

I Examined systems

I.1 Silicon

Silicon is the fundamental material in modern transistors and as such is one of the pillars of the digital revolution. Analogue to the Stone, Bronze or Iron Age, the current age of civilization can thus be called the Silicon Age [1]. Consequently, silicon is a well studied material from an experimental and theoretical standpoint. Combining this with the fact that DFT calculations on silicon are not particularly expensive makes it an ideal system for an introduction to DFT calculations as well as a good benchmarking system. Consequently, all benchmarks in this thesis were run on silicon first and with the information gained from that, benchmarks on a more expensive system were run.

The calculations in ?? and ?? were made with a plane-wave cutoff of 70 Ry and on a $40 \times 40 \times 40/6 \times 6 \times 6$ k -point grid respectively. All calculations use a PBE (Perdew-Burke-Ernzerhof [2]) XC-functional with a norm-conserving PP generated using Vanderbilt's method [3].

I.2 TaS₂

Tantalum Disulfide (TaS₂) belongs to the class of Transition Metal Dichalcogenide (TMDC)s. The most common stoichiometry of compounds in this class is MX₂, where M is a transition-metal and X is a chalcogen atom. TMDCs occur with different atomic coordinations. Fig. I.1 shows the structure of trigonal-prismatic TaS₂ (2H-TaS₂), which consists of a hexagonal transition-metal lattice between two hexagonal chalcogen lattices whose atoms are aligned on top of each other. Seen from above, they form a honeycomb lattice.

TMDC's were known and studied as a bulk material since more than five decades [5]. The more recent possibility to do experiments on freestanding monolayers [6] has brought these materials back into focus, as bulk TaS₂ shows superconductivity and formation of charge density waves, so the effect of the reduction of dimensionality on these phenomenon can be studied on them [7].

I.2.1 Charge-density waves

A charge density wave is a periodic modulation of the electronic charge density of a solid. This changes the potential acting on the nuclei, so a distortion of the lattice accompanies the formation of a charge-density wave, to the point where both terms are used interchangeably. Fig. I.2 shows a charge-density wave in 2H-TaS₂. TaS₂ forms charge-density waves in both the bulk case [8] and as a monolayer [9].

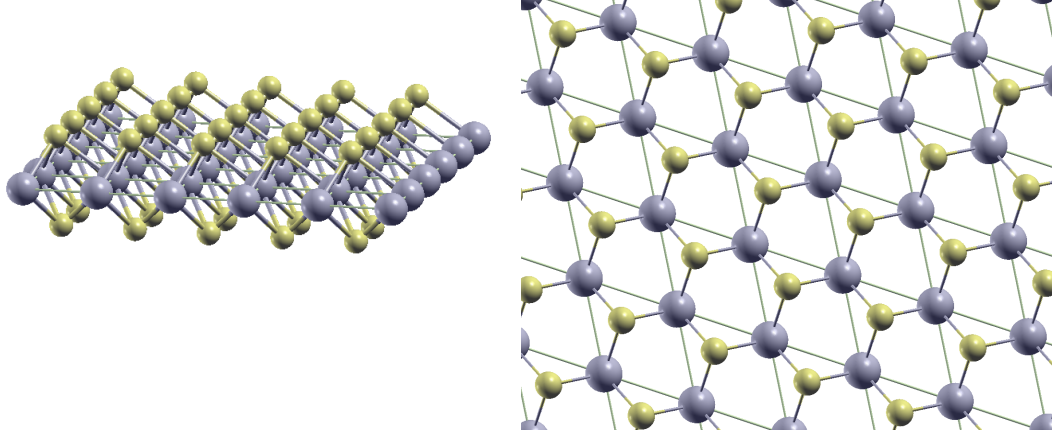


Figure I.1: Crystal structure of a 2H-TaS₂ monolayer as seen from the side (left) and from the top (right). Visualized using XCrySDen [4]

A simple model for the formation of charge-density waves in a one-dimensional case was given by Peierls in 1955. Following the review by Grüner [10], the argument is as follows: the formation of a periodic distortion of the lattice creates a unit cell twice as large as the original one, so the Brillouin zone becomes half as large. Thus, the bands fold back onto this smaller Brillouin zone and split due to an avoided crossing. This results in a gap at the Fermi level.

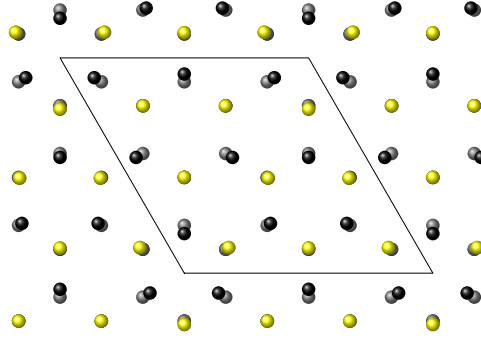


Figure I.2: TaS₂ charge density wave. Gray dots are atoms in the symmetric phase, yellow/black dots are the Tantalum/Sulfur atoms in the charge density wave phase.

I.2.2 Computational parameters

All calculations on the 2H-TaS₂ charge-density wave were made with a plane-wave cutoff of 100 Ry and on a 12×12 k -point grid. The calculations use a PBE XC-functional with a norm-conserving [PP](#) generated by Hartwigsen et al. [[11](#)]. Input files for both the symmetric and charge density wave phase were kindly provided by Dr. Jan Berges.