Effects of structural arrangements on thermoelectric properties of SiX (X=N,P,As,Sb,Bi) monolayers

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INTRODUCTION

Thermoelectric materials are being explored extensively in recent times as they are capable of generating electricity from waste heat of environmental sources. The efficiency of a thermoelectric material at a given temperature T is determined by its figure of merit $ZT = \frac{S^2\sigma}{\kappa}T$; S, σ, κ are Seebeck coefficient, electrical conductivity and thermal conductivity, respectively. The target of most research on thermoelectric materials is to increase ZT upto 3-4. In this work, we present first-principles based investigations into monolayers of hexagonal IV-V semiconductor SiX (X=N,P,As,Sb,Bi) family. We find that the transport properties are substantially affected by atomic arrangements in the planes of the monolayers, an effect observed earlier [1] in another family of two-dimensional materials.

METHODOLOGY

The band structure calculations are done using Density Functional Theory (DFT) as implemented in Vienna Ab initio Simulation Package (VASP) employing the Projector Augmented Wave (PAW) basis set with Generalised Gradient Approximation (GGA) exchange-correlation potential. The band structures are then used along with semi-classical Boltzmann Transport theory to compute the transport parameters S, σ and κ using BoltzTrap2 and ShengBTE packages.

RESULTS AND DISCUSSIONS

Two different stacking patterns, configuration(1) in which the Si atoms occupy the inner planes and configuration(2) where the X atoms occupy the inner planes are used to compute the effects of atomic arrangements on the thermoelectric properties. We find that the non-conventional stacking pattern (configuration(2)) leads to a maximum ZT of 3.23 at 800 K for electron doped SiP. Overall, the ZT obtained in configuration(2) are 2-6 times higher than those obtained in stacking pattern given as configuration(1). This is due to the combined effect of lower lattice thermal conductivity and higher electronic conductivity in configuration(2). The origin of this lies in the flatness of the bands, weaker bonds and increased anharmonicity in the low frequency acoustic modes when X atoms occupy the inner planes of the SiX monolayers.

The theromoelectric conversion efficiency η of the compounds which is related to the Carnot efficiency are then calculated using ZT

$$\eta = \frac{T_h - T_c}{T_h} \frac{\sqrt{1+ZT}-1}{\sqrt{1+ZT}+T_c/T_h} \qquad (1)$$

T_h, T_c are the temperatures of the hot and cold edges of the thermoelectric device, respectively. We find that SiP in configuration(2) has an efficiency of 27% when operated between 300-800K. This efficiency is comparable to that of a home refrigerator.

CONCLUSIONS

Hexagonal Si-X monolayers show substantial dependence of their thermoelectric efficiencies on the structural arrangements. This work demonstrates a route for experimentalists to tweak the stacking to effect significant gains in thermoelectric efficiencies in two-dimensional materials.

REFERENCES

[1] A. Majumdar, S. Chowdhury, and R. Ahuja, *Drastic reduction of thermal conductivity in hexagonal AX monolayers due to alternative atomic configuration*, Nano Energy **88**, 106248 (2021).

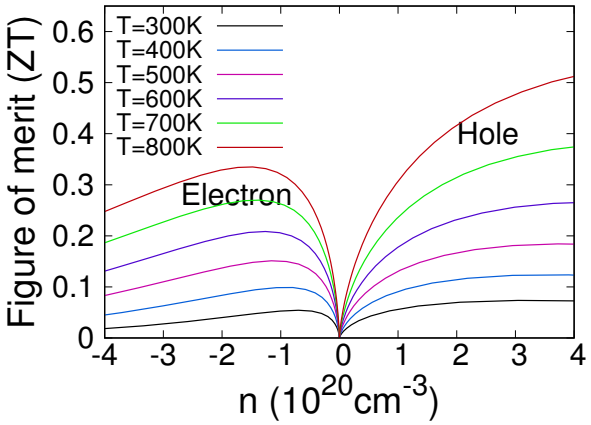

Fig. 1. Thermoelectric figure of merit for SiP in configuration(1)

