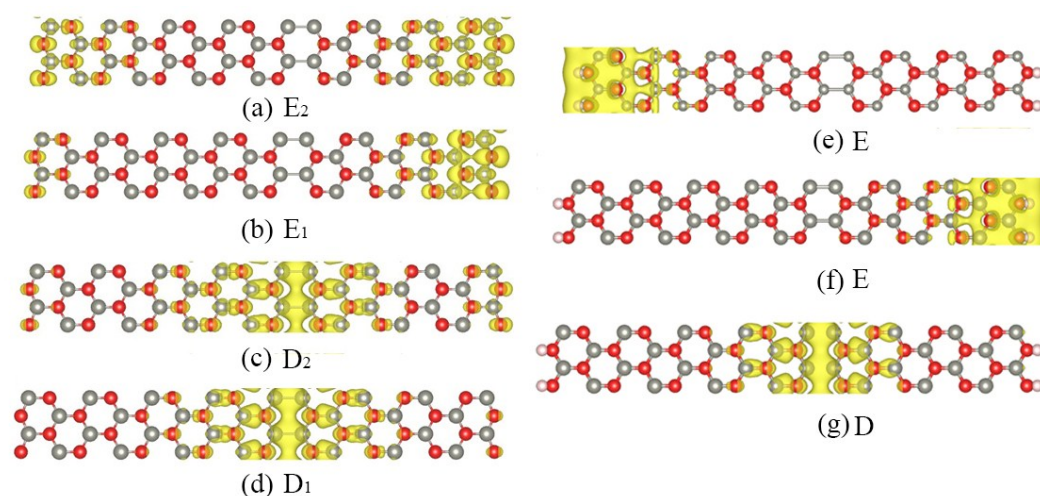
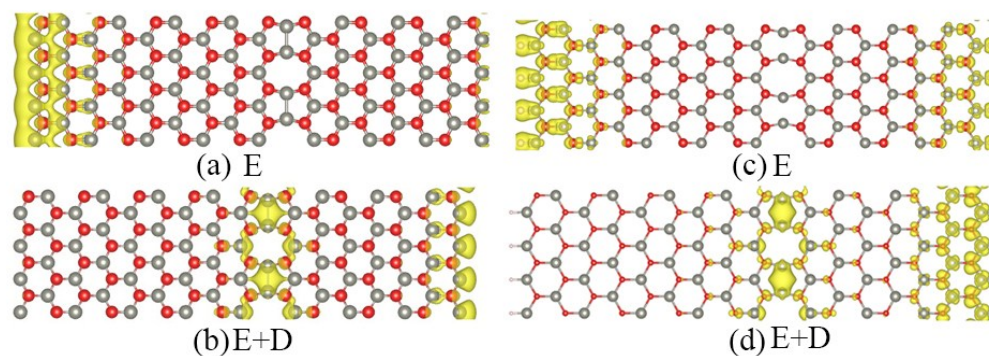


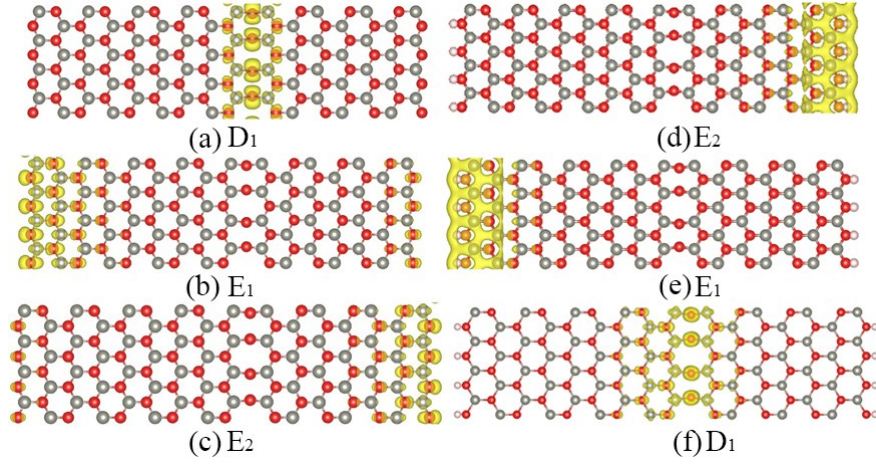
### Electronic Supplementary Information



