Na_2KSb: A promising thermoelectric material

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Na_2KSb: A Promising Thermoelectric Material

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Abstract. The present study reports the thermoelectric (TE) properties of Na_2KSb, and identified it as a promising material for TE applications. The electronic structure calculations are performed using the the full potential linearized augmented plane wave (FP-LAPW) within the frame work of density functional theory, and the transport properties are calculated by solving the Boltzmann transport equation within the constant relaxation time approximation. The obtained lattice parameters are in good agreement with the available experimental and other theoretical results. Thermoelectric properties like thermopower (S), electrical conductivity scaled by relaxation time (σ/τ) and power-factor ($S^2\sigma/\tau$) are calculated as functions of the carrier concentration and temperature, and both holes and electron doping are found to be favourable at ambient conditions. The effect of hydrostatic compressive and tensile strain on electronic and thermoelectric properties is examined. A significant enhancement in power-factor is observed in the case of electron doping around 10% compressive strain, which might enhance the TE performance.

INTRODUCTION

The research on thermoelectric materials is focused towards exploring novel thermoelectric material and to further improve the efficiency of existing TE materials [1,2]. Thermoelectric materials can directly convert the waste heat to electricity, which is very essential for the current world. The aptness of the TE materials can be understood from the dimensionless figure of merit $ZT=S^2\sigma T/\kappa$, where S, σ, κ, and T are thermopower, electrical conductivity, thermal conductivity, and absolute temperature respectively. κ includes both the electronic κ_e, and lattice contributions κ_l i.e, $\kappa=\kappa_e+\kappa_l$. In general, doped semiconductors are prospective materials for good TE performance. In the search of novel TE materials, we have focused on bi-alkali antimonide Na_2KSb, which possess semiconducting nature. Alkali and bi-alkali antimonides are investigated for several applications including photo-emissive and sensing devices [3,4]. In addition, these compound also fall into the well -known family of zintl phases. In general, these compound are stable dynamically and possess low thermal conductivity, which is beneficial for TE applications. The previous study on electronic structure of these compounds proposes the enhanced capability of photon absorption, which leads to photocathode applications [5]. The present study further focuses on the electronic and thermoelectric properties of Na_2KSb at ambient and strained state.

COMPUTATIONAL DETAILS

The FP-LAPW method as implemented in the Wien2k package [6] is used for the electronic structure calculations. The TB-mBJ [7] functional was used to obtain the electronic structure properties since the more traditional exchange correlation functional severely underestimate the band gap of semi-conductors. Total energy calculations were carried out for a k-grid of (36x36x36) which results in 1240 k-points in the irreducible part of the Brillouin zone. The optimized theoretical lattice parameters are used to get the thermoelectric properties such as thermopower (S), electrical conductivity scaled by relaxation time (σ/τ) and power-factor($S^2\sigma/\tau$) using the BoltzTraP code [8]. The lattice parameter is systematically changed for strain calculations.

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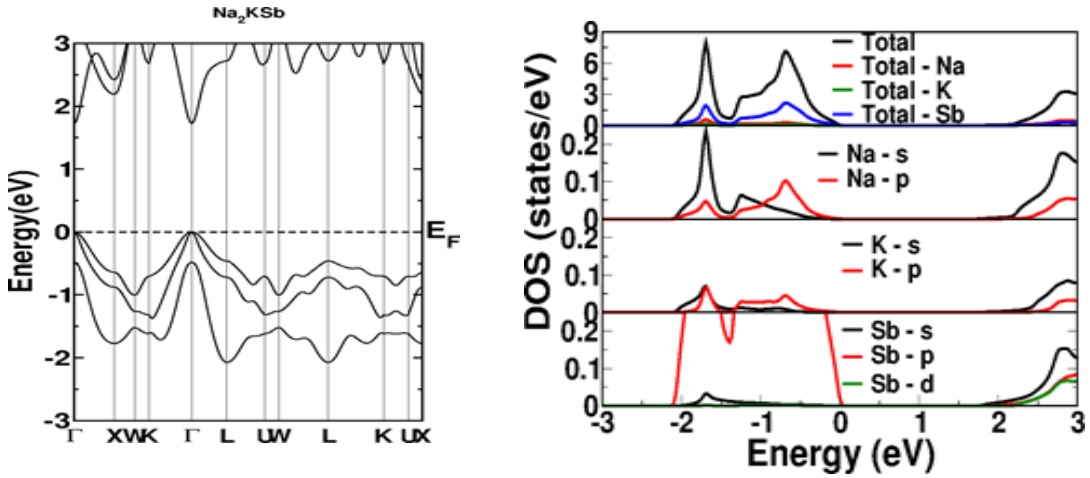
RESULTS AND DISCUSSION

