Thermoelectric properties of SiGe whiskers with various morphology

Anatoly Druzhinin[a,b], Ihor Ostrovskii[a,b], Natalia Liakh-Kaguy[a], Tomasz Zyska[c], Azhar Tuleshova[d],
Maksabek Satymbekov[e], Aigul Iskakova[f]
[a]Lviv Polytechnic National University, 12 Bandery Str., 79000, Lviv, Ukraine, [b]International Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw, Poland, [c]Lublin University of Technology, 38a Nadbystrzycka Str. 20-618 Lublin, Poland, [d]al-Farabi Kazakh National University,71 al-Farabi Str, Almaty, Kazakhstan, [e]Institute of Information and Computational Technologies, 125 Pushkin Str., Almaty, Kazakhstan, [f]Kazakh National Research Technical University after K. I. Satpayev, 22 Satpayev Str., Almaty, Kazakhstan,

ABSTRACT

Thermoelectric properties of $Si_{1-x}Ge_x$ (x = 0.03) solid solution whiskers doped with B impurities to the concentrations $10^{17} – 10^{19}$ cm^{-3} were studied in temperature range 300 - 420K. An influence of the whisker morphology, in particular their diameters and obliquity, on Seebeck coefficient, thermal conductivity and resistance was investigated. Seebeck coefficient and resistance was shown to increase, while thermal conductivity to decrease when the whisker diameter drops from 100 to 10μm, that is accompanied by a rise of figure of merit (up to 0,12 at 300K). Use of the whiskers with large obliquity leads to a small rise (of about 10 – 15%) increase of their Seebeck coefficient.

Keywords: SiGe, whiskers, thermoelectric properties, Seebeck coefficient, thermal conductivity generator

1. INTRODUCTION

$Si_{1-x}Ge_x$ solid solution whiskers with low germanium content have maximum ratio of mobility to the phonon thermal conductivity, which is promising for thermoelectrics [1 - 3]. Low values of thermal conductivity occur in nanowires due to presence of size effect [4] connected with phonon boundary scattering channel [5]. Despite of SiGe is good high temperature thermoelectric material, low thermal conductivity allows to approach high values of thermoelectric parameters even at low temperatures. For example, the thermal conductivity of silicon nanowires with diameters less than 30nm and up to 115nm were observed in the works [5] and [4, 6] in the temperature range 20 - 100K and up to 300K, respectively. The authors of work [6] have concluded that quantifying the surface roughness is crucial to studying of the phonon transport mechanisms and the thermoelectric device creation. High ZT values of 2D silicon structure could be obtained at room temperature by optimization of the doping level and effective surface passivation [7].

Use of facile conversion chemistry leads to enhanced thermoelectric performance [8] due to reduction of thermal conductivity. The reducation of thermal conductivity κ of thermoelectric materials is connected with possible their applications, it is also important for sensors of physical values that operate based on thermoelectric effects, because the temperature gradient should be maintained for their flawless operation. Thermal conductivity of $Si_{1-x}Ge_x$ solid solutions is determined by the element composition x [9 - 13], but it is still high (minimum of 12W/(m×K) for bulk $Si_{0.7}Ge_{0.3}$ samples [9]. The dimensional features of samples, even in micron-scale solids with large length-to-diameter ratios, as whiskers, the κ decreases for certain Ge content. Thermal conductivity of SiGe nanowires turns to be 5 to 10 times lower than that of their bulk counterparts, approaching the theoretical amorphous limit [14 – 17]. This is prospective for substantial improvement of thermoelectric figure-of-merit of the nanoscale SiGe and development of high efficiency thermoelectric microconverters [18, 19].

We have studied in our previous works [20 – 22] temperature behavior of Seebeck coefficient of $Si_{1-x}Ge_x$ solid solutions whiskers in low temperature range – from 4,2K to above room temperatures. The results showed a slight difference of the whiskers parameters from bulk materials. However, we have not considered the influence of the whisker geometry and shape on the thermoelectric properties, while it could be crucial as mentioned above [6, 7]. Certain results were obtained by us for Ge and Si whiskers, where it was shown that peculiarities of whisker shape could be successfully used for determination the whisker parameters [23, 24]. But investigations of the thermoelectric were not earlier conducted on solid solution SiGe whiskers.

The aim of present work is study of possible influence of $Si_{1-x}Ge_x$ whisker geometry on their thermoelectric parameters.

