Supplementary Information for:

Graphene bandgap induced by ferroelectric $Pca2_1$ HfO_2 substrate: a first-principles study

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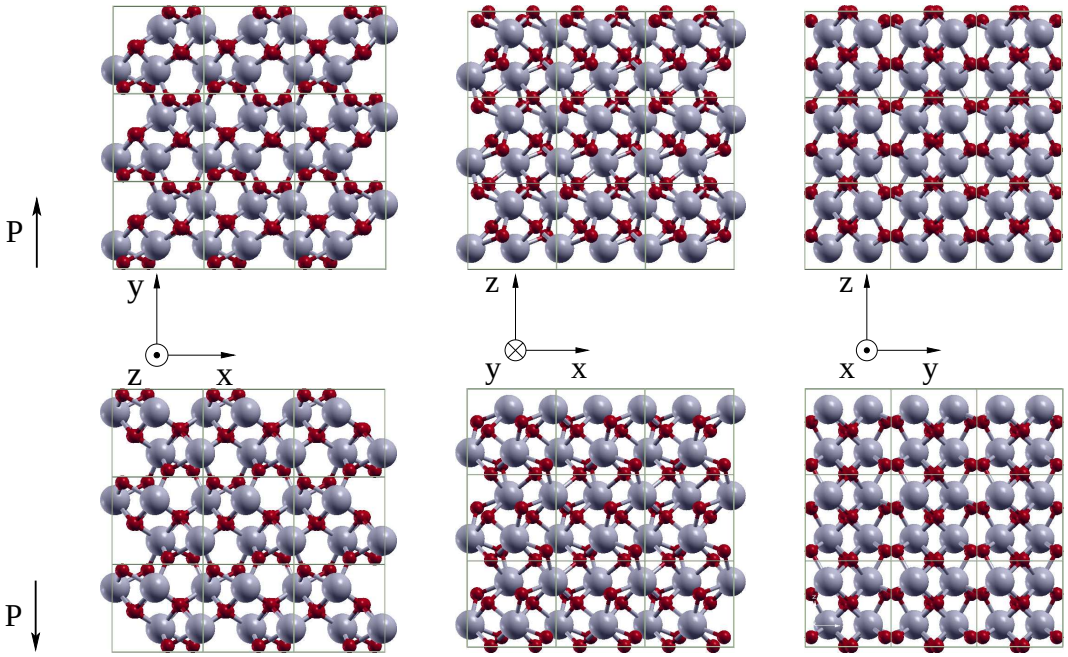
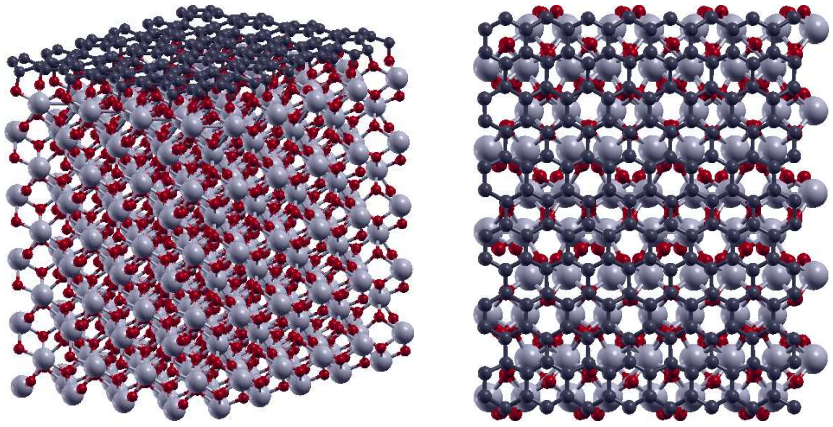
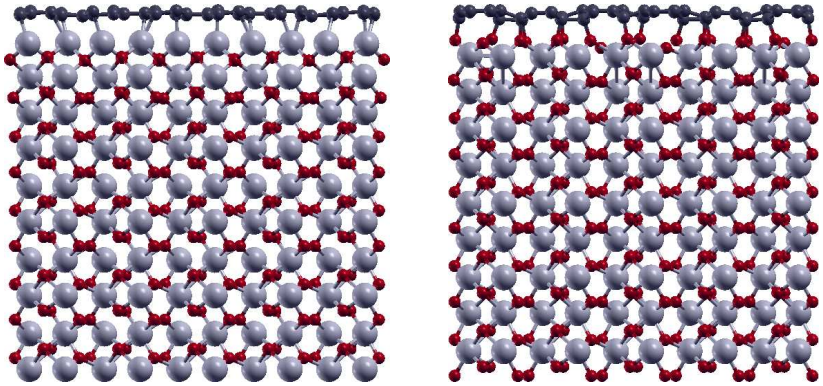


Figure S1: Atomic configurations of bulk HfO_2 with space group $Pca2_1$, exposing (x, y), (x, z) and (y, z) faces, for P and $-P$ polarizations. Color codes: Hf – gray, O– red.



