Supplemental Material: Thermoelectric properties of marcasite-type compounds MSb_2 (M = Ta, Nb): A combined experimental and computational study

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I. COMPUTATION DETAILS

In this Supplemental Material (SM), we provide the electronic structure calculations using spin-orbit coupling (SOC) implemented in Quantum Espresso code [1]. Transport properties are calculated using BoltzTraP package [2]. A heavy k-mesh of size $20 \times 40 \times 20$ is used for the transport calculations. The experimental transport properties are compared with the calculated results of non-SOC and SOC.

II. RESULTS AND DISCUSSION

Fig. S1(a) and (c) show the calculated band-structure of $TaSb_2$ and $NbSb_2$, respectively. A very small effect is observed by the inclusion of SOC. Fig. S1(b) and (d) exhibit the density of states (DOS) of said compounds, respectively using SOC. Fig. S2 represents the comparison of experimental Seebeck coefficient (S) with calculated one. For $TaSb_2$, the non-SOC and SOC give the similar trend of S, while for $NbSb_2$, the non-SOC give more closer match of S with experiment as compared to SOC. A significant change in the electrical conductivity per relaxation time ($\boldsymbol{\sigma}/\tau$) are observed for both compounds by inclusion of SOC in Fig. S3. For $TaSb_2$, the non-SOC electronic part of thermal conductivity (κ_e) provides closer match as compared to SOC as shown in Fig. S4(a). Fig. S5 shows the power factor per relaxation time as a function of chemical potential using SOC. The maximum power factors for p-type conduction are calculated as $\sim$1.01 and $\sim$1.12 mW m^{-1} K^{-2} at ~ -375 and ~ -320 meV, respectively at 300 K. These values are found to be $\sim$1.41 and $\sim$1.53 mW m^{-1} K^{-2} at $\sim$215 and $\sim$215 meV for n-type of $TaSb_2$ and $NbSb_2$, respectively at 300 K. Fig. S6(a) and (b) exhibits the calculated direction-dependent S for $TaSb_2$ and $NbSb_2$, respectively.

III. ACKNOWLEDGEMENTS

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[1] P. Giannozzi *et al.*, J. Phys. Condens. Matter **21,** 395502 (2009).
[2] G. K. H. Madsen and D. J. Singh, Comput. Phys. Commun. **175,** 67 (2006).

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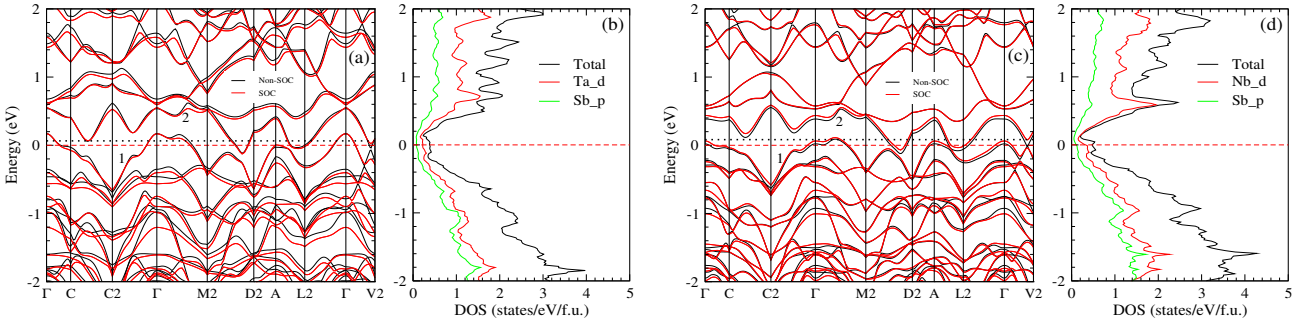

FIG. S 1: (a) Band-structure and (b) density of states (DOS) of $TaSb_2$; (c) band-structure and (d) DOS of $NbSb_2$. The coordinates of the high symmetry k-points are: Γ (0 0 0), C (0.27 0.29 0), C_2 (-0.29 0.7 0), M_2 (-0.5 0.5 0.5), D_2 (0.26 0.26 0.5), A (0 0 0.5), L_2 (0 0.5 0.5) and V_2 (0 0.5 0).