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2. METHODOLOGY OF EXPERIMENT

The Si-Ge whiskers were grown by CVD method in closed bromide system [25]. The method provides the growth of the whiskers with unique mechanical parameters that widely used in sensors [26 - 28].

2.1 Measurement of the thermal conductivity

To adequately assess thermoelectric parameters of $Si_{1-x}Ge_x$ whiskers in certain temperature range it is important to know the real value of the coefficient of thermal conductivity κ. As is known, the parameter is highly dependent on the composition, degree of solid solution perfection. Therefore, we have tested 3ω method [29] to determine the temperature dependence of the whisker's thermal conductivity. The method consists in the following. Electric current of a certain frequency is passed through the whisker attached as a bridge on a dielectric thermal conductive substrate. The current heats the center of the crystal. Accordingly, the thermal conductivity flows from the center to the ends along the whisker axis. Heat flux at the air in these conditions can be ignored because it does not exceed 1% of the heat in the dielectric substrate. Solution of continuity equation by imposing certain boundary conditions that limited sample sizes and geometric value of the current flowing through the sample, can be written as follows [29].

$$V_{3\omega} = \frac{\sqrt{2} I_0^{\,3} R R' L}{\pi^4 \kappa S}, \tag{1}$$

where I_0 is the current with frequency ω, $V_{3\omega}$ is a voltage with frequency 3ω, R is the whisker resistance, R' is a change in resistance with temperature, L is the length of the crystal, S is cross-sectional area.

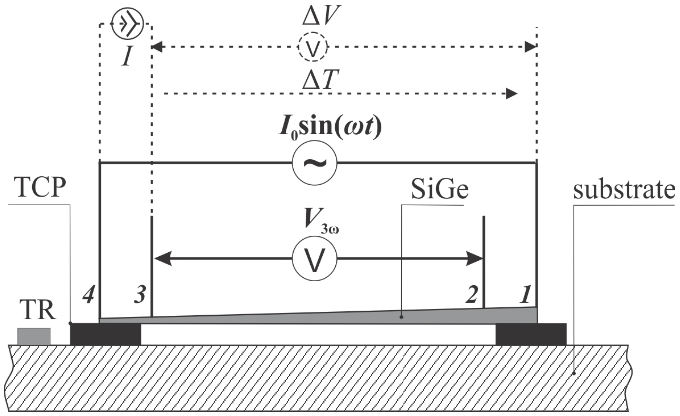

Figure 1. Schematic view of the setting for thermal conductivity measurements by 3ω-method (solid lines) and for measurements of Seebeck coefficient (dashed lines) of SiGe whiskers. In the latter case, the part of whisker is heated by current I, which is passed between two adjacent electrodes (here, 3 and 4), and the thermo-e.m.f. ΔV induced due to temperature difference ΔT is measured between the hot and cold ends (here, 3 and 1, respectively) 30.

Thus, the third harmonics voltage in the tested object inversely depends on its thermal conductivity, and the coefficient κ can be easily derived from the measurements of V3ω signal developed between electrodes 2 and 3 (Fig. 1).

Thus, the voltage measured in the sample at a frequency 3ω is inversely proportional to the thermal conductivity of the crystal. This method was implemented to measure the thermal conductivity of $Si_{1-x}Ge_x$ (x = 0,01 - 0,05) whiskers in the temperature range 300 - 400K [29].

2.2 Measurement of the Seebeck coefficient

The above-described setting has been used also for the measurements of Seebeck coefficient α of oblique p-type $Si_{1-x}Ge_x$ whiskers (x = 0,01 – 0,05) with boron concentrations ranging from 10^{17} cm^{-3} to 10^{20} cm^{-3}. The hot end has been heated by passing a current between two neighboring contacts, which induced the Joule heating of one of the ends, and the temperature was determined from the known *R(T)* dependencies for studied SiGe whiskers, as described in [21]. Since active direct heating of whisker end was applied and a whisker-to-substrate thermal contact area was negligible in comparison with dimensions of virtually infinite substrate, the temperature gradient ΔT along the crystal could be easily maintained, even though the substrate was thermally conductive. To elucidate the effect of obliquity on thermopower of whiskers, the measurements were performed in such a manner that in one case the thickest part (*1 – 2*) was heated and in another – the thinnest one (*3 – 4*), as shown in Fig. 1. Resistance of whiskers was determined by a two-probe method, in which the readings obtained upon passing a current in two directions were averaged, and the resistivity was derived accounting for an effective cross-sectional area of oblique crystals. For measurements of temperature dependencies of thermal conductivity, thermopower and resistance the designed setting was placed inside the resistive furnace, and the temperature of the inset was determined by a thermoresistor attached to the substrate.

