Electron Diffusion and Phonon Drag Thermopower in Silicon Nanowires

Kasala Suresha

Department of Physics, Government First Grade College, Hosadurga – 577 527, India

Email: kasalasuresha@rediffmail.com

Abstract— *The field of thermoelectric research has undergone a renaissance and boom in the fast two decades, largely fueled by the prospect of engineering electronic and phononic properties in nanostructures, among which semiconductor nanowires (NWs) have served both as an important platform to investigate fundamental thermoelectric transport phenomena and as a promising route for high thermoelectric performance for device applications. In this report we theoretical studied the carrier diffusion and phonon-drag contribution to thermoelectric performance of silicon nanowires and compared with the existing experimental data. We observed a good agreement between theoretical data and experimental observations in the overall temperature range from 50 – 350 K. Electron diffusion thermopower is found to be dominant mechanism in the low temperature range and shows linear dependence with temperature.*

Keywords— *Electron diffusion, Mott formula, Phonon drag, Silicon nanowire, Thermopower.*

I. INTRODUCTION

Transport properties such as hot electron energy relaxation and thermopower at low temperature depend purely on electron-phonon interaction and are used as tools to determine the electron-phonon coupling strength. Thermoelectrics describes direct conversion between thermal and electrical energy, which encompasses three separately identified physical phenomena: the Seebeck effect, the Peltier effect and the Thomson effect. Many physically interesting examples of carrier transport in mesoscopic structures occur in the low-temperature and low-carrier-energy regime. In this regime, [1-4] acoustic phonons account for a significant fraction of the carrier scattering and may dominate over the scattering of carriers by optical phonons. Moreover, in nanoscale structures where dimensional confinement is significant, it is possible that phase space restrictions may weaken or forbid optical phonon scattering processes that would normally dominate in bulk structures. Recently, there has been an extensive literature on the role of dimensional confinement in modifying longitudinal optical phonon modes and their interactions with charge carriers in nanoscale and mesoscopic semiconductor structures [5-7] and the

numerous papers referenced therein. However, there are relatively few treatments dealing with the role of dimensional confinement in modifying acoustic phonon modes and their interactions with charge carriers [1-2, 8-10]. Specifically, few efforts have been reported that formulate a theory of the electron-acoustic phonon interaction in nanoscale structures where the treatment of acoustic phonon confinement effects may be essential [11-12]. The importance of such theoretical treatments has been demonstrated recently by experimental studies [1-2, 5-8] providing both direct and indirect evidence of the significance of acoustic phonon confinement in reduced dimensional electronic structures. Motivated by these developments and the lack of appropriate theoretical treatments as well as by the advancing technology for fabricating nanoscale cylindrical structures [13], we investigate electron–acoustic phonon interactions in various cylindrical semiconductor quantum wires in this paper. Based on our earlier work on [100] directed GaAs quantumwires,16 the scattering rates due to the deformation potential interaction are obtained for confined phonons along several crystallographic directions for a few technologically important material systems; that is, the directional dependence of electron-acoustic-phonon

scattering rates for GaAs, InGaAs and InSb quantum wires is studied for electron transport along the [100],[110] and [111] directions as well as for both free and clamped boundary conditions. Using a parameterization of the acoustic phonon velocities, the directional dependence of both the longitudinal and transverse velocities and is taken into account for each of the cases considered. The principal findings in the present paper are that acoustic phonon confinement effects may be crucial for calculating accurate low-energy electron scattering rates in semiconductor quantum wires and that there may be significant directional dependence in the subject scattering rates.

