Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Supporting Information for:

Emulating Synaptic Behavior in Surface-Functionalized MoS_2 through Modulation of Interfacial Charge Transfer via External Stimuli

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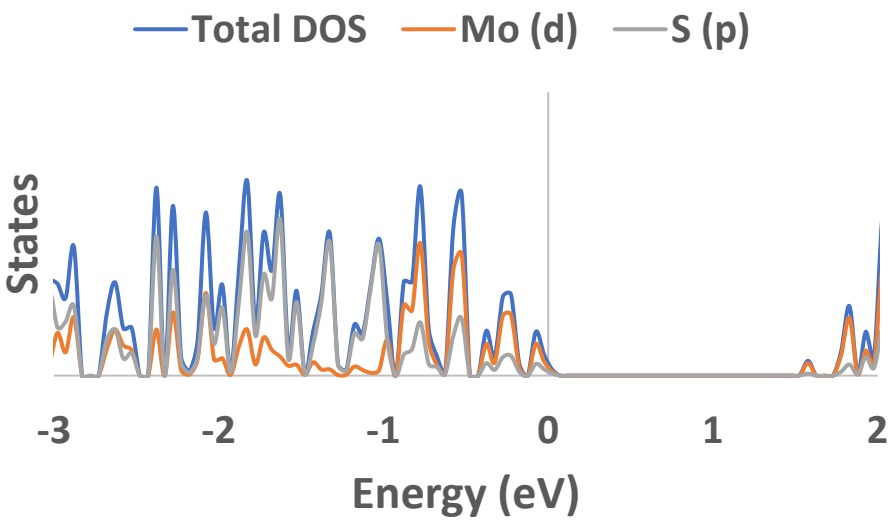

Figure S1. GGA-PBE calculated density of states (DOS) profile for the pristine basal plane of MoS_2.

[1]Penn State Greater Allegheny, Pennsylvania State University, McKeesport, PA 15132, USA

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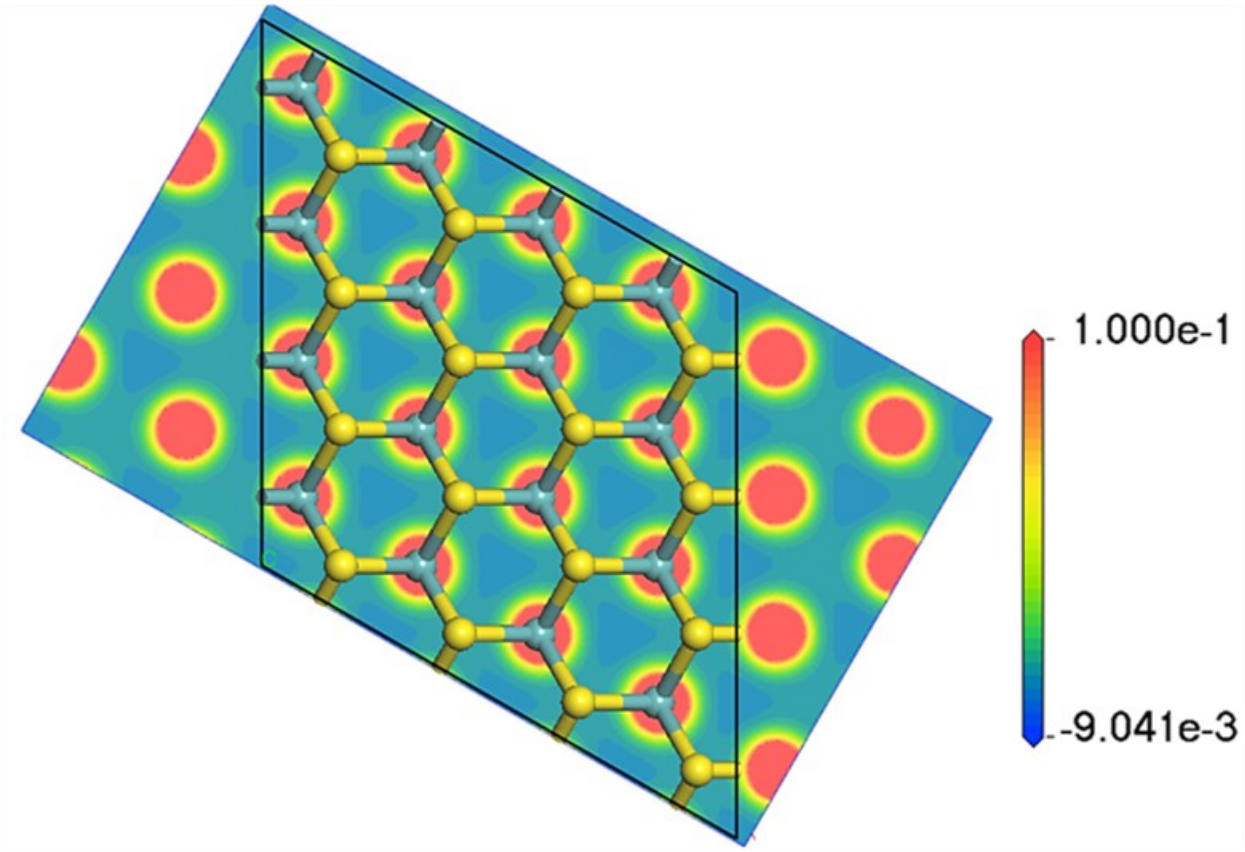


