Electronic Supplementary Material (ESI) for Nanoscale.

Supplemental Materials: Surface passivated and encapsulated ZnO atomic layer by high-κ ultrathin MgO layer

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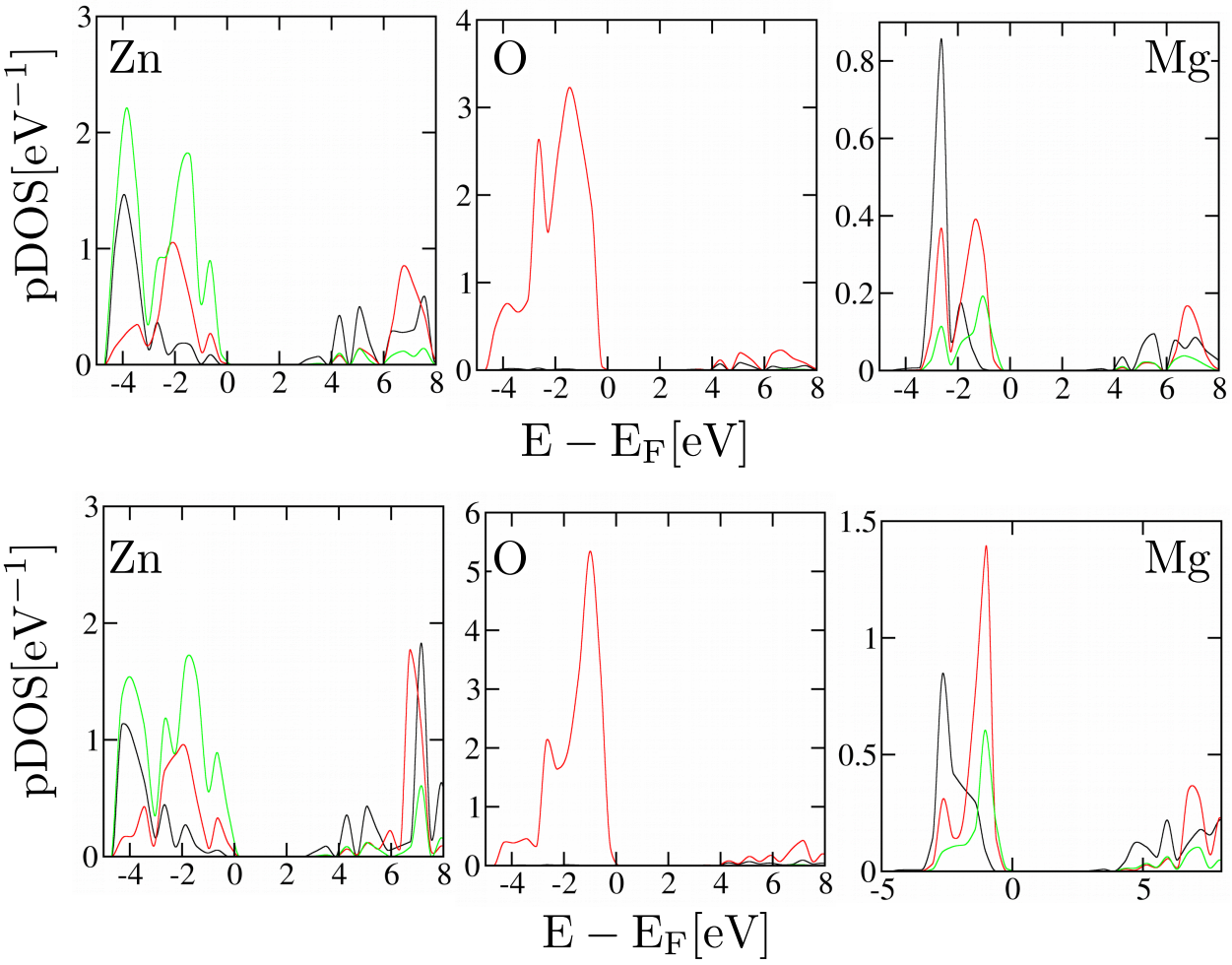

Figure 1: **Electronic structure of vdW heterostructures.** Partial density of states of Zn, O, and Mg. Note, the O atoms from ZnO and MgO have been plotted together. However, over 98% of the contributions of O to the vdW materials are from ZnO. The top panel is for MgO/ZnO while the bottom panel is for MgO/ZnO/MgO. Black, red, and green lines in the plots correspond to *s*, *p*, and *d* orbitals, respectively.

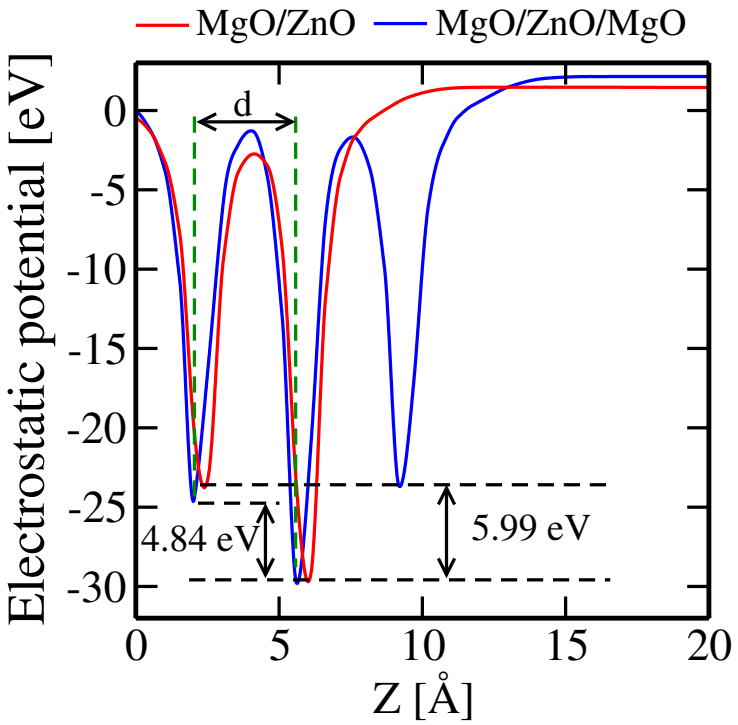

Figure 2: **Electronic band levels of ZnO-based vdW heterostructures.** Electrostatic potential profile along the z-direction of the crystallographic plane. For MgO/ZnO vdW heterostructure, the charge transfer across the heterojunction needs to overcome an energy barrier of 5.90 ± 0.02 eV, and a transfer distance of 3.72 ± 0.02 Å. For MgO/ZnO/MgO, the charge transfer across the heterointerface needs to overcome an energy barrier of 4.84 ± 0.02 (5.99 ± 0.02 eV) and a transfer distance of 3.59 ± 0.02 (3.72 ± 0.02 Å) for the top (bottom) interfaces. d is a typical transfer distance.

Below we present the crystal information in the primitive cell; the Crystallographic Information File format is used.

MgO passivated ZnO - MgO/ZnO crystal structure

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O5	1.0	0.166667	0.166667	0.238178	Biso	1.000000	O
O6	1.0	0.166667	0.666667	0.238178	Biso	1.000000	O
O7	1.0	0.666667	0.166667	0.238178	Biso	1.000000	O
O8	1.0	0.666667	0.666667	0.238178	Biso	1.000000	O
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MgO encapsulated ZnO - MgO/ZnO/MgO crystal structure

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Acknowledgment

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