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Thermoelectric Properties of InSb Nanowires Over a Wide Temperature Range

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The thermoelectric power and electrical conductance of bundles of indium antimonide nanowires with a diameter of about 5 nm have been measured over the temperature range of 80 K to 400 K. In the range from 80 K to 300 K, the temperature dependence of the conductance of nanowires is close to power law, while the thermopower increases linearly with temperature. The thermoelectric properties of the nanowires are discussed in terms of the Luttinger liquid theory, taking into account enhancement of the electron–electron interaction in one-dimensional conductors.

Key words: InSb, nanowire, thermoelectric properties, Luttinger liquid

INTRODUCTION

Low-dimensional structures are attracting growing attention due in particular to possible enhancement of their thermoelectric figure of merit compared with bulk materials. Among these structures, electronic properties of quasi-one-dimensional wires are of special interest because the enhanced electron–electron interaction results in the formation of the unusual state of the electronic subsystem called a Luttinger liquid.[1] The characteristic features of the transport properties of a Luttinger liquid were revealed in experimental results on electrical conductance and current–voltage characteristics of InSb,[2–4] $Bi_{1-x}Sb_x$,[5] $NbSe_3$,[6] and MoSe nanowires[7]; polymer nanofibers[8]; carbon nanotubes[9–11]; and fractional quantum Hall edge states.[12] Luttinger liquid behavior of the thermoelectric power was revealed in InSb nanowires[3,4] and carbon nanotubes.[11]

The important feature of the Luttinger liquid is a strong increase of power factor $S^2\sigma$ (where S is the thermopower and σ is the electrical conductivity) with increasing temperature, which is favorable for thermoelectric applications. However, the majority of measurements have been carried out at temperatures below 300 K. In this work, we measured the thermopower and electrical conductance of bundles of InSb nanowires embedded into an asbestos matrix over the temperature range of 80 K to 400 K. It has been shown that the thermoelectric power factor of the nanowires increases monotonically with temperature in this range.

RESULTS

The method of nanowire preparation is described elsewhere.[2,13] We recall only that the natural chrysotile asbestos, used for nanowire preparation, consists of long nanotubes with internal diameters depending on the origin of the mineral. We used asbestos with a channel diameter of about 5 nm. The channels in the asbestos matrix were filled with molten InSb under a pressure of ~15 kbar. The samples have a typical cross section of ~0.01 mm^2 and a length of ~1 mm along the nanowire direction.

The electrical resistance was measured by the two-probe method. In this method, the probe–sample contact resistance is included in the measured result. However, our samples consist of very long nanowires, and each nanowire, according to the estimation of Ref. 2, contains about 100 to 1000 strong defects (tunnel barriers). It seems likely that two additional barriers between the nanowire and

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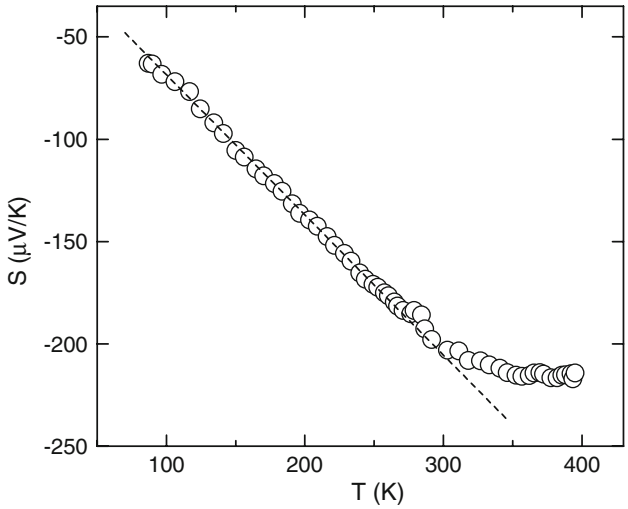

Fig. 1. The temperature dependence of the thermoelectric power of a bundle of InSb nanowires. The dashed line is the linear function $S \propto T$.

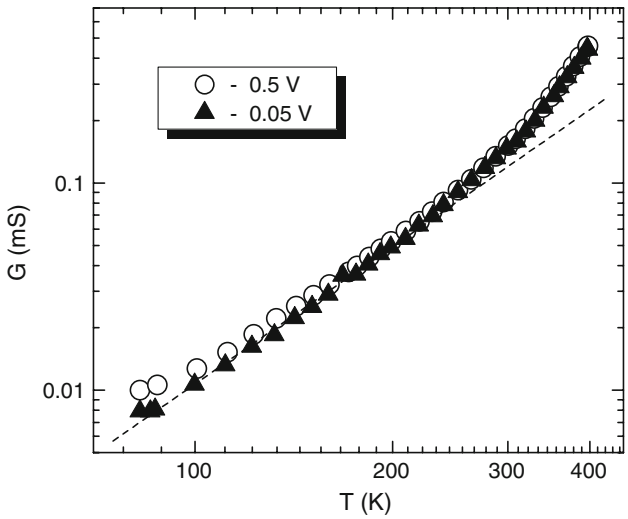

Fig. 2. The temperature dependence of the differential conductance of a bundle of InSb nanowires plotted for two applied voltages on a double-logarithmic scale. The dashed line is the power-law function $G \propto T^{2.2}$.

the bulk leads should not change considerably the total resistance of the structure. The results of the current–voltage characteristic measurements, as will be discussed later, give additional support in favor of the relative smallness of the contact resistance in our samples.