Figure S2. Electron density map for MoS_2's basal plane, parallel to the [001] direction and across Mo atoms. Color code: Yellow and green spheres represent S and Mo atoms.

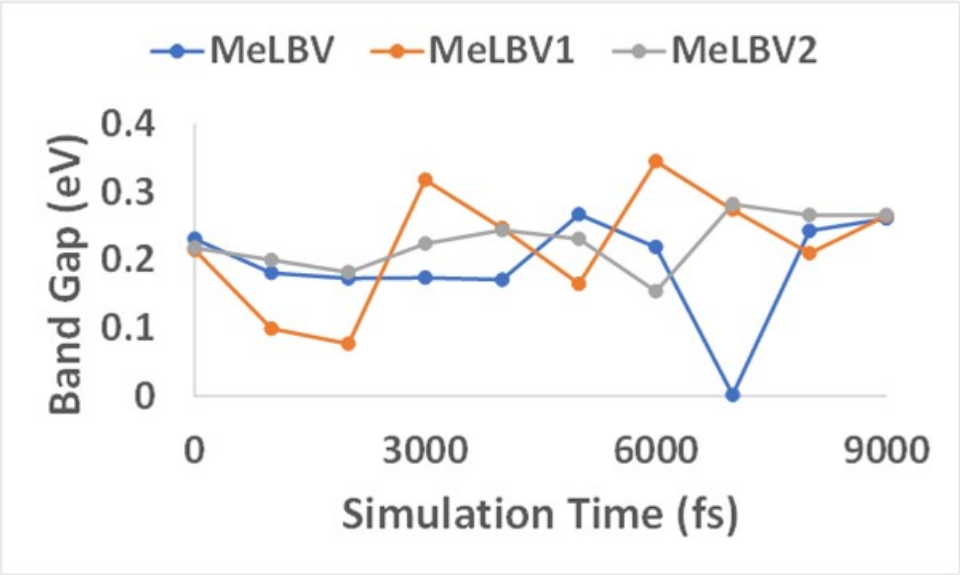


Figure S3. DFT/PBE calculated band gap evolution of the three MoS_2| RMVX systems shown in Fig. 1.