(a) Structural and Electronic properties

The crystal symmetry of the investigated compound is cubic with space group Fm-3m (225). The optimized lattice parameter 'a' is 7.79 Å, is in good agreement with the experimental value of 7.72 Å [9]. Further we have proceeded with the optimized value of lattice parameter. The calculated band structures along the high symmetry points of the Brillouin zone and the density of states (DOS) for Na_2KSb are presented in Figure 1. The calculated band gap using TB-mBJ functional is found to be direct with 1.73 eV, and reported experimental value is 1.1 eV [9]. The dispersion of bands in the conduction band is found to be higher than the same in valence band, which we have verified through effective mass calculations. The calculated effective mass in the unit of electron mass for holes is 1.44 me, and for electrons it is 0.36 me. Combinations of light and heavy band masses are observed near the Fermi level in valence band. To further understand the contributions of atoms near the Fermi level, we have plotted total and partial density of states of Na_2KSb, and presented the same in Figure 1 (b). From the figure, it is quite clear that, in valence band Sb -p states are dominating and in conduction band we could see almost similar contribution from Na, K, Sb 's' and 'p' states. One vital point to note is the variation of density of states near the Fermi level in valence band being steeper compared to the conduction band. Overall from the electronic structure, it is evident that the thermopower values of holes will be higher than the electrons due to the presence of more heavy bands, and electrical conductivity scaled by relaxation might be higher for electron doping. To tune the electronic structure and hence the TE properties, we have applied hydrostatic compressive and tensile strain to Na_2KSb. The band gap is found to be altered with strain, while the band profile remained the same. The band gap is found to increase with compressive strain and the same is found to decrease with tensile strain.



(a) (b)

FIGURE 1. (a) Calculated band structure and (b) density of states of Na_2KSb

(b) Thermoelectric properties

The TE properties of Na2KSb are calculated using the BoltzTrap code. The thermopower (S in μV/K), the electrical conductivity scaled by relaxation time (σ/τ in $\Omega^{-1}m^{-1}s^{-1}$) and the power-factor ($S^2\sigma/\tau$ in $Wm^{-1}K^{-2}s^{-1}$) are calculated as functions of carrier concentrations for both electrons and holes at different temperatures. Calculated thermopower for hole and electron doping are presented in Figure 2 (a, b). The magnitude of thermopower is found to decrease with carrier concentration for both hole and electron doping. Hole doping is found to possess more thermopower values than the electron doping, which was evident from the band structure and density of states near the Fermi level. We have analysed the TE properties for wide temperature range of 300 K to 900 K. Figure 2 (c, d) presents the variation of electrical conductivity scaled by relaxation time (will call as electrical conductivity) as a function of carrier concentration and temperature. The magnitude of electrical conductivity is found to be increased with carrier concentration and decreased with temperature. To understand the net contribution from electronic part for the thermoelectric properties, we have analysed the power factor, which is presented in Figure 2 (e, f). Overall the

power factor for holes and electrons are found to be almost same, and the range of power factor is comparable with reported TE materials. Next let us discuss the effect of hydrostatic strain on TE properties. For that, we have presented the thermopower value for different hydrostatic strain in Figure 3(a, b).

