

STRAIN INFLUENCE ON THE DIFFUSION THERMOPOWER IN Bi WIRES

E. Condrea

*Ghitu Institute of Electronic Engineering and Nanotechnologies, Academiei str. 3/3, Chisinau,
MD-2028 Republic of Moldova
E-mail: condrea@nano.asm.md*

(Received November 21, 2018)

Abstract

We present the results of studies of the thermopower in glass-coated Bi wires under the action of uniaxial deformation. At the liquid helium temperature, thin Bi wires exhibit large positive thermopower values, which are dominated by the diffusive transport mechanism of holes. The observed increase in the negative contribution to total thermopower under uniaxial strain testifies that the hole-dominated transport can be transformed into the electron-dominated transport. The observed trend to negative thermopower values under strain can be attributed partially to a slight increase in the electron mobility and partially to the occurrence of the phonon drag effect of electrons. The phonon drag effect prevails in the thermopower, if the phonons acquire a sufficient momentum to scatter the carriers across the Fermi surface changed after an Electronic Topological Transition.

1. Introduction

Currently, metallic and semimetallic wires are widely used as building elements in a variety of nanodevices. The emergence of studies concerned with the transport properties of Bi wires is motivated partially by the interesting thermoelectric and magnetotransport properties of bismuth nanowires (NWs), which make them potentially useful for device applications [1–4].

The prospect of the thermoelectric application of the wires will require the possibility of controlling their parameters. In this context, Bi wires embedded in an individual glass capillary allow us to perform measurements of thermoelectric parameters in wide ranges of temperatures and elastic strains. It is well known that the total thermopower (TEP), which is also referred to as the Seebeck coefficient (S), of bulk bismuth samples at the liquid helium temperature is the sum of the diffusion (S_d) and phonon drag terms (S_g):

$$S = S_d + S_g. \quad (1)$$

Systematic measurements of a set of bulk Bi samples with different sizes have shown the changes in the magnitude of phonon drag thermopower with a moderate variation in the size of the samples [5–8]. As concerns low-dimensional Bi samples, such as films, whiskers, and wires, there is a larger spread in the thermopower measurements. In addition, a severe discrepancy was observed between experimental results for Bi wires with identical diameters obtained by different technological methods [9–13] (see table).

In different cases of positive S , which indicate the p -type transport for Bi NW composites [10] and for individual polycrystalline NWs [3] or negative S of Bi/Al₂O₃ arrays [11], the authors suggest that uncontrolled impurities can easily affect the value and sign of S .

In general, in bulk Bi crystals oriented along the bisectrix axis, an intrinsically negative phonon drag contribution is dominant in the TEP at low temperatures [6, 7]. Our previous studies of individual glass-coated Bi wires oriented along the bisectrix axis revealed a large positive thermopower dominated by the diffusion effect [14]. The diffusive origin of the TEP was attributed to structural imperfections, which strongly diminish the phonon–electron contribution to the total TEP. The suppression of the electron contribution to TEP was attributed to a mechanism based on the model of selective carrier scattering by potential barriers, which was advanced by Ravich [15]. Some significant changes in the quality and parameters of Bi wires can be obtained using different external impacts, such as the electric field effect, deformation, and magnetic field.

Thermopower discrepancy observed between experimental results for Bi wires obtained by different technological methods

Technological methods	Thermopower (TEP)		Temperatures (T) K
	carrier dominated transport (d is diameter, nm)		
	positive (p -type)	negative (n -type)	
glass-coated Bi Nws ⁹	$d < 800$	$d > 800$	77
Bi NWs ¹⁰ and composites ¹¹ embedded in porous anodic alumina (Bi/Al ₂ O ₃)	positive		below 30
		negative	above 30
		$d = 40$ and 65	4.2–300

The goal of this work is to study the changes revealed in the TEP behavior under the action of uniaxial deformation at low temperatures. To draw detailed information on how the diffusive and phonon drag terms of TEP vary with strain, we measured the temperature dependences of thermopower $S(T)$ at various values of applied strain. The critical value of the uniaxial strain when the phonon drag effect becomes predominant in the total TEP was determined.

2. Results and Discussion

The studied single-crystalline Bi wires were obtained by the glass-coated melt spinning method [16]. The axis of the prepared wires is oriented at an angle of about 19° to the C_1 bisector axis in the bisector–trigonal plane.

