Supplementary Information: Electronic properties of bare and functionalized Two-Dimensional Tellurene structures

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Abstract

In this supplementary information, we present the electronic band structure of bare α and β-Te monolayers, MD snapshots for H_2, O_2, F_2 and H_2O adsorbed γ-Te. We also include the phonon dipersion curves and atomic movements for certain negative frequencies for functionalized α and β-Te monolayers and their corresponding electronic band structures. We present MD snapshots for H_2, O_2, and F_2 coverage on α and β-Te monolayers. We also present the optimized geometry and charge difference of H-β-Te and O-β-Te (full and half half coverage) and bare β-Te on a GaSe substrate.

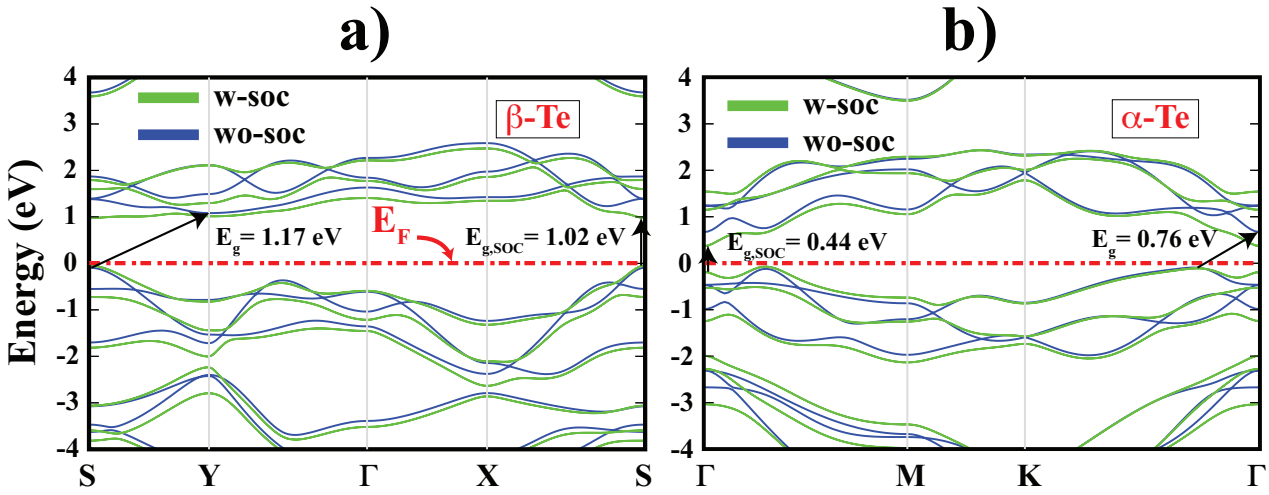

Figure 1: The electronic band structure of bare a) β-Te and b) α-Te with (green) and without (blue) spin-orbit coupling (SOC) effects.

