

# Ab-initio tight binding Hamiltonian generation for 1D and 2D MoS<sub>2</sub> based Devices

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## ABSTRACT

In this work electronic structure of mono layer MoS<sub>2</sub> is analysed by using density functional theory (DFT) and wannierised tight binding approach. The DFT analysis is performed by open source tool Quantum ESPRESSO and wannierised electronic band structure is computed by Wannier 90 tool. First the primitive cell calculations have performed to get the electronic band structure and further the wannier transformation has been carried out to get the band structure. The Cartesian co-ordinate system unit cell is being used to make the tight binding (TB) Hamiltonian which can be easily replicated for the complete device structure. In addition, the DFT calculations along with Wannier transformation are performed for 1D/2D MoS<sub>2</sub> based nano devices. The electronic band structure computed by Wannier transformation is accurately matched with the band structure computed by DFT. The results shows that TB hamiltonian generated by Wannier transformation for 1D/2D MoS<sub>2</sub> based nano devices can be further used in the non-equilibrium Green's function formalism.

**Keywords:** Density functional theory (DFT), Wannier functions, tight binding hamiltonian, MoS<sub>2</sub>

## 1 INTRODUCTION

Due to finite energy gap and comparable electron mobility with graphene, transition metal dichalcogenides particularly molybdenum disulfide (MoS<sub>2</sub>) has shown immense potential for the nanodevices [1-2]. However, to implement the MoS<sub>2</sub> based 1-dimensional (1-D) and 2-dimensional (2D) sub-7 nm nano-devices, simple and accurate model which explores the band structure and electronic properties of the MoS<sub>2</sub> is strongly needed. To investigate the electronic properties of mono-layer to multi-layer structures, ab-initio calculations have been used by various research groups [3-4]. The various group have reported the tight binding models for electronic band structure of monolayer MoS<sub>2</sub>[5] [6]. The few reports have been found on tight binding models for band structure of MoS<sub>2</sub> nanoribbon[7].

Although the reported ab-initio calculations based on the density functional theory (DFT) computes the valuable information about the electronic properties, due to computational limitations for large atom systems the reported ab-initio approaches are not suitable for 1D and 2D device structures. To overcome this problem, tight

binding approximations have been used to simulate the 1D and 2D device structures consisting of 10 to 10<sup>6</sup> numbers of atoms in the channel [8]. Although the tight binding approximation have been used by the researchers for investigating the electronic properties, the ab-initio DFT calculations along with tight binding approximation have not been reported for the MoS<sub>2</sub> based 1D and 2D device structures.

In this work, DFT calculations are performed on 1D/2D MoS<sub>2</sub> device structures. After performing the ab-initio DFT calculations, the Wannier tight binding approach has been used to generate the accurate Hamiltonian. The maximally localised Wannier functions (MLWF) are generated to get tight binding model. The generated Hamiltonian can be further used for the transport calculation performed by the non equilibrium green function (NEGF) for MoS<sub>2</sub> based 1D and 2D device structures.

## 2 SIMULATION SETUP

The DFT computations have been performed by using Quantum ESPRESSO [9-10], which uses self consistent plane wave pseudo potential total energy method. Further, generalized gradient approximation (GGA) proposed by Perdew and Wang (PW91) has been used for exchange correlation potential. The lattice constants considered for MoS<sub>2</sub> are  $a = 3.16 \text{ \AA}$  and  $c = 12.294 \text{ \AA}$  which are fully relaxed until the energy convergence of 10<sup>-9</sup> eV is reached. Further, wave function and charge density cut-offs of 40 Ryd and 300 Ryd have been used respectively. In addition, the optimized structure (coordinates) have been used to perform self consistent calculations with a Monkhorst-Pack 4×4×1 k-mesh followed by the non-self consistent calculations for band structures and density of states(DOS).