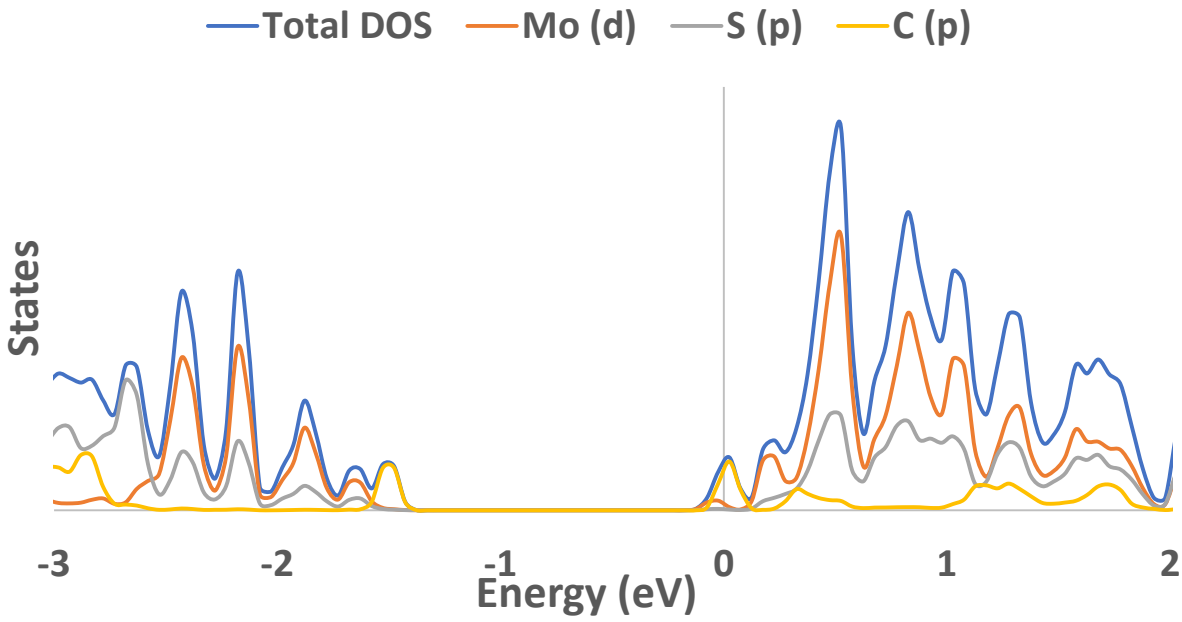
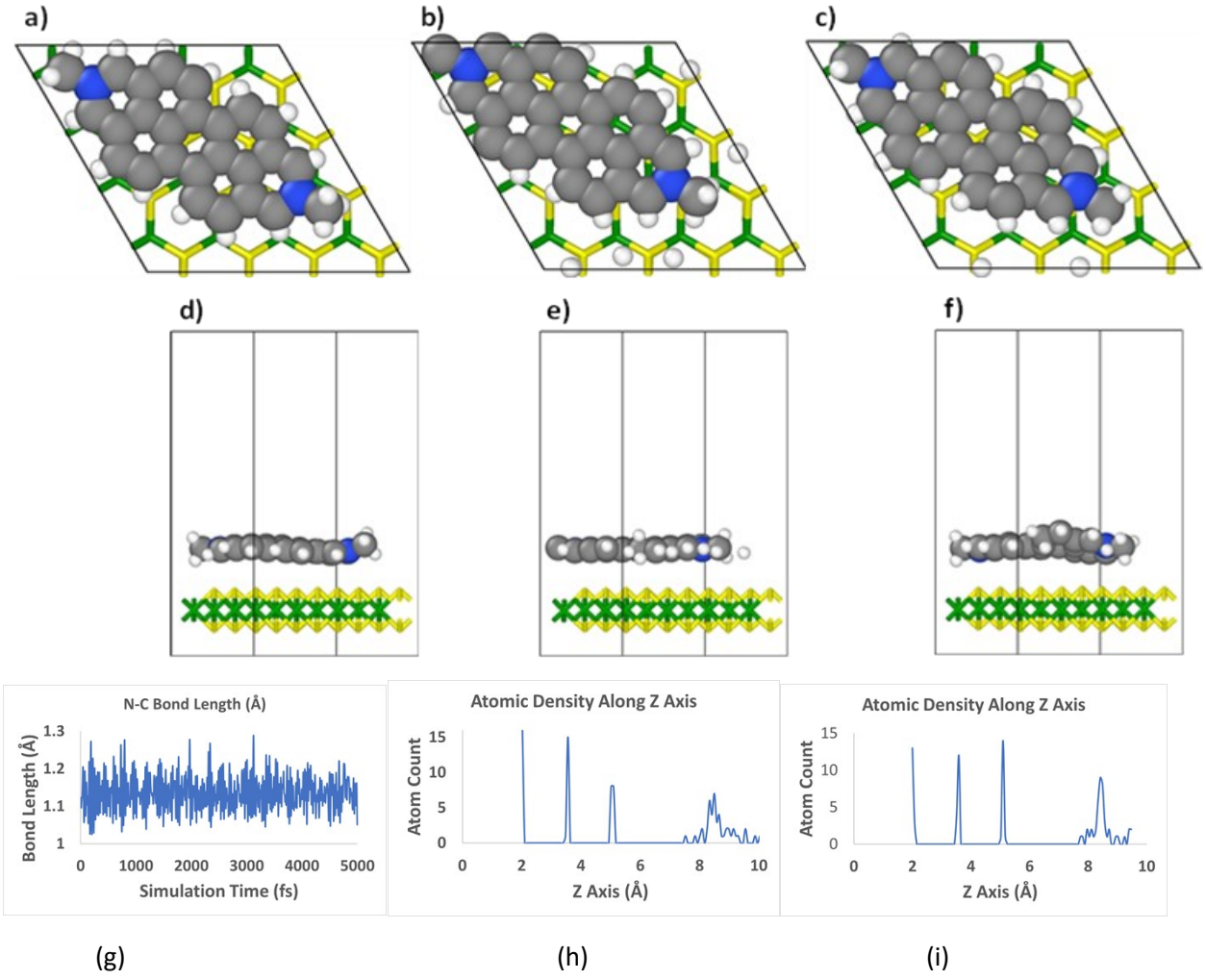


