

Functionalized TaS₂ for thermoelectric applications: an ab-initio investigation

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INTRODUCTION

Thermoelectricity (TE) is a very promising field of research where wasted heat is converted into electricity through the Seebeck effect. The TE efficiency is closely connected to the dimensionless figure of merit ZT that can be written as (1)

$$ZT = \frac{\sigma S^2}{k} T \quad (1)$$

ZT depends on the electrical conductivity (σ), the Seebeck coefficient (S) and inversely on the thermal conductivity (k). Therefore, efficient TE materials should have high σ and S , and low k . In this scenario, many efforts are currently ongoing to optimize ZT by increasing S and and/or by reducing k .

Among different strategies, 2D materials show promising features for TE applications due to their relatively high electronic conduction properties [1]. However, 2D materials still exhibit large thermal conductivities, hindering their possible TE applications. For this reason, great efforts have been recently devoted to reduce their thermal conductivity. A recent experimental study [2] showed that the 2D TaS₂ functionalization with specific covalently-bonded organic side-chains (CBOs) leads to a drastic thermal conductivity reduction without affecting the electronic properties. However, the role of the CBOs in affecting k has not fully yet been clarified. The focus of this work is to investigate this system by proposing a model that comprehensively explains the reduction of thermal conductivity.

METHODS

All the calculations have been performed by means of first principles DFT calculations using the Quantum Espresso package[3]. The characterization of the vibrational and thermal properties

has been performed by means of the Alamode package[4]. The thermal conductivity is obtained using the Boltzmann Transport Equation within the Relaxation Time Approximation (BTE-RTA).

CONCLUSIONS

The calculations showed a dramatic TaS₂ thermal conductivity reduction upon the CBOS functionalization. We attribute such a significant reduction to i) the increase of the TaS₂ inter-layer separation and ii) the role of CBOs as thermal energy-sinks. We finally explore the use of longer and/or chemically different CBOs to further reduce the thermal conductivity of 2D materials therofre increasing the overall ZT .

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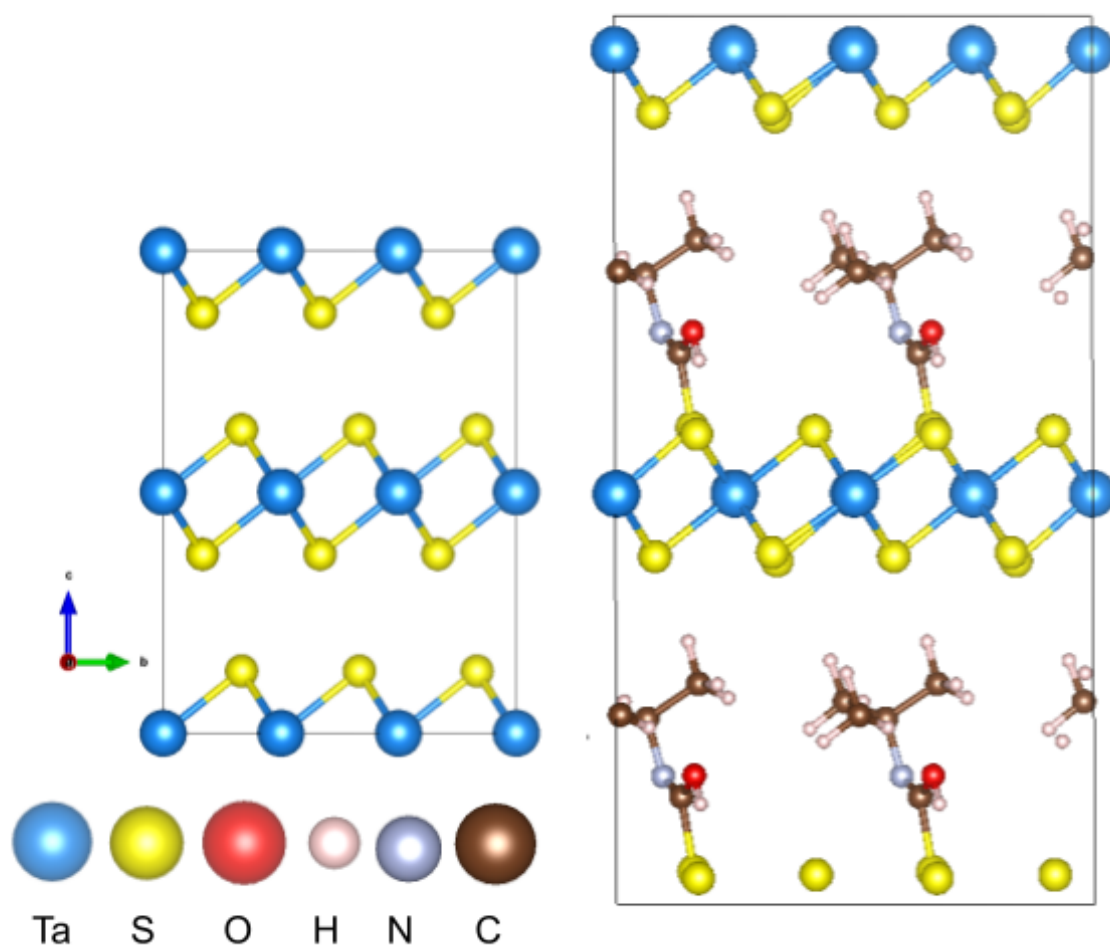


Fig. 1. Left: pristine 2D structure of TaS₂.
Right: CBOSSs-functionalized TaS₂ structure