Further the tight binding approximation of electronic band structure is accomplished by transforming plane wave basis into maximally localized wave functions (MLWF) [11]. The open source Wannier 90 tool implements this transformation to get tight binding Hamiltonian from plane wave DFT calculation performed in quantum ESPRESSO. In order to perform Wannier unitary transformation, 11 orbitals have been considered which are 7 highest valence bands and 4 lowest conduction bands consisting of d orbitals of metal atom and p orbitals for chalcogen atom. The remaining orbitals are excluded from the calculation as 11 wannier functions are used to in unitary transformation. The d/p orbitals are selected as initial projections so that Wannier functions are closer to localize atomic orbitals.

### 3 RESULT AND DISCUSSION

Fig. 1 (a) and Fig. 1 (b) shows the primitive cell in real space and Fig. 1 (c) shows irreducible Brillouin zone considered for DFT computation. The primitive cell has 1 Molybdenum (Mo) and 2 Sulphur (S) atoms. The DFT calculations have been carried out on primitive cell. Fig. 2(a) shows the band structure of mono layer MoS<sub>2</sub> along path  $\Gamma$ -M-K- $\Gamma$  and Fig. 2 (b) shows the density of state. It is found from Fig. 2 (a) that the mono layer MoS<sub>2</sub> has band gap of 1.8 eV which matches with the experimental results obtained in [12].

The Wannier transformation is performed over the primitive cell which has one Mo atom and two S atom. The Mo and S atoms are bonded by d and p orbitals respectively. The 11 orbitals consists of 6 p and 5 d orbitals. In the Wannier unitary transformation, 11 Wannier functions are implemented on projections which are composed of d and p orbitals. Fig. 3 shows the electronic band structure of monolayer MoS<sub>2</sub> computed by DFT which almost matches with Wannier unitary transformation. The 11 bands comprised of d-p orbitals have been matched with DFT computation. These Wannier transformed tight binding approach have considered first neighbour interaction with 11 MLWF.

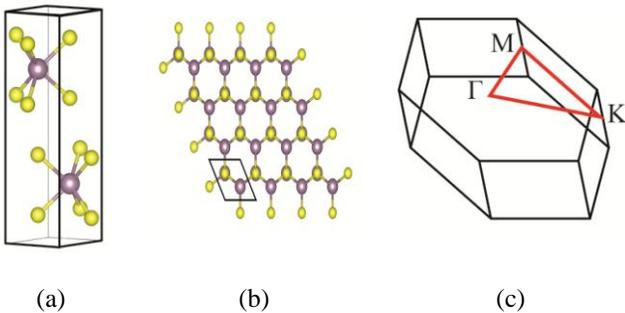


Fig. 1- Primitive cell of monolayer MoS<sub>2</sub> (a) vertical view, (b) top view of and (c) reciprocal space (Brillouin zone)

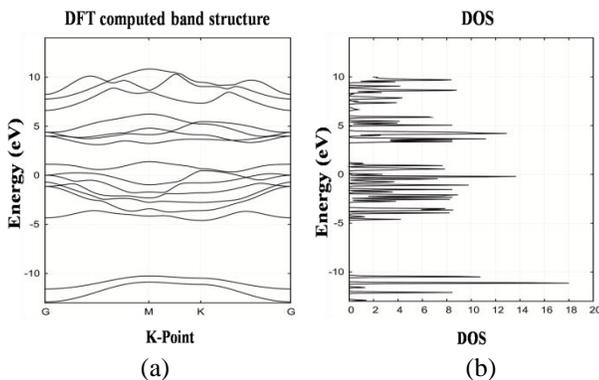


Fig. 2 (a) Electronic band structure (b) density of states of monolayer MoS<sub>2</sub>

