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Theoretical study of thermoelectric properties of SiC nanowires

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Abstract. We present a theoretical study of thermoelectric properties of silicon carbide (SiC) nanowires as a function of the wire cross section at high temperature (500K), based on non-equilibrium classical molecular dynamics simulations for the lattice thermal transport and the non-equilibrium green's function for the electrical transport. Our calculations show that figure of merit (ZT) increases with decreasing cross section area: ZT of SiC nanowire at 2x2 nm^2 has a maximum value in the range of 0.65 - 0.89 at 500 K, which is 7 - 8 times larger than the maximum ZT of a SiC thin film (0.125 at 973 K). These results show that SiC may be a promising material for thermoelectric applications operating at high temperature.

Introduction

The combination of the distinctive physical and chemical properties of silicon carbide (SiC) and unique advantages of nanowires (NWs) make them outstanding building blocks for the fabrication of nano-electronic devices, which can operate in harsh environment. Our group has demonstrated that SiC NWs field effect transistors (FETs) have similar electrical behavior compared with Si NWs FETs in the ballistic regime [1]. However, compared with the study of electronic properties of SiC NWs [1,2,3], much less has been done for thermal properties of SiC NWs [4]. To the best of our knowledge, there is no report of experimental or theoretical results about the thermoelectric properties of SiC NWs.

Bulk SiC is a poor thermoelectric material due to its high thermal conductivity (300 $Wm^{-1}K^{-1}$) [5]. However, one-dimensional structures, such as NWs, are theoretically predicted by Hicks and Dresselhaus to have a larger ZT compared with the bulk counterpart, due to their novel band structure [6]. In particular, SiC is expected to be promising at a higher temperature region due to wide gap semiconductor characteristics. Therefore, these unique properties of SiC, if coupled with the advantages of NWs, will make it an outstanding candidate for thermoelectric devices, which operate at high temperature.

In this work, we first attempt to predict the thermoelectric properties of SiC NWs with different square cross sections in the ballistic regime at high temperature (500K).

Background and Computational Methods

Figure of Merit (ZT). The performance of a thermoelectric device is characterized by the figure of merit ZT (a dimensionless value), which can be calculated by Eq (1):

$$ZT = \frac{TS^2G}{\kappa}, \quad (1)$$

where Z is a measure of a material's thermoelectric properties, T is the temperature [K], S is the Seebeck coefficient [V/K] , G is the electrical conductance [Ω^{-1}], and κ is the thermal conductance [W/K], which is the sum of the electron (κ_e) and phonon (lattice vibration) (κ_l) contributions.

In order to increase the ZT, G should be large to minimize joule heating losses and maximize the electron contribution for the thermoelectric effect. S also should be as large as possible to generate a large voltage difference between the two contacts even for a small temperature difference. In addition, it must have small κ to maintain the temperature gradient between two contacts [7]. However, the quantities S, G and κ are not mutually exclusive, but coupled with each other. Therefore, it is very difficult to control these variables independently.

Thermoelectric Power Factor (S^2G). The numerator of Z, which is (S^2G), is called the power factor. It quantifies the electrical performance of thermoelectric materials. The conductance of SiC NWs (in Eq. (2)) and the corresponding Seebeck coefficient (in Eq. (3)) were computed within the framework of the quantum-transport theory of Landauer–Büttiker [8]:

$$G = \frac{2q^2}{h}\int_{-\infty}^{\infty} T(E)(-\frac{df}{dE})dE \tag{2}$$

$$S \equiv -\frac{\Delta V}{\Delta T}\bigg|_{I=0} = -\frac{1}{qT}\int_{-\infty}^{\infty}(E-E_F)T(E)(f_1(E)-f_2(E))dE \Big/ \int_{-\infty}^{\infty} T(E)(f_1(E)-f_2(E))dE \tag{3}$$

where q is the electron charge, T is temperature, h is the Planck constant, $T(E)$ is the transmission coefficient, f_1 and f_2 are equilibrium Fermi–Dirac distributions for the contacts, and E_F is the Fermi level at both contacts.

Thermal Conductance (κ). The thermal properties of NWs are much different from those of the bulk material. The high surface to volume ratio of NWs may increase the boundary inelastic scatterings by the surface and hence dramatically decreases the thermal conductivity with respect to the bulk value.

