

Lower limit to the lattice thermal conductivity of nanostructured Bi_2Te_3 -based materials

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We investigate the lower limit to the lattice thermal conductivity of Bi_2Te_3 and related materials using thin films synthesized by the method of elemental reactants. The thermal conductivities of single layer films of $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ and multilayer films of $(\text{Bi}_2\text{Te}_3)_m(\text{TiTe}_2)_n$ and $[(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3]_m(\text{TiTe}_2)_n$ are measured by time-domain thermoreflectance; the thermal conductivity data are compared to our prior work on nanocrystalline Bi_2Te_3 and a Debye-Callaway model of heat transport by acoustic phonons. The homogeneous nanocrystalline films have average grains sizes $30 <d< 100$ nm as measured by the width of the (003) x-ray diffraction peak. Multilayer films incorporating turbostratic TiTe_2 enable studies of the effective thermal conductivity of Bi_2Te_3 layers as thin as 2 nm. In the limit of small grain size or layer thickness, the thermal conductivity of Bi_2Te_3 approaches the predicted minimum thermal conductivity of 0.31 W/m-K. The dependence of the thermal conductivity on grain size is in good

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agreement with our Debye-Callaway model. The use of alloy $(\text{Bi,Sb})_2\text{Te}_3$ layers further reduces the thermal conductivity of the nanoscale layers to as low as 0.20 W/m-K.

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Introduction

Currently, the highest performance materials for the direct conversion of thermal and electrical energy near room temperature are based on Bi_2Te_3 ¹. Recently^{2,3}, nanostructured Bi_2Te_3 prepared by ball-milling and spark-plasma-sintering have been demonstrated with significantly enhanced efficiency in comparison to materials prepared by traditional methods of powder processing. The improved efficiency can be mostly attributed to a reduction in thermal conductivity created by scattering of phonons at the boundaries between nanoscale grains. These observations raise the question of how small of grains are needed to significantly reduce the lattice thermal conductivity and ultimately what is the lower limit to the lattice thermal conductivity of Bi_2Te_3 -based thermoelectric materials.

To help answer these questions, we have prepared a variety of thin films of Bi_2Te_3 , related alloys, and multilayers with well-controlled structure on nanometer length scales. Specifically, we introduce disordered layers of TiTe_2 in multilayers to create strong phonon scattering on well-defined length scales of only a few nanometers.

Experimental Details

Bismuth telluride⁴ (Bi_2Te_3) and antimony telluride⁵ (Sb_2Te_3) have rhombohedral crystal structures with one chemical formula per unit cell. The most often used

description is the pseudo-hexagonal unit cell obtained by transformation of axes and containing three chemical formulae per unit cell with lattice parameters⁶: Bi_2Te_3 $a = 4.4$ Å, $c = 30.5$ Å; Sb_2Te_3 $a = 4.3$ Å, $c = 30.4$ Å. The hexagonal cell is formed by stacking of layers of like-atoms perpendicular to the c -axis and following the sequence (called quintet) Te-X-Te-X-Te ($X = \text{Bi}$ or Sb); each quintet is bonded to the next by Te-Te bonds and longer-ranged electrostatic^{7,8} interactions. Titanium telluride (TiTe_2) has a hexagonal crystal structure⁹ with one formula unit per unit cell ($a = 3.8$ Å, $c = 6.5$ Å). In this structure, one sheet is composed of a hexagonal plane of Ti atoms bonded to two Te layers by strong covalent–ionic bonds and each two-dimensional TiTe_2 sheet is bonded to adjacent sheets by weak van der Waals forces.

The films were prepared using the modulated elemental reactants technique¹⁰. Various sequences of Bi, Sb, Ti and Te layers (99.995% purity, Alfa Aesar) were deposited onto polished Si substrates. The base pressure in the deposition chamber was $\sim 6 \times 10^{-7}$ Torr. Bi, Sb, and Te were deposited using effusion cells and Ti was deposited with an electron beam source.

