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# Modeling heat transfer in $Bi_2Te_3$–$Sb_2Te_3$ nanostructures

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ABSTRACT

$Bi_2Te_3$–$Sb_2Te_3$ nanostructures are gaining importance for use in thermoelectric applications following the finding that the $Bi_2Te_3$–$Sb_2Te_3$ superlattice exhibits a figure of merit, $ZT = 2.4$, which is higher than conventional thermoelectric materials. In this paper, thermal transport in the cross-plane direction for $Bi_2Te_3$–$Sb_2Te_3$ nanostructures is simulated using the Boltzmann transport equation (BTE) for phonon intensity. The phonon group velocity, specific heat, and relaxation time are calculated based on phonon dispersion model. The interfaces are modeled using a combination of diffuse mismatch model (DMM), and the elastic acoustic mismatch model (AMM). The thermal conductivity for the $Bi_2Te_3$–$Sb_2Te_3$ superlattice is compared with the experimental data, and the best match is obtained for specularity parameter, $p$, of 0.9. The present model is extended to solve for thermal transport in 2-D nanowire composite in which $Sb_2Te_3$ wires are embedded in a host material of $Bi_2Te_3$. Unlike in bulk composites, the results show a strong dependence of thermal conductivity, temperature, and heat flux on the wire size, wire atomic percentage, and interface specularity parameter. The thermal conductivity of the nanowire is found to be in the range of 0.034–0.74 depending on the atomic percentage and the value of $p$.



## 1. Introduction

Thermoelectric energy conversion is a field that can greatly benefit from the nanoscale heat transport phenomenon. The efficiency of thermoelectric conversion for a material is measured by a non-dimensional figure of merit (ZT) defined as, $ZT = \sigma S^2 T/k$ where $\sigma$ is the electrical conductivity, $S$ is the Seebeck coefficient, $T$ is the temperature, and $k$ is the thermal conductivity [1]. During the last decade, advances have been made in increasing ZT using nanostructures [2–7]. This was achieved by reducing the phonon thermal conductivity more than the electrical conductivity [8]. Although the decrease in thermal conductivity may be due to several effects such as the phonon group velocity reduction caused by the spectrum change, the interface thermal resistance and interface scattering, phonon interference and tunneling, and dislocations, it is found that scattering of energy carriers at interfaces plays the most important role [9]. The reduction in the effective thermal conductivity enhances the thermoelectric energy conversion.

The nanostructure configuration that show potential for enhancing ZT consists of multilayered thin films of different materials with thickness ranging from a monoatomic layer to thousands of angstroms called superlattice. Recent experimental studies [5,6] have demonstrated significant enhancement of ZT with $Bi_2Te_3$/$Sb_2Te_3$ superlattice in the cross-plane direction and PbTe/PbTeSe quantum dot superlattice along the film plane direction. Venkatasubramanian et al. [6] has reported a ZT of 2.4 for $Bi_2Te_3$–$Sb_2Te_3$ superlattice. However, superlattice grown by thin-film deposition techniques is not suitable for large scale applications due to its high manufacturing cost and difficulty to scale up for large scale applications. Nanocomposites offer a more economical alternative to superlattice in the quest for high ZT materials [10]. The features that make nanowire composites attractive for thermoelectric application are the 'size-effect' and 'effect of atomic percentage' which imply that the effective thermal conductivity of the nanowire depends on the material dimension as well as the atomic percentage of the wire. This remarkable feature of nanocomposites can be used to tailor the mechanical, thermal, and electrical properties which are best suited for a particular application.

Despite the importance of nanocomposites for thermoelectric applications, only a few experimental and numerical studies exist in the literature on the thermal characteristics of nanocomposites. Most of the computational studies on thermal transport using the particle based Boltzmann transport equation (BTE) have focused mainly on thin films, superlattices, and crystals [11–14]. Yang and Chen [15] have performed numerical simulations of cross-plane heat transport in a two-dimensional Ge–Si nanocomposite with an array of silicon wires embedded in a host material of Germanium. Yang et al. [16] report numerical simulation of Ge–Si core shell and tubular nanowires with heat transport along the axis of the wire. In both cases, the interfaces are treated as totally diffuse and their results show the effective thermal conductivity of the Ge–Si nanowire to be more than the superlattice for Si atomic percentage of 0.2. Recently, Prasher [17] has developed an analytical method for estimating the longitudinal thermal conductivity in nanowires and nanopores.

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**Nomenclature**

*Chemical symbols*

| | |
|---|---|
| $Bi_2Te_3$ | bismuth telluride |
| Ge | germanium |
| $Sb_2Te_3$ | antimony telluride |
| Si | silicon |