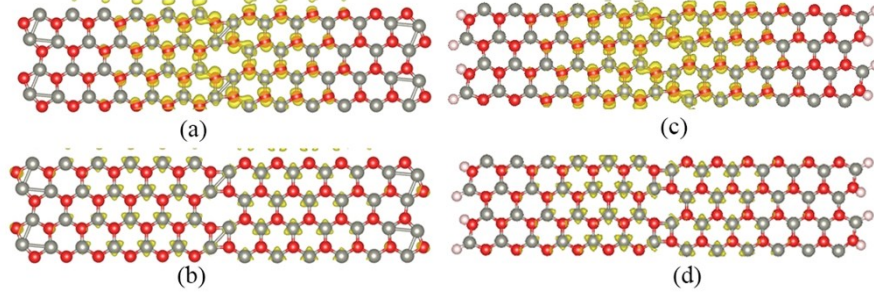
**Figure S1.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for "[OZn] - [ZnO]" (a-d); and hydrogen passivated "[OZn] - [ZnO]" (e-g). Note the E state is a degenerate state.



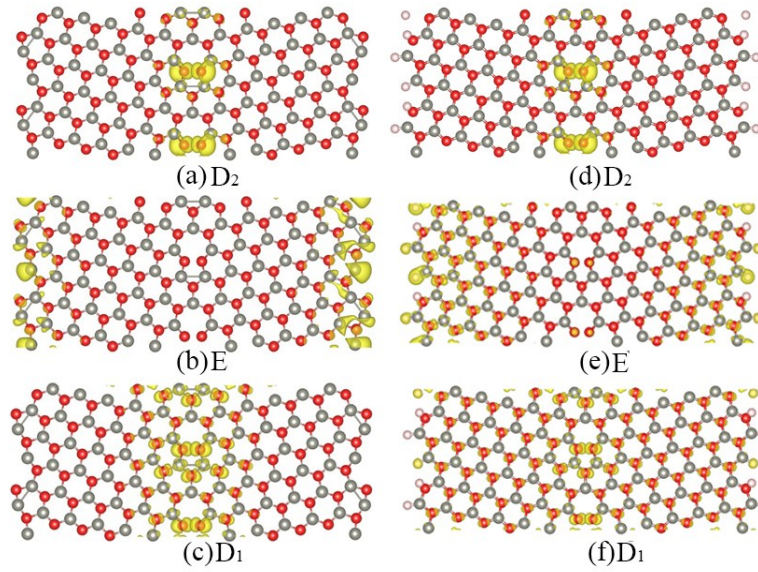
**Figure S2.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for "[ZnO] -  $Zn_2$  - [OZn]" (a-b); and hydrogen passivated "[ZnO] -  $Zn_2$  - [OZn]" (c-d).



**Figure S3.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for "[OZn] - O<sub>2</sub>- [ZnO]" (a-c); and hydrogen passivated "[OZn] - O<sub>2</sub>- [ZnO]" (d-f).

