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Electric field effect on thermoelectric properties of bismuth telluride square nanowires

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We theoretically studied the influence of the applied electric filed on thermoelectric properties of intrinsic bismuth telluride nanowires, which are grown along [110] direction. The electronic structure and wave functions were calculated by solving the self-consistent system of the Schrődinger and Poisson equations by means of both the linearization of Poisson's equation and the spectral element method. The thermoelectric parameters were calculated using a constant relaxation-time approximation. The external electric field can increase the Seebeck coefficient of a 7- and 15-nm-thick square nanowire by nearly a factor of two, and enhance the figure of merit by an order of magnitude.

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1. Introduction

Bismuth telluride and its solid solutions ($Bi_{2-x}Sb_xTe_3$, $Bi_2Te_{3-y}Se$) are one of the best known commercial thermoelectric materials. At room temperature, these materials possess notable properties like a high anisotropic multi-valley Fermi surface, a small value of the thermal conductivity, and an optimal value of a carrier concentration. Since the electron dimensional confinement and the enhancement of the photon boundary scattering control the electron transport, bismuth telluride based superlattices and quantum wires have advanced thermoelectric properties when compared to bulk bismuth telluride. For instance, the figure of merit of the Bi_2Te_3/Sb_2Te_3 superlattices achieves its maximum value of 2.4 at room temperature [1]. Due to a larger confinement effect, for thermoelectric applications the bismuth telluride nanowires (NW) are better than the superlattices. The bismuth telluride NW can be obtained by electrochemical deposition of the material in the nanopores of anodized alumina membranes, by Taylor-Ulitovsky technique, and by high pressure injection of the melt into capillaries [2,3].

Electric field effect (EFE) is a powerful tool to control electrical properties of low dimensional structures. Experimental and theoretical studies showed that the EFE can significantly improve thermoelectric properties of the Bi nanowires and PbTe films [4-6]. While dimensional confinement of electrons leads to a modification of their density of states, the Fermi level can be changed due to the EFE control of the electron (hole) concentration in the nanowire. The EFE on the nanowire thermoelectric properties is also a result of the energy spectrum modification and the local dependence of the electron distribution function. Also, the applied electric field causes additional quantum confinement. The EFE on the nanowire thermoelectric properties is also due to the energy spectrum modification because of the additional quantum confinement caused by the applied electric field and the local dependence of the electron distribution function. These factors, affected by the side gate potential, lead to an improvement of the nanowire thermoelectric properties.

In this paper we theoretically studied how the electric field effect influences the thermoelectric properties of the intrinsic square bismuth telluride NWs with thicknesses 7 and 15 nm at room temperature. The electron mobility in the NWs is supposed to coincide with the electron mobility in the bismuth telluride bulk material. We used the carrier effective mass components calculated previously for the NW with growth direction of [110] [7]. The transport parameters were estimated using the semi-classical Boltzman approximation [8]. We used a constant-relaxation-time approximation to calculate the transport parameters. The justification of applying this approach to the calculation of the transport properties of bismuth telluride NW material is given in our previous paper [7].

2. Mathematical model

We considered the Bi_2Te_3 nanowire connected to the source and the drain and separated from the lateral metallic gate by a dielectric SiO2 layer with a thickness dSiO2=300 nm. A similar device based on the Bi nanowire with a thickness of 28 nm has been experimentally studied recently [6]. The following derivations were applied. To estimate the nanowire transport properties we found a solution of a self-consistent system of Schrődinger and Poisson equations

$$-\frac{\hbar^2}{2m_x}\frac{d^2\Psi}{dx^2}-\frac{\hbar^2}{2m_z}\frac{d^2\Psi}{dz^2}+\left[V_c(x,z)+E_c-e\varphi(x,z)\right]\Psi(x,z)=E\Psi(x,z) \quad (1)$$

$$-\varepsilon\varepsilon_0\Delta\varphi(x,z)=\rho(x,z). \quad (2)$$

where E_c is the bottom of the conduction band, φ is the electrostatic potential, and V_c is the confinement potential which corresponds to an infinitely high cylindrical quantum well. After replacing the sign in front of the second derivatives in equation 1 by an opposite one, we obtained the Schrodinger equation for holes. The applied electric field was assumed to be directed along the z axis. We did not take into considerations the local exchange-correlation energy because the electron (hole) density in the intrinsic semiconductor nanowires is supposed to be small. Integrating the both sides of the Poisson equation, we obtained the charge neutrality equation for the NW in the form [9]