(a) (b)

Figure S2: Graphene on top of the HfO_2 slab, depicting a $4 \times 5 \times 5$ supercell for a better visualization of the G@O-Hf system: (a) 3D view and (b) top view. In this case, graphene interacts with the O-terminated HfO_2 slab.



(a) (b)

Figure S3: Graphene on top of HfO_2 slabs with symmetric terminations: (a) G@Hf-Hf and (b) G@O-O.

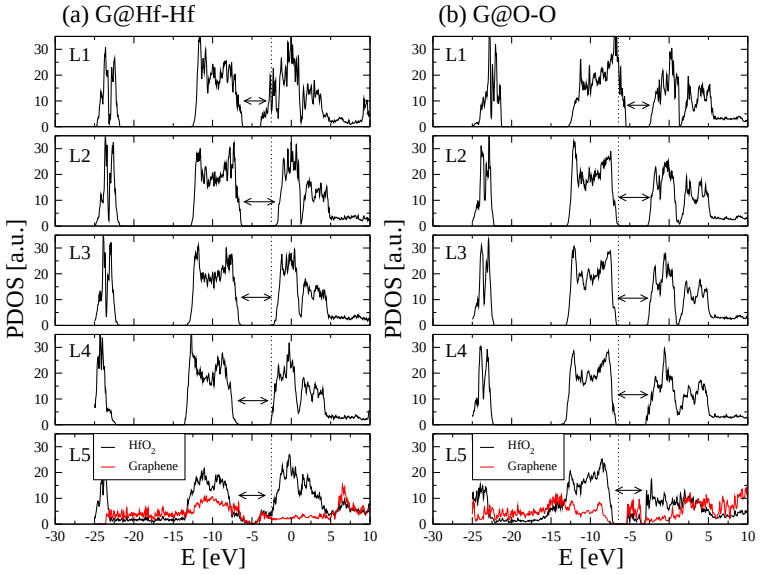


Figure S4: Partial density of states (PDOS) for (a) G@Hf-Hf and (b) G@O-O. Compared to the asymmetrically terminated slabs, G@Hf-O and G@O-Hf, the large shift induced by the polar surfaces is not anymore present. However, there is a similar behavior in PDOS for the interfacial HfO_2 layer and graphene, concerning the energy gaps. One notable difference is found in the slightly shifted Fermi level compared to the G@O-Hf system, which now enters the gap, suggesting that the bottom terminations of the HfO_2 slab or the base substrate can influence the transport properties of graphene.

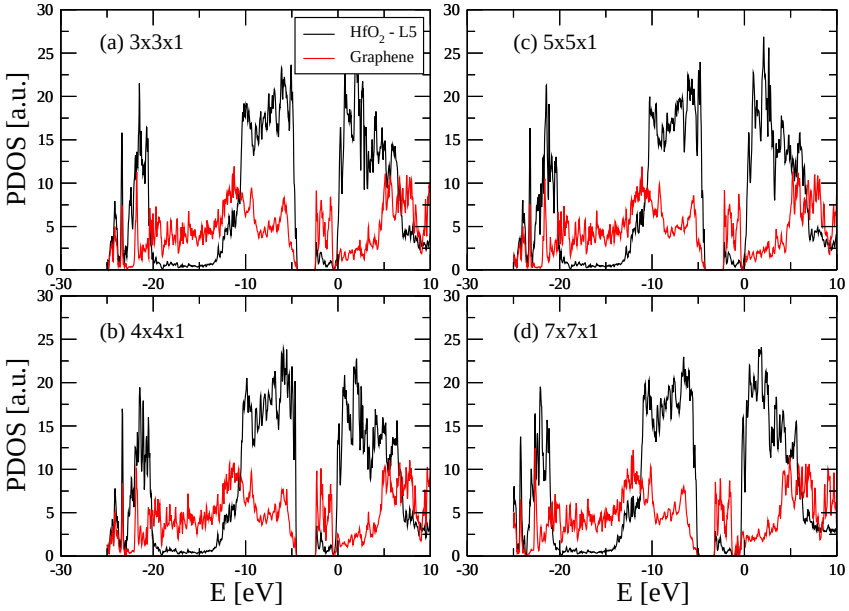


Figure S5: PDOS of the HfO_2 interfacial layer and graphene in G@O-Hf system, for several Monkhorst-Pack k-grids: (a) 3×3 (reference), (b) 4×4, (c) 5×5 and (d) 7×7.

