Supplementary Information

Tuning electronic properties and work functions of graphane/ fully hydrogenated h-BN heterobilayer *via* heteronuclear dihydrogen bonding and electric field control

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Figure S1. The atomic charge population of G/fHBN bilayer in eight stacking patterns. The numbers are given in |e|.



Figure S2. Band structures of (a) graphane and (b) fHBN, respectively.



Figure S3. Band structures and DOS of other patterns from CHB family (patterns II, III and IV) and CHN family (patterns VI, VII and VIII) of G/fHBN bilayer.



Figure S4. Schematic diagram of the G/fHBN bilayer with the E-field, the band gap and binding energy as functions of E-field belong to the CHB family (patterns II, III and IV) and CHN family (patterns VI, VII and VIII), respectively.

E-field	-0.008	-0.007	-0.006	-0.005	-0.004	-0.003	-0.002	-0.001	0	0.001
Δe (e /unit cell)	-0.032	-0.024	-0.016	-0.008	0	0.004	0.012	0.02	0.024	0.032

Table S1. Charge transfer from fHBN monolayer to graphane as a function of E-field.