2.3 Measurement of a ratio of Seebeck coefficient to thermal conductivity κ/α

One can propose another method for thermoelectric parameters determination. The special conditions of the whisker growth (temperature regimes, oversaturation in gas phase, etc.) lead to creation of various aggregates with cross-like and X-like shape. In the paper we propose to use the whisker structures for estimation of certain thermoelectric parameters.

First of all, one can determine a type of conductance of semiconductor. For this purpose I-U curve of such cross-like aggregate should be measured. A growth of two whiskers with asymmetrical cruciform shape can be used. Applying a current to the longitudinal and measuring the voltage on the tansverse shoulders of the growth one can obtain C-type I-U characteristics (see Fig. 2).

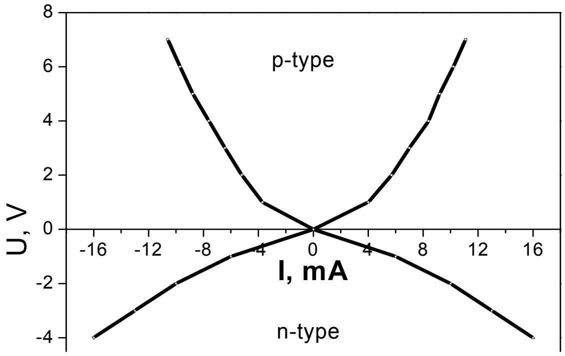

Figure 2. I-U characteristics of Si-Ge whisker of p-type and n-type conductance.

Interestingly, the potentials measured at certain distances from node growths were positive in materials of p-type, or negative in n-type materials. Thus, the resulting I-U characteristics of cross-shaped growths can be used to determine the conductivity type of the whisker material.

Based on the studies it was developed method of determining the thermoelectric parameters of whiskers, which allows us to define other thermoelectric parameters of the crystals, in particular a ratio of Seebeck coefficient to thermal conductivity κ/α.

According to the proposed method, current is passed through a heating branch, namely two adjacent cross-shaped contacts of growth; two others contacts *1* and *2* serve as a measuring branch. The main measurable parameters were:

1) the potential difference U_1 and U_2 between the ends of measuring branch and the node of growth,

2) the impedance of measuring branch R_3,

3) the current through measuring branch I_3.

Therefore, warming up the node of growth by current I_3 creates two heat flows from the growth middle to points *1* and *2* of the measuring branches that could be recorded as

$$W_1 = \frac{\kappa S}{l_1}\Delta T_1 \; W_2 = \frac{\kappa S}{l_2}\Delta T_2$$

(S - sectional area of the growth, l_1 and l_2 are the length of measuring branch, respectively).

Taking into account that temperature gradients ΔT_1 and ΔT_2 can be expressed due to value of thermopower $U_1=\alpha\Delta T_1$ and $U_2=\alpha\Delta T_2$, and the difference in heat flow creates between points *1* and *2* an electrical power, we get the equation

$$I_3{}^2 R_3 = \frac{\kappa S}{\alpha l_1}(nU_2 - U_1), \tag{2}$$

where n = l_1/l_2.

The equation (2) allows us to determine the ratio of κ/α. The calculation were firstly successfully checked for Si whiskers. The results are presented in the Table 1.

Table 1. Thermoelectric parameters of cross-like Si whisker growths

I, mA Heating current	U_1, mV voltage diference 0-1	U_2, mV voltage diference 0-2	R_3, Ohm resistance of measuring branch	I_3, μA heating current	κ/α, A/cm	α/κ, cm/A
50	49.0	33.8	224	9.6	4.2	0.24
60	73.5	57.5	242	11.6	4.05	0.26

The data of Table 1 shows that obtained value of κ/α consists of 4.1. The value was compared with data of [31] for bulk silicon at room temperature, which was equal to 4.6. Therefore, the results evidences that good agreement of our estimations with literature data was obtained.