The composition of individual building blocks of Bi_2Te_3 , Sb_2Te_3 and TiTe_2 were calibrated as described previously^{11,12}. We measured the composition using electron probe microanalysis (EPMA). EPMA data are acquired using a Cameca SX-50 or SX-100 operating at 20 nA current, 1 μm spot size, and 8, 12 and 16 kV beam energies at multiple locations on the sample. The data are refined using the STRATAGEM software package^{13 14}. The oxygen content of the samples is a few atomic percent.

Three series of $[(\text{Bi}_2\text{Te}_3)_m (\text{TiTe}_2)_n]$ ($m, n = 2-6$) multilayer films were prepared. In the first series, the repeat unit contained $m = 3$ bilayers of Bi and Te with 2:3 atomic

ratio, followed by $n = 2 - 6$ bilayers of Ti and Te with 1:2 atomic ratio. In the second series, the repeat unit contained $m = 2-6$ bilayers of Bi and Te with 2:3 atomic ratio, followed by $n = 3$ bilayers of Ti and Te with 1:2 atomic ratio. The third series consisted of multiples of the repeat unit consisting of alternating equal numbers of Bi/Te and Ti/Te bilayers with $m = n = 2, 5, \text{ and } 6$. After annealing at 300 °C for 30 minutes, the films were 50-100 nm thick.

The alloy and multilayer alloy films were prepared by depositing alternating bilayers of Bi/Te, Sb/Te and Ti/Te. Films with chemical composition $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ were deposited and then annealed in sealed quartz ampoules evacuated to 1×10^{-6} Torr in the presence of Te-rich Bi_2Te_3 powder. The films were subsequently annealed at 150 °C for 30 min, at 250 °C for 10 min, or at 300 °C for 4 and 8 min and after annealing were ≈ 60 nm thick. Films with chemical composition $[(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3]_m (\text{TiTe}_2)_n$ ($x = 0.55 \pm 0.09$, $m = 2-4$, $n = 3$ and 6) were deposited using the same recipe but annealed in a dry N_2 atmosphere at 250 °C. After annealing, the films had a thickness between 40 and 60 nm.

We employed x-ray diffraction (XRD) and x-ray reflectivity (XRR) to determine the phase-formation, grain-size, film thickness, multilayer-period and the crystallographic orientation of the as-deposited and annealed samples. The x-ray studies were performed using $\text{Cu K}\alpha_1$ radiation source with a Goebel mirror to collimate the beam. Thickness of the films was evaluated from the Kiessig fringes of low angle XRR scans. The period of the multilayers was determined from the position of superlattice Bragg diffraction maxima in the high angle XRD data. The grain size was calculated using the Scherrer equation after accounting for the contribution of instrumental broadening, $\Delta (2\theta) \approx 0.05^\circ$, to the width of the (003) diffraction peak.

Grazing incidence in-plane x-ray diffraction (GIIXRD) and high-angle x-ray diffraction (HAXRD) data of the as-deposited alloy and multilayer alloy samples indicate the incipient formation of alloy composition with nanometer size grains. Subsequent annealing promoted grain growth of the respective alloy composition, as noted from the narrowing of the diffraction maxima. The $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ alloy films are highly textured in the $[00L]$ crystallographic direction. Post deposition annealing of the multilayer $[(\text{Bi}_2\text{Te}_3)_m (\text{TiTe}_2)_n]$ samples results in superlattice formation as determined from the presence of superlattice Bragg diffraction past the 30th order corresponding to the superlattice period.

Additional structural characterization was obtained using synchrotron x-ray diffraction at beam line 33-ID-D at the Argonne Lab Advanced Photon Source. We evaluated the degree of turbostratic disorder in the annealed samples by scanning the diffraction intensity through reciprocal space where the $(I\ 0\ L)$ reflection intersected the Ewald sphere. These studies revealed that TiTe_2 exhibits turbostratic disorder; i.e., the annealed films have a disordered layered structure with precise stacking of the a - b planes relative to the substrate and a small, ~ 1 nm, domain size in $[I\ 0\ L]$ directions.

Synchrotron x-ray diffraction studies of the $[(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3]_m (\text{TiTe}_2)_n$ films showed the formation of high quality multilayers with precise stacking of the a - b planes of the superlattice relative to the substrate. Bragg diffraction maxima were observed past the 50th order, indicating the high quality of the alloy multilayer. Diffraction data obtained using an in-plane geometry revealed diffraction corresponding to TiTe_2 and $(\text{Bi,Sb})_2\text{Te}_3$ with no diffraction observed corresponding to either Bi_2Te_3 or Sb_2Te_3 , confirming the alloy nature of the superlattices.