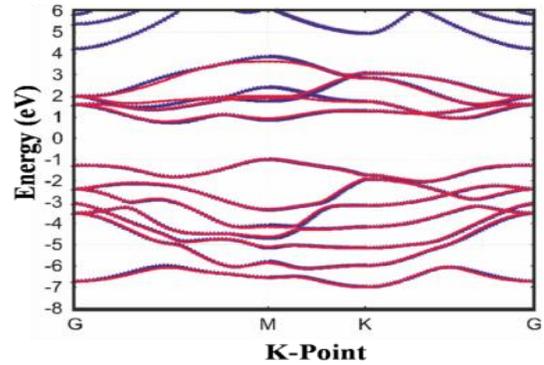


Fig.3- The electronic band structure of monolayer MoS<sub>2</sub> (primitive cell) calculated by DFT (blue) and Wannier transformation (red)

Generally electronic band structure calculations are performed on primitive cell as it has less number of atoms and smallest geometric representation of material due to available inherent periodicity. In order to study the transport of nano devices, there is need to investigate the electronic band structure properties of whole device. Hence it is need of device simulator which can compute electronic band structure of whole device similar to primitive cell. In this work we have considered rectangular co-ordinate system which can be easily implemented to get the geometry of whole device. Fig. 4 shows the schematic view of monolayer MoS<sub>2</sub> based 2-D device structure. The unit cell can be implemented for monolayer MoS<sub>2</sub> based 2D device structure to get tight binding Hamiltonian. Hence by using periodicity of channel material, the tight binding hamiltonian can be implemented for whole device by using the smaller unit cell. This has advantage of reduced computaional effort. Similarly Fig. 5 shows the MoS<sub>2</sub> nanoribbon based 1D nano device structure.

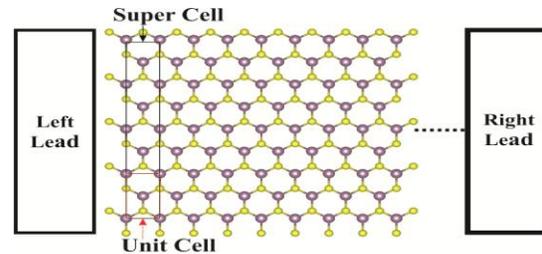


Fig. 4- Schematic view of monolayer MoS<sub>2</sub> based 2-D device structure

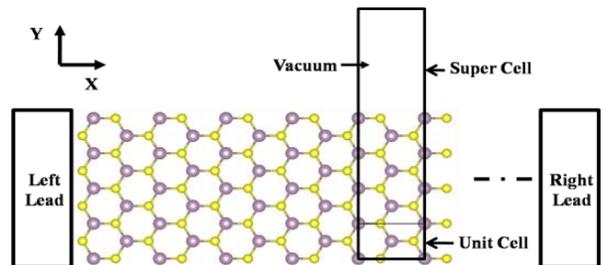


Fig. 5- Schematic view of monolayer MoS<sub>2</sub> based 1-D device structure

Fig. 6(a) and Fig. 6(b) shows the unit cell representation in real space and Fig. 6(c) shows irreducible Brillouin zone used in device structure shown in Fig. 4. The DFT calculations have been carried out for relaxed unit cell. Fig. 7 shows the electronic band structure for this unit cell along K path  $\Gamma$ -Y-S-X- $\Gamma$  shown in Fig 6(c). It has been found from Fig. 7 that the band gap for unit cell have found to be 1.8 eV which is equal to DFT calculation for primitive cell. Further the Wannier unitary transformation calculations are performed for unit cell. The unit cell is composed of two pair of equivalent primitive cell atoms. The twice of primitive cell atoms in unit cell doubles the MLWF required for Wannier unitary transformation. The 22 MLWF are used for Wannier transformation which gives the d orbitals and p orbitals bands. Fig 7 shows that electronic band structure computed by Wannier unitary transformation exactly matches with DFT calculated band structure.

