APPENDCES C

PUBLICATIONS

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Electronic structure and thermoelectric properties of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) by first principle calculation

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Abstract

The electronic structure and thermoelectric properties of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) were systematically investigated by the first-principles calculations and Boltzmann transport theory based on QUANTUM ERPREESO and BoltzTraP. According to the calculated electrical conductivity, Seebeck coefficient and thermal conductivity with respected to empirical scattering by substituted Bi by Sb for rhombohedral structure exhibit diverse anisotropic thermoelectric properties. It is found that the $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) show n- and p-type substituting better thermoelectric performance compared to Bi_2Te_3. We would like to be beneficial to stimulate further theoretical and experimental works.

Selection and/or Peer-review under responsibility of SACT 2016.

Keywords: Thermoelectric properties; Boltzmann transport theory; $Bi_{2-x}Sb_xTe_3$; density functional theory; semiconductor

1. Introduction

Thermoelectric (TE) materials have been earned heat into electricity as well as it much attention for their potential application in power generate and refrigerate [1]. The quality of TE materials is related to a parameter called the dimensionless figure of merit: $ZT = S^2\sigma T / (\kappa_e + \kappa_L)$, where S is the Seebeck coefficient, T is absolute

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temperature, σ is electrical conductivity, κ_e is the thermal conductivity and κ_L is the lattice thermal conductivity [2]. Bismuth Telluride, (Bi_2Te_3) has been significant role thermoelectric technology due to its low Seebeck coefficient, high electrical conductivity and low thermal conductivity [3]. We proposed to improve performance of this material by substitution Antimony in Bi-site. In addition, the TE properties of TE materials can be investigated by the Boltzmann transport theory [4, 5]. In this work, we studies. Theoretical substituted Bi by Sb to investigate the electronic structure and enhanced TE properties of Bi_2Te_3 material by using density functional theory and BoltzTraP simulation.

2. Computational details

The $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) cluster models were designed by using a space group number 166, lattice parameters a = 4.38 Å, b = 4.38 Å and c = 30.49 Å, as show in Fig. 1. The electronic structure was calculated by the density functional theory based on QUANTUM ESPRESSO [6]. The exchange correlation function, energy convergence limit set as 10^{-8} Ry, and energy cutoff 40 Ry are performed [7]. The density of states was calculated using 8×8×1 k-mesh and 4000 k-points in the Brillion zone. The TE properties, such as, Seebeck coefficient, electrical conductivity, and thermal conductivity was calculated using Boltzmann transport theory based on BoltzTraP [4, 6], the equation can be written as;

$$\sigma(T;\mu) = \frac{1}{\Omega}\int \sigma(\varepsilon)\left[-\frac{\partial f_\mu(T;\varepsilon)}{\partial \varepsilon}\right]d\varepsilon\,, \tag{1}$$

$$\nu(T;\mu) = \frac{1}{eT\Omega}\int \sigma(\varepsilon)(\varepsilon-\mu)\left[-\frac{\partial f_\mu(T;\varepsilon)}{\partial \varepsilon}\right]d\varepsilon \tag{2}$$

$$S = E(\nabla T)^{-1} = (\sigma^{-1})\nu \tag{3}$$

where Ω is unit cell, ν is the band velocity, μ is the chemical potential, ε is band energy, f_μ is the Fermi function and E is electromotive force. Since they are no experimental data, we only give the electrical conductivity, electronic thermal conductivity and thermoelectric power factor in terms of τ. The electronic thermal conductivity has similar outlines to electrical conductivity, which is because that the electrical thermal conductivity is connected with electrical conductivity by the Wiedemann-Franz law;

$$\kappa_e = \sigma LT = \frac{\kappa_e}{\tau_r}\times\tau \tag{4}$$

$$\kappa = \kappa_e + \kappa_L \tag{5}$$

where L is the Lorenz number, κ is total thermal conductivity τ is empirical scattering time and τ_r is true scattering time.

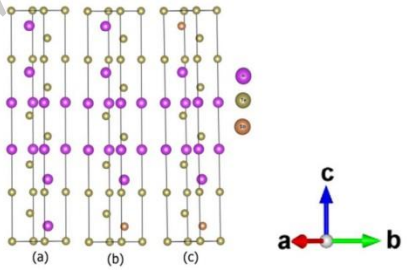

Fig. 1. Cluster atom model (a) Bi_2Te_3, (b) $Bi_{1.67}Sb_{0.33}Te_3$, and (c) $Bi_{1.33}Sb_{0.67}Te_3$.

3. Results and discussion

The total density of states of $Bi_{2-x}Sb_xTe_3$ (0, 0.33, and 0.67) are shows in Fig. 2. The formation energy of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) as shown in Table 1. Both results show semiconductor behavior composed of electron and hole motive, which effected to thermoelectric properties.

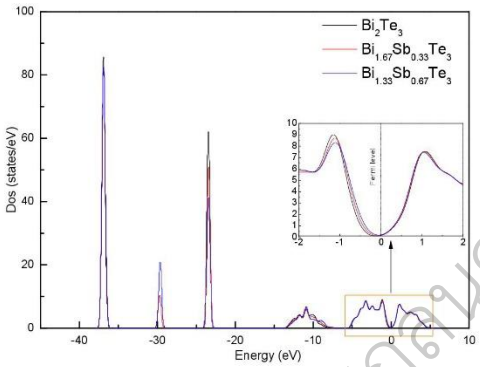

Fig. 2. Total density of states of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67).