Thermal conductivity of the annealed samples was measured at room temperature using time-domain thermoreflectance (TDTR)^{15,16}. The measurement procedure for closely related samples was described¹¹ previously.

Results and discussion

Figures 1 and 2 show that the thermal conductivity Λ in the cross-plane direction of the multilayer samples has a strong dependence on the average composition of the film but only a weak dependence on the density of interfaces. As described in our previous work on closely-related PbSe-based multilayers¹¹, we attribute the low thermal conductivity in the cross-plane direction to strong phonon scattering and anisotropic elastic constants of the turbostratic disordered material, in this case TiTe₂. The (Bi,Sb)₂Te₃ / TiTe₂ multilayer alloy samples have a lower thermal conductivity ($\Lambda_{\text{avg.}} = 0.17$ W/m-K) than the Bi₂Te₃ / TiTe₂ multilayer films ($\Lambda_{\text{avg.}} = 0.24$ W/m-K), see Figs. 1b and 2b.

The average thermal conductivity of our homogeneous TiTe₂ thin film samples is extremely low, 0.12 W/m-K. In bulk form, TiTe₂ is a semimetal with an electrical conductivity along the *c*-axis that is a factor of 35-40 smaller than the conductivity in the *a-b* plane^{17 18 19}. The electrical conductivities in the *a-b* plane of our thin film samples is 4100-4600 ohm⁻¹ cm⁻¹, a factor of ≈ 2.3 smaller than *a-b* conductivity of bulk¹⁷ TiTe₂. If the anisotropy of electrical conductivity our thin film samples were the same as in bulk TiTe₂, we would expect an electronic contribution to the *c*-axis thermal conductivity of 0.09 W/m-K. Given the turbostratic microstructure of our thin film samples, however, we expect that the anisotropy of electrical conductivity will be significantly larger than in

the bulk, and the electronic contribution to the c -axis thermal conductivity of TiTe_2 will be significantly smaller than 0.09 W/m-K.

To gain insight into the effect of grain size on the lattice thermal conductivity of Bi_2Te_3 , we follow previous work²⁰ and construct a Debye-Callaway (D-C) model using the procedure described by Morelli et al.²¹. To constrain the parameters of the D-C model, we estimate the cutoff frequencies ($\theta_L = 96$ K and $\theta_T = 62$ K) by the acoustic phonon frequencies at the zone boundary ($f_L = 2.0 \times 10^{12}$ Hz; $f_T = 1.3 \times 10^{12}$ Hz) from calculations of the phonon density of states^{7,22}. We calculate the relative ratio of the anharmonic scattering strengths of umklapp and normal processes, B_U and B_N , using equations 10 and 25 (with $a = 2$, $b = 1$) from Reference 21: $B_U = \hbar \gamma^2 / (M v^2 \theta)$ and $B_N = k_B \gamma^2 V^{1/3} / (M v^3)$ for the longitudinal and transversal propagation modes. Here \hbar is the reduced Planck constant, k_B is the Boltzmann constant, $\gamma_L = 1.0$ and $\gamma_T = 0.7$ are the mode Grüneisen constants, $M = 2.66 \times 10^{-25}$ kg and $V = 3.38 \times 10^{-29}$ m³ are the average mass and volume per atom, v is the speed of sound and θ is the cutoff frequency. Phonon scattering by isotope disorder calculated with equation 16 of Reference 21 is negligible ($\Gamma = 8.21 \times 10^{-5}$). A boundary scattering rate $\tau^{-1} = v / h$ describes phonon scattering by grain boundaries and hetero-interfaces; v is the speed of sound and h is the grain size. We estimated the polycrystalline average rigidity modulus G and bulk modulus K from elastic properties²³ of single crystal Bi_2Te_3 using the Voigt-Reuss-Hill method^{24,25}, i.e., the arithmetic mean of G and K moduli calculated by the Voigt and Reuss approaches. We calculated the transversal and longitudinal polycrystalline average speeds of sound as $v_T = \sqrt{G / \rho} = 1590$ m/s and $v_L = \sqrt{(K + 4G / 3) / \rho} = 2840$ m/s, where $\rho = 7.86$ g/cm³ is the mass density of Bi_2Te_3 .