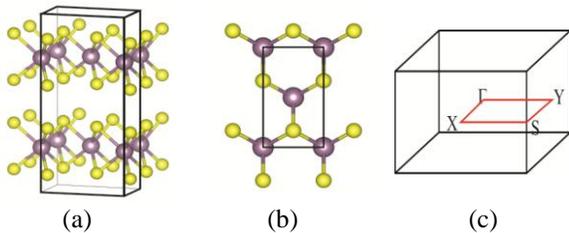


Fig. 6 – Monolayer MoS<sub>2</sub> with unit cell (a) vertical view, (b) top view, (c) reciprocal space (Brillouin zone)

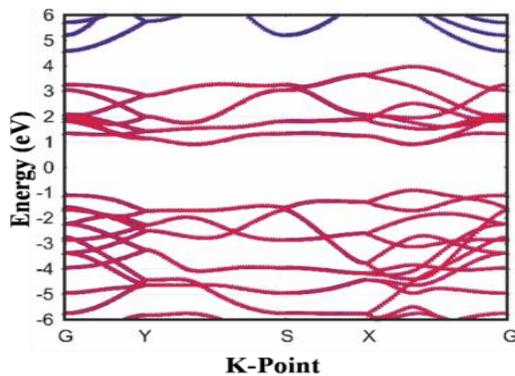


Fig.7- The electronic band structure of monolayer MoS<sub>2</sub> (unit cell) calculated by DFT (blue) and Wannier transformation (red)

Fig. 8 (a) and Fig. 8 (b) shows the unit cell representation in real space and Fig. 8 (c) shows the irreducible Brillouin zone used in device structure shown in Fig. 5. The vacuum of 2 nm is added along y-direction such that interaction of periodic images of nanoribbon is neglected. The Mo atoms at the edges are passivated by Oxygen atom and S atoms present at edges are passivated by Hydrogen atoms[13]. Further the atoms are fully relaxed in DFT calculation before performing band calculations. The supercell for armchair 1D MoS<sub>2</sub> nanoribbon is

composed of 8 equivalent primitive cells. Due to this, 88 MLWF are used for wannier transformation of armchair MoS<sub>2</sub> nanoribbon to get tight binding hamiltonian. Fig. 9 shows the electronic band structure for armchair 1D MoS<sub>2</sub> nanoribbon computed by DFT and Wannier transformation. It has been found that it has bandgap of 1.2 eV.

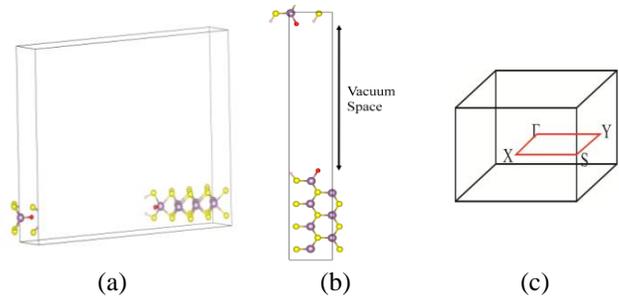


Fig.8 – Armchair MoS<sub>2</sub> nanoribbon super cell (a) 3D view, (b) top view, (c) reciprocal space (Brillouin zone)

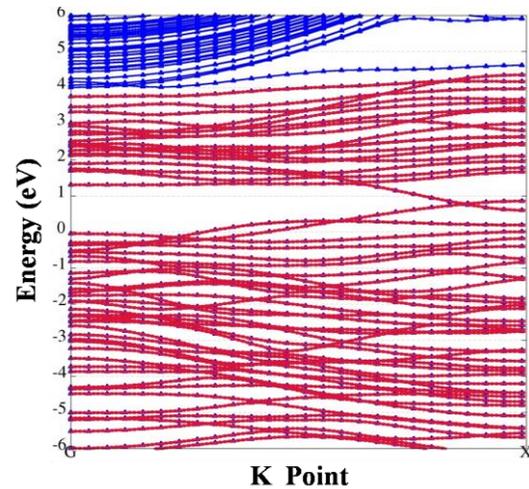


Fig. 9- The electronic band structure of armchair MoS<sub>2</sub> (supercell) calculated by DFT (blue) and Wannier transformation (red)

