Supporting Information

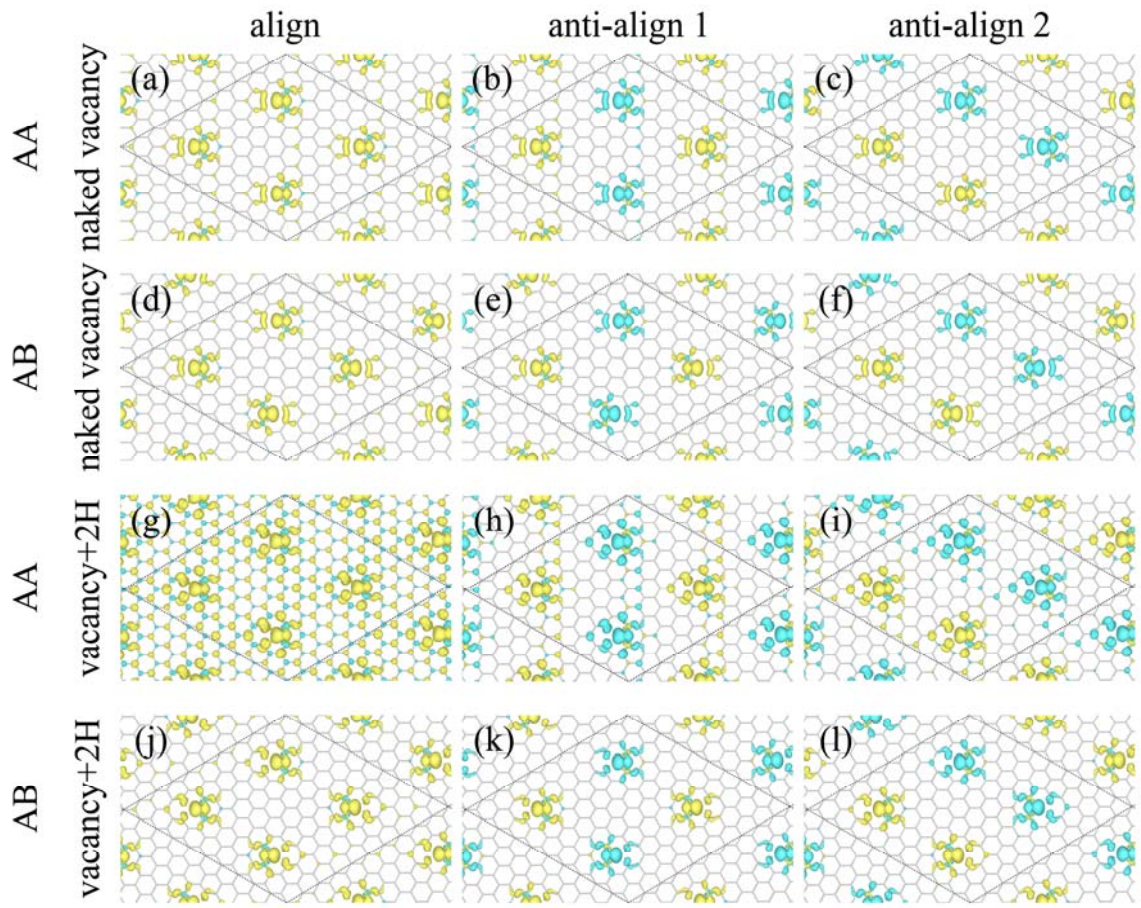

Figure s1. Spin density (isosurface=0.002μ_B/Å^3) of certain spin coupling configurations of (a)-(c) AA naked vacancy, (d)-(f) AB naked vacancy, (g)-(i) AA vacancy+2H (j)-(l) AB vacancy+2H. Vacancy distribution patterns are indicated on the left, while spin coupling configurations are labeled on the top. Yellow colored isosurface stands for spin up while cyan stands for spin down.

