

Ab initio finite-temperature lattice dynamics calculations of thermal properties in TMDs materials

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Since graphene's first isolation, two dimensional (2D) materials have received extensive attention due to their unique atomically thin structure and novel physical properties. In particular, various Transition Metal Dichalcogenides (TMDs), such as MoS₂, WS₂, MoSe₂ and WSe₂, have been exploited for a wide range of applications from electronic and optoelectronic devices, such as transistors, to energy storage applications [1].

Of particular interest is the change in electrical and optical properties with the reduction of the material thickness from bulk to monolayer [2]. Alongside electronic/optical properties, their thermal transport properties have drawn considerable interest as well.

A deep understanding of the thermal properties of 2D materials is indeed crucial to their implementation in electronic and optoelectronic devices and to provide new directions for thermal energy management. Although the thermal properties of these materials are widely studied, the effect of flake thickness is still under scientific debate, with discrepancies in the thermal conductivity values both in experiments and theoretical works [2] [3].

In this study, we investigated the in-plane thermal conductivity (κ) and the heat capacity of TMDs, key thermal parameters, analyzing in detail the effect of the thickness on thermal properties and comparing with experiments.

We performed first principles calculations within the density functional theory framework as implemented in the SIESTA program [4] and the Temperature Dependent Effective Potential package (TDEP)[5] for finite temperature lattice dynamics calculations. The most important novelty is the investigation of thermal properties using an *ab initio* approach that rigorously includes finite temperature effects, thus allowing to obtain room-temperature transport parameters. The method identifies effective harmonic and cubic interatomic force constants consistent with the thermal displacements and forces of a canonical ensemble at a specified temperature which represents the best possible representation of the Born-Oppenheimer potential energy surface for atomic displacements around the equilibrium positions [6].

By employing this method, we present a systematic study of the specific heat capacity and the thermal conductivity in TMDs at room temperature for different number of layers, from the monolayer up to the bulk structure. Our results are compared to the values measured experimentally [7].

References

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