Improved Accuracy on Empirical Lattice Thermal Conductivity Model of Bi_2Te_3

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INTRODUCTION

Thermoelectric engineering has drawn notable attention in the last decades hoping to generate a certain extent of energy from wasted heat in an efficient way. The efficiency of a thermoelectric system is measured through a dimensionless quantity, thermoelectric figure of merit $ZT = S^2\sigma T/\kappa$, where S, σ, and κ are the Seebeck coefficient, electrical conductivity, and thermal conductivity, respectively [1]. Due to the large difference between phonon and electron wavelengths, scaling the thermoelectric materials down to the nanoscale leads to significant reduction in thermal conductivity with relatively less affected electrical properties, causing an improvement of ZT. Among thermoelectric materials, bulk bismuth telluride (Bi_2Te_3) and its alloys have been widely used for the commercial application, and the Bi_2Te_3/Sb_2Te_3 superlattices have achieved the best ZT value of ~ 2.4 [2].

While some experimental studies have been conducted on measuring thermal conductivity of bulk and nanostructured Bi_2Te_3, very few intensive theoretical predictions have been reported [3]-[7]. It is, therefore, interesting to apply the simple and amendable thermal conductivity models to Bi_2Te_3 and to predict its behaviors with varying parameters. Unlike computationally expensive simulation tools, the thermal conductivity models introduced by Klemens, Callaway, and Holland are relatively simple and cheap as representing the phonon scattering processes by frequency-dependent relaxation times [8]-[10]. Later work extended the simple model for making up for its failure to capture the anisotropy by solving the relaxation time approximation with the full phonon dispersions [11].

STUDY

In this study, the lattice thermal conductivity model of Bi_2Te_3 has been established with full phonon dispersion and directional momentum, taking account of the phonon scattering due to van der Waals (vdW) interactions between neighboring tellurium (Te) atoms. The Te layers bonded through vdW forces form weak bonding, thus leading to a reduction in thermal transport [12]. For vdW interface between identical materials, the phonon transmissivity can expressed in terms of the phonon frequency and the material properties [13]. To bring the phonon transmissivity into play, the theoretical model for the thermal transport in superlattice nanowires has been benchmarked [14]. Because Bi_2Te_3 forms a structure of quintuple layers (Te-Bi-Te-Bi-Te) attached on top of each other through vdW bonding, one can assume its structure as superlattice structure with the vdW interfaces taken place between the layers. Adopting the superlattice thermal conductivity model in [14] in which the phonon transmissivity plays an important role, the branch-dependent phonon relaxation time due to vdW interfaces can be defined as a function of phonon frequency, momentum, and the quintuple layer thickness.

The total relaxation time has been rewritten including the van der Waals term, τ_V, and the resulting thermal conductivities for bulk and 3 and 9 quintuple-layer (QL) Bi_2Te_3 were computed. Figure 1 shows the thermal conductivities in a cross-plane direction with temperature as an independent variable. The solid lines and dashed lines are the thermal conductivities with and without the vdW effect taken into account, respectively. Although, it does not make a large difference for 3-QL and 9-QL, it demonstrates a reasonable reduction of thermal conductivity from ones without the vdW effect. Figure 2 shows comparison of the calculated thermal conductivity with other theoretical and experimental data. Due

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to the lack of available data on Bi_2Te_3, they are not all measured or predicted under the same condition, but it confirms that the trend of each data set, especially for bulk, matches the thermal conductivity calculated in this study.

Once the simple model that agrees with experiments, it is straightforward and efficient to apply this model to different materials with varying size. Especially there will be no limitation on scaling in direction wise because this model has taken care of phonon momentum in three different directions in Cartesian coordinate system. Later, exploring other materials having the same structure as Bi_2Te_3 or Bi_2Te_3 supported by insulators would be also very interesting in seeking for better thermoelectric sources.

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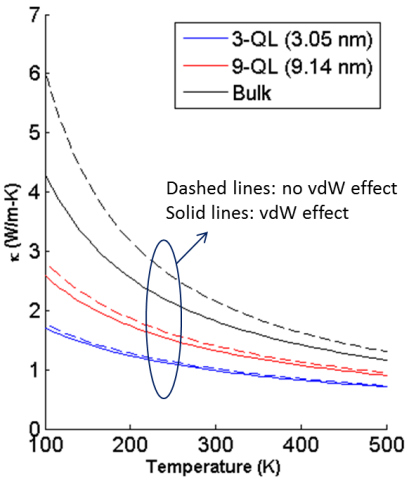

Fig. **1**. Thermal conductivities of bulk, 3 and 9 QL Bi_2Te_3 with (solid lines) and without (dashed lines) vdW effect taken into account.

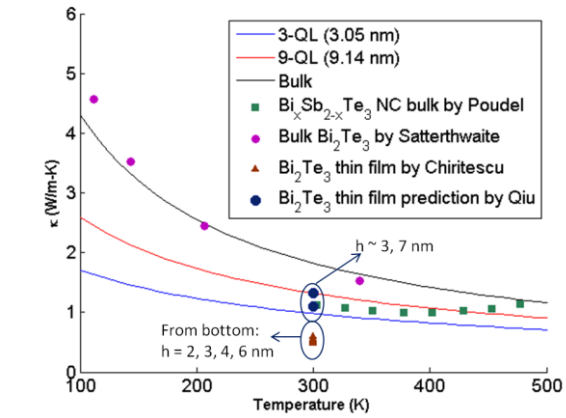

Fig. **2**. Thermal conductivity comparison with reported experimental and theoretical data. h implies a thickness of a thin film.