Experimental and theoretical results show NW structure to be effective at reducing the lattice thermal conductivity of SiC. From experimental measurements, it is shown that the thermal conductivity of Si NWs is about 2 orders of magnitude smaller than that of bulk crystals [9]. SiC NWs present lower thermal conductivity (82 ± 6 $Wm^{-1}K^{-1}$) with diameter around 60 nm, compared to bulk SiC (300 $Wm^{-1}K^{-1}$) [10]. From theoretical calculations, N. Papanikolaou has estimated the lattice thermal conductivity of SiC NWs at high temperature (500 K) by using non-equilibrium classical molecular dynamics simulation [4]. According to their simulation results, SiC NWs with diameter from 2 to 4 nm show a thermal conductivity of 3 to 6 $Wm^{-1}K^{-1}$ at 500K. We implemented this simulated lattice thermal conductivity into the calculation of thermoelectric properties of SiC NWs.

The electron thermal conductance (κ_e) was calculated by means of the quantum-transport theory of Landauer–Büttiker, which can be calculated according to Eq (4):

$$\kappa_e = \frac{2}{hT}\int_{-\infty}^{\infty} T(E)(E-E_F)^2(-\frac{df}{dE})dE - TS^2G \tag{4}$$

Results and Discussion

Fig. 1 shows the power factor (S^2G) and thermal conductance (κ, κ_e and κ_l) in squared cross-section SiC NWs as a function of the wire cross section. The power factor has its maximum value near the threshold voltage and increases with increasing wire cross section.

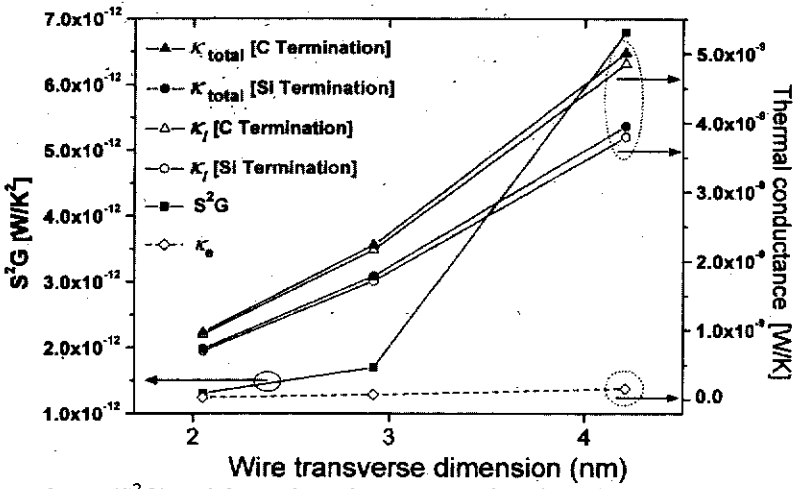

Figure 1. Power factor (S^2G) and thermal conductance as a function of the wire transverse dimension.

An interesting observation is that the smaller wire cross section increases the surface to volume ratio, therefore more phonons are diffusely reflected at the surface. As a result, thermal conductance was gradually reduced with decreasing cross-sectional area. The electron contribution for thermal conductivity is much lower than the phonon (lattice) contribution. The lattice thermal conductivity depends sensitively not only on the cross section of the nanowire but also on surface terminations (Si and C) of the NW. A Si terminated SiC NW has a lower thermal conductance than a C terminated SiC NW. This might be due to stronger scattering on a more disordered Si surface compared to the C one [4].

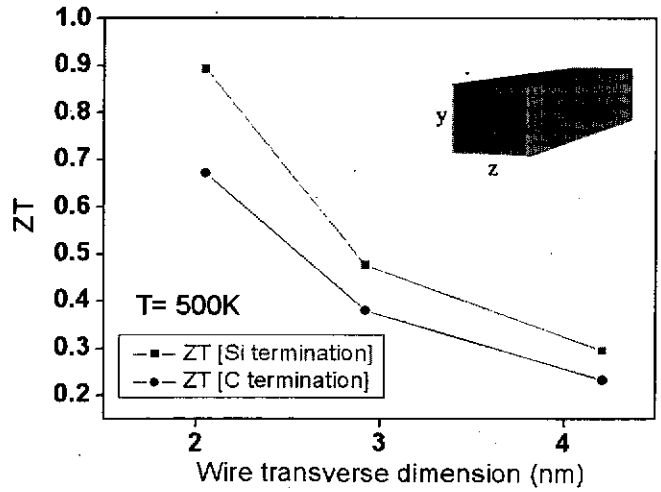

Figure 2. Figure of merit (ZT) in square SiC NWs as a function of the wire transverse dimension (inset: schematic structure of the simulated square NWs)

