Supporting Information for: Fundamental properties alkali-intercalated bilayer graphene nanoribbons

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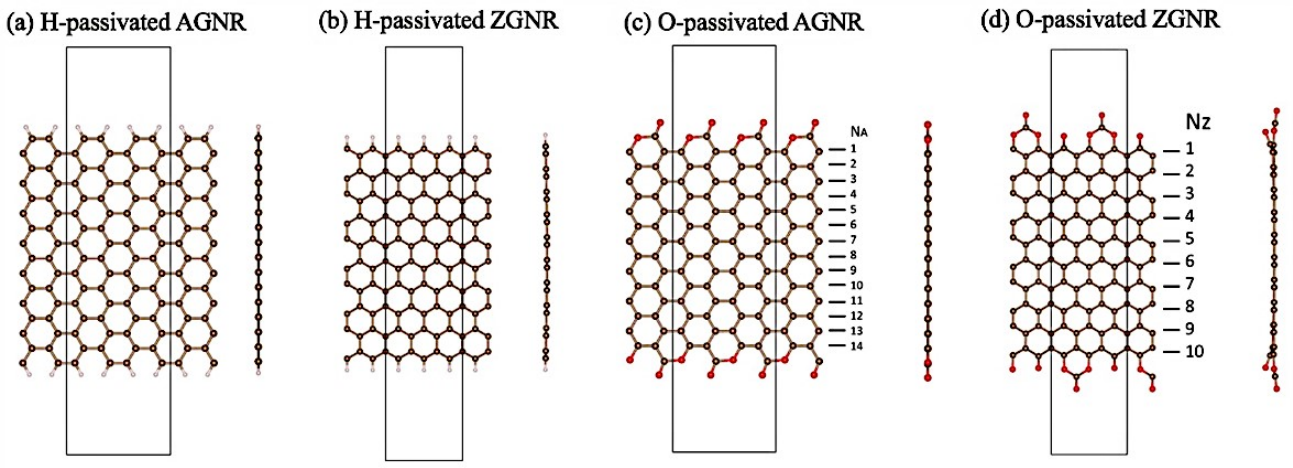

Fig. S1 The geometry of monolayer H-passivated (a) AGNR, (b) ZGNRs and O-passivated (c) AGNR, (d) ZGNR with the number of dimer lines (N_A) and zigzag lines (N_Z) are 14 and 10, respectively.