Experimental procedure and methods are presented in detail in our previous papers concerning the galvanomagnetic properties of Bi wires [14, 17]. In this paper, we discuss the results of the thermopower measurements extended in a temperature range of 4.2–50 K and focus on the explanation of the unusual behavior of TEP at a high value of uniaxial strain.

The measured temperature dependences of the thermopower for wires with diameters of 100–350 nm exhibit a nearly linear behavior with a positive slope in a temperature range of 5–20 K (Fig. 1a, curves 1–3). Considering the quasi-linear dependence of $S(T)$, the observed TEP was

associated with the diffusion transport mechanism. According to [14], the diffusive origin of the TEP was attributed to the limited diameter and structural imperfections within the nanowire, which strongly diminish the contribution from electrons or from the phonon–electron interaction to the total thermopower. The suppression of the electron contribution to TEP was attributed to a mechanism based on the model of selective carrier scattering by potential barriers, where the electrons are scattered much stronger than holes, which eventually affects the electron mobility: $\mu_e < \mu_h$ [15]. The predominance of the phonon drag term was observed for the thicker wires with $d > 600$ nm (Fig. 2). This finding can be attributed to an increase in the electron to hole mobility ratio, which in turn can be associated either with a diminished effect of the surface scattering of electrons (due to an increase in the wire diameter) or with a decrease in the concentration of structural imperfections (due to technological conditions in the preparation of thicker wires).

In the continuation of the studies of the strained Bi wires, the dependences of thermopower on strain ($0 \leq \varepsilon \leq 3\%$) and temperature ($4.2 \text{ K} \leq T \leq 300 \text{ K}$) were measured. Note that an increase in the contribution of electrons to TEP up to a sign change from positive to negative was observed in our earlier studies on Bi wires under a uniaxial strain [17]. The TEP dependences on strain are fairly different for thinner and thicker wires in the studied range of diameters (Fig. 1).

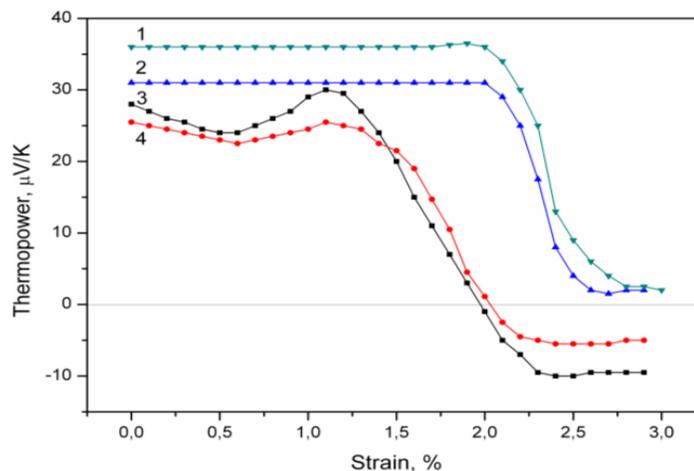


Fig. 1. Strain dependence of TEP for Bi wires of different diameters at 4.2 K: (1) 2.4, (2) 1.0, (3) 0.18, and (4) 0.35 μm .

Two types of behavior for two sets of samples were revealed: the first type for NWs with diameters $0.1 \leq d \leq 0.35 \mu\text{m}$ and the second type for wires with $0.35 < d < 3.0 \mu\text{m}$. The application of strain upon the wires of the second type ($d > 0.35 \mu\text{m}$) does not produce any variation in the $S(\varepsilon)$ dependence up to a strong deformation (Fig.1, curves 1, 2). Only above $\varepsilon = 2.0\%$, the $S(\varepsilon)$ decreases and trends to zero. The second type of behavior is developed for the wires when the phonon drag contribution is dominant in the total TEP. In the case of the first type wires ($d \leq 0.35 \mu\text{m}$), the $S(\varepsilon)$ dependences exhibit a nonmonotonous behavior, which indicates a complex charge transport mechanism (Fig. 1, curve 3, 4). The initial positive TEP smoothly decreases, goes through fine minima, and increases up to a peak around strain of 1.1% with the subsequent sharp decrease, a change in sign, and the nearly saturation behavior under a higher strain. Note that a nonmonotonous behavior is inherent in the wires in which TEP is dominated by the diffusion transport mechanism. This complex behavior is representative of a competition

among the different scattering mechanisms for the carriers and balances between the diffusive transport of electron or hole parts of the Fermi surface that changed during straining. The emergence of a peak on $S(\varepsilon)$ around $\varepsilon \approx 1.1\%$ (Fig. 1, curve 3, 4) is indicative of a change in the Fermi surface topology under strain known as the Electronic Topological Transition (ETT), or the Lifshitz transition.