3. EXPERIMENTAL RESULTS AND DISCUSSION

Seebeck coefficient was investigated for $Si_{1-x}Ge_x$ (x = 0,03) whiskers doped with B impurity to concentration ranging from 10^{16} cm^{-3} to 10^{20} cm^{-3}. Seebeck coefficient values changes from 80 to 450μV/K at room temperature dependent on the impurity concentration. Maximum value of α is found for whiskers with least carrier concentration. In the whiskers doped to the concentration 10^{16} cm^{-3} Seebeck coefficient increases from 440 to 500μV/K in the temperature range 300 - 390K. Seebeck coefficient of samples with concentration 10^{17} cm^{-3} consists of about 400μV/K at room temperature. When temperature increases weak enlargement of α is observed. Minimum α value is found for the whiskers with holes concentration 10^{20} cm^{-3}. At 300K α = 90μV/K.

The above methodological aspects allows us to estimate κ and α for $Si_{1-x}Ge_x$ (x = 0,03) whiskers in the temperature range 280 - 400K. Firstly we presented the temperature dependence of Seebeck coefficient for the whiskers with 10^{17} cm^{-3} (see Fig. 1). Then taking into account the measured parameters of the whiskers growth we have obtained the following temperature dependences of coefficient of thermal conductivity (see Fig. 1).

As it is obvious from Fig.1, the samples doped with B impurity show temperature rise of Seebeck coefficient in the temperature range 300 – 400K, slop and magnitude of α being dependent on impurity concentration. Atoms of B are known to create in $Si_{1-x}Ge_x$ (x = 0,03) forbidden gap shallow donor levels with ionization energy 0,042eV. At room temperature these levels are completely ionizated. That is why character of $\alpha(T)$ dependencies in the measurable temperature range is corresponded to weak change of Fermi level position in the whiskers at temperature rise.

The obtained data of the whisker thermal conductivity (Fig. 3) are 3 times smaller than the correspondent parameters for bulk silicon. It is unexpectedly large change due to rather small composition of solid solution (x = 0,03). So, the obtained data should be checked by other direct measurement, in particular by 3ω method.

The observed rather small values of the whisker thermal conductivity are likely connected with peculiarities of the whisker geometry. To investigate the possible influence of the whisker shape we have obtained a size dependence of $Si_{1-x}Ge_x$ whisker Seebeck coefficient as well as its dependence on the whisker obliquity.

The dimensional dependency of Seebeck coefficient for SiGe whiskers was measured for whiskers with diameters ranging from 20 to 100μm.

As it is obvious from Fig. 4, Seebeck coefficient of the whiskers rises at the decrease of their diameters. We have investigated the dimensional dependencies of the whisker resistance. The dependency shows that resistance rises at the whisker diameter decrease (Fig. 4).

As follows from our previous consideration enlargement of α is observed in the whiskers with a decrease of their impurity concentration (increase of their resistance). Thus, at the decrease of the whisker diameters their impurity content decreases and correspondingly their Seebeck coefficient rises as shown in Fig. 4.

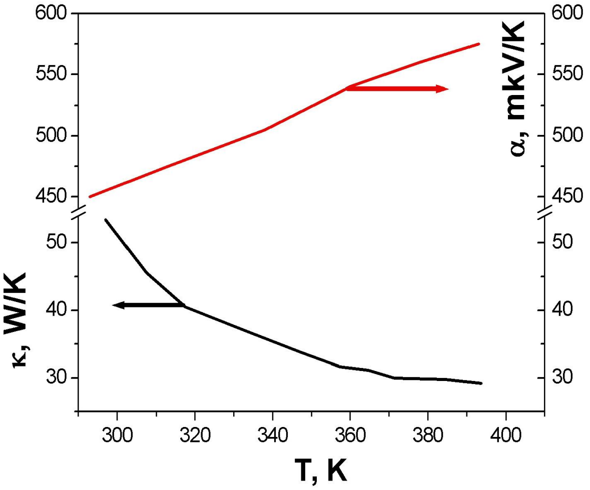

Figure 3. Seebeck coefficient and thermal conductivity versus temperature dependency for Si1-xGex (x = 0,03) whiskers with p = 1017cm-3.