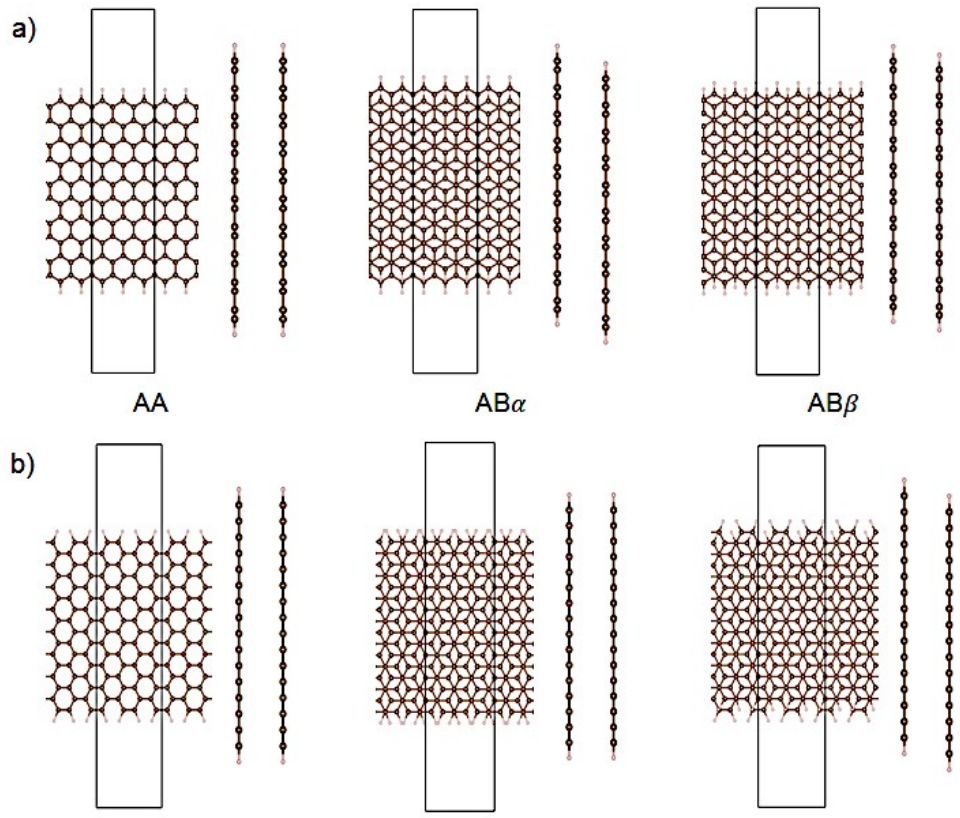

Fig. S2 Structural bilayer H-passivated (a) ZGNRs and (b) AGNRs in AA, $^{AB}{}_{\alpha}$ and $^{AB}{}_{\beta}$, respectively.

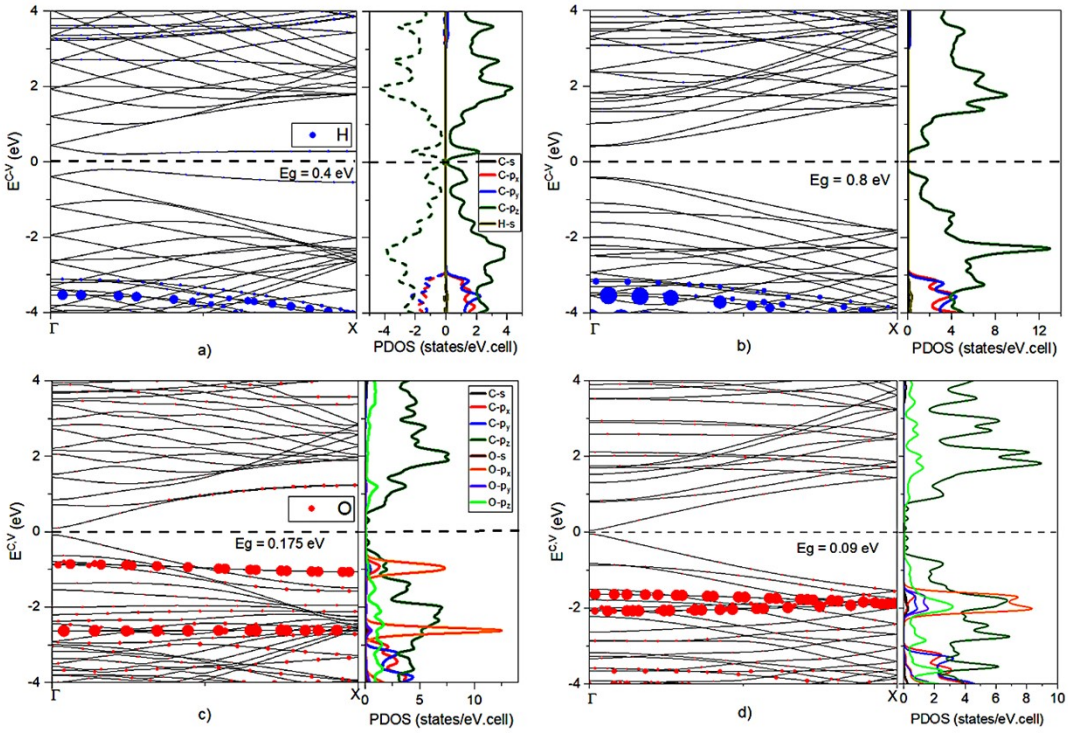

Fig. S3 Band energy spectra and corresponding orbital-projected DOSs of H-passivated (a) ZGNRs, (b) AGNRs and O-passivated (c) ZGNR, (d) AGNR monolayer. Blue and red circles represent hydrogen and oxygen atoms which dominate the band energy spectra, respectively.