Analyzing the nonmonotonous behavior of $S(\varepsilon)$ (Fig. 1, curves 3, 4) under a strain higher than the ETT point ($\varepsilon \geq 1.1\%$), we observed a sharp decrease in TEP and the sign change from positive to negative. Taking into consideration some predicted anomalies arising in the phonon spectrum for the materials undergoing an ETT of the Fermi surface [18], we can suppose that the increase in the negative contribution to TEP is attributed to the occurrence of the phonon drag effect of electrons under strain. The phonon drag effect prevails in the TEP, if the phonons acquire a sufficient momentum to scatter the carriers across the Fermi surface changed after the ETT.

To draw more detailed information on how the diffusive and phonon drag terms of TEP vary with strain, we measured the temperature dependences of thermopower $S(T)$ at various values of applied strain. Figure 4 shows the temperature dependences of TEP from the nondeformed state (curve *a*) up to maximal strain (curve *g*).

A quasi-linear $S(T)$ dependence for thermopower in the nondeformed state persistent up to a strain of $\varepsilon \approx 1.1\%$ indicates the diffusive origin of it (Fig. 4, curves 1–3). The temperature dependence of thermopower measured after an ETT in the region of the abrupt drop of $S(\varepsilon)$ develops into a nonlinear dependence. The observed nonlinear dependence with a negative temperature coefficient ($\partial S/\partial T < 0$) indicates the dominance of the phonon drag effect in a narrow temperature range of 5–18 K (Fig. 4, curve 4). In correlation with the predictions made in [18], the phonon drag effect becomes dominant in the thermopower if the phonon momentum is sufficient for interacting of the phonons with the electrons after an ETT.

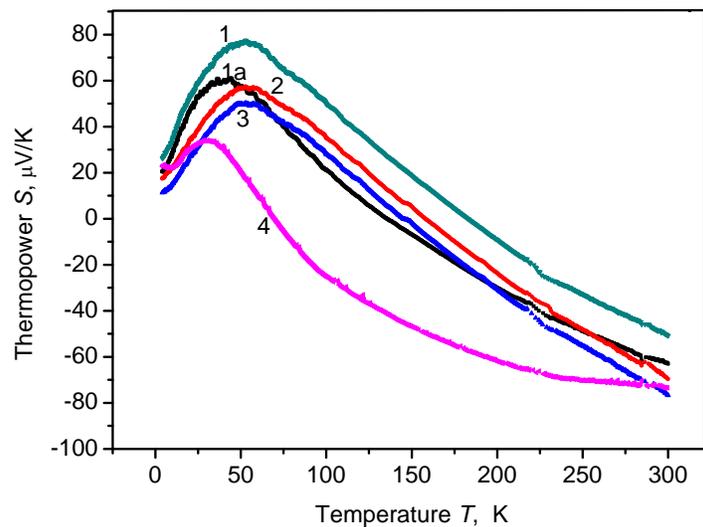


Fig. 2. Temperature dependences of the thermopower for as-prepared and thermally annealed Bi wires of various diameters: (1) 120 nm (as-prepared); (1a) 120 nm (after thermal treatment); (2) 250 nm; (3) 350 nm; and (4) 650 nm.