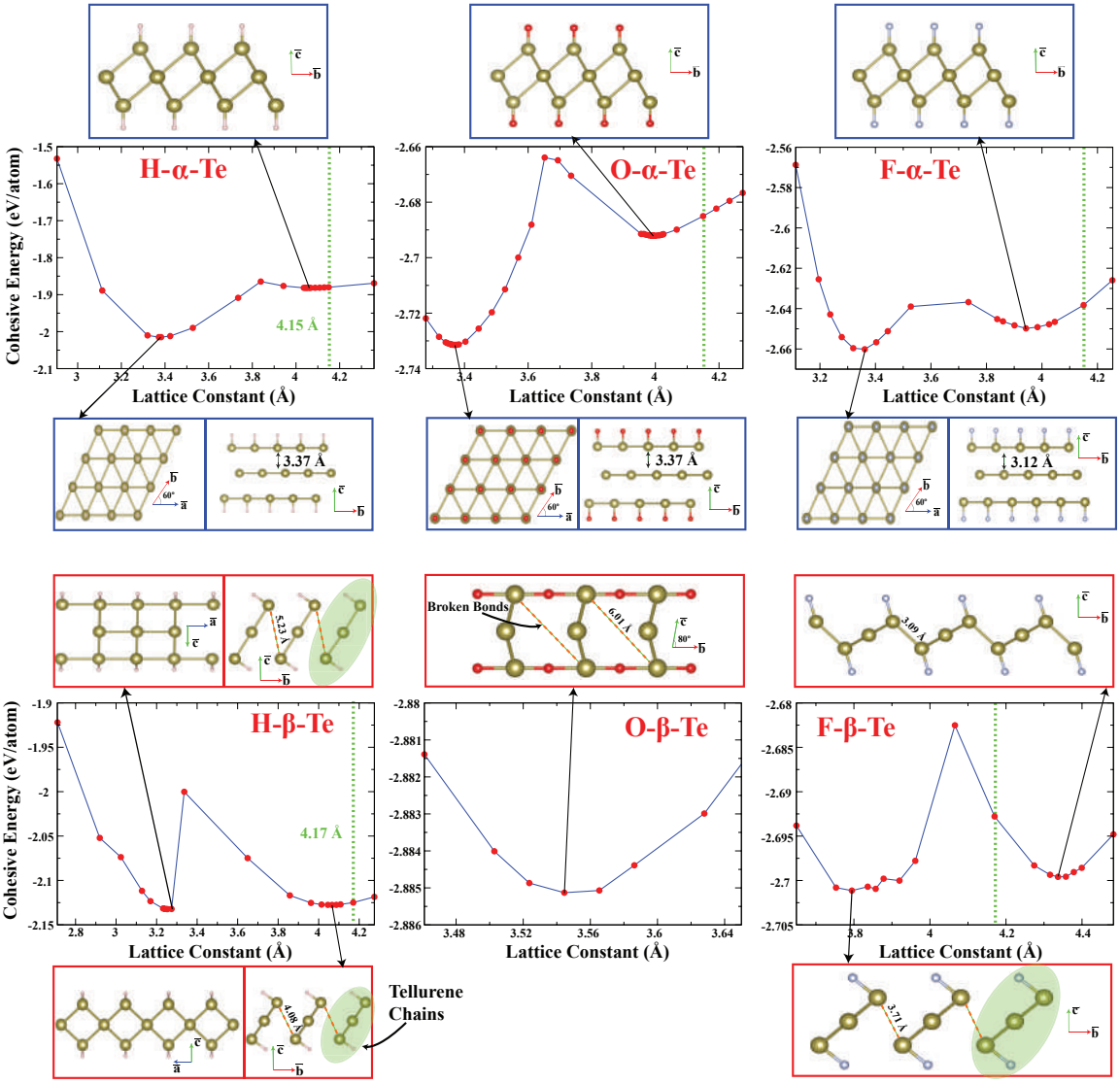

Figure 2: The cohesive energy (in eV/atom) as a function of lattice constant (for the $\vec{a}$ direction) for 2D α and β-Te functionalized with H, O, and F. We observe one local and one global cohesive energy minimum for all functionalized structures except O-β-Te, which contains a single global minimum. The optimized geometry of the functionalized Tellurene structures at each minimum are depicted above and below the corresponding energy landscape. Bond breaking for O, H and F-β-Te is indicated by the orange dashed lines. The green dashed lines on cohesive energy plots represent the lattice constant (in the $\vec{a}$ direction) for bare monolayer α-Te (top row) and β-Te (bottom row).

