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Supporting Information : Regulation of Transport Property by Polytypism: A computational study on bi-layer MoS_2

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1 Computation Details

The projector-augmented-wave pseudopotential[1] with an energy cutoff of 60 Ry for the plane-wave basis sets was adopted, and gamma-centred 11 x 11 x 1 Monkhorst-Pack k-point meshes are used for structural optimisation of bilayer slab calculations. Finer k-mesh (30 x 30 x 1) with an energy cutoff of 80 Ry and density cutoff of 640 Ry have been used to achieve converged results of band dispersion to find the transport parameters. All atomic positions and lattice constants are optimized using the Broyden-Fletcher-Goldfarb-Shanno (BFGS) algorithm, with the total energy and atomic forces minimised. The convergence for total energy was chosen to be less than 1 x 10^{-5} eV between two consecutive steps, and the maximum Hellmann-Feynman forces acting on each atom were less than 0.02 eV $Å^{-1}$ upon ionic relaxation. For bilayer slabs, atomic positions are fully relaxed after considering a vacuum region of $\sim$ 30 Åalong the z-direction, which is large enough to make the mirror interaction between neighbouring images negligible. The optimised lattice parameters for each of the polytypes are as follows, $2H_c$: a = b = 3.11892; $2H_a$: a = b = 3.12155; 3R: a = b = 3.12087 (in every cases $\alpha = 90°$, $\beta = 90°$, $\gamma = 120°$). The effect of Spin orbit coupling (SOC) has also been considered. Although inclusion of SOC leads to a splitting of the valence band and the conduction band, it does not affect the location of the band extrema resulting in a insignificant shift in absolute band-gap (smaller than 19 meV).[2] So, transport calculation without SOC is reasonably accurate and computationally efficient while considering MoS_2 polytypes.

2 Results & Discussion

Importantly, dispersive interaction varies for different polytypes (1-3 meV/atom) which is also associated with dissimilar polarisation of electron cloud at the interlayer space(see Figure 1).

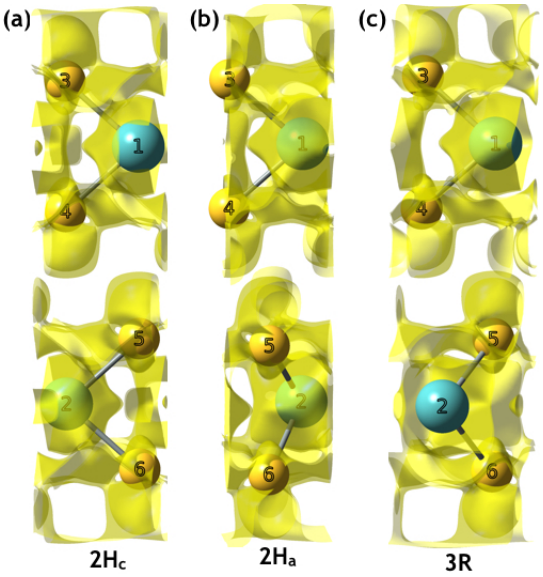

Fig. 1 Net overlap for different MoS_2 bilayers with different stacking orders in (a) $2H_c$, (b) $2H_a$ and (c) 3R bilayers.

References

1 P. E. Blöchl, *Physical Review B*, 1994, **50**, 17953.
2 J. Padilha, H. Peelaers, A. Janotti and C. Van de Walle, *Physical Review B*, 2014, **90**, 205420.