The most efficient thermoelectrics have historically been heavily doped semiconductors because the Pauli principle restricts the heat carrying electrons to be close to the Fermi energy [14] for metals. The Wiedemann–Franz law, $\frac{\kappa_e}{\sigma T} = \frac{\pi^2}{3}\left(\frac{k_B}{e}\right)^2$, where κ_e is the electronic contribution to κ, constrains $ZT = \frac{S^2 \sigma T}{\kappa}$, where S is the Seebeck coefficient (or thermoelectric power), and σ and κ are the electrical and thermal conductivities, respectively. Semiconductors have a lower density of carriers, leading to larger S values and a κ value that is dominated by phonons $\left(\kappa_{ph}\right)$, implying that the electrical and thermal conductivities are somewhat decoupled [14]. κ can be reduced by using bulk semiconductors of high atomic weight, which decreases the speed of sound. However, this strategy has not yet produced materials with ZT>1. For a metal or highly doped semiconductor, S is proportional to the energy derivative of the density of electronic states. In low-dimensional (nanostructured) systems the density of electronic states has sharp peaks [15-17] and, theoretically, a high thermopower. Harnessing this electronic effect to produce high ZT materials has had only limited success [18-19]. However, optimization of the phonon dynamics and heat transport physics in nanostructured systems has yielded results [20-21]. Nanostructures may be prepared with one or more dimensions smaller than the mean free path of the phonons and yet larger than that of electrons and holes. This potentially reduces κ without decreasing σ [22]. Bulk silicon (Si) is a poor thermoelectric (ZT at 300K) [23], and this phonon physics is important for our Si nanowires, in which the electronic structure remains bulk-like.

The thermoelectric properties of any new materials can be realized by studying with its possible applications in thermoelectric devices as their efficiency depends upon the thermoelectric figure of merit Z, Phonon-drag thermopower as well as electron diffusion thermopower.

Theoretically S_{drag} is studied in graphene [24], 3D Dirac semimetals [25]. Diffusion thermopower S_{diff} is predicted to show linear dependence agreeing with Mott formula and experimental results. The measured value of S_{diff} at room temperature is about 62µV/K. In the present theoretical work, we tried to show the comparison between existing experimental observations [26] with theory and obtained a good agreement with experimental thermopower of Si nanowire in the overall temperature range.

II. THEORY

It is assumed that, the temperature is high enough to maintain an equipartition regime of electron-phonon interaction, in which the number of occupation of the phonon modes contributing to carrier diffusion and phonon drag effect. The most efficient Thermoelectrics have been heavily doped semiconductors because the Pauli principle restricts the heat carrying electrons to within $k_B T$ of the Fermi level for metals. Semiconductors have a lower density of carriers, leasing to larger thermopower values implying that the electrical and thermal conductivities are somewhat decoupled. Thermal conductivity can be reduced by using bulk semiconductors of high atomic weight, which decreases the speed of sound. However, this strategy has not yet produced materials with figure of merit greater than unity. For metal or highly doped semiconductors, thermoelectric power S is proportional to the energy derivative of the density of electronic states. In low-dimensional (nanostructured) systems the density of electronic states has sharp peaks [17] and theoretically a high thermopower.

We consider separately the electron diffusion S_{diff} and phonon drag S_{drag} contribution to the thermopower $S = S_{diff} + S_{drag}$ for the Si nanowire over the temperature T from 50 K to 400K. Charge carriers dissipate heat to the lattice through a process that first involves momentum conserving (non-dissipative) electron-phonon collisions. The phonons that contribute to phonon drag cannot have a wavelength shorter than λ_{min} which is determined by the size of the Fermi surface. Phonon drag is observed in metals only at low temperature because the Fermi surface is large and the heat carrying short-wavelength phonons have short lifetime at lower temperature ($T < 20$K). Umklapp (non-momentum-conserving) phonon-phonon scattering processes determine the rate of phonon heat dissipation. The Debye energy θ_D sets the energy scale for umklapp scattering. The number of umklapp phonons available to dissipate

long wavelength phonons is given by Bose-Einstein function

$$N_{ph} = \frac{1}{\exp\left(\frac{\theta_D}{T}\right) - 1}$$

(1)

Leading to a scattering rate, $\frac{1}{\tau_{ph}} \propto N_{ph}$, when $T \gg \theta_D$, so that $\frac{1}{\tau_{ph}} \propto T$ because $\theta_D = 640K$ for Silicon, the Bose-Einstein expression must be applied for $T \leq 350$ K. The electronic contribution S_{diff} is estimated using Mott formula [27-28];

$$S_{diff}(T) = \frac{\pi^2 k_B^2 T}{3e}\left(\frac{\partial \ln \sigma(E)}{\partial E}\right)$$

(2)

where the conductivity derivative equals the reciprocal of the energy scale over which it varies.