The temperature dependence of the thermoelectric power S and the differential electrical conductance G of a bundle of InSb nanowires in an asbestos matrix are presented in Figs. 1 and 2, respectively. The thermopower is a linear function of temperature up to 300 K, however it saturates at higher temperatures. The conductance increases rapidly with increasing temperature. The dependence presented in Fig. 2 is close to a power-law function of temperature in the range of ~80 K to 300 K (see also Ref. 2). At $T > 300$ K, the rate of the conductance temperature variation increases; therefore the power factor of nanowires rapidly increases with temperature in spite of saturation of the thermopower. The crossover temperature can differ from 300 K by several tens of degrees in different samples.

DISCUSSION

The obtained results are difficult to interpret rigorously. However, there is no doubt that Coulomb interaction has a pronounced effect on electron transport in one-dimensional conductors.[1,14] The simplest version of the Luttinger liquid theory, taking into account electron–electron interaction in a one-dimensional conductor, describes spinless electrons, which are scattered by a single potential barrier. According to the theory, in the limiting case $k_B T \gg eV_1$, the electrical current $I \propto V_1 T^{\alpha}$ and the conductance $G \propto T^{\alpha}$ (see Refs. 9, 10, 14). Here V_1 is the voltage drop across the single potential barrier, T is the temperature, $-e$ is the electron charge, k_B is the Boltzmann constant, and the exponent α (>0) depends on the strength of the electron–electron interaction. The value of V_1 coincides with the total voltage drop across a nanowire V, if there is only one potential barrier (we do not take into account the contact resistances). If there are N identical independent barriers, $V = NV_1$. In this limit the current–voltage characteristics are linear and the conductance is described by a power-law function of temperature. In the opposite case, $eV_1 \gg k_B T$, the current $I \propto V_1 |V_1|^{\beta}$ and the differential conductance $G \propto |V_1|^{\beta}$ (see Refs. 9, 10, 14), where $\beta \equiv \alpha$. In other words, in this limit, the current–voltage characteristics are nonlinear, while the conductance is independent of temperature.

These relations describe electron transport in nanowires with a small number of independent defects (impurities, constrictions, etc.). In the opposite case of a large density of defects, the nanowire can be modeled as a chain of quantum dots divided by potential barriers.[15] In this model, the electrical current is described by power-law dependences $I(V,T)$, which are similar to those mentioned above. However, the exponents α and β depend on the degree of disorder and satisfy the condition $\alpha \gg \beta$. This approach takes into account the electron–electron interaction in the framework of Coulomb blockade theory.

Much less is known about realistic one-dimensional systems, which require closer inspection of both the interaction and the disorder. It is hoped that the dependences $I(V,T)$, describing realistic one-dimensional conductors, will also be close to power-law functions at small bias voltages or temperatures. However, the exponents α and β should depend on both the interaction and the disorder.[16]

In the framework of Luttinger liquid theory, the thermoelectric power is a linear function of

temperature.[17–19] Note that, according to this theory, the thermopower of nanowires can be enhanced by the electron–electron interaction in comparison with that calculated in terms of the Fermi-gas model.

The temperature dependence of the thermoelectric power presented in Fig. 1 agrees with the theoretical prediction up to room temperature. Moreover, the thermopower of 5-nm nanowires under study exceeds the thermopower measured in the ~40-nm InSb nanowire[4] by about seven times. It is known that the electron–electron interaction (and the exponent α) increases with decreasing nanowire diameter.[7] Therefore, the higher thermopower of our nanowires compared with that measured in Ref. 4 can arise because of larger electron–electron interaction.

The temperature dependence of the electrical conductance shown in Fig. 2 can be described by the simplest Luttinger liquid theory at temperatures up to about 300 K. This part of the curve is well fitted by the power-law function $G \propto T^{2.2}$, i.e., the exponent $\alpha = 2.2$ for this sample. At temperatures of ~80 K, a small deviation of the upper curve from power-law behavior appears, likely because of violation of the inequality $k_B T \gg eV_1$.

A physical reason for the saturation of the $S(T)$ dependence and for the increase of the slope of the $G(T)$ dependence at temperatures above 300 K (Figs. 1 and 2, respectively) is not clear at the moment. One possible explanation of these peculiarities is a violation of the condition $k_B T \ll E_F$ (where E_F is the Fermi energy), which determines the region of validity of the low-energy Luttinger-liquid theory. The studied nanowires were not intentionally doped, therefore their Fermi energy should be relatively small. Unfortunately, it is impossible to analyze the obtained results at temperatures above 300 K because there is no theory describing high-energy excitations in one-dimensional conductors with strong electron–electron interaction. The indicated features of the dependences $S(T)$ and $G(T)$ can be caused also by the rise of the intrinsic electrical conductivity of semiconductor nanowires at high temperatures. Finally, these peculiarities may arise because of filling of the second subband of the size-quantized electron spectrum at high temperatures.[4] The additional conducting channels can increase the electrical conductance and change the temperature dependence of the thermopower of nanowires.