*Roman symbols*

| | |
|---|---|
| $a$ | lattice constant |
| AP | atomic percentage |
| $C$ | volumetric specific heat (J/m$^3$ K) |
| $D_p$ | density of states per unit volume (m$^{-3}$) |
| $f$ | phonon distribution function |
| $\hbar$ | Planck's constant divided by $2\pi$ ($1.054 \times 10^{-34}$ J s/phonon) |
| $I$ | phonon intensity (W m$^{-2}$ sr$^{-1}$) |
| $I^*$ | non-dimensional phonon intensity |
| $k$ | thermal conductivity (W/mK) |
| $k_B$ | Boltzmann constant ($1.38 \times 10^{-23}$ J/K phonon) |
| $Kn$ | Knudsen number ($\Lambda/L$) |
| $L$ | material dimension |
| $L_p$ | superlattice period thickness (m) |
| $L_w$ | nanocomposite wire size (m) |
| $\vec{n}$ | normal vector |
| $N_\phi$ | number of discrete azimuthal angular divisions |
| $N_\theta$ | number of discrete polar angular divisions |
| NC | nanocomposite |
| $p$ | interface specularity parameter |
| $q$ | heat flux (W/m$^2$) |
| $Q_x$ | heat flow in the $x$-direction (W) |
| $q_x^*$ | non-dimensional heat flux $[q_x/vC(\overline{T}(x = L_{Bi_2Te_3}) - \overline{T}(x = 0))]$ |
| $q_s$ | heat flux per solid angle (W m$^{-2}$ sr$^{-1}$) |
| $\vec{r}$ | position vector |
| $R_d$ | diffuse reflectivity |
| $R_s$ | specular reflectivity |
| $\vec{s}$ | phonon direction vector |
| $S$ | Seebeck coefficient (μV/K) |
| SL | superlattice |
| $T$ | absolute temperature (K) |
| $\overline{T}$ | average temperature (K) |
| TBR | thermal boundary resistance |
| $T_r$ | reference temperature (K) |
| $T^*$ | non-dimensional temperature $[(T - T_r)vCL_{Sb_2Te_3}/Q_x]$ |
| $t$ | time (s) |
| $T_d$ | diffuse transmissivity |
| $T_s$ | specular transmissivity |
| $U$ | volumetric internal energy (J/m$^3$) |
| $V$ | volume (m$^3$) |
| $v$ | average phonon group velocity (m/s) |
| $w_\theta$ | quadrature weight corresponding to the polar angle |
| $w_\phi$ | quadrature weight corresponding to the azimuthal angle |
| $x$ | coordinate |
| $y$ | coordinate |
| $x^*$ | non-dimensional $x$-coordinate $[x/L_{Bi_2Te_3}]$ |
| $y^*$ | non-dimensional $y$-coordinate $[y/L_{Bi_2Te_3}]$ |
| ZT | non-dimensional figure of merit |

*Greek symbols*

| | |
|---|---|
| $\beta$ | grid compression factor |
| $\delta$ | interface roughness (m) |
| $\eta$ | $y$-directional cosine |
| $\theta$ | polar angle (rad) |
| $\Lambda$ | phonon mean free path (m) |
| $\lambda$ | phonon wavelength (m) |
| $\mu$ | $x$-directional cosine |
| $\sigma$ | electrical conductivity (S/m) |
| $\tau$ | frequency independent phonon relaxation time |
| $\phi$ | azimuthal angle (rad) |
| $\omega$ | phonon frequency (Hz) |
| $\zeta$ | acoustic impedance |

*Subscripts*

| | |
|---|---|
| b | boundary |
| c | phonon scattering |
| d | diffuse interface |
| eq | equilibrium |
| g | phonon generation |
| h | host ($Bi_2Te_3$) |
| i | interface |
| p | polarization mode |
| r | phonon reflection |
| s | specular interface |
| U | umklapp scattering |
| w | wire ($Sb_2Te_3$) |
| $x$ | $x$-coordinate |
| $y$ | $y$-coordinate |

The main objective of the present work is to extend the general framework of thermal modeling of superlattice to simulate heat transport in $Bi_2Te_3$–$Sb_2Te_3$ nanowire composite. The heat transport in the cross-plane direction of $Bi_2Te_3$–$Sb_2Te_3$ superlattice is modeled and its thermal conductivity is compared with the experimental result [18]. The simulations are extended to $Bi_2Te_3$–$Sb_2Te_3$ nanowire composites, to understand the temperature and heat flux distributions as well as the effective thermal conductivity of such nanocomposites. The effects of the wire size, interface treatment, and atomic percentage on the thermal properties are also studied.

## 2. Problem description and modeling

The schematics of the superlattice and the 2-D nanowire composite chosen for the present simulations are shown in Fig. 1. The superlattice consists of periodically repeating stacks of $Bi_2Te_3$ on $Sb_2Te_3$ layers with heat transport across the layers. It is useful in studying the role of interface in the reduction of thermal conductivity.

The nanowire composite consists of an array of $Sb_2Te_3$ wires aligned parallel to each other and embedded in the host material of $Bi_2Te_3$, as shown in Fig. 1(b). Both the wires and the host are assumed to have a square cross-section. The heat transport is applied across the direction of the wire and there is no heat flow along the wire direction. This configuration mimics a thermoelectric device in which the wires are aligned for the device to transport the maximum heat flux perpendicular to the wire axis. Thus, a 2-D thermal energy transport is considered. For the ease of computations, the unit cell approach [19] shown in Fig. 1(c) is used to simulate the effect of a single wire and the surrounding host material within the cell.

To investigate the heat transfer in nanostructures, the main energy carriers in semiconductor materials namely phonons are modeled. Since the present study is conducted at room temperature, it is assumed that the short wavelength acoustic phonons contribute to the heat transfer. Hence, the wave nature of phonons is neglected and phonons are modeled as particles [20]. The