Supplementary Information for

Edge Functionalized Graphene Nanoribbons with Tunable Band Edges for Carrier Transport Interlayer in Organic-Inorganic Perovskite Solar Cells

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Supplementary Fig. 1 Conduction band edge and valence band edge levels relative to the vacuum level by varying the size of width from 5 to 10 in F-GNR. Red and blue lines indicate the supercells enlarged by 6 times and 3 times, respectively.



Supplementary Fig. 2 Probability of Pb and I atomic orbitals in clean MAPbI₃(001) slab and extracted main peaks of occupied and empty surface states with the contribution greater than 50%.



Supplementary Fig. 3 Probability of Pb and I atomic orbitals against the energy levels relative to electrostatic potential. Yellow and black arrows indicate main peaks of surface states for each heterostructure.



Supplementary Fig. 4 Electrostatic potential of each clean MAPbI₃(001) slab, {H,F}-GNR@MAPbI₃(001) along the z-direction.



Figure S5. PBE band structures of H-GNR@MAPbI₃(001) with the lattice constant adjusted to (a) MAPbI₃(001) and (b) of H-GNR. The energy levels are in reference with the average electrostatic potential at the center layers. The orbital contributions of Pb, I atoms at the surface layer and C atoms at the ribbon, denoted by the blue, red and magenta open circles, respectively.



Figure S6. PBE band structures of F-GNR@MAPbI₃(001) with the lattice constant adjusted to (a) MAPbI₃(001) and (b) of F-GNR. The energy levels are in reference with the average electrostatic potential at the center layers. The orbital contributions of Pb, I atoms at the surface layer and C atoms at the ribbon, denoted by the blue, red and magenta open circles, respectively.

Table S1. Band edge levels (eV) by PBE for H-GNR@MAPbI₃(001) as indicated by the arrows in Figure S5, and the variation ($\Delta \epsilon$) caused by the strain.

| H-GNR@MAPbI ₃ (001) | $a^{\text{het}} = 12.45 \text{ Å}$ | $a^{\text{het}} = 12.78 \text{ Å}$ | Δε |
|---------------------------------|------------------------------------|------------------------------------|-------|
| FSS of MAPbI ₃ (001) | 1.25 | 1.15 | -0.10 |
| ESS of MAPbI ₃ (001) | 3.35 | 3.30 | -0.05 |
| VBM of H-GNR | 2.05 | 2.00 | -0.05 |
| CBM of H-GNR | 3.65 | 3.50 | -0.15 |

Table S2. Band edge levels (eV) by PBE for F-GNR@MAPbI₃(001) as indicated by the arrows in Figure S6, and the variation ($\Delta \epsilon$) caused by the strain.

| F-GNR@MAPbI ₃ (001) | $a^{\text{het}} = 12.45 \text{ Å}$ | $a^{\text{het}} = 12.87 \text{ Å}$ | Δε |
|---------------------------------|------------------------------------|------------------------------------|-------|
| FSS of MAPbI ₃ (001) | 1.30 | 1.15 | -0.15 |
| ESS of MAPbI ₃ (001) | 3.45 | 3.40 | -0.05 |
| VBM of H-GNR | 1.75 | 1.70 | -0.05 |
| CBM of H-GNR | 2.80 | 2.75 | -0.05 |