Figure S4. GGA-PBE calculated DOS profile for the MoS_2|RMV3 system.



(g) (h) (i)

Figure S5. Panels a), b) and c) show top view images of the MoS_2|RMV3 heterostructure at the initial structure, 4500 fs and 9000 fs, respectively. Panels d), e) and f) show top view images of the MoS_2|RMV3 heterostructure at the initial structure, 4500 fs and 9000 fs, respectively. Color code: green, yellow, gray, blue and white spheres represent Mo, S, C, N and H atoms, respectively. Panels (g), (h), and (i) display the time evolution changes of the NC bond length of the RMV3 molecule, the average atomic density for the MoS2/ RMV3 system along the Z axis at -0.3 V/Å, and at zero electric field respectively. The last peak between 8 and 10 Å in (h) reveal variations in the molecular density under the applied field.

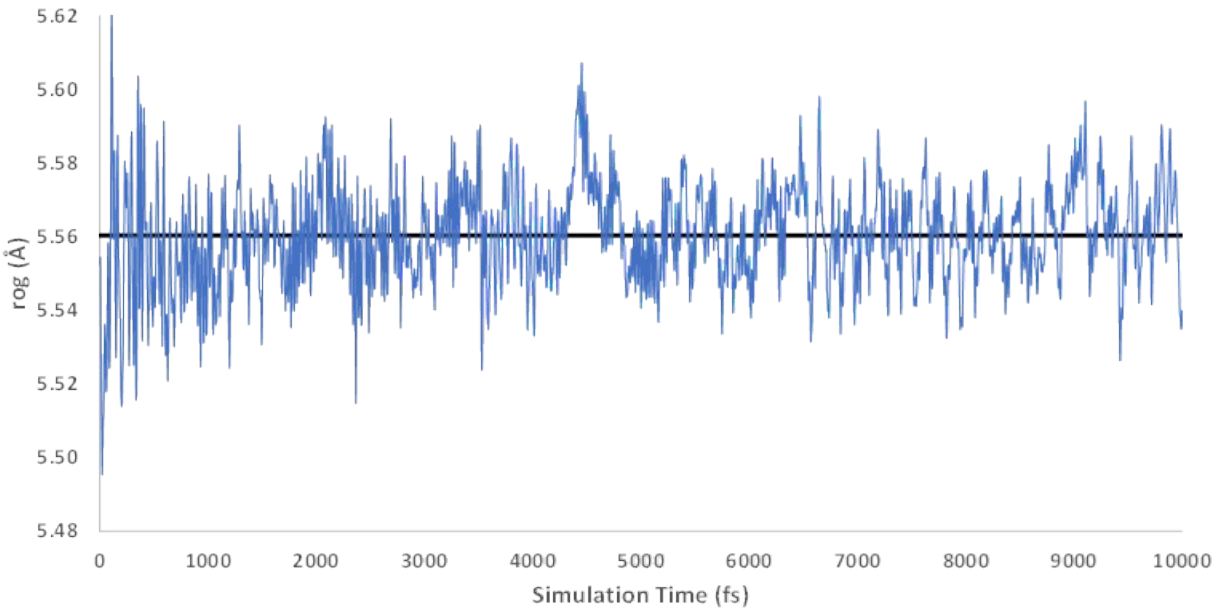


Figure S6. RoG evolution at 300 K of the MoS_2|RMV3 heterostructure.

