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Lorenz Function of Bi_2Te_3/Sb_2Te_3 Superlattices

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Combining first-principles density functional theory and semiclassical Boltzmann transport, the anisotropic Lorenz function was studied for thermoelectric Bi_2Te_3/Sb_2Te_3 superlattices and their bulk constituents. It was found that, already for the bulk materials Bi_2Te_3 and Sb_2Te_3, the Lorenz function is not a clear function of charge carrier concentration and temperature. For electron-doped Bi_2Te_3/Sb_2Te_3 superlattices, large oscillatory deviations of the Lorenz function from the metallic limit were found even at high charge carrier concentrations. The latter can be referred to quantum well effects, which occur at distinct superlattice periods.

Key words: Thermoelectric transport, heterostructures, DFT, electronic structure

INTRODUCTION

For many decades, thermoelectric (TE) energy conversion has successfully enabled self-supporting energy devices for outer-space missions or integrated electronics.[1,2] However, poor conversion efficiency prohibited the breakthrough use of thermoelectrics as an alternative energy source. The conversion performance of a TE material is quantified by the figure of merit (FOM)

$$ZT = \frac{\sigma S^2}{\kappa_{\mathrm{el}} + \kappa_{\mathrm{ph}}} T = \frac{S^2}{L + \frac{\kappa_{\mathrm{ph}}}{\sigma T}}, \qquad (1)$$

where σ is the electrical conductivity, S is the thermopower, and $\kappa_{\mathrm{el}} = L\sigma T$ and κ_{ph} are the electronic and lattice contributions to the thermal conductivity, respectively. L denotes the Lorenz function, which becomes the Lorenz number $L_0 = \frac{(\pi k_B)^2}{3e^2}$ in the highly degenerate, metallic limit.

In recent years, nanostructuring concepts[3,4] have enabled higher values for ZT by increasing the numerator, called the power factor (PF = σS^2), or decreasing the denominator of Eq. 1. The latter is obtained by phonon blocking at superlattice (SL) interfaces or grain boundaries[5–8] and leads to a reduced lattice thermal conductivity κ_{ph}. Here, the Lorenz function is particularly important for thermoelectrics, providing a measure to separate the electronic and lattice contribution to the thermal conductivity.[9] Deviations $L \neq L_0$ already occur in the degenerate limit for simple metals, semimetals, and semiconductors.[10] Hence, assuming incorrect values for the Lorenz number leads to incorrect values for κ_{el} and κ_{ph} and can even result in non-physical negative values for κ_{ph}.[11] To the best of our knowledge, investigations on the Lorenz function of thermoelectric SLs at *ab initio* level are lacking so far.

In the present work we analyze the anisotropic Lorenz function for Bi_2Te_3/Sb_2Te_3 SLs, as well as for the bulk constituents. The two telluride single crystals and the composed *p*-type SL show the highest values for bulk and nanostructured TE so far.[12] On the basis of *ab initio* density functional theory (DFT) and semiclassical Boltzmann transport equations (BTE), the Lorenz function is in particular studied for different charge carrier concentrations and SL periods at room temperature.

METHODOLOGY

For both Bi_2Te_3 and Sb_2Te_3, as well as for the composed SLs, we used the experimental lattice parameters and relaxed atomic positions[13] as

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