Thermoelectric Transport Properties of Tin Selenide from First-principles Calculations

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Introduction

- Thermoelectricity is the direct conversion of heat into electricity and vice versa, i.e Seebeck and Peltier effect.
- Seebeck effect – phenomenon in which temperature difference creates an electric potential: used in thermoelectric generators (TEGs).
- Peltier effect – is the evolution or absorption of heat at the junctions of two dissimilar materials due to a potential difference.
- The efficiency of the thermoelectric materials is determined by dimensionless figure of merit given by:

$$ZT = \frac{S^2 \sigma T}{k_l + k_e}$$

- S is the Seebeck coefficient, σ is the electrical conductivity, T is the operating temperature, k_e and k_l are the electron and lattice thermal conductivities, respectively.
- For high ZT a thermoelectric material should have:
  -high Seebeck coefficient (S)
  -high electrical conductivity (σ)
  -low thermal conductivity ($k_e + k_l$)

Purpose of the Research

- This study aims to investigate the thermoelectric properties of SnSe at different carrier concentration levels between 300 K and 800 K using theoretical methods.
- The results from this work will aid in complementing the experimental observations.

Why Tin Selenide (SnSe)

- Lead chalcogenides are the best TE materials at mid-high temperatures while Bismuth chalcogenides perform best at room temperature (Kumar et al., 2016). However, lead is toxic limiting large scale application while Bismuth and Tellurium are expensive.
- Tin Selenide (SnSe) which is lead free hence non-toxic and composed of relatively abundant elements could be an ideal alternative for thermoelectric applications.

Methodology

- Package: Viena Abinitio Simulation Package (VASP) for all electronic structure calculations
- Functional: Generalized Gradient Approximation (GGA) in the form of Perdew-Burke-Enzerhof (PBE) exchange correlation functional was used.
- Projector-Augmented Wave Pseudopotentials for treating the Sn & Se valence electrons.
- Kinetic energy cutoff was set at 520 eV. The brillouin zones were intergrated by a 10 x 10 x 2mesh grid.
- The thermoelectric properties were then calculated using BoltzTraP code that solves the Boltzmann transport equation under constant relaxation time (Madsen & Singh, 2006).
- Relaxation time, $\tau = 1 \times 10^{-15}$ s.
- Phono3py code: Lattice thermal conductivity, K_l calculations