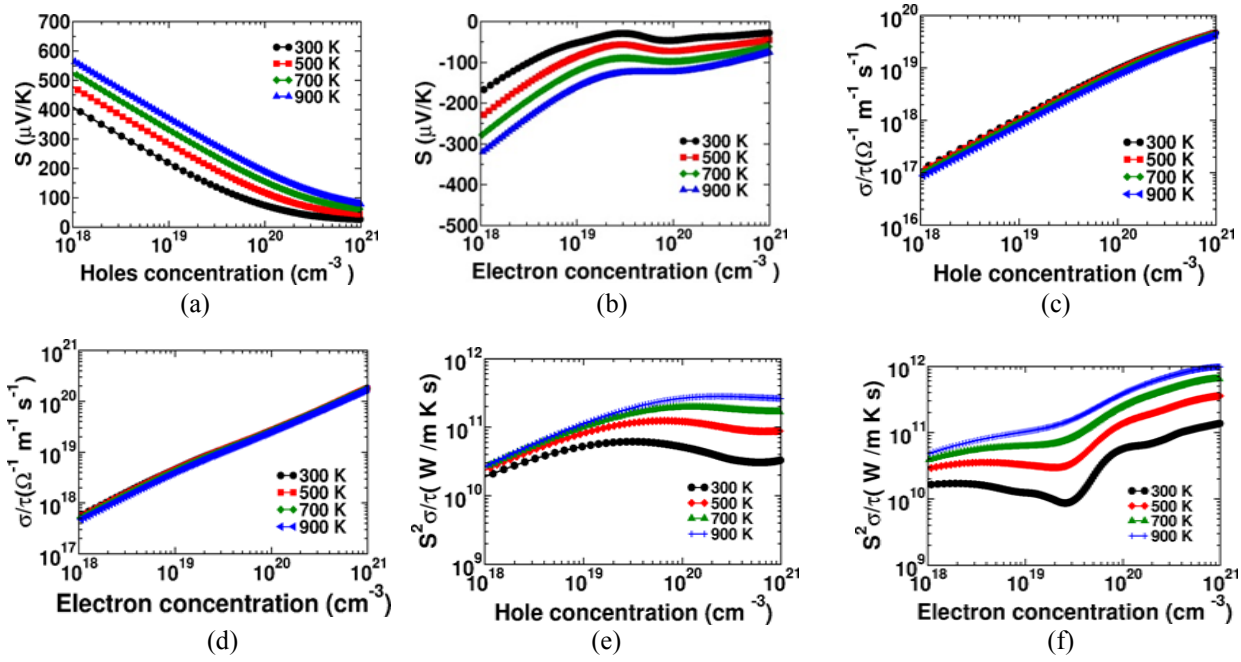


FIGURE 2. (a, b) Variation of thermopower, (c, d) electrical conductivity scaled by relaxation time and (e, f) power-factor with holes and electrons.

For hole doping we could not see any significant change in transport properties, except small shift in the value of thermopower. But for electron doping, for compressive strain around 10 %, we could see an enhancement in power-factor, and this might be due to the formation of less dispersed band as a function of strain. For electrical conductivity we have observed a detrimental effect. Overall the power factor for electron doping is found to be enhanced in the compressive strained state. For a wide concertation range the power factor is found to be similar, which is beneficial for device applications. The magnitude of thermopower for hole doping is found in the range of 550μV/K, which is comparable with established TE material. For electron doping the magnitude is found around 300 μ V/K. In the case of electrical conductivity, electron doping has secured higher values, and the magnitude is comparable with other TE material. Overall present study proposes a novel material for TE application for a wide temperature range.

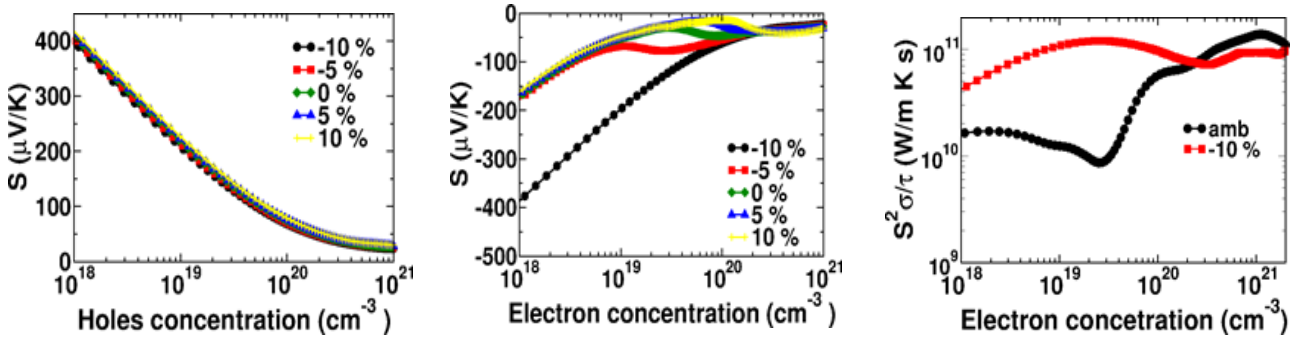


FIGURE 3**, (**a) thermopower for holes, (b) thermopower for electrons, (c) Power–factor for electron as a function of strain

CONCLUSIONS

The electronic and thermoelectric properties of Na2KSb were calculated within the frame work of density functional theory. To our knowledge the thermoelectric properties of this compound is analyzed for the first time. Promising thermoelectric properties were observed at ambient. Further we have enhanced the power factor, which is found to be almost constant for a wide concentration range using compressive hydrostatic strain in Na2KSb.

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