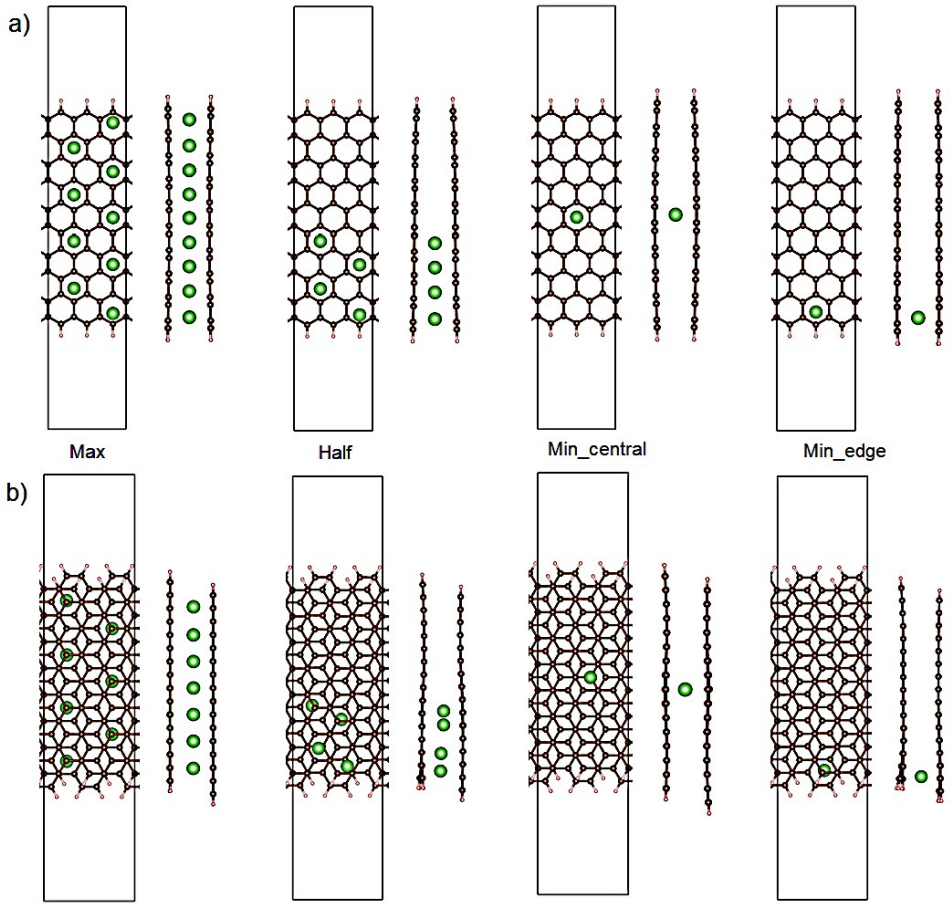

Fig. S4 Structural optimization of alkali-intercalated GNRs in H-passivated (a) zigzag and (b) armchair edges with different concentrations and intercalated positions.