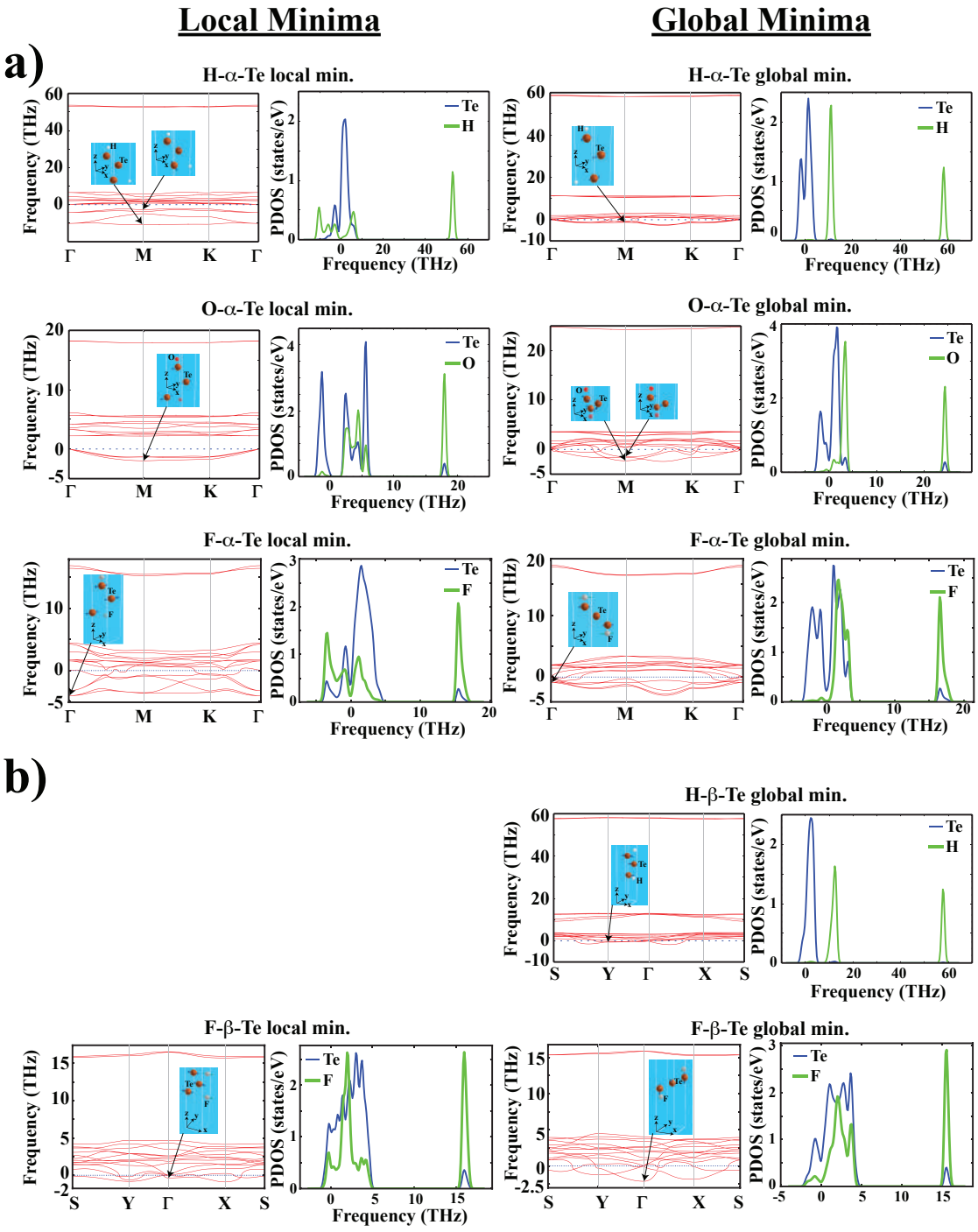

Figure 3: The phonon dispersion curves and phonon projected density of states (PDOS) of the additionally considered functionalized a) α-Te and b) β-Te structures at the local and global minima. These dispersion curves and PDOS plots indicate that all of the depicted structures are unstable in free standing form. Atomic movements for certain negative frequencies at the special high symmetry points are given in the insets.