Thermal conductance decreases faster than a power factor with decreasing wire cross section. Hence, ZT gradually increases with decreasing wire cross section, as shown in Fig. 2. The ZT of a SiC NW at 500 K can reach values of up to 0.6 - 0.89 at 2x2 nm^2, which is 5 - 7 times larger than the maximum ZT value of a SiC thin film (0.125 at 973 K) [11]. This value is also comparable to the highest experimental ZT value of a Si NW (0.6) at room temperature [9]. This ZT enhancement mainly comes from a drastic reduction of thermal conductivity through the enhanced scattering of phonons, without significant reduction of the Seebeck coefficient (S) and electrical conductance (G).

Summary

We have investigated the size dependence of thermoelectric properties of SiC NWs. When cross sectional area of the NW decreases, the power factor decreases slowly while the thermal conductance reduces remarkably. Hence, ZT is gradually increasing with decreasing wire cross section. ZT reaches a value of 0.65 - 0.89 for a wire of cross section 2x2 nm^2, which is 5 - 7 times larger than the highest experimentally reported ZT value for a SiC thin film (0.125 at 973K). Based on simulated results, SiC NWs could be promising structures for thermoelectric applications operating at high temperature.

References

[1] K. Rogdakis, S. Poli, E. Bano, K. Zekentes, M. G. Pala, Phonon and surface roughness limited mobility of gate-all-around 3C-SiC and Si nanowire FETs, Nanotechnology 20 (2009) 295202.

[2] W. M. Zhou, F. Fang, Z. Y. Hou, L. J. Yan, Y. F. Zhang, Field-Effect Transistor Based on β-SiC Nanowire, IEEE Elec. Dev. Lett. 27 (2006) 463.

[3] Chan-Oh Jang, Tae-Hong Kim, Seung-Yong Lee, Dong-Joo Kim, Sang-Kwon Lee, Low resistance ohmic contacts to SiC nanowires and their applications to field-effect transistors, Nanotechnology 19 (2008) 345203.

[4] N Papanikolaou, Lattice thermal conductivity of SiC nanowires, J. Phys.: Condens. Matter 20 (2008) 135201.

[5] Ju Li, Lisa Porter, Sidney Yip, Atomistic modeling of finite-temperature properties of crystalline β-SiC, J. Nucl. Mater. 246 (1997) 53.

[6] L. D. Hicks, M. S. Dresselhaus, Thermoelectric figure of merit of a one-dimensional conductor, Phys. Rev. B 47 (1993) 16631.

[7] Allon I. Hochbaum, Peidong Yang, Semiconductor Nanowires for Energy Conversion, Chem. Rev. 110 (2010) 527.

[8] Raseong Kim, Supriyo Datta, Mark S. Lundstrom, Influence of dimensionality on thermoelectric device performance, J. Appl. Phys. 105 (2009) 034506.

[9] Allon I. Hochbaum, Renkun Chen, Raul Diaz Delgado, Wenjie Liang, Erik C. Garnett, Mark Najarian, Arun Majumdar, Peidong Yang, Enhanced thermoelectric performance of rough silicon nanowires, Nature 45 (2008) 163.

[10] K. M. Lee, T. Y. Choi, S. K. Lee, D. Poulikakos, Focused ion beam-assisted manipulation of single and double β-SiC nanowires and their thermal conductivity measurements by the four-point probe 3ω method, Nanotechnology 21 (2010) 125301.

[11] X. H. Wang, A. Yamamoto, K. Eguchi, H. Obara, T. Yoshida, Thermoelectric properties of SiC thick films deposited by thermal plasma physical vapor deposition, Sci. Technol. Adv. Mat. 4 (2003) 167.