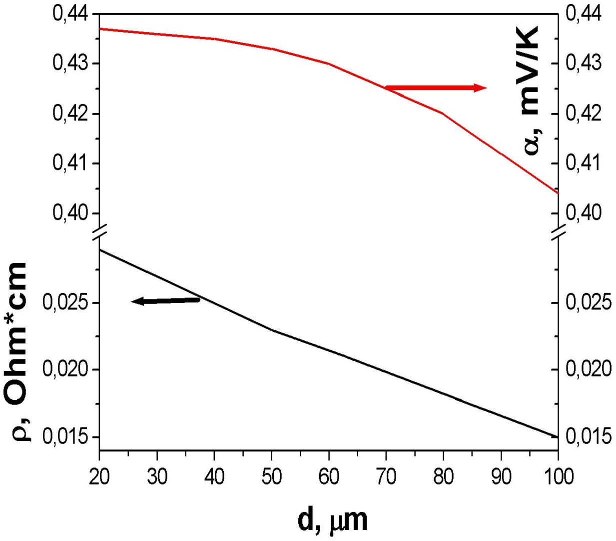

Figure 4. Dimensional dependence of Seebeck coefficient and resistivity of Si1-xGex (x = 0,03) whiskers (T = 300K, p = 1017cm-3).

One can obtain the similar dimensional effect when the samples with various obliquities are used. So we measured the Seebeck coefficient for the whiskers with different diameters of their ends. Heating of thick end of the whiskers gives substantially greater Seebeck coefficient as compared with the coefficient at heating of thin end of the whiskers. The obtained results of Seebeck coefficient change at heating of different ends of the whiskers (ΔT = const = 80°C in the temperature range 20 - 100°C) are shown in Table 2.

As a rule, at heating of the thicker end of the whisker an obliquity impact (positive sign of $\Delta U/U_c$ parameter) was more significant. Dependence of the thermopower on the obliquity value K for different whisker sets could not observable - decisive factor plays a degree of the whiskers doping level.

Thus Table 2 shows that the greatest impact on the relative change of thermopower should not caused by relative size of crystals but their resistivity. In fact the biggest changes of thermopower (up to 15%) were found in crystals with the highest resistivity of 0,06Ωcm, while the lowest one are their characteristic obliqueness of $1{,}50\cdot10^{-3}$.

Instead, the sample with the greatest obliquity of $2{,}98\cdot10^{-3}$, but with the lowest resistivity of 0,002Ωcm have rather small relative change in thermopower – only 5%.

Table 2. Seebeck coefficient change (ΔT = 80°C) in Si1-xGex (x = 0,03) whiskers of various resistance and obliquity

No	ρ, Ωcm	Δl, μm	Δd, μm	$K=\Delta d/2\Delta l$	$\Delta U/Uc \times 100\%$
1	0,06	13400	40	$1{,}50\cdot10^{-3}$	14,1
2	0,03	14500	40	$1{,}39\cdot10^{-3}$	11,3
3	0,03	6500	22	$1{,}74\cdot10^{-3}$	9,6
4	0,002	5600	31	$2{,}81\cdot10^{-3}$	5,1
5	0,002	4850	29	$2{,}98\cdot10^{-3}$	4,6
6	0,008	12200	45	$1{,}85\cdot10^{-3}$	4,6

For further analyze of the impact of size effects one can performe theoretical calculations of heat flow components W, that could be presented as superposition of the following contributions: W_1 is a flow through the whisker, W_2 is a flow in the substrate, W_3 is a convective flow in the air:

$$W = \frac{\kappa_1 d_1 + \kappa_2 d_2}{l} b_1 \Delta T + \alpha_T b_2 l (T_{ef} - T_o), \tag{3}$$

where κ_1 is coefficient of $Si_{1-x}Ge_x$ thermal conductivity, κ_2 is coefficient of thermal conductivity of substrate (0,233W/mK) [8], αT is convective coefficient of heat transfer (100W/m^{2}K) [20], To is ambient temperature (20 °C), T_{ef} is an effective temperature of the whisker surface (60°C), d_1 – is an effective whisker diameter (~ √S), b_1 is an effective width of the whisker b_2 facet ($b_1 = 2{,}6a$, where a is the width of the whisker facet, $b_2 = 5a$), l is the whisker length, d_2 is substrate thickness ($2{,}10^{-3}$m)

One can neglect by third component associated with thermal radiation. According to the Stefan-Boltzmann effect its average value amounted to ~ $3{,}10^{-9}$W, which is rather small value as compare with other contributions. The results of calculation are presented in Fig. 5.