Results

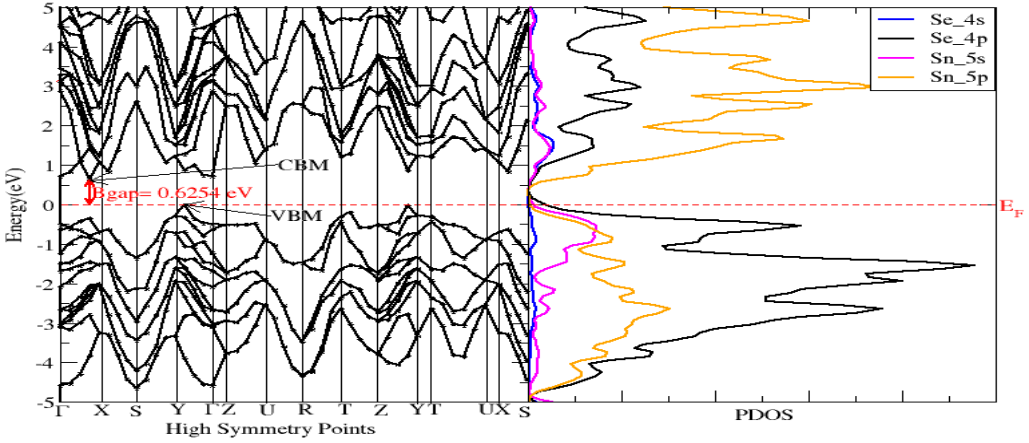

Fig.1: Band structure and pdos structure of SnSe

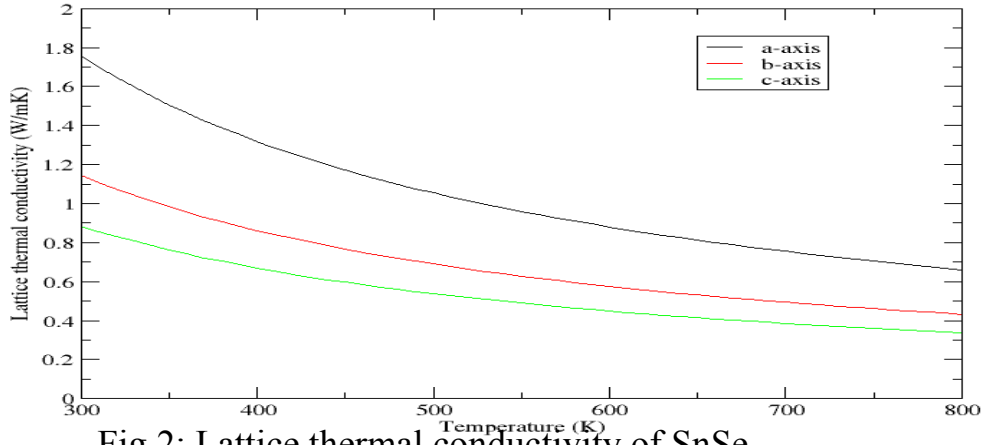

Fig.2: Lattice thermal conductivity of SnSe

- An indirect band gap of 0.625 eV between Γ-X and Y-Γ high symmetry points. This suggests semiconducting properties.
- Experimental band gap value is 0.9 eV (Huang et al., 2017).
- The conduction band is mainly contributed by the Sn-5p and Se-4p states while the Sn-5s states have almost no contribution to the conduction band.
- Se-4p states are more dominant the valence band than the Sn-5p while the Sn-5s and Se-4s states offer very little contribution to the valence band

- Lattice thermal conductivity is anisotropic and deacreases with temperature increase from 1.756, 1.146 and 0.0885 W/mK at 300 K to 0.66, 0.432 and 0.338 W/mK along a, b and c axis, respectively.
- Experimental k_l values reported to be as low as 0.20 W/mK