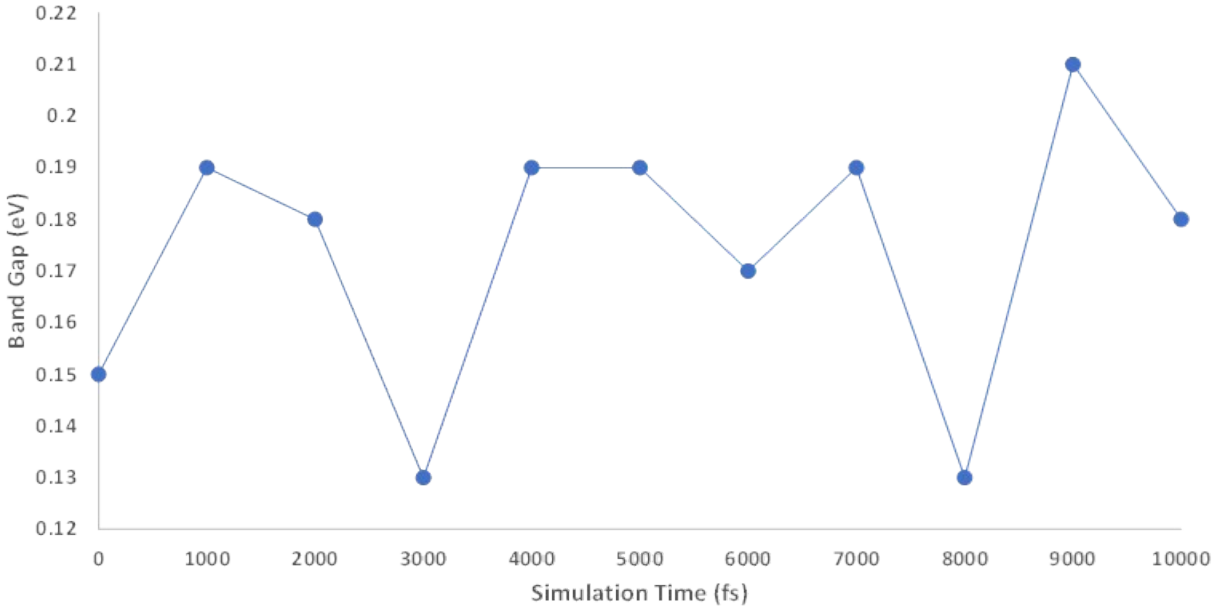


Figure S7. Band gap evolution at 300 K of the MoS_2| RMV3 heterostructure.