$$n_{1D}(E_F)=p_{1D}(E_F)+a_x\sigma \;. \quad (3)$$

Here the surface electric charge $-e\sigma$, induced by the gate electric field in the interface SiO_2/Bi_2Te_3, is associated with a Fermi-level-pinning boundary condition. Condition (3) means that the number of electrons and holes populating the subbands is equal to the number of charge carriers occupying surface states. The surface states do not contribute to the transport because of the strong surface roughness scattering. The applied electric field induces a surface charge on the SiO2/Bi2Te3 interface. The effective (induced) surface charge doping concentration σ can be estimated in the gate–SiO_2–Bi_2Te_3 capacitance structure as a function of the gate voltage V_g [6]

$$\sigma=\frac{\varepsilon_0\varepsilon_{SiO2}}{ed_{SiO2}}V_g\;. \quad (4)$$

Where ε_{SiO2}=3.9 is the permittivity of the SiO_2 layer. To solve the system of equations 1 and 2 we use both the Linearized Poisson Equation (LPE) approach and the Spectral Element Method (SEM) [9-11]. In the LPE approach, the mathematical definitions of the Seebeck coefficient, the electrical and thermal coefficients, and the figure of merit coincide with those given for the nanowires in the absence of the gate voltage [7]. In the framework of the SEM, the local dependence of the transport parameters is defined by the electron (hole) wave function [9]. For example, using the notations from our previous paper [6], the electron concentration can be written as

$$n_{1D}(x,z;E_F)=N_{c,v}^{1D}\sum_{n=1}^{n_{\max}}\sum_{l=1}^{l_{\max}(n)}\Phi_{-\frac{1}{2}}(\eta_{n,l})\left|\Psi(x,z)\right|^2 \quad (5)$$

Here, factor $N_{c,v}^{1D}=\left(2m_y^{e,h}k_BT/\pi\hbar^2\right)^{\frac{1}{2}}$ denotes the effective density of electron (hole) states. The thermoelectric parameters are effectively macroscopic parameters. Hence, with a side gate electric field, the figure of merit ZT and the Seebeck coefficient S are defined by averaging as [5]

$$ZT=\frac{\langle\sigma(z)S(z)\rangle^2}{\langle\sigma(z)\rangle\langle\kappa(z)\rangle}T\;, \quad (6)$$

$$S=\frac{\langle\sigma(z)S(z)\rangle}{\langle\sigma(z)\rangle}. \quad (7)$$

3. Gate voltage dependence of the thermoelectric parameters

Fig. 1 shows the dependence of the electron and hole concentrations in the nanowire on the gate voltage at room temperature. The results obtained using the LPE approach and those obtained using the SEM agree well. At the zero gate voltage, the charge carrier concentrations in the bismuth telluride nanowires are $n=p=3.11\times10^{-16}$ cm^{-3} and $n=p=3.03\times10^{-17}$ cm^{-3} for the thicknesses of 7 and 15 nm, respectively. Under an applied electric field, the carrier concentration increases by several orders of magnitude. At the positive (negative) gate voltage the electrons (holes) dominate. Hence, it is possible to manage the type of NW conductivity by applying the gate voltage. This fact was experimentally confirmed for the Bi nanowires [6].

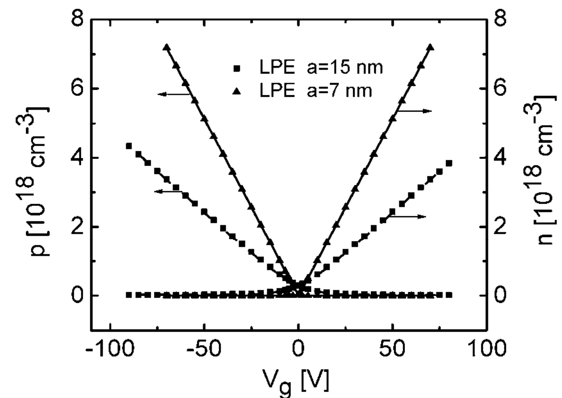

Fig. 1. The gate voltage dependence of the electron (n) and hole (p) concentrations of the Bi_2Te_3 NWs with the thicknesses a=7 nm (dashed line) and 15 nm (solid line) at room temperature.

The gate voltage dependence of the carrier concentration increases with a decrease of the NW thickness. For the NW thickness of 15 nm, the dependence of the electron concentration on the gate voltage is less pronounced compared to that for holes. This is, because the splitting between the energy subbands decreases for nanowires with large thicknesses due to a lesser confinement effect. Thus the number of subbands, involved in the transport, decreases with a decrease of the NW thickness. Since the hole effective mass is greater than the electron effective mass, the hole concentration increases faster with the gate voltage compared to the electron concentration.