The Seebeck coefficient with respected to scattering time of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) at temperature of 300 K as shown in Fig. 3(a). The maximum value of Seebeck coefficient is 536, 436 and 327 μV K^{-1} occurs at μ = −0.22, −0.26 and −0.29 eV. The Sb substitutes at Bi has been decreased Seebeck coefficient and showed the *p* and *n*-type semiconductors. Electrical conductivity of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) was almost independent of temperature as shown in Fig. 3(b). The maximum value of electrical conductivity of 1.502 × 10^{20}, 1.35 × 10^{20} and 1.41 × 10^{20} $\Omega^{-1}m^{-1}s^{-1}$ occurs at μ = −2.4, −2.0 and −1.9 eV at temperature of 300 K. The entire substitutes Sb at Bi is decrease electrical conductivity from 1.505 × 10^{20} $\Omega^{-1}m^{-1}s^{-1}$ to 1.39 × 10^{20} and 1.41 × 10^{20} to 1.34 × 10^{20} $\Omega^{-1}m^{-1}s^{-1}$ (at 300 K).

Table 1. The formation energy of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67).

Formation energy	Bi_2Te_3 (Ry)	$Bi_{1.67}Sb_{0.33}Te_3$ (Ry)	$Bi_{1.33}Sb_{0.67}Te_3$ (Ry)
Total energy	−100048.04	−9617.47	−9186.90
Total all-electron energy	−380673.68	−350575.03	−320476.37
One-electron contribution	−1281.75	−1314.63	−1347.50
Hartree contribution	771.13	785.60	800.06
XC contribution	−657.69	−674.83	−691.96
Ewald contribution	−1648.60	−1648.60	−1648.60
One-center paw contribution	−7231.13	−6765.02	−6298.92

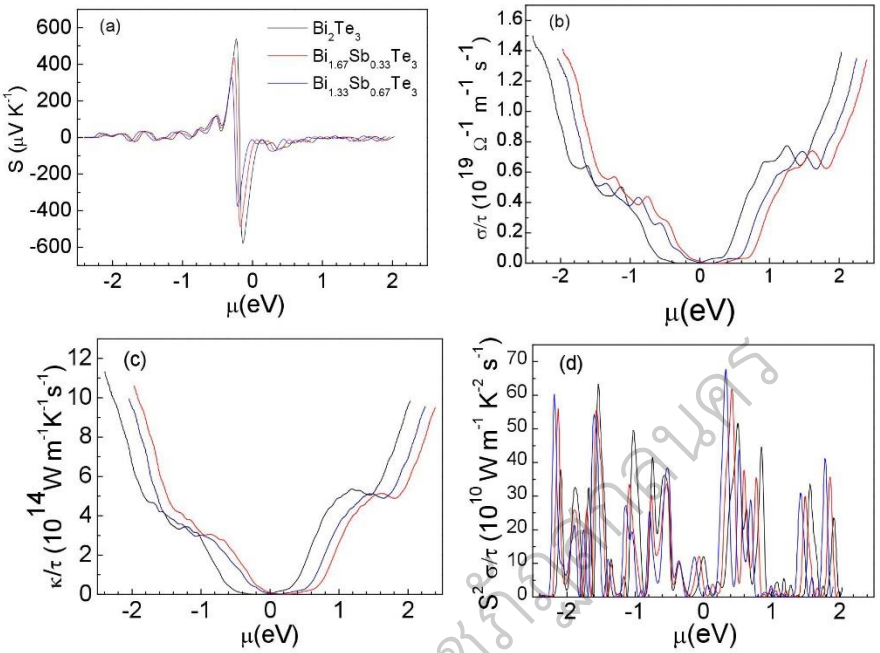

Fig. 3. (a) The (a) Seebeck coefficient, (b) electrical conductivity, (c) thermal conductivity and (d) power factor of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) with respected scattering time.

The substitution Sb at Bi as 1 atom and 2 atom are found to value electronic thermal conductivity decrease from 11.32×10^{14} to 9.82×10^{14} W m $^{-1}$ K s, 10.58×10^{14} to 9.50×10^{14} W m $^{-1}$ K s and 9.92×10^{14} to 9.54×10^{14} W m^{-1} K s at (300 K) as shown in Fig. 3(c). The thermoelectric power factor of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) plot at temperature 300 K as shown in Fig. 3(d). The maximum value of thermoelectric power factor of (~63.33×10^{10}, 61.81×10^{10} and 67.752×10^{10} W m^{-1} K^{-2} s^{-1}) at $\mu = -1.54$, 0.41, 0.32 eV.

4. Conclusion

The theoretical calculated electronic structure and thermoelectric properties of $Bi_{2-x}Sb_xTe_3$ (x = 0, 0.33, 0.67) have been investigated by using first principle calculation and Boltzmann transport theory. The thermoelectric properties substitutes Sb at Bi have been increased electrical conductivity and electronic thermal conductivity, while it decreased power factor at 300 K.

Acknowledgements

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