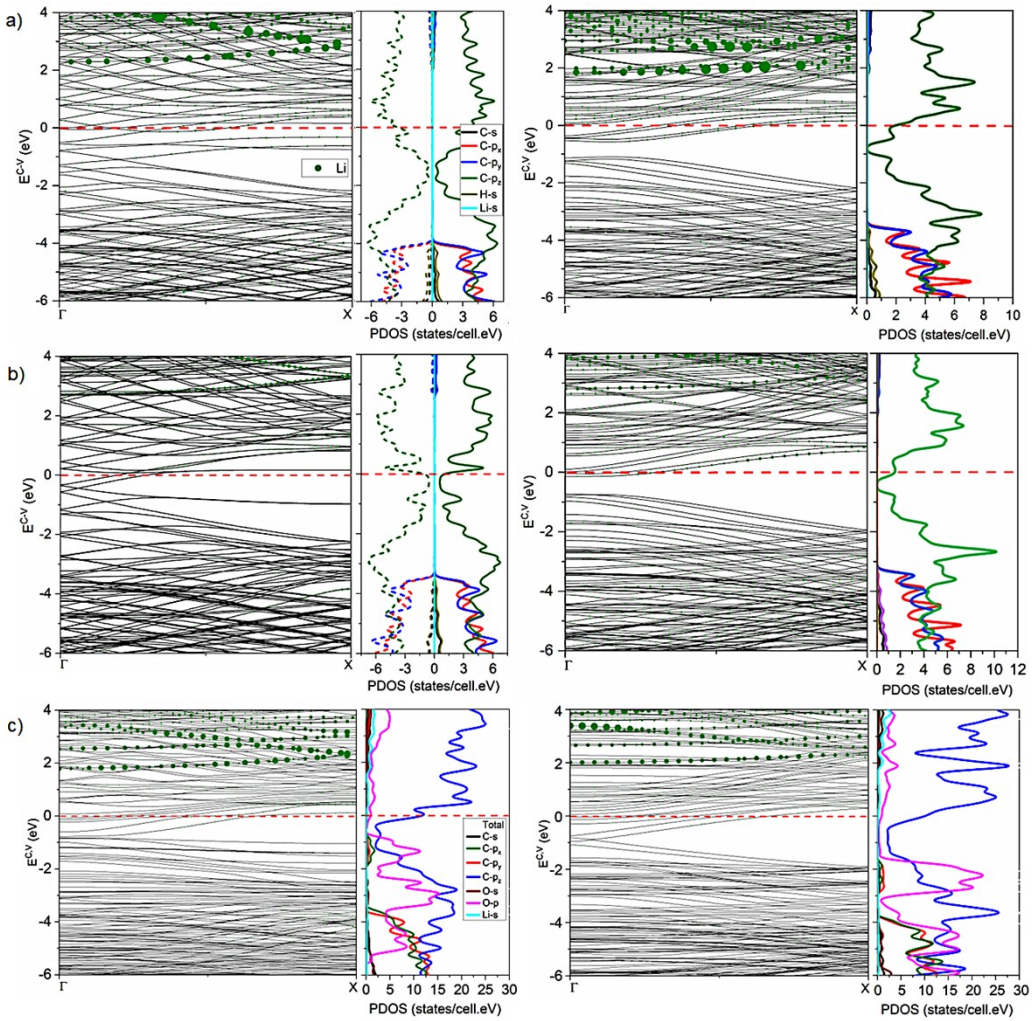

Fig. S5 Band energy spectra and corresponding orbital-projected DOSs of Li-intercalated bilayer H-ZGNR, H-AGNR in (a) half, (b) min_edge intercalation, and (c) O-ZGNR, O-AGNR in half intercalation.

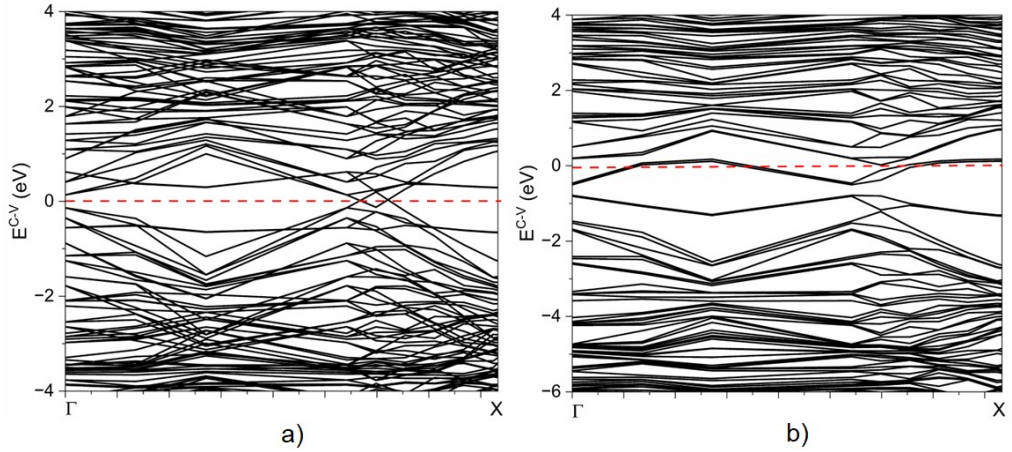

Fig. S6. Band energy spectra of a) bilayer H-ZGNR and b) Li-intercalated H-ZGNR in the min_central case through HSE calculations.