Figure 2 shows the dependence of the NW Seebeck coefficient on the gate voltage. For the NW with the thickness of 15 nm, a small disagreement between the LPE and SEM data appears at the large voltage, owing to the great number of the subbands involved in the transport. At the zero gate voltage, the Seebeck coefficient is equal to -167 and -252 μV/K for the NW thicknesses of 15 and 7 nm correspondingly. For the NW thickness of 7 nm, the maximum absolute value of the Seebeck coefficient approaches the value of 478 μV/K (494 μV/K) at the gate voltage equal to -3 V (2 V). For the thickness of 15 nm, the maximum absolute value of 333 μV/K (344 μV/K) is achieved at the greater gate voltage equal to -29 V (15 V). Since the electron mobility in the bismuth telluride is equal to 1200 cm^2/Vs and it is more than twice larger than the hole mobility, which is equal to 510 cm^2/Vs [7], the maximum absolute values of the Seebeck coefficient corresponding to the opposite gate polarities are achieved at the different gate voltages.

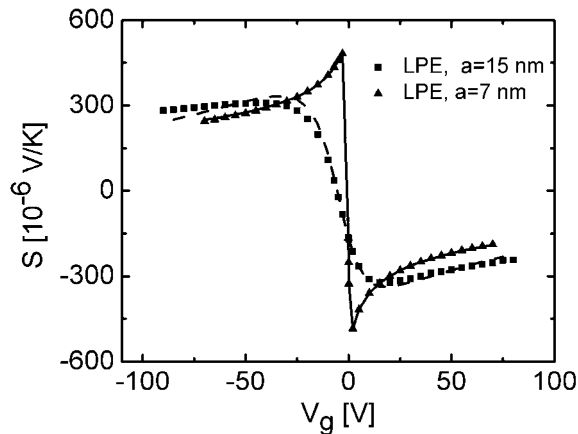

Fig. 2. The gate voltage dependence of the Seebeck coefficient S of the Bi_2Te_3 NWs with the thicknesses of a =7 nm (dashed line) and a = 15 nm (solid line) at room temperature.

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The EFE causes the Seebeck coefficient to increase as much as twice. Thus both the confinement effect and the electric field effect considerably improve the NW Seebeck coefficient. The calculations showed that the dependence of the NW thermal conductivity on the gate voltage is not monotonic. At the zero gate voltage, the thermal conductivity is equal to 0.248 $WK^{-1}m^{-1}$ (0.702 $WK^{-1}m^{-1}$) for the NW thickness 7 nm (15 nm). The minimum value of the thermal conductivity of 0.18 $WK^{-1}m^{-1}$ (0.39 $WK^{-1}m^{-1}$) is achieved at the gate voltage of 2 and -5 V (30 and -50 V) for the NW thickness of 7 nm (15 nm).

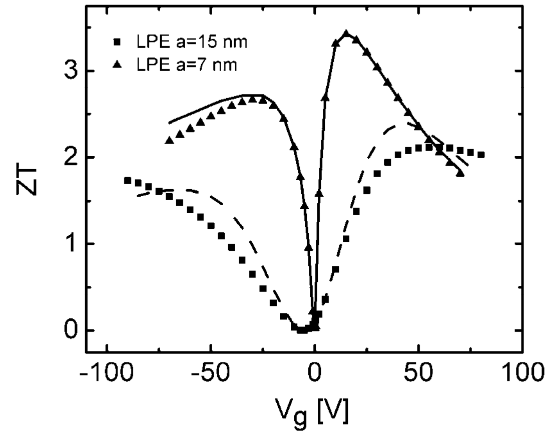

Fig. 3. The gate voltage dependence of the figure of merit ZT of the Bi_2Te_3 NWs with the thickness of a=7 nm (dashed line) and a = 15 nm (solid line) at room temperature.

Fig. 3 presents the gate dependence of the NW figure of merit at room temperature. At the zero gate voltage, the figure of merit is equal to 0.065 and 0.099 for the NW thicknesses of 7 and 15 nm, respectively. For the NW thickness of 7 nm (15 nm), the maximum value of the figure of merit ZT=3.4 (2.3) is achieved at the gate voltage 15 V (40 V). For the opposite gate polarity, when the holes dominate, the maximum value of the figure of merit achieves the lower value, because of the difference between the hole mobility and the electron mobility, as well as due to the difference in the effective masses of electrons and holes. Since the mathematical expression for the NW figure of merit is rather complex, the discrepancy between the LPE and the SEM data increases with the gate voltage for the NW thickness of 15 nm.

4. Summary

The results obtained by the SEM and LPE approaches agree rather well for the NWs with small thicknesses. The EFE can effectively control the type of the nanowire conductivity. Both the confinement effect and the electric field effect considerably improve the intrinsic nanowire thermoelectric properties. The EFE increases the Seebeck coefficient nearly as much as twice. The maximum value of the figure of merit achieves values of 3.4 and 2.3 for the 7- and 15-nm-thick NWs, respectively.

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