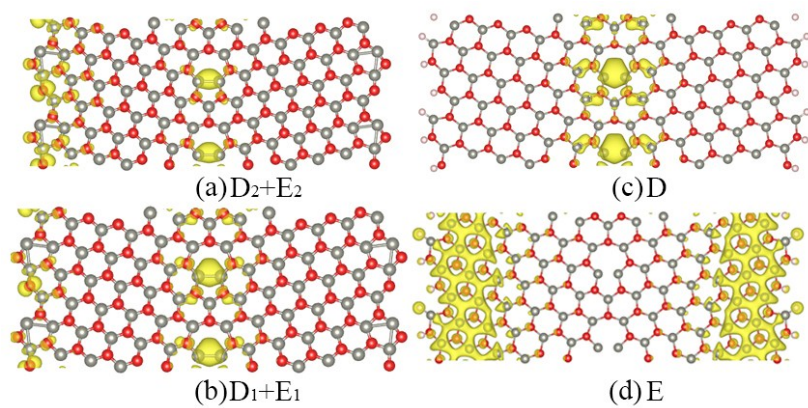


**Figure S4.** Profiles of the VBM (a,c); and CBM (b,d) of 4-8 aZnO-NRs with bare edge or hydrogen-passivated edge.

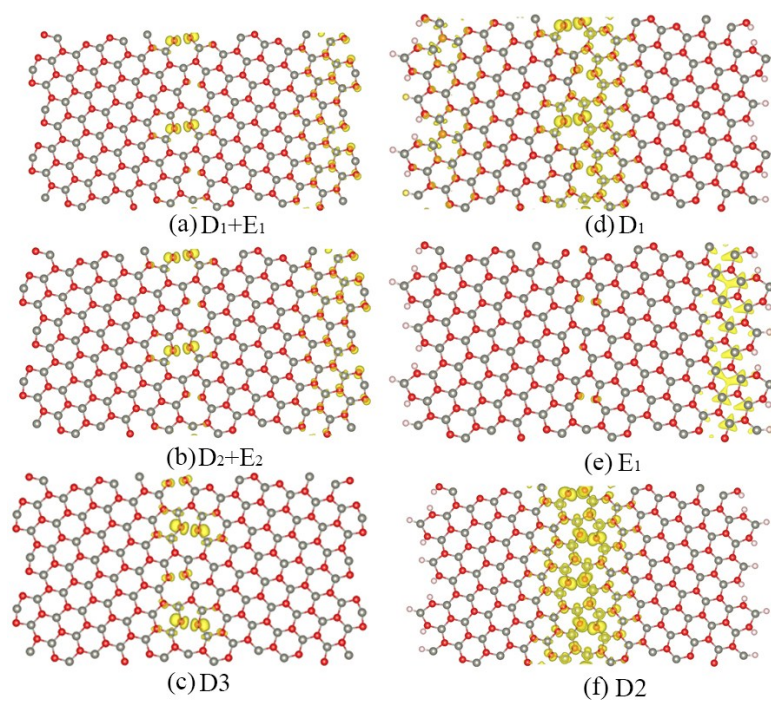


**Figure S5.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for zigzag oriented LAGB1 (a-c) ;and hydrogen passivated zigzag oriented LAGB1 (d-f).

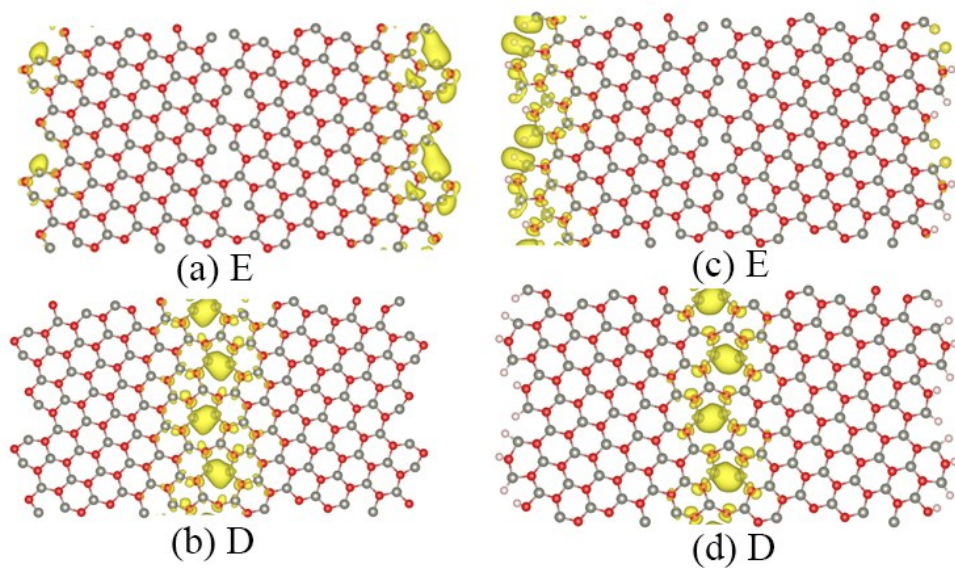




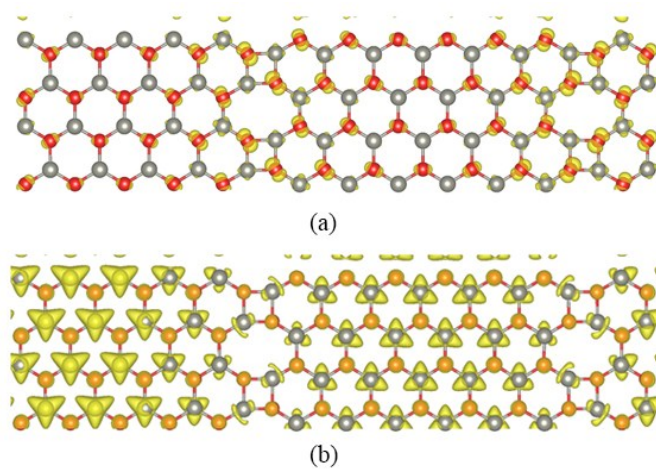
**Figure S6.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for zigzag oriented LAGB2 (a-b); and hydrogen passivated zigzag oriented LAGB1 (c-d).