Our implementation of the D-C model does not include heat transport by optical phonons and, because of the relatively low cut-off frequencies for the acoustic branches, greatly restricts the number of acoustic phonons that contribute to heat transport in the model; in fact, <10% of the $3N$ vibrational modes of the crystal are included in this approach. To account for heat transport by phonons that are neglected by the model, we estimate the thermal conductivity of these phonons using the calculated minimum²⁶ thermal conductivity Λ_{\min} of Bi_2Te_3 .

To fit the strength of anharmonic phonon scattering in the D-C model, the calculations of the D-C model are added to the minimum thermal conductivity, $\Lambda_{\min} = 0.31 \text{ W/m-K}$, and then compared to data for bulk²⁷ Bi_2Te_3 , see Fig. 3. For the calculations, we vary the coefficients B_U and B_N while keeping their relative ratio fixed and set the grain size of the sample to a large value $h = 4 \text{ mm}$ (i.e. single crystal sample). The values we obtain thus are $B_{UL} = 9.7 \times 10^{-18} \text{ s/K}$, $B_{UT} = 23 \times 10^{-18} \text{ s/K}$; $B_{NL} = 14 \times 10^{-18} \text{ s/K}$, $B_{NT} = 39 \times 10^{-18} \text{ s/K}$.

In Figure 4, we compare the thermal conductivity Λ measured for the various Bi_2Te_3 layers to the predictions of the D-C model as a function of grain size. This plot includes data for homogeneous, nanocrystalline Bi_2Te_3 that we previously presented as a function of annealing temperature; see Fig. 1 of Ref. 11. For comparison, we also include in Fig. 4 examples of other previous work^{28,29} on small grain Bi_2Te_3 .

To extract the effective thermal conductivity of the nanoscale Bi_2Te_3 layers in the multilayer $[(\text{Bi}_2\text{Te}_3)_m(\text{TiTe}_2)_n]$ samples, see Fig. 1b, we ignore possible contributions to the thermal resistance from the $\text{Bi}_2\text{Te}_3/\text{TiTe}_2$ interfaces and apply a simple effective medium model that treats the thermal resistance of the multilayer as the sum of the

thermal resistances of the individual layers; i.e., we solve for the thermal conductivity of the Bi₂Te₃ layers Λ -Bi₂Te₃ in the equation

$$\frac{x+y}{\Lambda - \text{multilayer}} = \frac{x}{\Lambda - \text{Bi}_2\text{Te}_3} + \frac{y}{\Lambda - \text{TiTe}_2}, \quad (1)$$

where x and y are the individual layer thicknesses in the multilayer repeat period and Λ -TiTe₂= 0.12 W/m-K.

The Bi₂Te₃ data closely follow the trend in the lattice thermal conductivity predicted by the D-C model. As the grain size is reduced toward 2 nm, the lattice thermal conductivity approaches the lower limit predicted by the model of the minimum thermal conductivity, 0.31 W/m-K. Similar values are observed³⁰ in short period superlattices of Bi₂Te₃/Sb₂Te₃. For grain sizes on the order of ~50 nm, our data and modeling indicate that the reduction in thermal conductivity compared to the reported bulk values²⁷ is observable but small, \approx 13%. This conclusion was also reached by Shi^{31,32} and co-workers from their studies of the thermal conductivity of 50 nm diameter Bi₂Te₃ nanowires.