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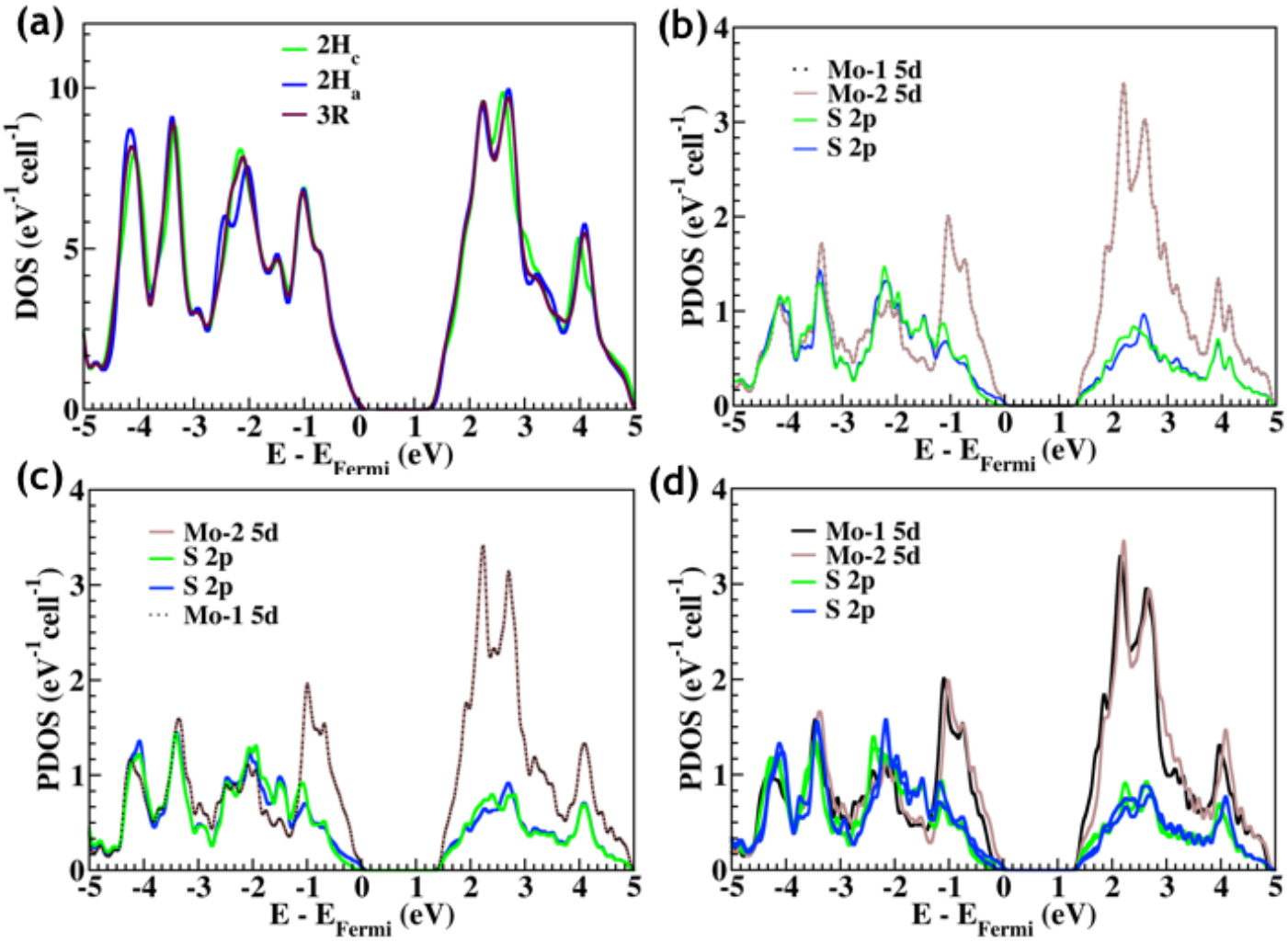

Fig. 2 (a) Total Density of states for different MoS_2 bilayers with different stacking orders. Projected density of states for (b) $2H_c$, (c) $2H_a$, and (d) 3R bilayers are shown.