For $T > 200$ K, the experimental thermopower data of silicon nanowire fit to the theoretical expression,

$$S = S_{diff} + S_{drag} = aT + b\left[exp\left(\frac{\theta_D}{T} - 1\right)\right]$$

(3)

where a, b and θ_D are varied to obtain the best fit to experimentally observed data [24].

III.   RESULTS AND DISCUSSION

Using the above final expression (3) for thermopower, we successfully explain the theoretical data with the experimental observations [26]. Figure 1 shows the best fit (black solid line) with the experimental data for the temperature $T > 200$ K by considering both contribution of thermopower from electron diffusion (S_{diff}) and phonon drag (S_{drag}). To obtain a best fit, a, b and θ_D are used as adjustable parameters. In our calculations, we used $a = 0.337$ µV/K, $b = 22.1$ µV/K and $\theta_D = 534K$. But for the temperature below 200K, we didn't get best with the experimental data of thermopower. Since for lower temperature $T < 200$ K, the contribution towards thermopower is only due to electron diffusion thermopower. With the above mentioned adjustable parameter a, we will not observe best with the experimental data. So we used adjustable parameter $a = 2.0$ µV/K to obtain a best fit in figure 1 (pink dash-dot line) for the temperature below 200K where diffusion thermopower is dominant mechanism. This is good agreement with predicted linear temperature dependence for $k_B T$ less than Fermi energy, which is about 200 meV agreeing with Mott formula and experimental results [29].
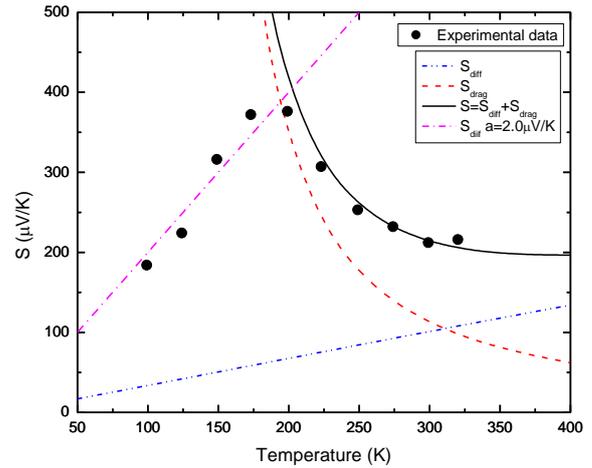
Fig. 1: Comparison of experimental data (solid dots) [26] with theory. Blue dash-dot-dot line represents for S_{diff}. Red dash line for S_{drag} and black solid line represent for total S. Pink dash-dot line represents for S_{diff} with $a = 2.0$ µV/K.

Figure 2 shows the comparison between experimental observations with theory. In this figure we obtain a best fit over the entire temperature range from the electron diffusion contribution only with adjustable parameter $a = 0.46$ µV/K. In this figure we don't observe contribution from phonon drag effect. The decrease of S to zero as T tends to zero occurs because the phonon mean free path reaches the sample size and the specific heat tends to zero according to third law of thermodynamics. It is also interesting to observe that a linear dependence of carrier diffusion thermopower with temperature which satisfies the experimental observations [29]. Without considering the phonon drag effect mechanism, the calculated values of diffusion thermopower contribution is enough to get best fit with experimental observations.
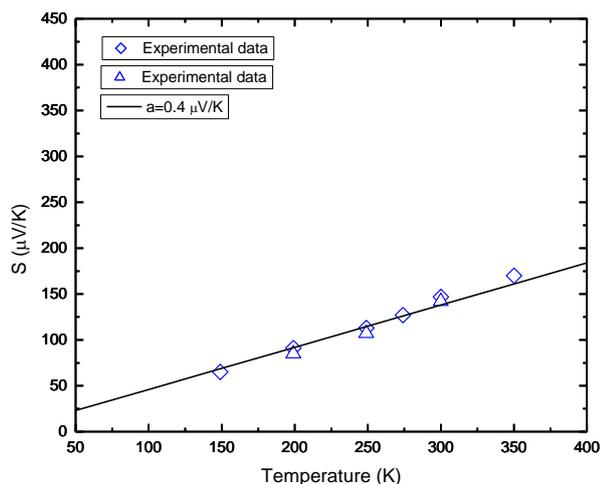
Fig.2: Comparison of experimental data (diamonds and triangles) [26] with theory. Solid line represent for S_{diff} with α =0.46 µV/K.