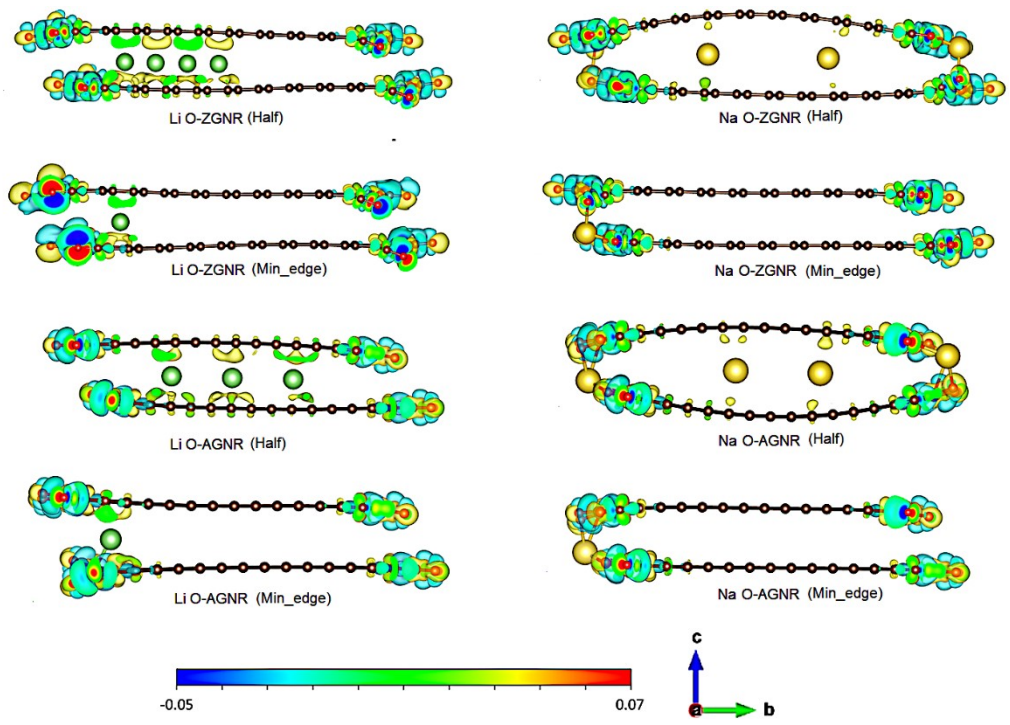

Fig. S7 The charge density of alkali-intercalated bilayer OGNRs in the half and min_edge cases.