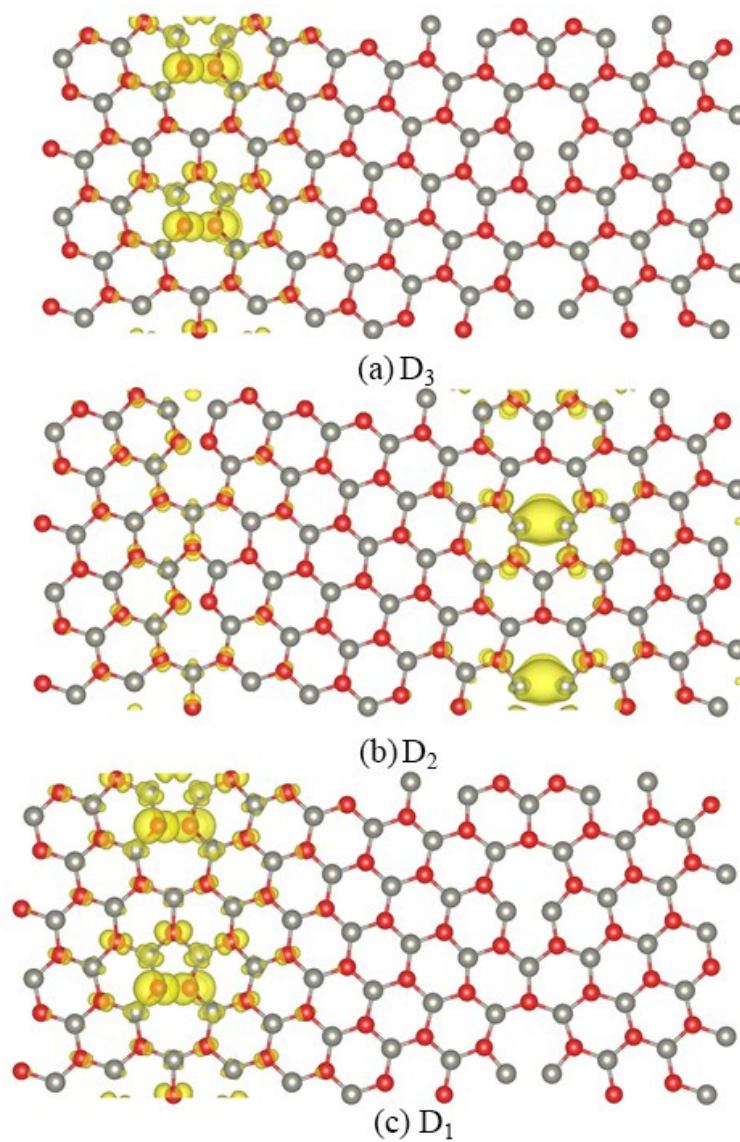
**Figure S7.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for zigzag oriented LAGB2 (a-c); and hydrogen passivated armchair oriented LAGB1 (d-f).



**Figure S8.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for zigzag oriented LAGB2 (a-b); and hydrogen passivated armchair oriented LAGB2 (c-d).