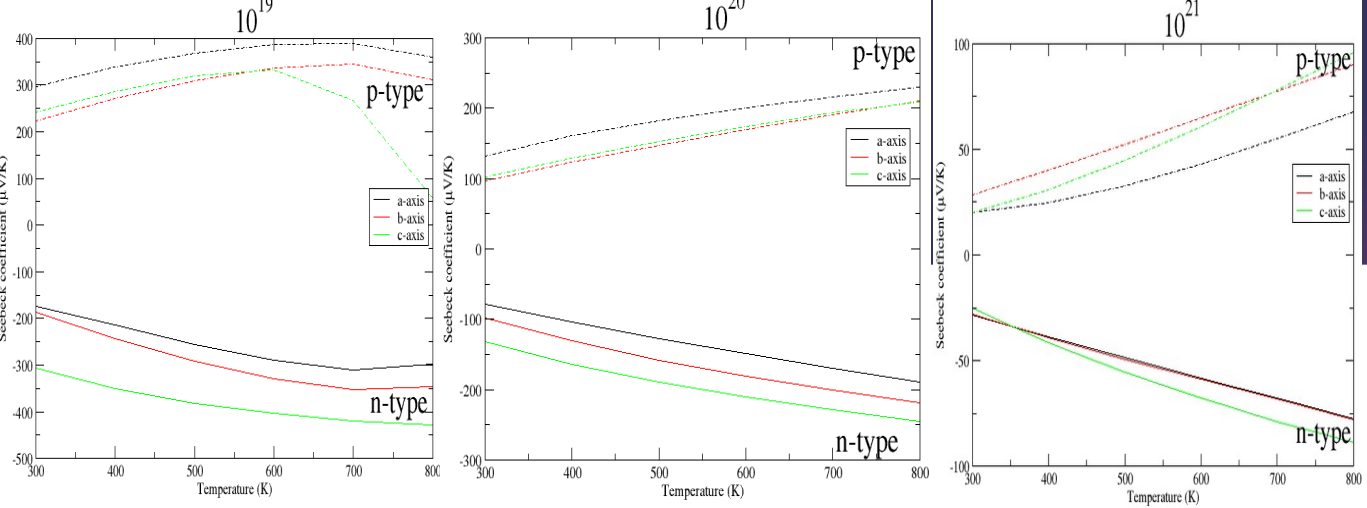

Fig. 3: Seebeck Coefficient (μV/K) for p-type (dotted lines) and n-type (solid lines) SnSe at different carrier concentration levels.

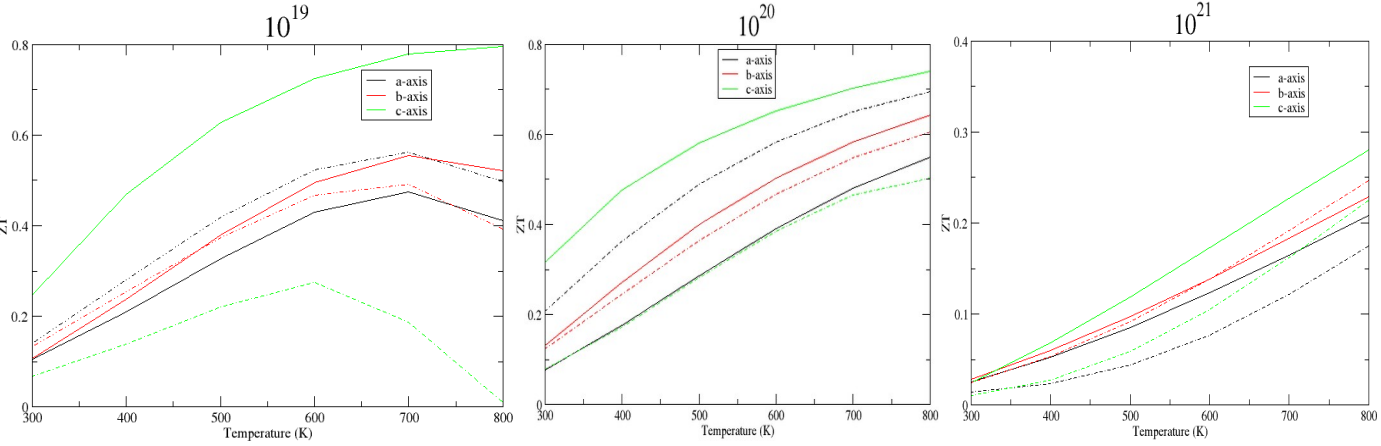

Fig. 4: Calculated figure of merit (ZT) as a function of temperature at different carrier concentration levels

- From Fig. 3, The Seebeck Coefficient increases with temperature for both p-type and n-type SnSe.
- The values were found to be as high as 428μV/K at 800 K in n-type SnSe.
- For the p-type SnSe the highest value of Seebeck coefficient was found to be +390μV/K at 600 K along the a-axis which then decreases gradually as the temperature increases to 800 K.

- In Fig. 4 ZT increases with temperature along all axes for both p and n-type SnSe.
- N-type (solid lines) SnSe exhibits higher ZT values than p-type(dotted lines).
- ZT values higher at 10^{19} cm^3 carrier concentration levels.
- Upper limit values of ZT found at 0.795 (c_axis) at 800K for n-type and 0.562 (a-axis) at 700 K for the p-type SnSe.

Conclusion

- The first-principles calculations for structural, electronic and thermoelectric properties of SnSe have been insightful. The lattice constants were found to be in good agreement with the experimental values and the computed band structure shows that the material is an indirect band gap semiconductor with the band gap (0.625 eV) found to be in close agreement with other DFT reported values but slightly lower than experimental value (0.9 eV).
- The results of transport properties indicate that SnSe has desirable thermoelectric properties and hence is an ideal candidate for low cost and environmentally benign thermoelectric applications.

THANK YOU!