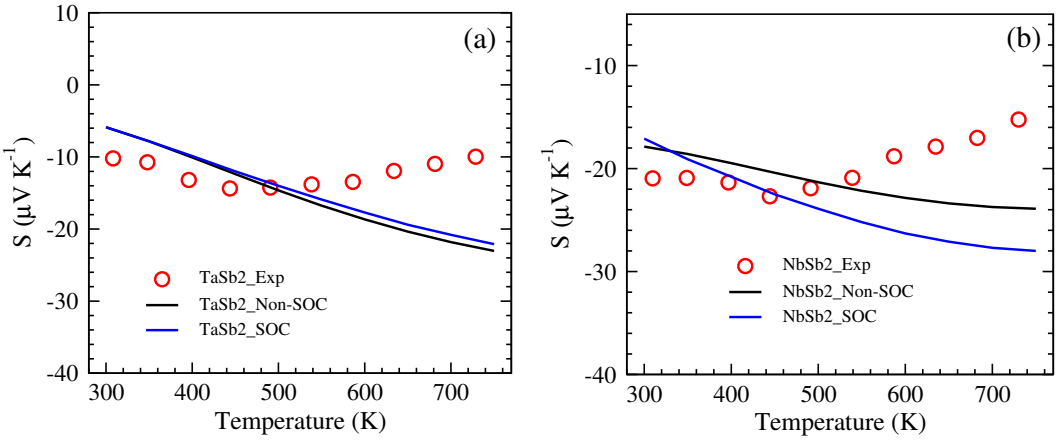

FIG. S 2: Comparison of experimental and calculated Seebeck coefficients, S of (a) $TaSb_2$ and (b) $NbSb_2$.

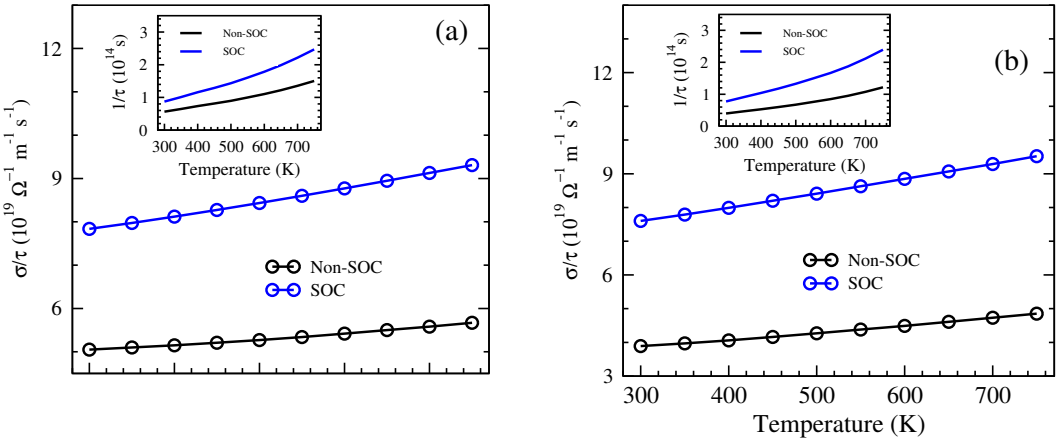

FIG. S 3: Calculated electrical conductivity divided by relaxation time, σ/τ of (a) $TaSb_2$ and (b) $NbSb_2$. Inset shows $1/\tau$ as a function of temperature, which is estimated by comparing the calculated σ/τ with experimental σ.