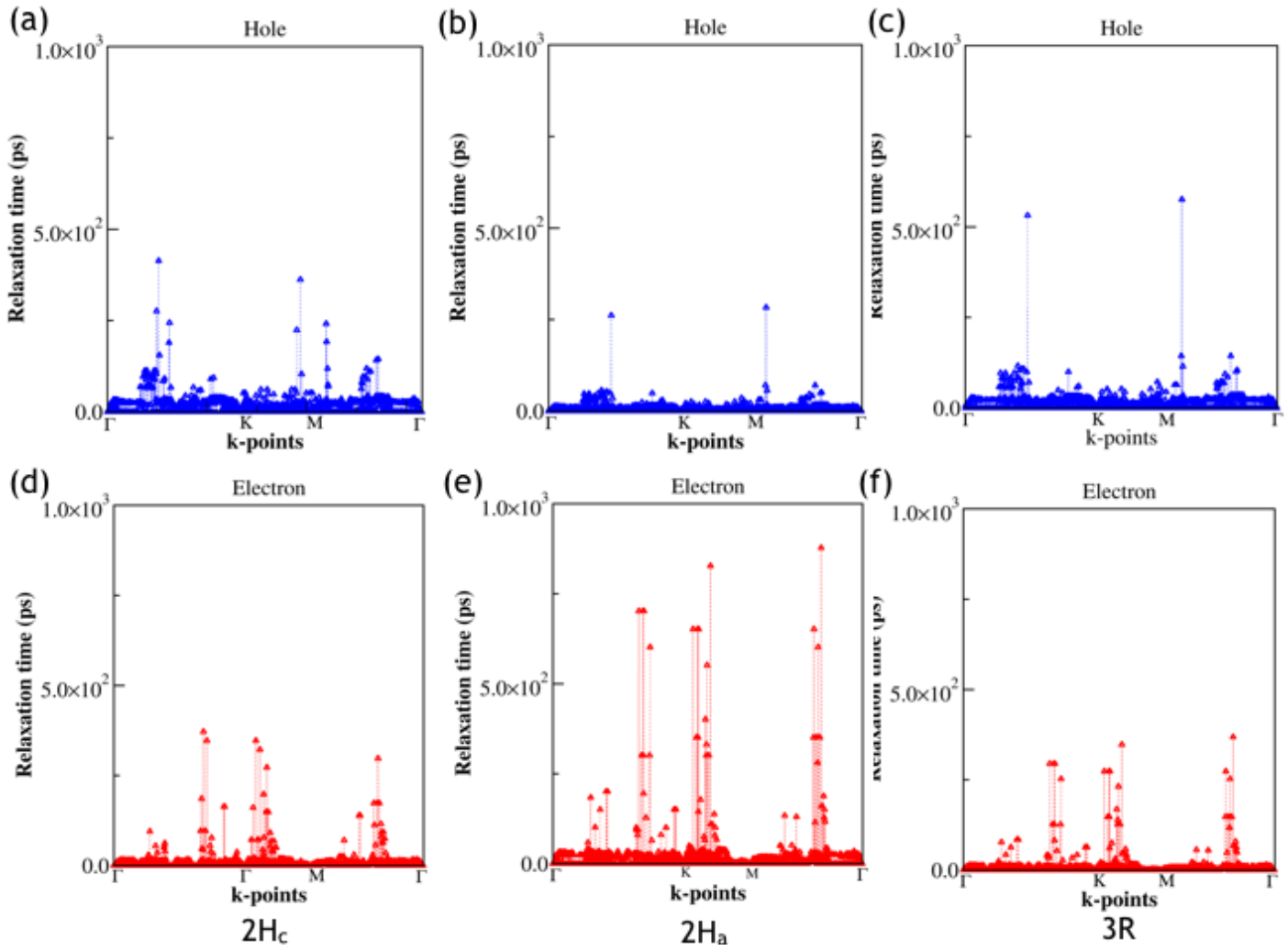

Fig. 3 Relaxation time (τ) versus corresponding carrier-states (in k-space) along the $\Gamma - K - M - \Gamma$ band line is shown for (a) holes and (b) electrons in $2H_c$, $2H_a$ and 3R polytypes.