IV.   CONCLUSION

In this theoretical study, we conclude that our calculations for thermopower in Silicon nanowire are in good agreement with the experimental observations. At low temperature, carrier diffusion thermopower shows a dominant mechanism for thermopower with linear temperature dependence. At higher temperature both carrier diffusion and phonon drag effects are effective to obtain a good agreement with the experimental observations.

REFERENCES

[1]   J. Seyler and M. N. Wybourne, Phys. Rev. Lett. **69**, 1427, (1992)

[2]   Z. V. Popvic, J. Spitzer, T. Ruf, M. Cardona, R. Notzel, and K. Ploog Phys. Rev. B **48**, 1659 (1993)

[3]   A. Tanaka, S. Onari, and T. Arai, Phys. Rev. B **47**, 1237 (1993)

[4]   P V Santos, A K Sood, M Cardona, K Ploog, Y Ohmori, and H Okamoto, Phys. Rev. B **37**, 6381 (1988)

[5]   N Mori and T Ando, Phys. Rev. B **40**, 6175 (1989)

[6]   H Rucker, E Molinari, and P Lugli, Phys. Rev. B **45**, 6747 (1992)

[7]   M A Stroscio, G J Iafrate, K W Kim, M A Littlejohn, A R Bhatt,and M Dutta, in *Integrated Optics and Optoelectronic*, edited by K.-K. Wong and M. Razeghi~SPIE, Bellingham, Vol. **CR45**, p. 341 (1993)

[8]   B Hillebrands, S Lee, G I Stegeman, H Cheng, J E Potts and F Nizzoli, Phys. Rev. Lett. **60**, 832 (1988),

[9]   H Benistry, C M Sotomayor-Torre´s, and C Weisbuch, Phys. Rev. B **44**, 10945 (1991)

[10] N Nishiguchi, Phys. Rev. B **50**, 10970 (1994)

[11] N Bannov, V. Mitin, and M. A. Stroscio, Phys. Status Solidi B **183**, 131 (1994)

[12] S Yu, K W Kim, M A Stroscio, G J Iafrate, and A Ballato, Phys. Rev. B **50**, 1733 (1994)

[13] M Watt, H E G Arnot, C M Sotomayor-Torre´s, and S P Beaumont, in *Nanostructure Physics and Fabrication*, edited by M A Reed and W P Kirk ~Academic, Boston, p. 89. (1989)

[14] D K C MacDonald,. Thermoelectricity: An Introduction to the Principles (Wiley, New York, 1962).

[15] L D Hicks and M S Dresselhaus, Phys. Rev. B **47**, 16631–16634 (1993)

[16] G D Mahan and J O Sofo, Proc. Nat. Acad. Sci. USA **93**, 7436–7439 (1996)

[17] T E Humphrey, and H Linke, Phys. Rev. Lett. **94**, 096601 (2005)

[18] A Boukai, K Xu, and J R Heath, Adv. Mater. **18**, 864–869 (2006)

[19] L Yu-Ming, Appl. Phys. Lett. **81**, 2403–2405 (2002)

[20] T C Harman et al,. Science, **297**, 2229 (2002)

[21] K F Hsu et al., Science **303**, 818–821 (2004)

[22] A Majumdar, Science **303**, 777–778 (2004)

[23] L Weber and E Gmelin, Appl. Phys. A **53**, 136–140 (1991)

[24] S S Kubakaddi, Phys. Rev. B, 79, 075417 (2009)

[25] R Landgren, P Laurell and G A Fiete, Phys. Rev. B, 90, 105115 (2014)

[26] Akram I Boukai, Yuri Bunimovich, Jamil-Kheli, Jen-Kan Yu, William A Goddard and James R Heath, nature LETTERS, **451**, 168 (2008)

[27] Mott N F, Davis E A, Electronic processes in Non-crystalline Materials, Oxford:Clarendon, (1979)

[28] Kasala Suresha, Int. J. Res. Appl. Sci. and Engg. Tech. **9**, 39 (2021)

[29] A Parari, N Khan and P Mandal, arXiv:1502.02264 (2015)