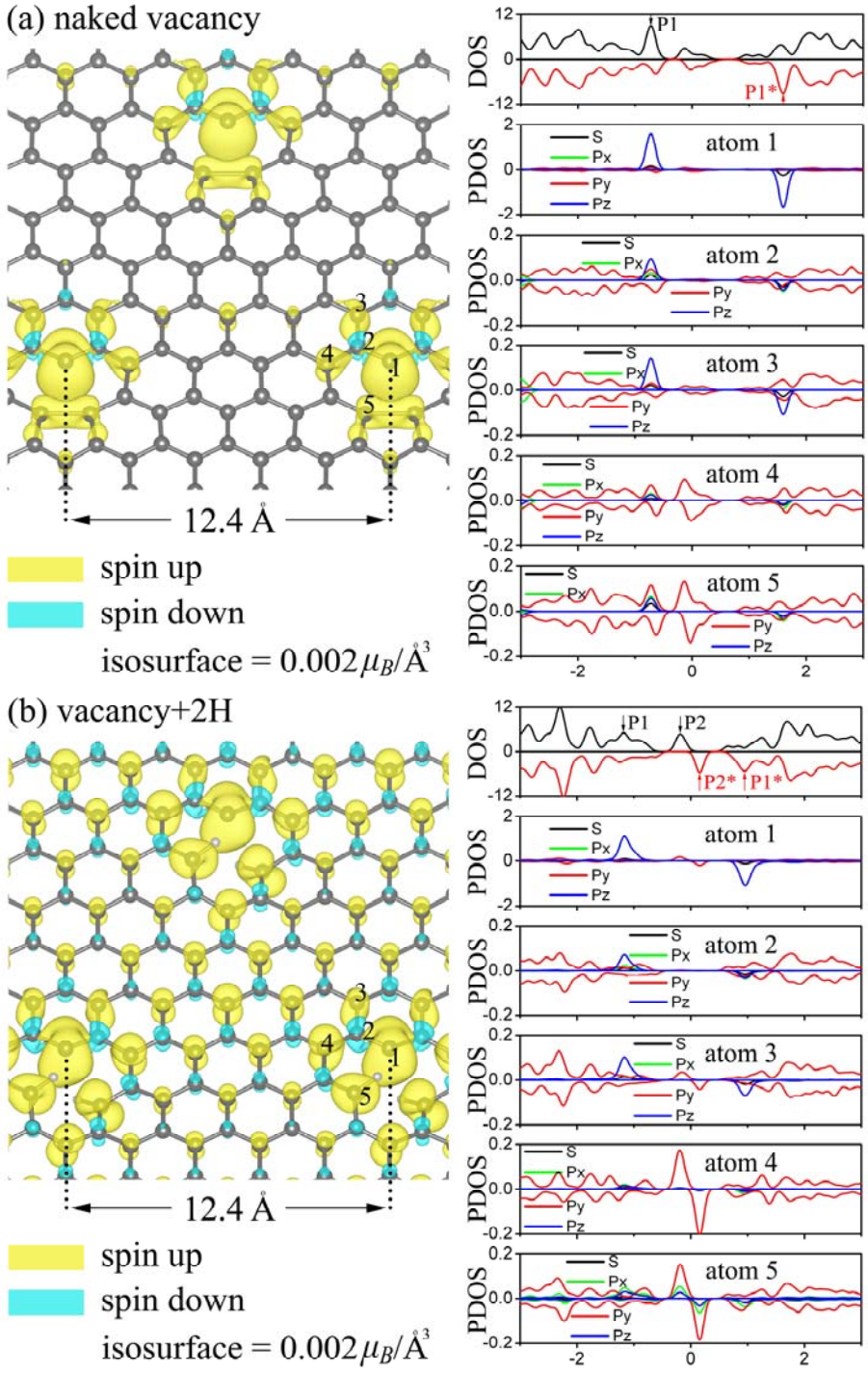

Figure s2. Spin density, DOS and projected DOS (gaussian smoothed) on atoms of AA patterned (a) naked vacancy and (b) vacancy+2H. Peaks labeled with P1, P2, P1* and P2* are discussed in the paper. Atomic serial number shown in the PDOS can be found in the right-bottom in the spin density figure.