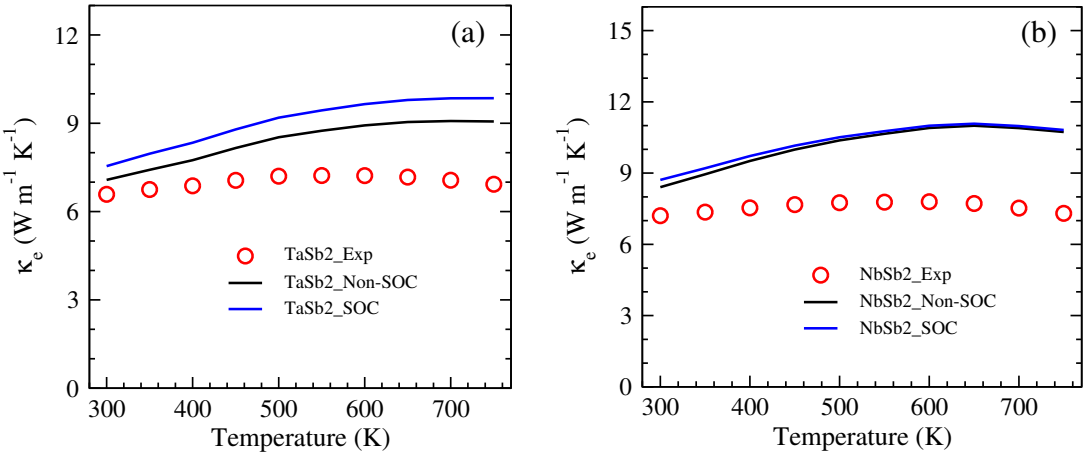

FIG. S 4: Comparison of experimental (estimated using Wiedemann-Franz law) and calculated electronic part of thermal conductivity, κ_e of (a) $TaSb_2$ and (b) $NbSb_2$.

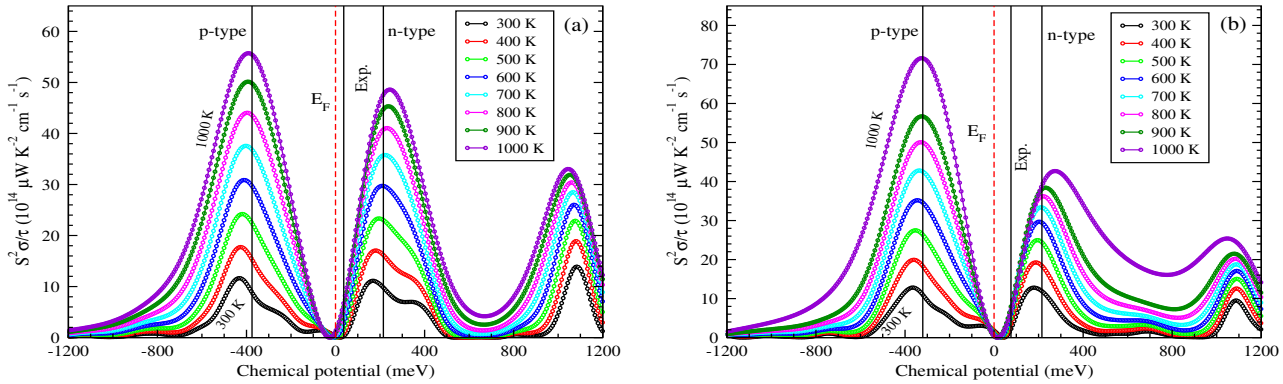

FIG. S 5: Variation of power factor ($S^2\sigma$) per relaxation time with chemical potential at different temperatures of (a) $TaSb_2$ and (b) $NbSb_2$.

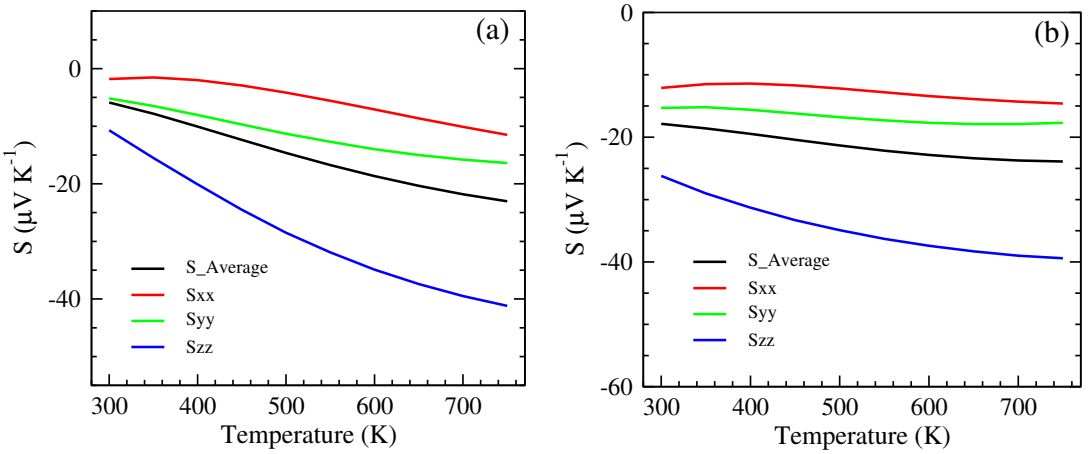

FIG. S 6: Calculated direction-dependent Seebeck coefficient, S for (a) $TaSb_2$ and (b) $NbSb_2$, obtained using Non-SOC.