Layered dichalcogenides as efficient Thermoelectric Materials

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Experimental studies on TiS_2 demonstrate a thermopower of -251 μV/K [1], which is commensurate with the thermopower of bulk Bi_2Te_3. Motivated by the recent focus on layered dichalcogenide materials and studies on two-dimensional crystals [2] we derive the thermoelectric parameters of TiS_2 of one trilayer (TL) thickness.

Our theoretical model uses *ab-initio* electronic structure calculations as implemented in the VASP software package combined with a Landauer approach to calculate thermoelectric transport coefficients. We employ an energy independent electron mean free path for the scattering within the device.

TiS_2 crystallizes in a simple layer-type structure, it consists of three atomic layers (S - Ti - S) arranged along the z-direction. Each trilayer is separated by weak van der Waals forces. Figure 1 shows the atomic trilayer structure being simulated. Figure 2 shows the ab initio electronic band structure of the TL film. The optimized lattice parameter is 3.418Å and the optimized thickness for the TL film is 3.15Å.

Figure 3 shows the density of modes calculated for the TL using the VASP electronic structure calculations. The density of modes, i.e the distribution of conducting channels in energy, is calculated by integrating over the first Brillouin zone. Thermoelectric coefficients are then calculated for the TL film at room temperature. The results for the Seebeck coefficient (S) and the thermoelectric figure of merit (ZT) of the trilayer are shown in Figure 4 and 5 respectively. These results are obtained using an electron mean free path of 20 nm and hole mean free path of 11 nm.

Bulk TiS_2 has a reported in-plane lattice thermal conductivity of 6.78 W/mK. Applying this to our Landauer evaluation of the thermoelectric coefficients provides a maximum ZT of 1.02. However, thermal conductivity measurements of thin film dichalcogenides have demonstrated a reduction in the lattice thermal conductivity by a factor of 3 compared to the bulk [3]. Applying similar measurements to TiS_2 trilayers could possibly reveal an improvement in ZT.

Imai at. al report a ZT of 0.16 for bulk TiS_2. Our DFT/Landauer approach shows an approximate factor of 10 improvement in ZT for trilayer TiS_2 while assuming bulk lattice thermal conductivity. ZT in the trilayer film can be enhanced by taking into consideration changes in the phononic band structure, which leads to a reduction in the lattice contribution to thermal conductivity.

REFERENCES

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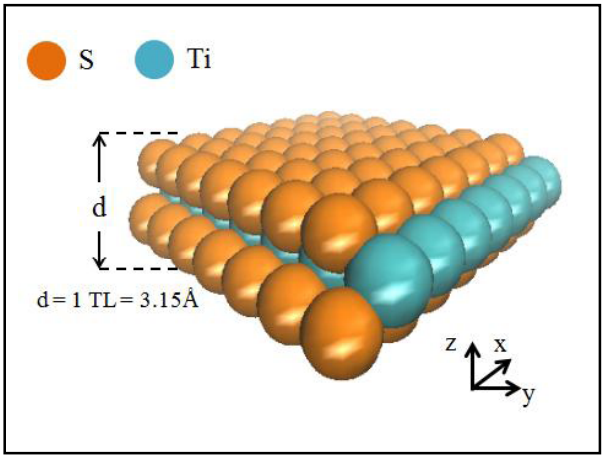

Fig. 1. Atomic structure of free-standing TiS_2 trilayer

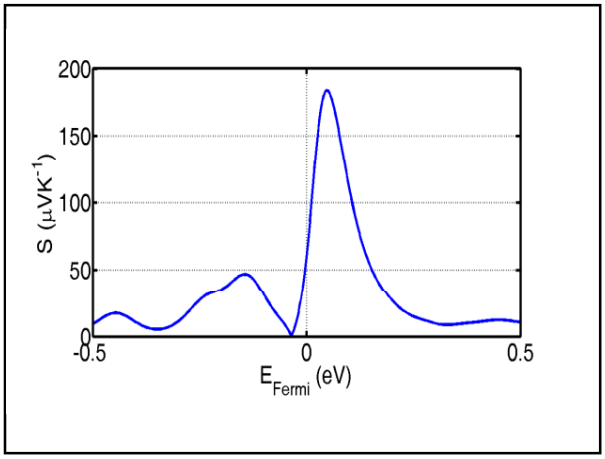

Fig. 4. Seebeck coefficient (S) at 300K for TiS_2 trilayer

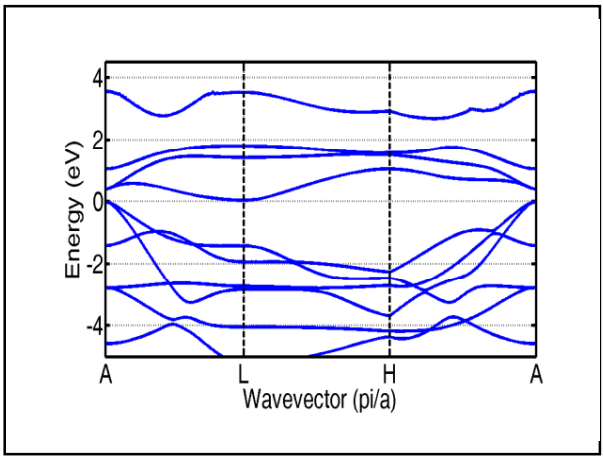

Fig. 2. Electronic structure for TiS_2 trilayer

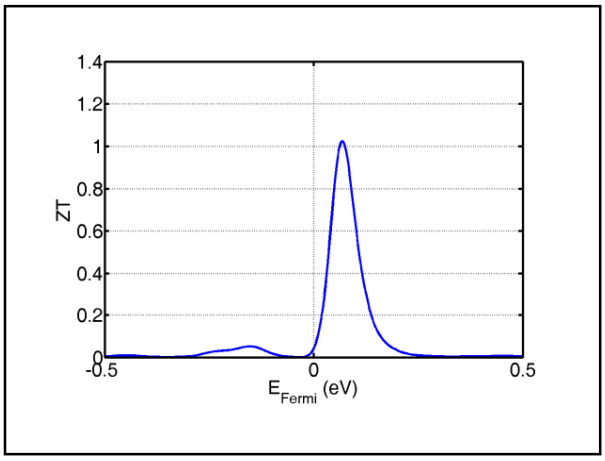

Fig. 5. Figure of Merit (ZT) at 300K for TiS_2 trilayer

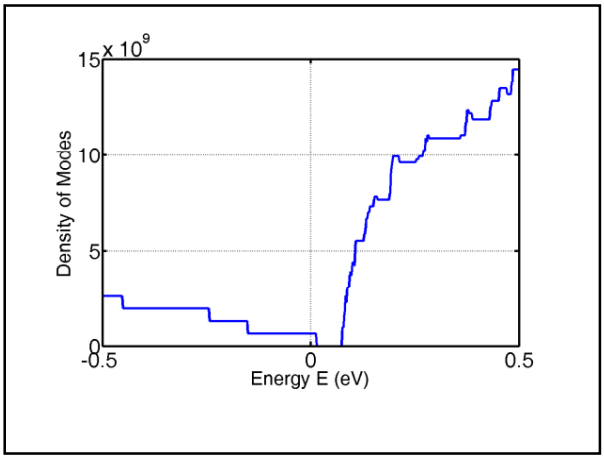

Fig. 3. Density of modes M(E) for TiS_2 trilayer