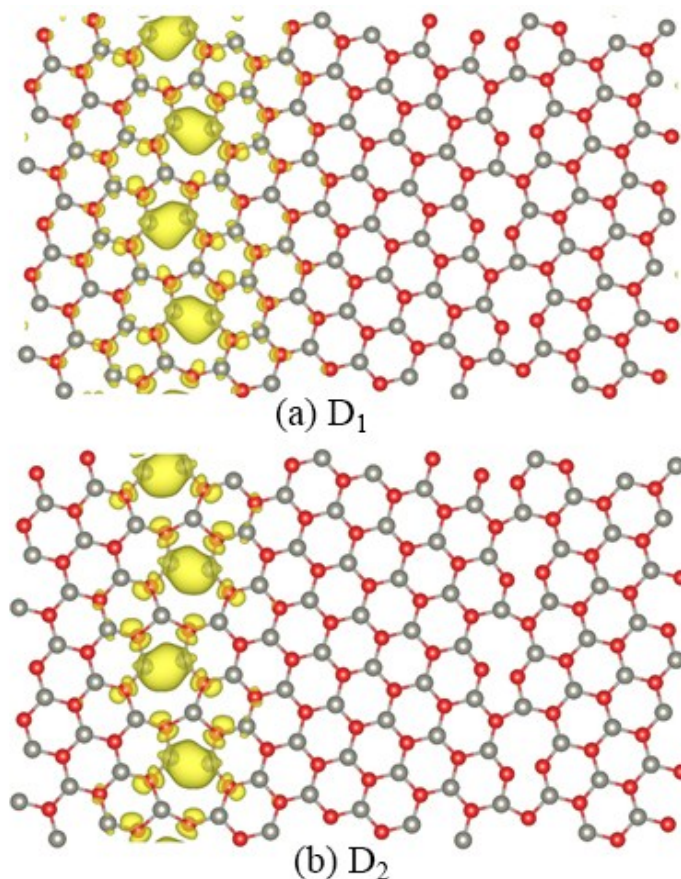


**Figure S9.** Profiles of the VBM (a); and CBM (b) of the ZnO monolayer sheet containing the 4-8 line defect.

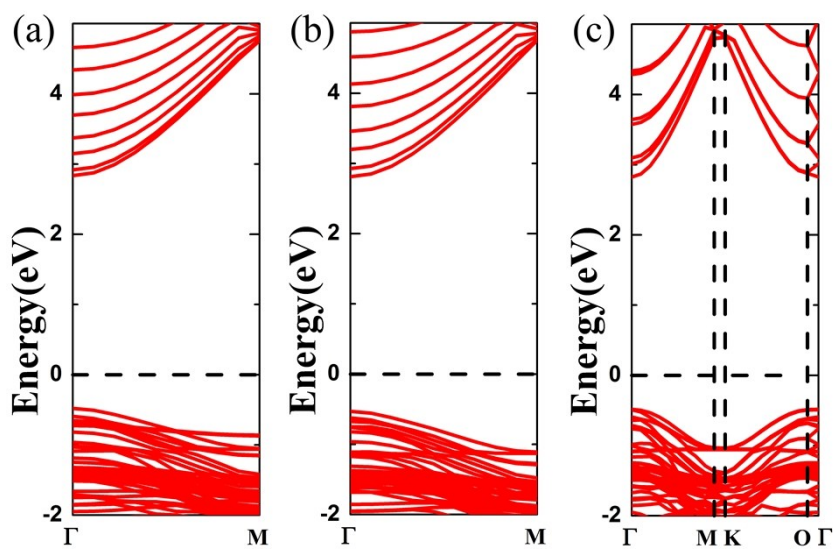


**Figure S10.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for 2D ZnO monolayer sheet with zigzag oriented LAGB.

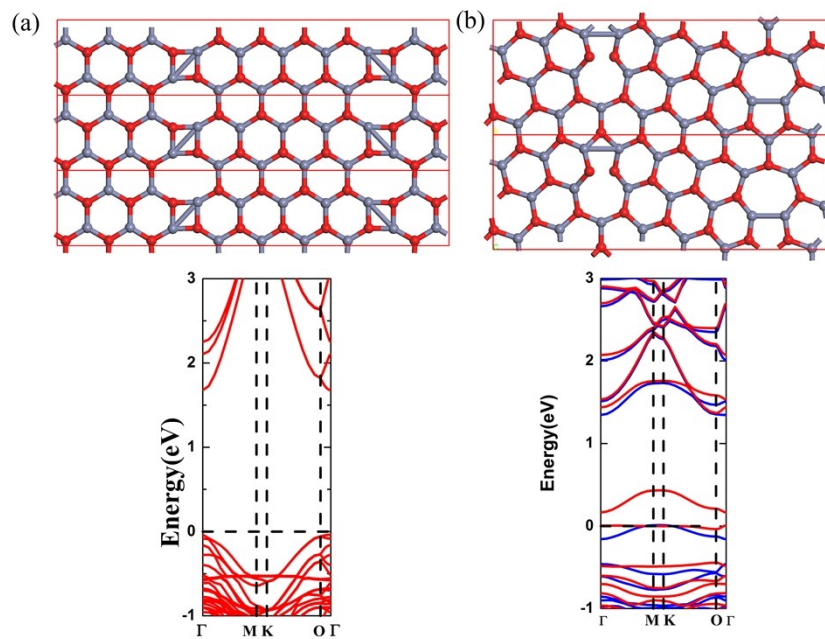




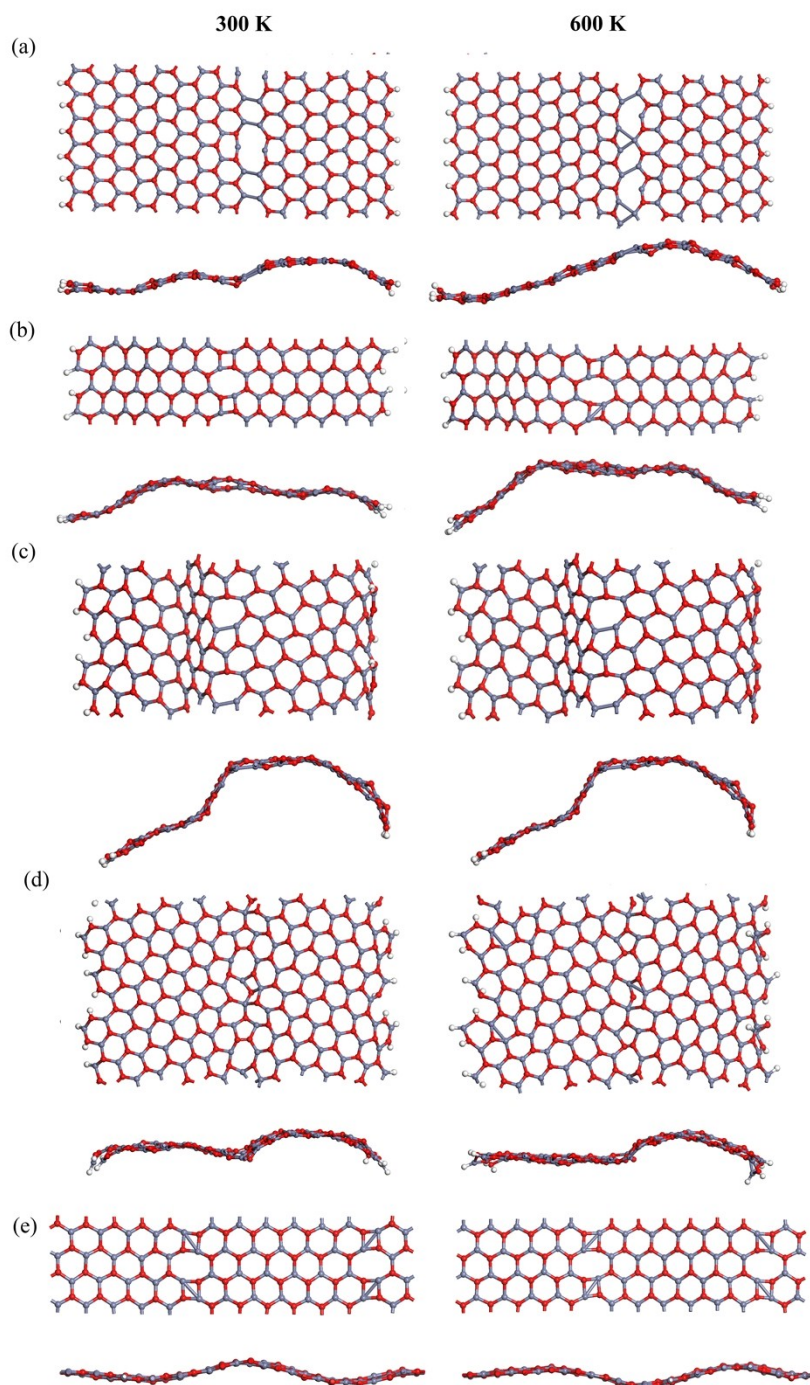
**Figure S11.** Profiles of the edge (E) and defect (D) states at the  $\Gamma$  k-point for 2D ZnO monolayer sheet with armchair oriented LAGB.



**Figure S12.** Computed HSE06 band structures of (a) aZnO-NRs embedded with a 4-8 line defect along the horizontal direction; (b) hydrogen passivated aZnO-NRs embedded with a 4-8 line defect; (c) 2D ZnO sheet embedded with 4-8 line defect, respectively.



**Figure S13.** The optimized structure and computed band structures with higher line defect concentration in 2D ZnO sheet: (a) 4-8 line defect with smaller cell size (29.88 Å, in contrast of 42.99 Å) along  $x$ , (b) zigzag oriented large-angle grain boundary (zLAGB) with smaller cell size along  $x$  (25.55 Å, in contrast of 38.42 Å).



**Figure S14.** Snapshots of BOMD simulations at 5 ps for (a) hydrogen passivated "[OZn]-[ZnO]", (b) hydrogen passivated 4-8 LD-aZnO-NR, (c) hydrogen passivated z-LAGB2, (d) hydrogen passivated a-LAGB1 and (e) 4-8 extended line defects embedded ZnO sheet.