Supporting Information

Spin Polarization and Tunable Valley Degeneracy in Monolayer MoS_2 by Proximity Coupling to Antiferromagnetic Cr_2O_3 Substrate

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Discussion on the reasonability of results calculated by DFT approach.

It is known that many-body perturbation theory in the GW approximation is currently one of the most accurate approaches for describing band structure of extended systems. It is found that the fundamental band gaps of the bulk TMDs can be well described by the GW approach (J. Phys. Chem. C 2012, 116, 7664-7671).[1] However, Comparing with the experimentally measured energy gap of 1.8 eV for monolayer MoS_2 (NanoLett. 2012. 12, 3695-3700, PRL 105, 136805 (2010)),[2, 3] the band gap which calculated by GW approach are equal to 2.8 eV are largely overestimated (Appl. Phys. Lett. 103, 042106 (2013), Phys Rev B. 86, 115409 (2012)).[4, 5] Further, the heterojunctions in our model are consist of 127 atoms, the GW calculations are almost not feasible. So, the GW approach may not a good choice for study these systems.

Since the DFT calculation of the band gaps for extended structure is widely used and the results are acceptable by researchers. In this work, we predict a slightly underestimated direct bandgap of 1.68 eV which is very close to the experimentally values.[2, 3] The work function of semiconductors, which is another factor to determine the band alignment, had been calculated by DFT approach in previous study, and the PBE functional provides comparable results with B3PW91 hybrid functional, which generally gives much more accurate band gaps as well as band offsets (J. Am. Chem. Soc. 2016, 138, 15853-15856).[6] So, the DFT approach could give a reasonable description of band alignment for heterojunctions, and the energy level position of VBM and CBM for monolayer MoS_2 in our calculation are agree well with previous work (Sci. Adv. 2016; e1600069).[7] Further, the previous work also indicates the PBE functional calculated valence-band spin-orbital splitting value are very close to single-shot G_0W_0 approach.[5]

Consequently, even though the band alignment and the corresponding charge transfer and Schottky barrier in our work may not accurate as advance GW approach or hybrid functional, the fundamentally properties and its variation trend are basically well described by the DFT approach.

Table. S1 The computed band gaps (Eg) and work function (W) of MoS_2 and Cr_2O_3 (0001) with different lattice constant.

System	*a* (Å)	Eg (eV)	W (eV)
MoS_2	**3.19**	**1.68 (direct)**	**5.45**
	3.31	**0.88 (indirect)**	**5.14**
Cr_2O_3 (0001)	**4.95**	**2.10**	**4.41**
	4.79	**2.18**	**4.19**

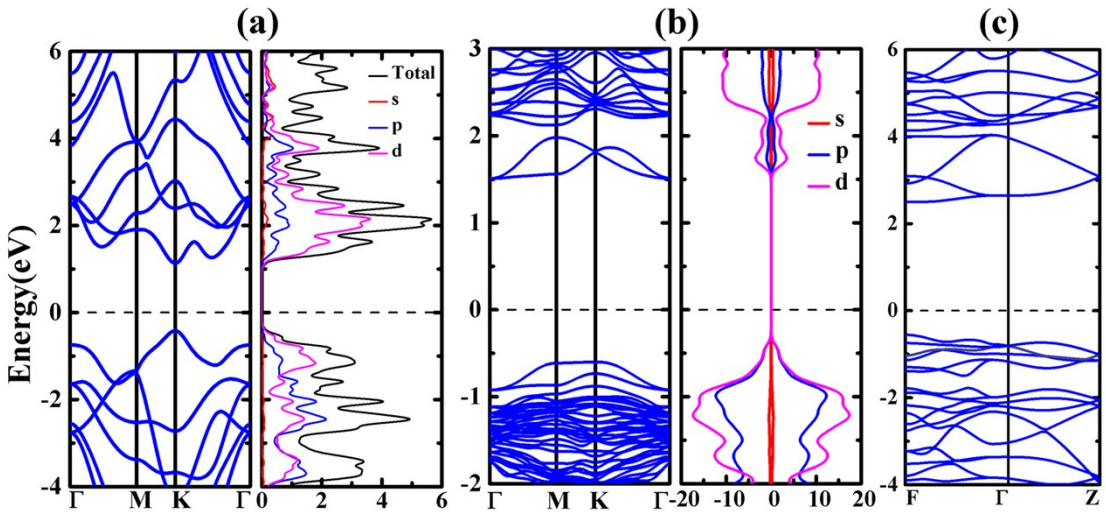

Figure. S1 The band structures and density of states of the (a) monolayer MoS_2, (b) Cr_2O_3 substrate and (c) bulk Cr_2O_3.

References:

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