We emphasize that our measurements are for the total thermal conductivity but the D-C model does not include the electronic contribution. We estimate the electronic contribution to the thermal conductivity of the nanocrystalline films from measurements of the in-plane electrical conductivity of Bi₂Te₃ films deposited on fused-quartz substrates using the same recipe as the films that were deposited on Si for the thermal conductivity measurements. (TDTR measurements require high thermal conductivity substrates.) Electrical measurements in the a - b plane showed the films are n -type Bi₂Te₃; Seebeck coefficient and electrical conductivity varied with increasing annealing time

from -83 to -137 $\mu\text{V/K}$ and 436 to 1482 $(\text{ohm-cm})^{-1}$, respectively. We estimate the electronic component of the thermal conductivity Λ_e using the Wiedemann Franz law with a non-degenerate Lorenz number $L = 2(k_B/e)^2 = 1.45 \times 10^{-8} \text{ V}^2/\text{K}^2$ and electrical conductivity equal to $1/4$ the average of the a - b plane values^{33,34}. We add the result, $\Lambda_e \approx 0.11 \text{ W/m-K}$ to the D-C model and plot the sum as the dashed line in Fig. 4. The data fall below this curve indicating either that the anisotropy in electrical conductivity is greater than a factor of 4 or that the D-C model overestimates the lattice thermal conductivity of our nanocrystalline Bi_2Te_3 films.

Figure 4 also summarizes our measurements of the thermal conductivity of $(\text{Bi,Sb})_2\text{Te}_3$ layers as a function of grain size. As before, we extracted the effective thermal conductivity of nanoscale $(\text{Bi,Sb})_2\text{Te}_3$ layers from measurements on multilayer $[(\text{Bi,Sb})_2\text{Te}_3]_m(\text{TiTe}_2)_n$ films (Figure 2b) using an effective medium model and the measured thermal conductivity of TiTe_2 :

$$\frac{x + y}{\Lambda - \text{multilayer}} = \frac{x}{\Lambda - (\text{Bi,Sb})_2\text{Te}_3} + \frac{y}{\Lambda - \text{TiTe}_2}. \quad (2)$$

Average measured thermal conductivity of the $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ films is 0.36 W/m-K , a factor of 1.9 smaller than that of homogeneous Bi_2Te_3 layers with similar grain size. Electrical measurements in the a - b plane of the $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3$ films deposited on fused-quartz substrates indicate n -type conduction: Seebeck coefficient and electrical conductivity varied with increasing annealing time from -23 to -113 $\mu\text{V/K}$ and 109 to 211 $(\text{ohm-cm})^{-1}$, respectively. As before, we estimate the electronic component of the thermal conductivity using the Wiedemann-Franz law with a non-degenerate Lorenz number and electrical conductivity equal to $1/4$ the average of the a - b plane values³⁵. $\Lambda_e = 0.02 \text{ W/m-K}$. The average lattice thermal conductivity obtained by subtracting the small

electronic component from the measured thermal conductivity is a factor of ≈ 2 smaller than the lattice thermal conductivity of Bi_2Te_3 layers predicted by the D-C model. The magnitude of this reduction is comparable to what is observed in bulk $(\text{Bi,Sb})_2\text{Te}_3$ alloys^{36,37}. Our observations are confirmed by recent reports on p-type $\text{Bi}_{0.5}\text{Sb}_{1.5}\text{Te}_3$ nanocrystalline bulk alloys^{38,39} that estimate lattice thermal conductivity ≈ 0.3 W/m-K in the limit of low grain size ($d < 10$ nm).

The reduction in lattice thermal conductivity of $(\text{Bi,Sb})_2\text{Te}_3$ alloys is typically attributed to phonon Rayleigh scattering created by variations in atomic mass and chemical bonding when Bi atoms are replaced by Sb. Calculations of point defect scattering by mass disorder are relatively straightforward but scattering rates created by variations in bond-length or bond-strength disorder are difficult to estimate; furthermore, even if the cross sections could be estimated, we do not know if the total cross sections for the variations scattering terms (mass, bond-length, bond-strength) should be added or if interference between the various scattering terms needs to be taken into account. Therefore, to gain insight in the magnitude of the reduction in thermal conductivity that might be created by point defect scattering in a $(\text{Bi,Sb})_2\text{Te}_3$ alloy, we add to the D-C model what we believe is a reasonable upper limit to the point defect scattering rate^{40,41}, $\Gamma = 0.3$. The result is included in Fig. 4. In this calculation, the lattice thermal conductivity is suppressed by a factor of 1.3 for grain sizes $d \approx 50$ nm; adding point defect scattering to the model has little effect in the limit of small grain size, $d < 10$ nm, where the thermal conductivity has already been reduced close to the minimum value.

