Tight-binding modeling of the band structures of monolayer, bilayer, and bulk MoS_2

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ABSTRACT

Molybdenum disulfide (MoS_2) belongs to a family of layered transition metal dichalcogenides (TMDC) in which the layers are held together by weak van der Waals forces, and it can be exfoliated mechanically to a single layer thickness. In its bulk form MoS_2 is an indirect band gap semiconductor which turns into a direct band gap semiconductor for monolayer structure [1]. This intrinsic semiconducting nature of MoS_2 is a major advantage over graphene as a channel material in field-effect transistors (FET). Recently, FET devices based on MoS_2 monolayer and bilayer have been fabricated in the experimental labs and demonstrated to have useful device performances [2]. Several theoretical studies of MoS_2 FET devices have also been reported [3,4], most of which are based on simplified description of the electronic structures within an effective mass approximation. However, we believe that due to the complex nature of the layered TMDC materials it may require more reliable, accurate, and atomistic treatment of the electronic structures.

Here, we report a parameterized non-orthogonal tight-binding (TB) model with sp^3d^5 orbitals, nearest-neighbor interactions, and spin-orbit coupling for bulk, monolayer and bilayer MoS_2. For the TB parameterization of the band structures we employed a recently developed software package named Nanoskif. Our TB scheme is based on the Slater-Koster model [5]. For the calculations of the target band structures we employ density functional theory (DFT) with the screened hybrid functional of Heyd, Scuseria, and Ernzerhof (HSE) [6] within a projector augmented-wave (PAW) pseudopotential plane-wave method as implemented in the VASP software package. It has been shown in previous studies that accurate band structures for a wide range of semiconductors can be obtained with the screened hybrid functional. Spin-orbit coupling is included self-consistently in all the calculations. For the structure relaxation we have taken into account of the van der Waals interactions between the layers of MoS_2. Excellent agreement with the experimental data for the band gaps is achieved by adjusting the screening parameter of the hybrid functional. In our tight-binding model with a same set of parameters (in total 96) we are able to reproduce accurately the band structures for three different forms of MoS_2: bulk, monolayer and bilayer. Another important feature of our tight-binding scheme is that it can be easily extended to other layered TMDC materials showing electronic characteristics similar to MoS_2 such as WS_2, WSe_2, and $MoSe_2$. The accuracy of our tight-binding fitting is validated by comparing the band structures (Fig.1), band gap values (Table 1) and the shape of the bands at the band edges (Fig.2) with our calculated *ab initio* results. The root-mean-square deviation of the fitting is within about 25 meV.

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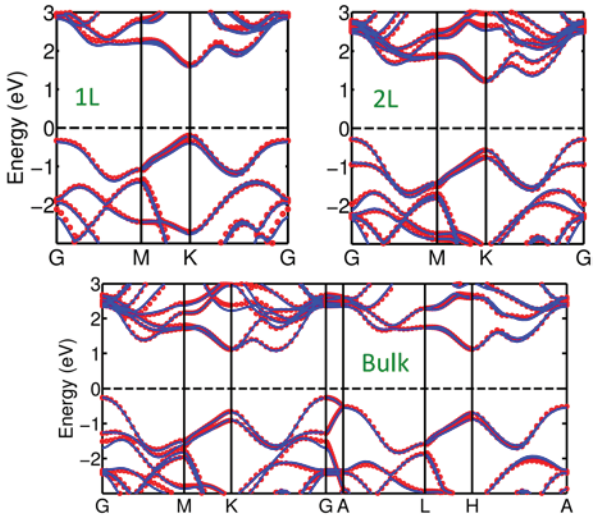

FIG. 1: Band structures for monolayer (1L), bilayer (2L), and bulk MoS_2 using screened hybrid density functional theory (blue lines) and our empirical tight-binding fitting (red dots). The zero in the energy axis is set at the Fermi level as shown by the dashed line. Spin-orbit coupling is included in the calculations.

Structures	Transitions	HSE (target)	TB (fitted)	Deviation (%)	Experiment
Monolayer	K_{v1} to K_c	1.786	1.805	1.06	1.90
	K_{v2} to K_c	1.974	1.969	0.24	2.05
Bilayer	Γ_v to K_c	1.480	1.516	2.41	1.60
	K_{v1} to K_c	1.779	1.792	0.76	1.88
	K_{v2} to K_c	1.980	1.987	0.35	2.05
Bulk	Γ_v to Σ_c	1.328	1.331	0.22	1.29
	K_{v1} to K_c	1.776	1.749	1.46	1.88
	K_{v2} to K_c	1.960	2.009	2.46	2.06

TABLE 1: Values of the band gap energies obtained from HSE calculations (target) and the tight-binding (TB) parameterization (fitted). Experimental data are obtained from Ref. 7 (for bulk) and Ref. 1 (for monolayer and bilayer). All the energies are in the unit of eV. The subscripts 'v' and 'c' stand for valence band and conduction band, respectively. The splitting of the valence band maximum at K point is given by K_{v1} (top) and K_{v2} (bottom), whereas Σ is the midpoint of the line joining the Γ and the K points.

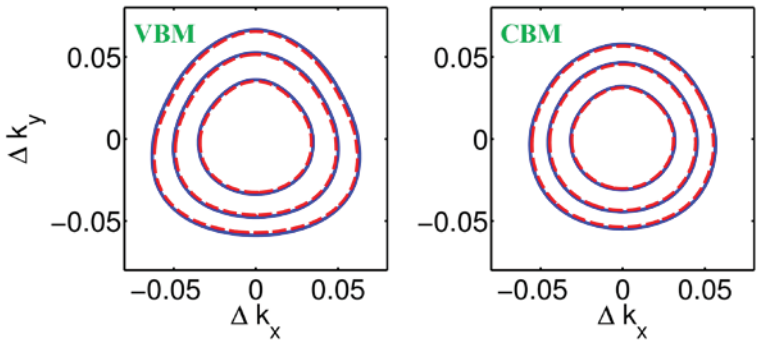

FIG. 2: Contour plots of the energies around the valence band maximum (VBM) and the conduction band minimum (CBM) for monolayer MoS_2 from screened hybrid density functional theory (blue solid lines) and empirical tight-binding fitting (red dashed lines). The contour lines are associated with $\Delta E = \pm(1/30)$, $\pm(1/15)$, and $\pm(1/10)$ eV, respectively, and the Δk_x and the Δk_y are in the unit of $2\pi/a$, where 'a' is the cell length.