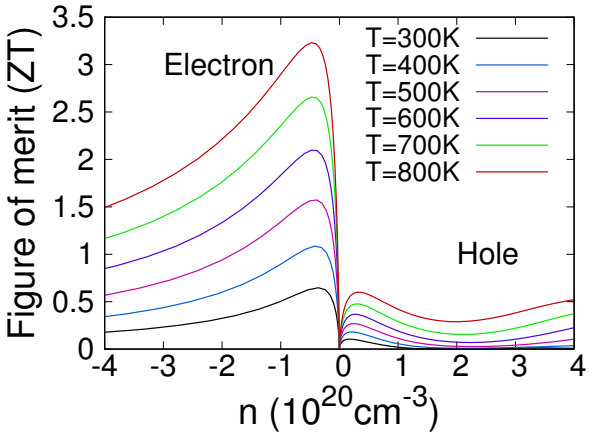

Fig. 2. Thermoelectric figure of merit for SiP in configuration(2)

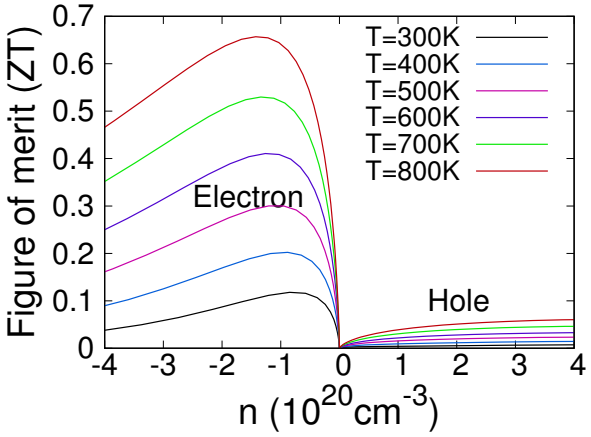

Fig. 3. Thermoelectric figure of merit for SiAs in configuration(1)

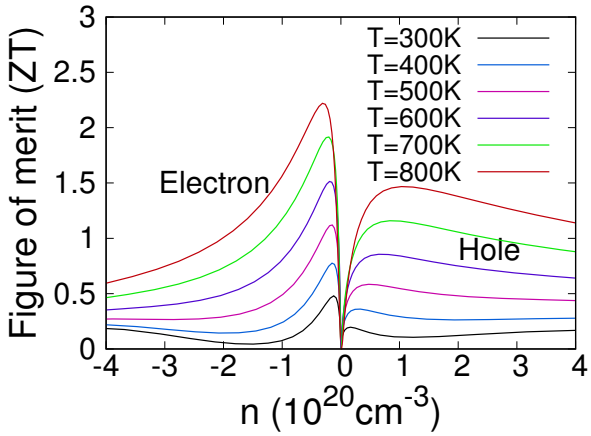

Fig. 4. Thermoelectric figure of merit for SiAs in configuration(2)

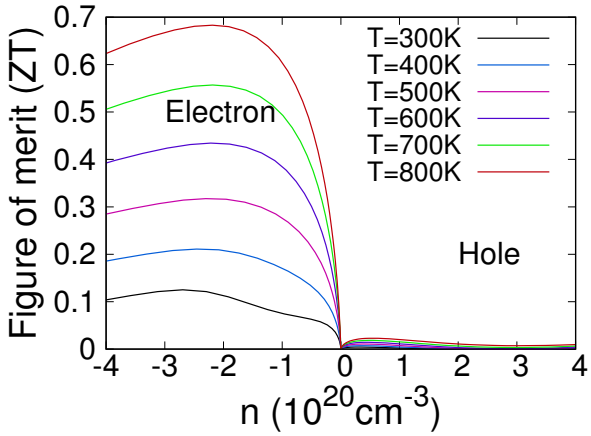

Fig. 5. Thermoelectric figure of merit for SiSb in configuration(1)

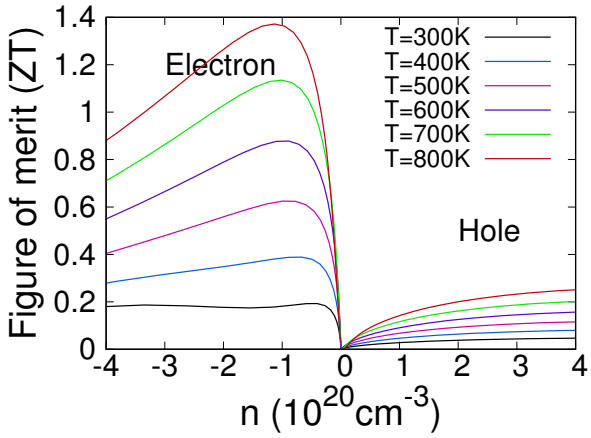

Fig. 6. Thermoelectric figure of merit for SiSb in configuration(2)