Data for the alloy layers, however, fall significantly below the calculation in the limit of small grain size and therefore also fall well below the prediction of the model of

the minimum thermal conductivity. We do not yet understand the difference in conductivity between Bi_2Te_3 and $(\text{Bi,Sb})_2\text{Te}_3$ alloy layers at small grain sizes, $d < 10$ nm, where the effects of point defect scattering should be small; we can only speculate at this time that the effective thermal conductivities of the $(\text{Bi,Sb})_2\text{Te}_3$ layers in the alloy multilayers $[(\text{Bi,Sb})_2\text{Te}_3]_m(\text{TiTe}_2)_n$ are being suppressed by the effects of interface resistance that are stronger in the alloy multilayers than in the $(\text{Bi}_2\text{Te}_3)_m(\text{TiTe}_2)_n$ multilayers.

Conclusions

Reducing the grain size of homogenous Bi_2Te_3 to a few nanometers causes the thermal conductivity to approach the minimum thermal conductivity limit. A Debye-Callaway model for the phonon scattering rates fits the experimental data for a range of grain sizes 1-100 nm reasonably well. Extremely low thermal conductivities can be achieved by combining the effects of alloying and turbostratic disorder in a multilayer structure.

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Figures

Figure 1. Cross-plane thermal conductivity of $[(\text{Bi}_2\text{Te}_3)_m(\text{TiTe}_2)_n]$ ($m = 2-6, n=2-6$) films. Data are plotted as a function of (a) TiTe_2 atomic fraction (calculated as $3n/(5m+3n)$); and (b) thickness of the multilayer repeat unit. Each data point is labeled by a $(m;n)$ index for the film where m is the number of Bi_2Te_3 quintet layers and n is the number of TiTe_2 sheets in the repeat unit. Also included is the average measured thermal conductivity for the pure TiTe_2 samples.

Figure 2. Cross-plane thermal conductivity of $[(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3]_m (\text{TiTe}_2)_n$ ($x = 0.55 \pm 0.09, m = 2-4, n= 3$ and 6) films. Data are plotted as a function of (a) TiTe_2 atomic fraction (calculated as $3n/(5m+3n)$); and (b) thickness of the multilayer repeat unit. Each data point is labeled by a $(m;n)$ index for the film where m is the number of $(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3$ quintet layers and n is the number of TiTe_2 sheets in the repeat unit. Also included is the average measured thermal conductivity for the pure TiTe_2 samples.

Figure 3. Plot of experimental lattice thermal conductivity of single crystal Bi_2Te_3 (Reference 27, filled circles) fitted with predictions of the Debye-Callaway (D-C) model described in the paper (solid line).

Figure 4. Thermal conductivity Λ measured for homogeneous Bi_2Te_3 (full circles) and $(\text{Bi,Sb})_2\text{Te}_3$ alloy (full triangles) plotted as a function of grain size or layer thickness h . Also included are literature values for small grain polycrystalline Bi_2Te_3 samples (References 28 – open square and 29 – filled squares). The open circles and triangles represent effective Λ for Bi_2Te_3 and respectively $(\text{Bi,Sb})_2\text{Te}_3$ alloy layers calculated from measured Λ of multilayer $[(\text{Bi}_2\text{Te}_3)_m(\text{TiTe}_2)_n]$ (Figure 1b) and $[(\text{Bi}_x\text{Sb}_{1-x})_2\text{Te}_3]_m (\text{TiTe}_2)_n$ (Figure 2b) films. The solid line is the D-C model calculation of lattice thermal conductivity of Bi_2Te_3 with the assumption of a boundary scattering length that is equal to the thickness of the sample. Minimum thermal conductivity of Bi_2Te_3 is added in the model calculation to account for heat transport by high frequency acoustic and optical phonons. The dashed line represents the calculated total thermal conductivity of Bi_2Te_3 with contributions from D-C model and the electronic thermal conductivity. The dash-dot line is the D-C model calculation of lattice thermal conductivity of $(\text{Bi,Sb})_2\text{Te}_3$ alloy with the addition of strong point-defect phonon scattering, $\Gamma=0.3$.