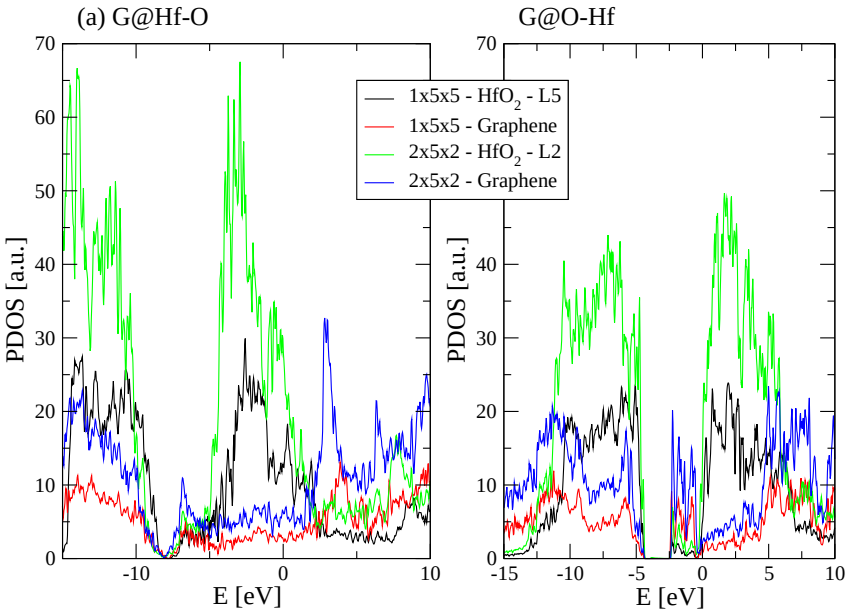


Figure S6: Comparison between two different supercells: PDOS of the HfO_2 interfacial layers and graphene in (a) G@Hf-O and (b) G@O-Hf systems, for the reference $1 \times 5 \times 5$ system (solid lines) and the $2 \times 5 \times 2$ system (dashed lines). Energy gap sizes in the range of 0.1-0.25 eV for G@Hf-O and $\sim$ 1.8 eV for G@O-Hf are observed. Due to the double size along the x direction for the $2 \times 5 \times 2$ system, the number of states corresponding to graphene and L2 interfacial layer is two times larger.

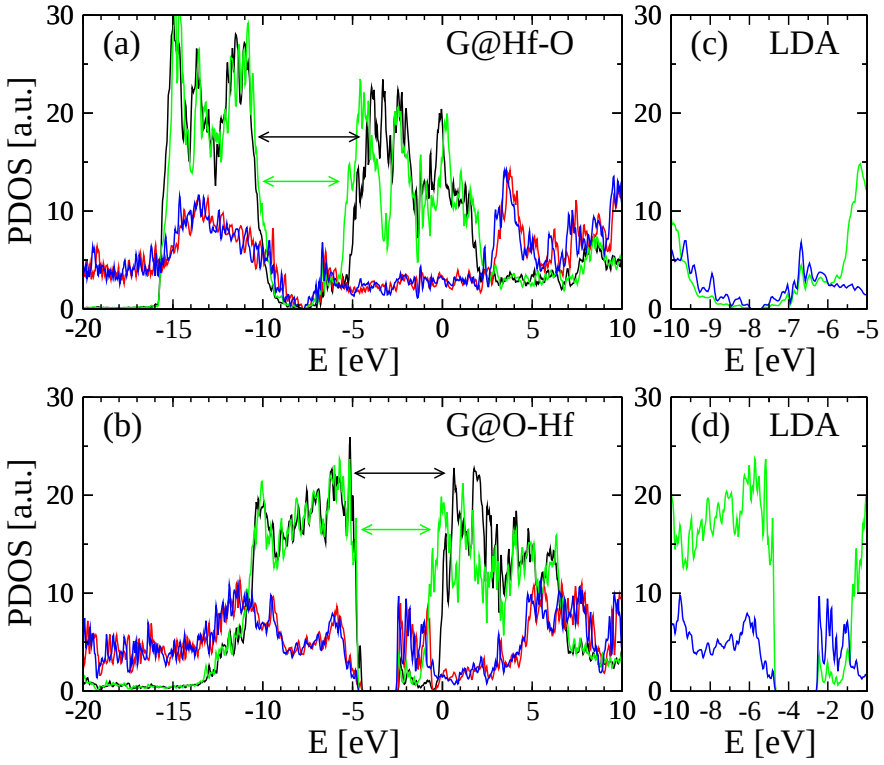


Figure S7: A comparison between LDA+U and LDA for (a) G@Hf-O and (b) G@O-Hf, with a detailed representation of the LDA PDOS indicated in (c) and (d) plots, respectively. Contributions corresponding to HfO_2 L5 interfacial layer and graphene are depicted: LDA+U – HfO_2 (black), graphene (red); LDA – HfO_2 (green), graphene (blue). Using LDA the bulk-like gap of the L5 layer is reduced with ≈ 1 eV, but the contribution to the in-gap states is rather similar and consequently, also the gaps induced in graphene are about the same magnitude as for the LDA+U approach.