The distinctive feature of a Luttinger liquid is a power-law behavior of the current–voltage characteristics in the region where $eV_1 \gg k_B T$. A strong nonlinearity of these characteristics of InSb nanowires was observed[2] near the liquid-helium temperature, even at relatively small bias voltage (~0.1 V). In the temperature range of 80 K to 400 K, for most of our InSb samples, the measured current–voltage dependences were linear at a bias voltage less than ~1 V. A small nonlinearity was

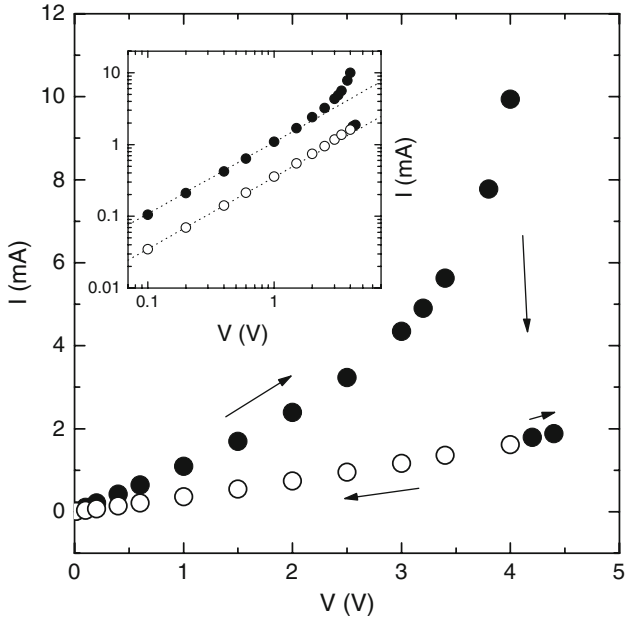

Fig. 3. The room-temperature current–voltage characteristic of a bundle of InSb nanowires with increasing (solid circles) and decreasing (open circles) bias voltage. The arrows indicate the consecutive order in which the experimental values were measured. The inset shows the dependence plotted on a double-logarithmic scale. The dashed lines are linear functions $I \propto V$.

detected only at greater biases but the attempts to increase considerably the applied voltage led to irreversible jumps of the sample resistance. However, some samples with low resistance (i.e., the samples with a small number of defects) revealed a pronounced nonlinearity of current–voltage characteristics at a relatively small bias because the average voltage drop across one defect V_1 was greater in this case.

In Fig. 3, the current–voltage characteristic of such a low-resistance sample is shown (upper curve). One can see that the characteristic is strongly nonlinear in the range from 2 V to 4 V. Note that the exponent $\beta \sim 2$ to 3 at a bias voltage ~4 V, i.e., in accordance with Luttinger liquid theory, it is close to the value of the exponent α of the dependence $G(T)$ presented in Fig. 2. With a further increase of the voltage, the sample resistance rose sharply at $V \approx 4$ V. It seems likely that most low-resistance nanowires were destroyed because of the electromigration of impurities or melting by Joule heat. The average density of defects in the remaining nanowires increased. As a result, the condition $eV_1 \gg k_B T$ was violated and the subsequent reduction of bias revealed the almost linear current–voltage characteristic (lower curve in Fig. 3).

These data suggest that the contact resistance between current leads and the sample is small. Otherwise, the applied voltage would drop mainly across two potential barriers between a nanowire and the leads. In this case, $V_1 \approx V/2$ and the room-temperature current–voltage characteristics would

be nonlinear already at $V > 0.05$ V. This conclusion is obviously in conflict with the experimental results presented in Fig. 3. It follows that the applied voltage is distributed mainly between intrinsic defects in the nanowire and the contribution of the contact resistance is small.

CONCLUSIONS

We have shown that the electrical conductance and the absolute value of the thermopower of InSb nanowires increase monotonically with increasing temperature in the range from 80 K to 300 K. In this region, the thermopower varies almost linearly with temperature. The temperature dependence of the conductance is close to power law with an exponent $\alpha > 2$. The current–voltage characteristics of the nanowires are nonlinear in general case. The nonlinearity depends on the applied voltage and the defect structure of the nanowires. The analysis of the experimental results indicates that many features of transport properties of InSb nanowires can be, at least qualitatively, described by Luttinger liquid theory. This theory, in particular, predicts enhancement of the thermopower due to electron–electron interaction and our results confirm this prediction. The simple Luttinger liquid model is inapplicable to InSb nanowires at temperatures above about 300 K. The thermoelectric power factor of the studied structures rapidly increases with increasing temperature in the range from 80 K to 400 K.

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