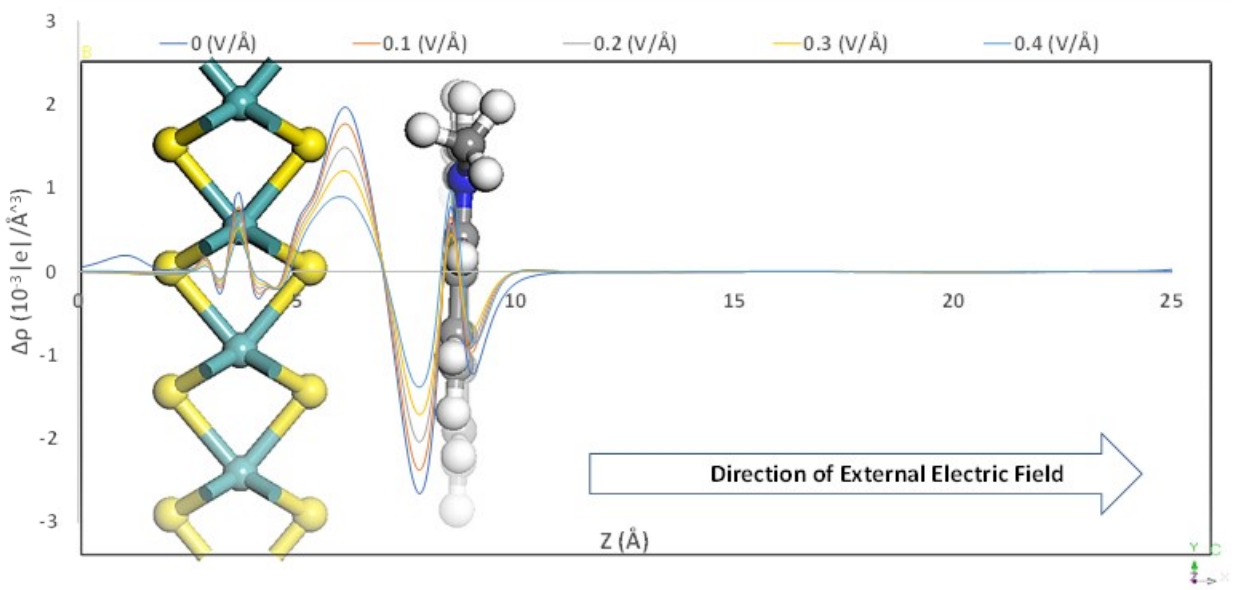


Figure S8. Plane-averaged charge density differences along the z direction for MoS_2| RMV3 heterostructure. Color code: Yellow, green, gray, blue and white spheres represent S, Mo, C, N and H atoms, respectively.

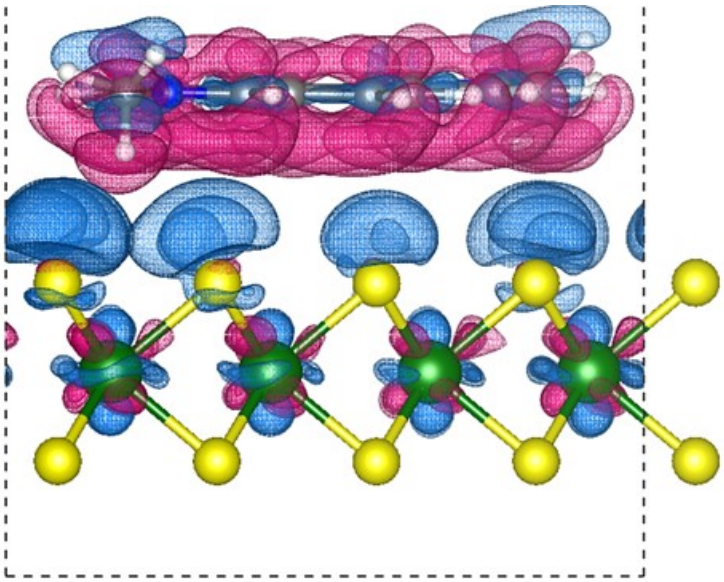


Figure S9. Charge density difference isosurface at 0.0005 e/Å^3 for the MoS_2|RMV3 heterostructure optimized at an electric field strength of 0.3 V/Å pointing inwards towards the basal plane. Color code: Yellow, green, gray, blue and white spheres represent S, Mo, C, N and H atoms, respectively.

