dependences of resistance and thermopower of *Bi* wires with d = 320 nm and d = 75 nm at different temperatures.

As was shown in [12, 20], anisotropic deformation of crystal lattice of *Bi* wires leads to a strong nonequivalent change in the volume of individual parts of the Fermi surface, causing no essential change in the anisotropy and slope angles of increasing electron ellipsoids.

Resistance decrease on $R(\xi)$ at deformation in tensile region 0 – 1 % and formation of minimum at $\xi = 0.8 - 1.1$ % is explained in terms of electron topological transition, whereby carriers from electron ellipsoid L_1 elongated along the wire axis with less mobile carriers flow over to $L_{2,3}$ electron ellipsoids with more mobile charge carriers up to electron topological transition $3e^L+1h^T \rightarrow 2e^L+1h^T$. Exactly at these values of ξ a minimum is formed on the deformation curve of resistance $R(\xi)$ at 4.2 K (Fig. 6 *a*, *b*, *c*, *d*, *e*, *f*, Fig. 7).



(a) *(b)*

Fig. 7. Deformation dependences of resistance $R(\xi)$ (1) and thermopower $\alpha(\xi)$ (2) in Bi nanowires Bi, d = 70 nm, at different temperatures: a)curve 1 – T = 4.2K, curve 2- T = 6.6 K);b)T = 55 K.

For the first time the availability of transition under anisotropic deformation of bulk *Bi* crystals was mentioned in [12, 20], and under tension of whiskers - in [21]. In the deformation area prior to vanishing of S^e, carrier concentration of electron ellipsoid L_1 at extreme point is so small that contribution to electric conductivity is observed no more. In the bulk samples of *Bi*, critical deformation whereby the electron ellipsoid L_1 disappears $\xi_k \approx 0.5\%$, which is in good agreement with the value of $\xi_k = 0.4$ % in *Bi* whiskers [21]. According to estimates made in [20], such electron topological transition should occur at $\xi = 0.4$ %.

At 200 K the resistance is reduced in the entire area of elastic tension, the absolute value $\Delta R/R$ in the temperature range 250 to 300 K being not more than 10 – 15 %. The temperature whereby there is a quantitative change in dependence $R(\xi)$ is a function of the wire diameter d. A nonmonotonous dependence $R(\xi)$ is observed in the wires of almost all diameters under study (d > 80 nm) and takes place up to temperatures 100 to 200 K, shifting towards higher temperatures with decreasing diameter.

Conspicuous is the fact that maximum on the deformation curves of resistance at different temperatures coincides with the point of inversion of thermopower sign from (+) to (-) on the deformation curves of thermopower (Fig. 6 *a* – *d*, Fig. 7).

The most precise information on the change in the Fermi surface (FS) and its variation with lattice deformation was obtained from the Shubnikov - de Haas effect (SdH).

As already mentioned above, SdH oscillations from T - holes (from the hole section close to maximum) are poorly seen on the wires at $B \parallel I$ on $R(B)$, i.e. on the longitudinal magnetoresistance, particularly if ellipsoid is reduced under deformation and there is a sharp drop of oscillation amplitude. However, experiments on the bulk *Bi* samples on recording $\partial R/\partial B(B)$ derivatives both in $\perp$ and $\parallel$ magnetic fields have shown that under tensile deformation along bisector axis there is a reduction in the volume of hole ellipsoid at T - point [12, 20]. Experiments on the wires doped with *Sn* acceptor impurity, when the Fermi level is determined down the energy scale, and concentration of T - holes is increased on doping, lead to manifestation of ShH oscillations from T - holes on the longitudinal magnetoresistance even on $R(B)$ (Fig. 8), so it is easy to trace a change in the period of ShH oscillations from T - holes (Fig. 8) under deformation of the wires with $(10\underline{1}1)$. orientation.

For clarity, Fig. 8 shows ShH oscillations on the longitudinal magnetoresistance $R(B)$ of *Bi* – 0.07at % *Sn* wire with $d = 600$ nm under different values of tensile deformation. The insert in Fig. 8 shows a change in the frequency of ShH oscillations from T - holes, L_1 and $L_{2,3}$ hole ellipsoids at L - point of the Brillouin zone $f = [\Delta(1/H)]^{-1} = \dfrac{S_{ex}c}{eh}$, where S_{ex} is extreme section of the Fermi surface normal to magnetic field direction (shaded cross - sectional area of the Fermi surface in Fig. 1). From Fig. 8 (insert) it follows that the volumes of the Fermi surface of T - holes (f_1) and $L_{2,3}$ – hole ellipsoids at point L (f_3) are reduced, and the volume of hole ellipsoid L_1 (f_2) is increased.