As you can see, heat flux W_2/W, called by heat conductivity in the substrate, is almost independent on the whisker width. The other components are changed as follows. The heat flux due to the whisker thermal conductivity W1/W substantially drops when the whisker width decrease from 100 to 10μm. Convective heat flux W_3/W has inverse dimensional dependence - it decreases exponentially with the increase in the whisker size.

Thus, our calculations show that at decrease of the whisker diameter from 100 to 10μm the total heat flow should decrease at 50% due to strong drop (in 3,5 times) of W_1/W and an increase (in 2,3 times) of W_3/W components.

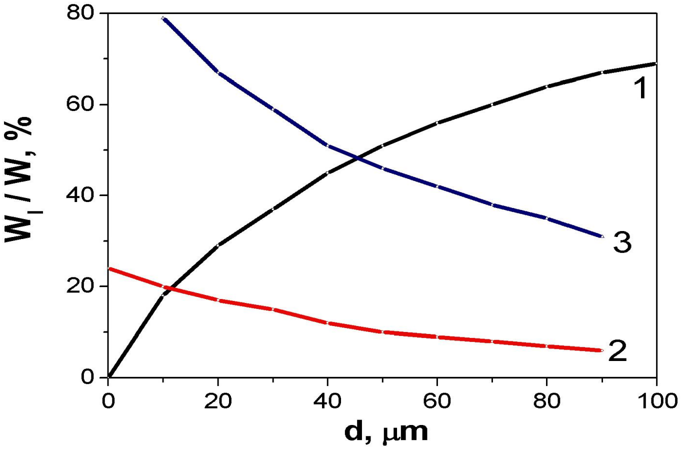

Figure 5. Dependencies of heat flow components versus the width of the whisker: 1 - W1/W is the heat flux due to whisker thermal conductivity, 2 - W2/W is a part of the heat flux due to heat conductivity in the substrate (the magnitude is x5), 3 - W3/W is the heat flux due to convective flow in air.

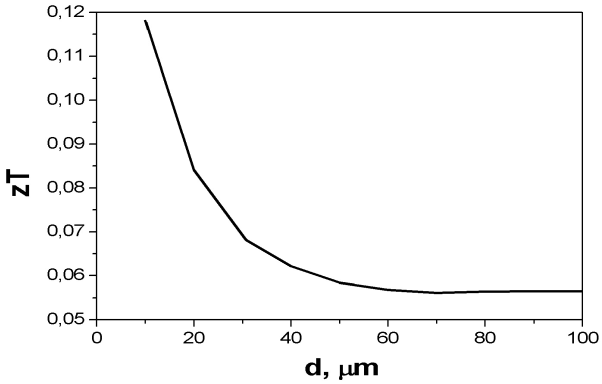

Figure 6. ZT parameter versus diameter in Si1-xGex (x = 0,03) whiskers (T = 300K, n = 1019 cm-3).

Taking into account the obtained parameters one can calculate ZT value for the whiskers. The dimensional dependence of ZT for $Si_{1-x}Ge_x$ (x = 0,03) whiskers with hole concentration $p = 1\cdot10^{19}$ cm^{-3} at T = 300K is presented at Fig.6.
As you can see from Fig. 6, a significant rise of ZT occurs at decrease of the whisker diameter down 20μm. This fact is promising for further increase of the whisker figure of merit when diameter becomes of about nanometer scale.

4. CONCLUSIONS

The manuscript presents a study of influence of $Si_{1-x}Ge_x$ (x = 0,03) whisker geometry on their thermoelectric parameters. The whiskers have the diameters ranged from 10 to 100 μm, besides the whiskers with obligaty $\Delta d/2\Delta l$ of about $(1{,}5\text{-}3{,}0)\cdot10^{-3}$ have been investigated. The obtained results have shown that the whisker resistance and Seebeck coefficient increases when their diameters drops. The calculation of the distribution component of the heat flux along the whiskers have shown that thermal conductivity decreases with the diameter drop. The obtained results allow us to calculate a figure of merit, that for thin (of about 10μm) whiskers with high boron concentration $1\cdot10^{19}$ cm^{-3} approaches to 0,12 at room temperature. The value is rather high for Si-Ge solid solution: on one side, one can approach ZT almost to 1,0 in the high temperature range (900 - 100K); on another side, one can suppose substantial growth of the whisker figure of merit using the nanoscale whisker diameters.

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