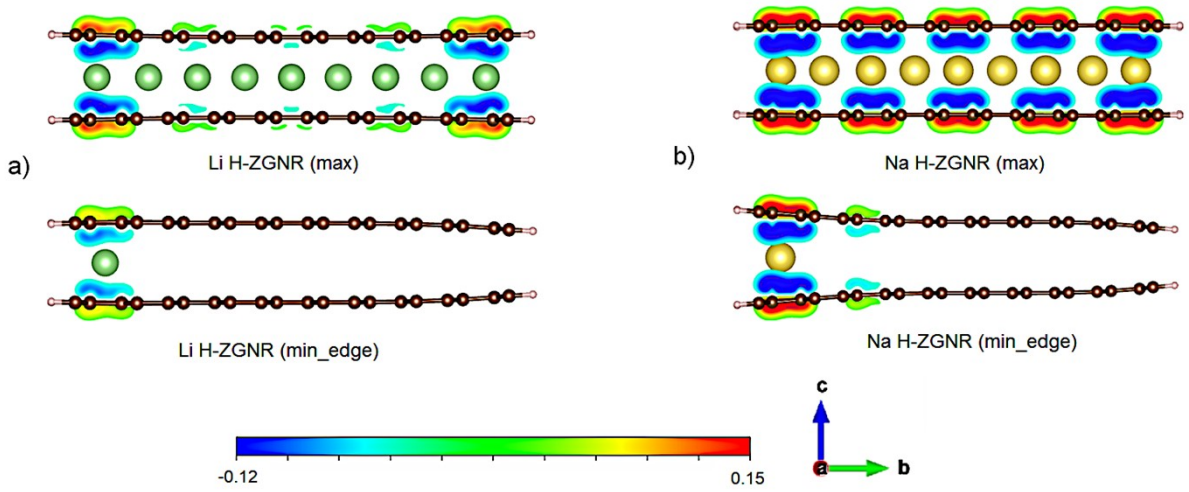

Fig. S8 The charge density of alkali-intercalated bilayer HGNRs in (a) the max and (b) min_edge cases.

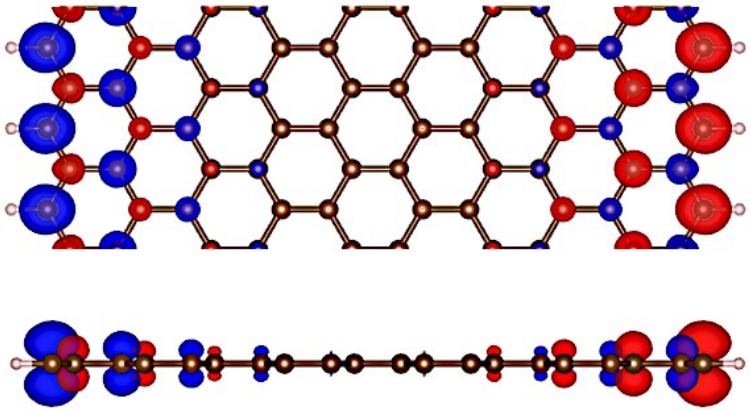

Fig. S9 Spin distribution of monolayer H-ZGNR.

Table S1. The ground state energies (eV) of four types of spin configurations including AFM-AFM, FM-AFM, AFM-FM, and FM-FM in bilayer H-ZGNRs.

	AFM-AFM	FM-AFM	AFM-FM	FM-FM
AA	-115.827	-115.8261	-114.465	-114.4647
AB_{α}	-115.8112	-115.811	-114.4603	-114.425
AB_{β}	-115.792	-115.801	-114.4678	-114.4262

Table S2. The magnetic moment (μB) at the edge atom (outermost carbon edge atom), the total magnetic moment in single layer and bilayer GNR-based systems.

System	Edge atom (edge1, edge2) (layer1/layer2)	Single layer (layer1/layer2)	Bilayer	Magnetism (Single layer-Bilayer)
2L-AA (Fig. 9a)	-0.142, -0.142/0.142, 0.142	-0.79/0/79	0	FM-AFM
2L-AA (Fig. 9b)	0.141, -0.141/-0.141,0.141	0/0	0	AFM-AFM
2L-$^{AB_{\alpha}}$ (Fig. 9c)	-0.144, -0.144/0.144, 0.144	-0.828/0.828	0	FM-AFM
2L-$^{AB_{\alpha}}$ (Fig. 9d)	-0.129, 0.129/0.129, -0.129	0/0	0	AFM-AFM
2L-$^{AB_{\beta}}$ (Fig. 9e)	-0.151, -0.151/0.151, 0.151	-0.75/0.75	0	FM-AFM
2L-$^{AB_{\beta}}$ (Fig. 9f)	-0.15, 0.15/0.15, -0.15	0/0	0	AFM-AFM
Li HzAA half (Fig. 9g)	0.129, -0.02/-0.129,0.02	0.156/-0.156	0	FM-AFM
Li HzAA min_edge (Fig.	0.066, 0/0.066, 0	0.217/0.217	0.434	FM-FM

9h) Na HzAA min_edge (Fig. 9i)	0.074, 0/ 0.074, 0	0.238/0.238	0.476	FM-FM