Fig. 10 (a) and Fig. 10 (b) shows the unit cell representation in real space and Fig. 10 (c) shows the irreducible Brillouin zone used in device structure shown in Fig. 5. The vacuum of 2 nm is added along y-direction such that interaction of periodic images of nanoribbon is neglected. The atoms are fully relaxed in DFT calculation before performing band calculations. The supercell for zigzag 1D MoS<sub>2</sub> nanoribbon is composed of 5 equivalent primitive cells. Due to this, 55 MLWF are used for Wannier transformation of armchair MoS<sub>2</sub> nanoribbon to get tight binding Hamiltonian. Fig. 11 shows the electronic band structure for zigzag 1D MoS<sub>2</sub> nanoribbon computed by DFT and Wannier transformation. It has been observed that it does not have bandgap.

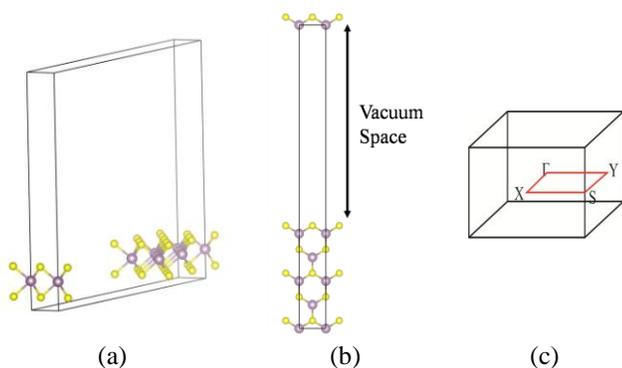


Fig.10 – Zigzag MoS<sub>2</sub> nanoribbon super cell (a) 3D view, (b) top view, (c) reciprocal space (Brillouin zone)

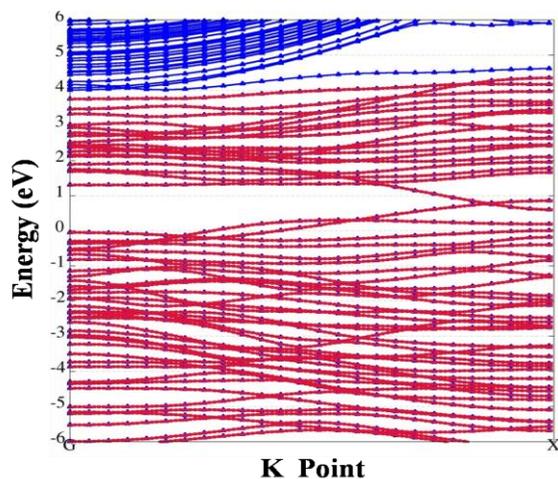


Fig.11- The electronic band structure of zigzag MoS<sub>2</sub> nanoribbon (supercell) calculated by DFT (blue) and Wannier transformation (red)

## 4 CONCLUSION

In this work, we have accurately modelled the MoS<sub>2</sub> based nano device structures by using ab-initio tight binding approach. In order to get tight binding Hamiltonian for 1D/2D MoS<sub>2</sub> based nano devices, the smallest periodic block of channel is considered. First, the ab-initio calculations are performed for 2D/1D MoS<sub>2</sub> based structures. Further the Wannier unitary transformation is carried out to get the tight binding Hamiltonian from unit cell. In this work the MLWF considered for MoS<sub>2</sub> gives the most accurate representation of electronic band structure. The approach of getting tight binding Hamiltonian from ab-initio calculation by using Wannier unitary transformation is well suited for the investigation of 2D/1D MoS<sub>2</sub> based nano devices. Hence accurately overlapped MLWF based tight binding Hamiltonian generated for both devices can be further used for device analysis by using NEGF approach.

## 5 ACKNOWLEDGMENTS

We are thankful to Ministry of Electronics and Information Technology, Government of India for providing support through Visvesvaraya PhD scheme.

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