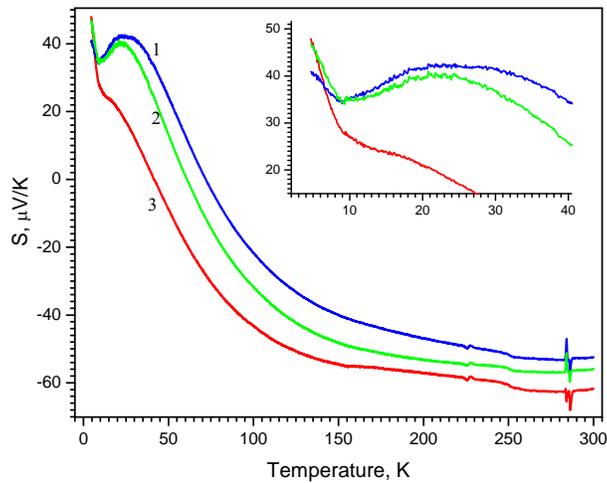


Fig. 3. Temperature dependences of the thermopower for second type of Bi wires when the phonon drag contribution is dominant in the total TEP for various diameters: (1) 0.8, (2) 1.1, and (3) 3.0 μm . Inset (b): expanded version of $S(T)$ dependences in a temperature range of 4.2–20 K.

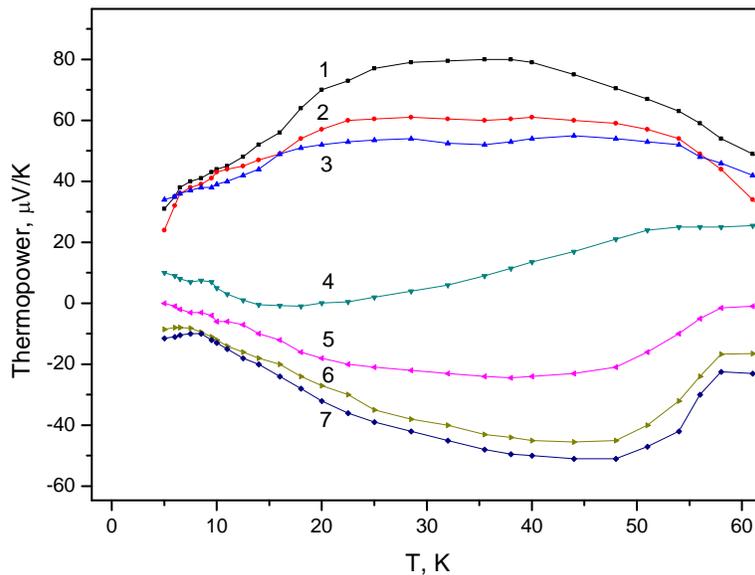


Fig. 4. Temperature dependence of thermopower of the 300-nm Bi wire at various strain values (1) 0, (2) 0.6, (3) 1, (4) 1.6, (5) 1.9, (6) 2.2, and (7) 7 %.

The increase in negative contribution under strain to the total TEP (equation (1)) suggests that a phonon drag term with a negative sign is added to the initial positive diffusion term. The predominance of the phonon drag effect of electrons implies that required condition $q \leq 2k_F$ (q is the phonon momentum, $2k_F$ is the maximum dimension of Fermi surface) for the phonons

interacting with a certain type of charge carriers is satisfied for electrons after an ETT in the region of the abrupt drop of $S(T)$.

Some information on the Fermi momentum p_F of the electrons was provided by the set of curves of the LMR measurements under different strains. In accordance with the galvanic magnetic size effect theory, the extreme p_F value of the electrons in the plane perpendicular to the magnetic field was determined in the nondeformed state and at a strain value of $\varepsilon \approx 1\%$: p_F ($\varepsilon = 0\%$) = 1.1×10^{-21} g cm s⁻¹ and p_F ($\varepsilon \approx 1\%$) = 1.25×10^{-21} g cm s⁻¹. These rough estimates indicate an increase in p_F of heavy electrons, which can contribute to the phonon drag effect.