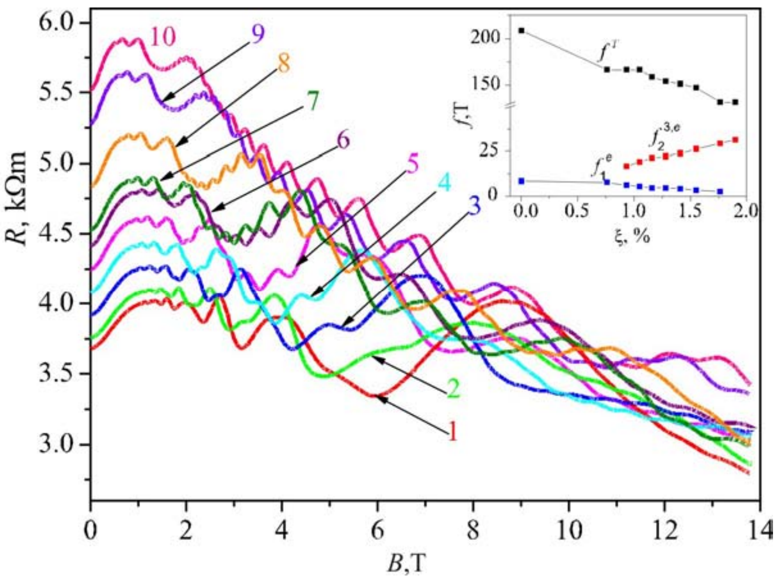


Fig. 8. ShH oscillations on longitudinal magnetoresistance R(B) (B || I) of Bi wire - 0.07at % Sn with $(10\underline{1}1)$ orientation along the axis of wire d = 600 nm with fixed values of elastic tension from ξ = 0 to ξ = 1.9 %, T = 4.2 K. On the insert: Deformation dependences of ShH oscillation frequencies on the sections of the Fermi surface: T- holes (f_1), L_1 - hole (f_2) and $L_{2,3}$ - hole ellipsoids (f_3) at T = 4.2 K.

Estimation of elastic deformation whereby T - ellipsoid will disappear was done with regard to the data on ShH oscillations on the bulk *Bi* samples (method of extrapolation to 2 % of relative elongation) yielded the value of $\xi = 1.2\%$, when a tensile force is directed along the bisector C_1 axis. Taking into account that the axis of *Bi* wires is deflected by 20 % from the bisector C_1 axis and that topological transition $3e^L+1h^T \rightarrow 2e^L+1h^T$ occurs in *Bi* wires at the values of $\xi = 0.8 – 1$ %, i.e. twice those in the bulk samples of *Bi* ($\xi_k = 0.4$ %), oriented along C_1. Thus, the value of relative elongation

ξ = 2.2 % is a real one, indicating the disappearance of T - ellipsoid in pure *Bi* wires at maximum point on $R(\xi)$ dependence. Exactly at this point there occurs inversion of thermopower sign from (+) to (-).

From the standpoint of practical applications, in the context of thermoelectricity it would be interesting to trace as a whole a change in resistance $R(\xi)$, thermopower $\alpha(\xi)$ and power factor $\alpha^2\sigma$ due to elastic tensile deformation in the temperature range 200 to 300 K. In Fig. 6 *a* – *f* (inserts) are shown calculated values of power factor as a function of tension of *Bi* wire with d = 320 nm at different temperatures. It is established that power factor growth occurs only in the temperature range where resistance is reduced, i.e. at high temperatures. In the low temperature region, where $R(\xi)$ is of nonmonotonous nature, power factor is reduced as well. A simultaneous resistance decrease and increase in the absolute thermopower value at tensile deformation in *Bi* wires in the high temperature range T > 250 K results in power factor duplication $P.f. = |\alpha|^2\sigma$ (Fig. 6, *a*), which achieves the value 7.2*10^{-5} W/cm*K^2 at temperatures 250 – 300 K, at ζ = 0.6 % for a wire with d = 330 nm. Restriction of power factor growth is related to restriction of the area of elastic tension of *Bi* wires to 1.5 % in the temperature range T > 250 K.

Taking into account the fact that thermal conductivity in *Bi* nanowires with d = 200 nm at 300 K is at least 3 times lower [5, 10] than in the bulk samples, one should expect the value of ZT more than unity at 300 K in *Bi* wires at elastic tension deformation along the bisector axis.

Conclusions

Integrated studies of the deformation dependences of resistance and thermopower of glass-coated single crystal *Bi* wires of different diameters in the temperature range 4.2 to 300 K have been performed. It is shown that anomalies on the deformation dependences of resistance $R(\xi)$ and thermopower $\alpha(\xi)$ correspond to deformation-induced topological transitions.

The method of investigation of Shubnikov - de Haas oscillations in the pure and acceptor impurity - doped bismuth has proved conclusively that the point of thermopower sign inversion to $\alpha(\xi)$, which coincides with the position of maximum on $R(\xi)$ in low temperature region is related to electronic topological transition induced by strong (up to 6 GPa) elastic deformation whereby holes at T - point of the Brillouin zone disappear. The thermoelectric figure of merit is increased to Z > 1 at 300 K with elastic deformation of single crystal *Bi* wires with (10$\underline{1}$1) orientation along the axis.

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