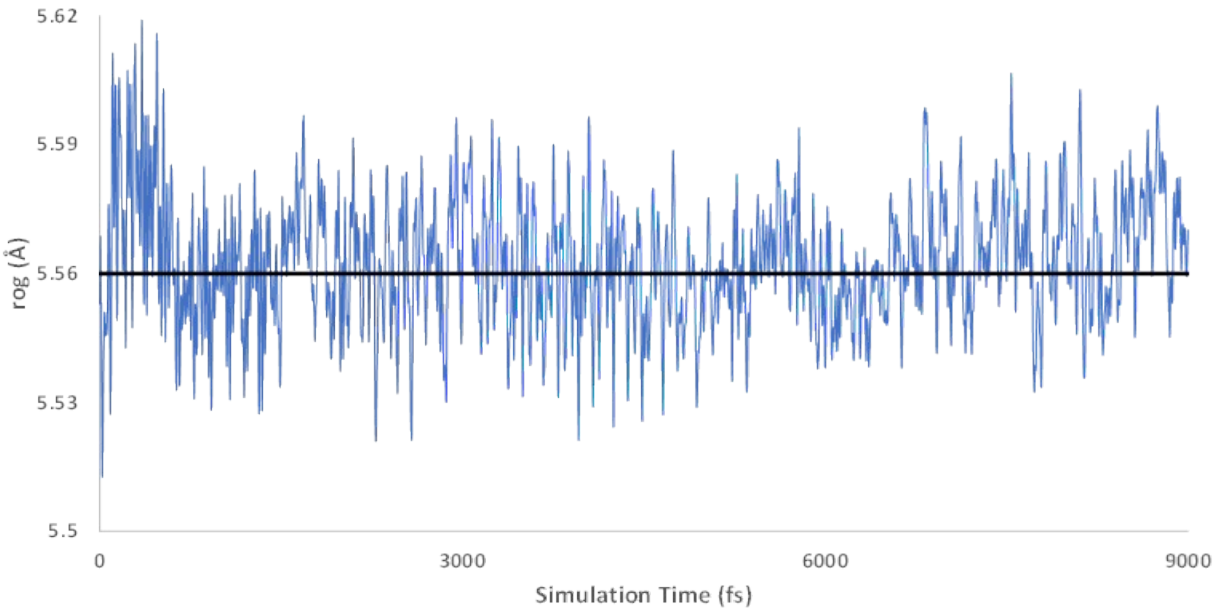


Figure S10. The rog evolution at 300 K of the MoS_2|RMV3 heterostructure with an external stimulus of -0.3 V/Å.

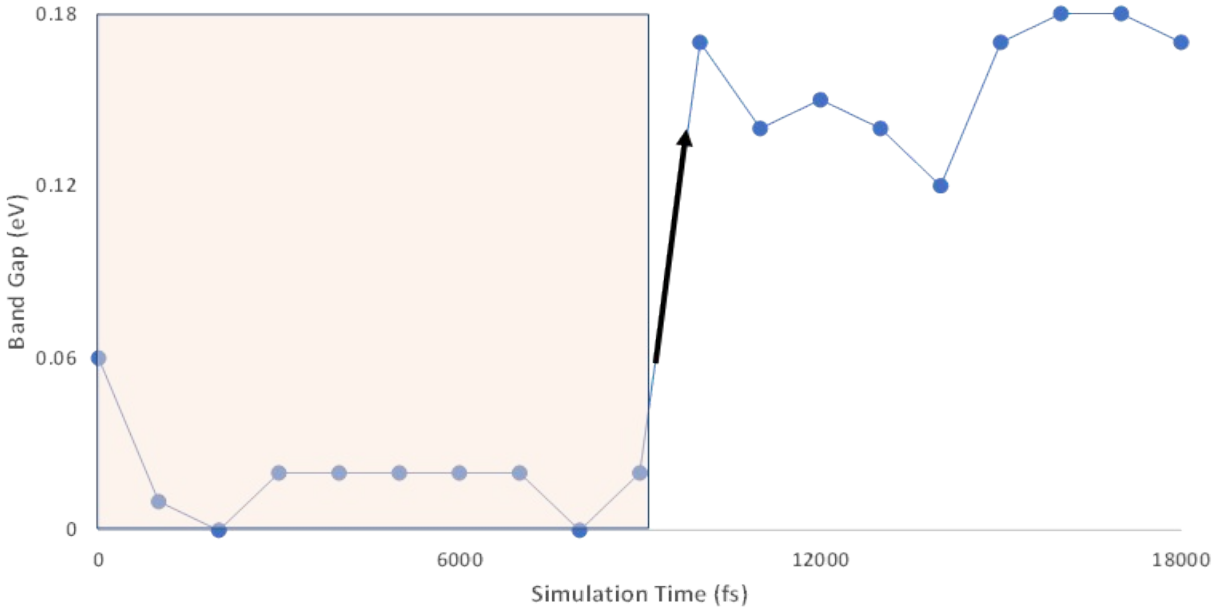


Figure S11. Bandgap evolution at 300 K of the MoS_2| RMV3 heterostructure with applied field (-0.3 V/Å) from 0 to 9000 fs and with the field switched off from 9000 fs to 18000 fs.

Table S1. Rog values and band gap of the optimized MoS_2|RMV3 system with the applied electric field values of -0.5 V/Å and 0.5 V/Å.

Electric Field	Radius of Gyration	Band Gap (eV)
-0.5 V/Å	4.10	0.05
0.5 V/Å	4.11	0.24