After the change in the sign of the TEP, a further increase in strain does not have an obvious effect on the $S(\varepsilon)$ dependence under a higher strain (i.e., a nearly saturation behavior above $\varepsilon \geq 2\%$ in curves 3, 4 in Fig. 1); however, it revealed an unusual evolution of the $S(T)$ temperature dependence started after the ETT and shown in curves 4–7 in Fig. 4. At the first glance, the $S(T)$ dependence under a high strain (Fig. 4, curves 5–7) represents a standard behavior of TEP in metals and becomes more metallic as the applied strain increases. However, the occurrence of minima around 40–50 K and the following trend to positive values do not correlate with this assumption. For a bismuth-like semimetal, in which electrons and holes can simultaneously contribute to TEP, it is necessary to take into account how each contribution to the phonon scattering mechanism acts and how they combine under a given strain to yield the sign and the magnitude of the total TEP. If we assume that a modification of all parts of the Fermi surface, both electrons and holes, takes place as a result of an ETT, we do not exclude that, at a certain strain value, one can expect the occurrence of a hole phonon drag effect. A relative enhancement of the hole phonon drag effect can be also caused by a change in the anisotropy of the hole Fermi surface and in the respective ellipsoid of the interacting phonons. To univocally interpret the origin of the temperature dependence of TEP under a high strain, we should extend the measurements of $S(T)$ under strain to a temperature range below 4.2 K for determining the position and magnitude of the phonon drag peak.

3. Conclusions

Thermopower of Bi wire of Bi wires is examined in the light of the strain-induced Electronic Topological Transition in the band structure under uniaxial strain.

At the liquid helium temperature, the diffusive transport mechanism of holes is dominant in the total TEP. However, the hole-dominated transport can be transformed into the electron-dominated transport through smooth manipulations with the phonon spectrum and the Fermi surface by applying strain. The observed trend to negative thermopower values under strain is representative of a slight increase in the electron mobility after an ETT, and the saturation-like behavior of the negative TEP under a high strain is the result of competition between the different scattering mechanisms for the heat carriers and balances between the electron and hole phonon drag contributions.

References

- [1] N.P. Armitage, R. Tediosi, F. Levy, E. Giannini, L. Forro, and D. van der Marel, Phys. Rev. Lett. 104, 237401 (2010).
- [2] T.W. Cornelius, M.E. Toimil Molares, R. Neumann, and S. Karin, J. Appl. Phys. 100, 114307 (2006).
- [3] F. Chen and K.L. Stokes, R. Funahashi, Appl. Phys. Lett. 81, 2379 (2002).

- [4] S.V. Ovsyannikov, V.V. Shchennikov, G.V. Vorontsov, A.Y. Manakov, A.Y. Likhacheva, and V.A. Kulbachinskii, *J. Appl. Phys.* 104, 053713 (2008).
- [5] J.-P. Issi and J.H. Manges, *Phys. Rev. B* 6, 4429 (1972).
- [6] C. Uher and W.P. Pratt Jr., *J. Phys. F: Met. Phys.* 8, 1979 (1978).
- [7] J. Boxus and J.-P. Issi, *J. Phys. C: Solid State Phys.* 10, L397 (1977).
- [8] J. Boxus, C. Uher, J. Heremans, and J.-P. Issi, *Phys. Rev. B* 23, 449 (1981).
- [9] P.P. Bodiul, D.V. Gitsu, V.A. Dolma, M.F. Miglei, and G.G. Zegrea, *Phys. Status Solidi (a)* 53, 87 (1979).
- [10] J. Heremans and C.M. Thrush, *Phys. Rev. B* 59 12579 (1999).
- [11] Y.-M. Lin, O. Rabin, S.B. Cronin, J.Y. Ying, and M.S. Dresselhaus, *Appl. Phys. Lett.* 81, 2403 (2002).
- [12] Yu. P. Gaidukov, *Sov. Phys.-Usp.* 27, 256 (1984) [*Usp. Fiz. Nauk* 142, 5710 (1984)].
- [13] J.P. Heremans, C.M. Thrush, D.T. Morelli, and M.C. Wu, *Phys. Rev. Lett.* 88, 216801 (2002)
- [14] A.D. Grozav and E. Condrea, *J. Phys.: Condens. Matter*, 16, 6507 (2004)
- [15] Y.I. Ravich, in: *CRC Handbook of Thermoelectrics*, edited by D.M. Rowe, CRC Press, New York, 1995, ch. 7, pp. 67–73.
- [16] M. Hagiwara and A. Inoue, *Production Techniques of Alloy Wires by Rapid Solidification in Rapidly Solidified Alloys*, edited by H.H. Liebermann, Dekker, New York, 1993, p.141.
- [17] E. Condrea, A. Nicorici, A. Gilewski, and S. Matyjasik. *J. Low Temp. Phys.* 174, 232 (2014).
- [18] L. Dagens. *J. Phys. F: Met. Phys.* 8, 2093 (1978).