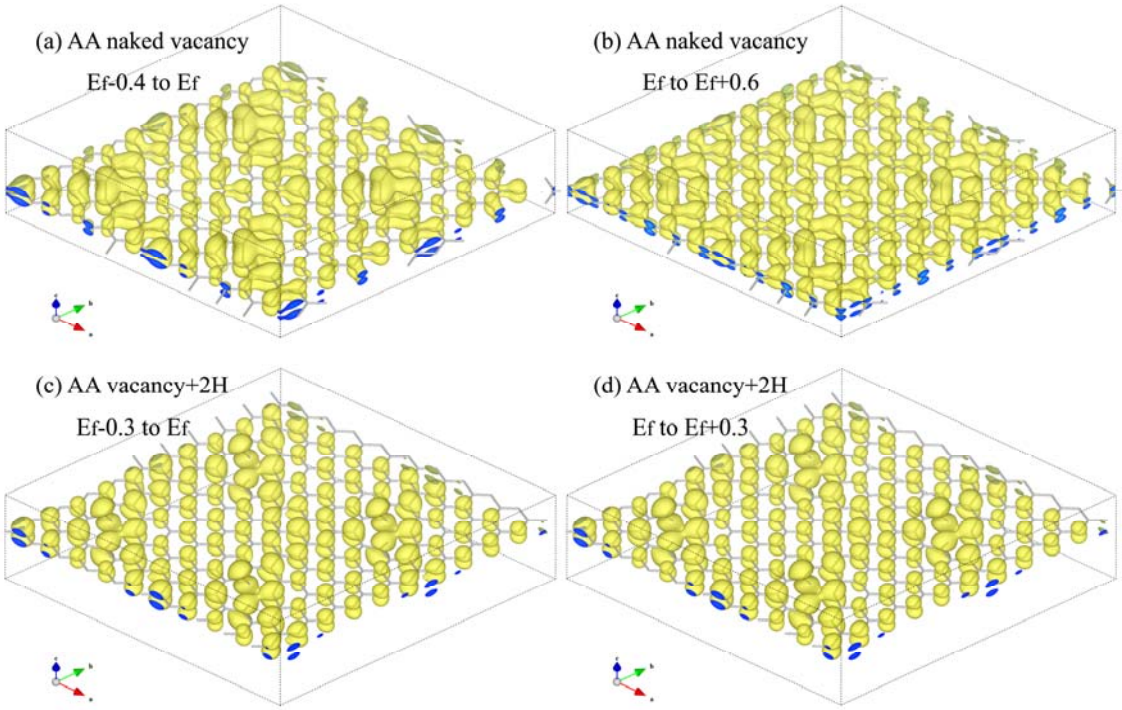

Figure s3. Partial charge density (spin up plus spin down, isosurface=0.0004|e|/Å^3) of frontier orbital . The energy ranges that are used for the evaluation of the partial charge are corresponding to that in Fig. 5 in the paper.

For naked vacancy (Fig. s3a, s3b), *Pz* orbital of one A-site carbon overlaps efficiently with its B-site neighbor, so that π bond of a pristine graphene is highly reserved. On the contrary, for vacancy+2H, frontier electron is only localized at B-site carbons. The breaking of π bond makes these *Pz* electrons indiffusible so that the system is semiconductor.

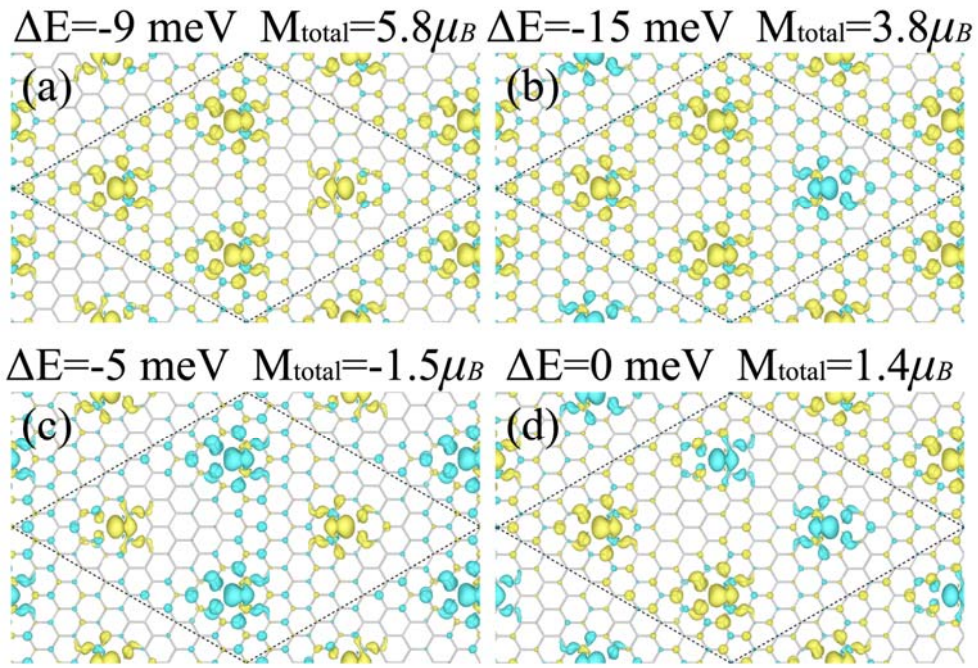

Figure s4. Spin density (isosurface=0.002μ_B/Å^3) of A_3B patterned vacancy+2H defects in a (10×10) supercell: (a) align spin configuration, (b)$3A_{up}B_{down}$ spin configuration, (c) anti-align 1 spin configuration and (d) anti-align 2 spin configuration. The relative energy per defect (taking anti-align 2 as reference state) and total magnetic moment of the supercell are labeled on the top. Yellow colored isosurface stands for spin up while cyan stands for spin down.