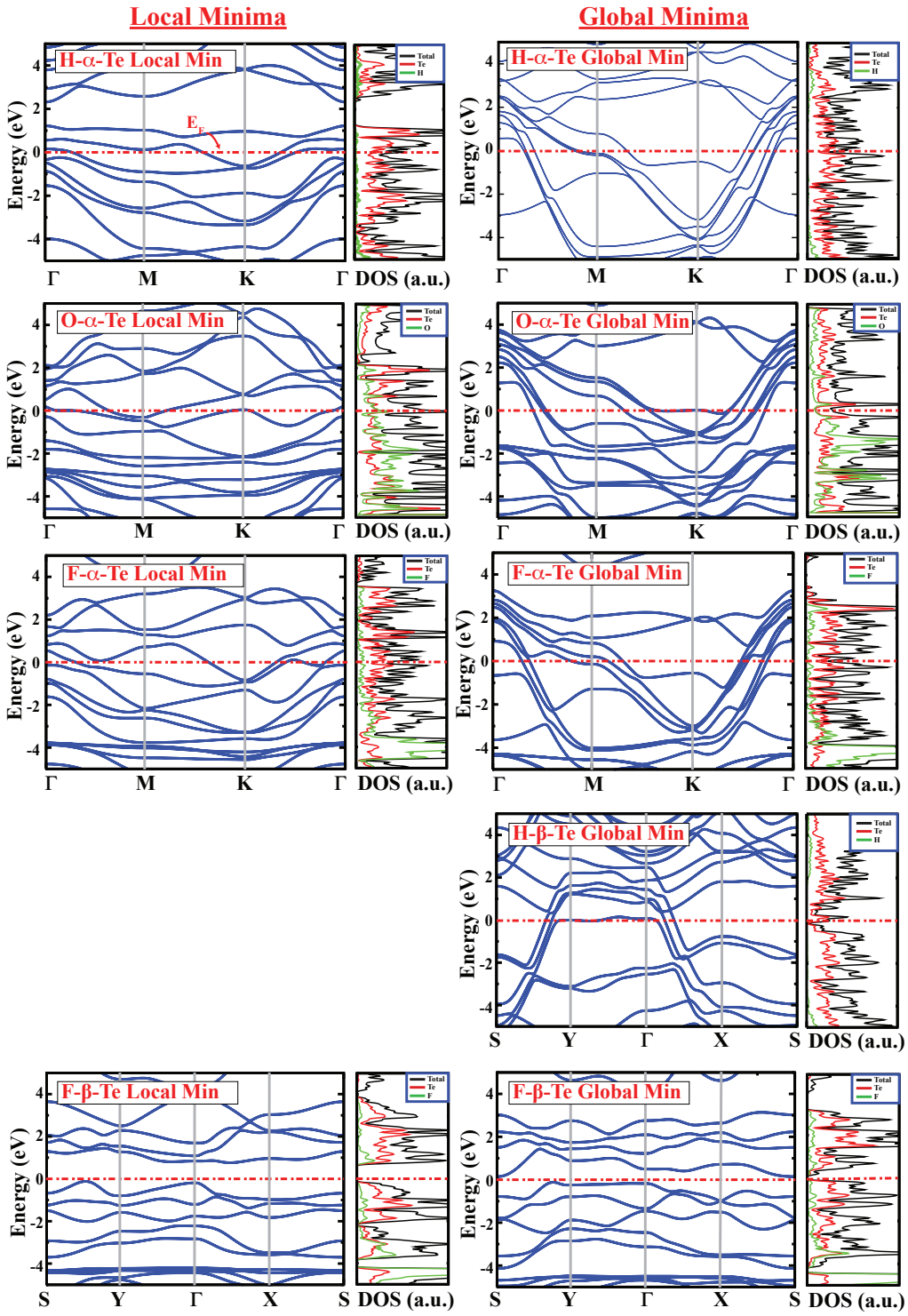

Figure 4: The additionally considered electronic band structures and orbital projected density of states (PDOS) of functionalized Te structures at their local minimum and global minimum.

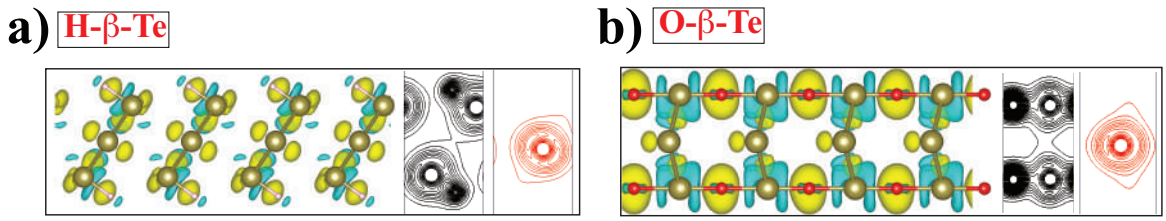

Figure 5: The charge difference for H-β-Te (local min.) and O-β-Te (yellow isosurfaces correspond to charge accumulation and blue isosurfaces correspond to charge depletion) and the total charge density (black contours correspond to the [100] plane at the origin while red contours correspond to the [100] plane cutting through the halfway point of the unit cell). For the charge difference plots, the isosurface value is set to 0.005 e/Å^3 and for the 2D total charge plots, a contour density of 0.01 e/Å^3 was used.

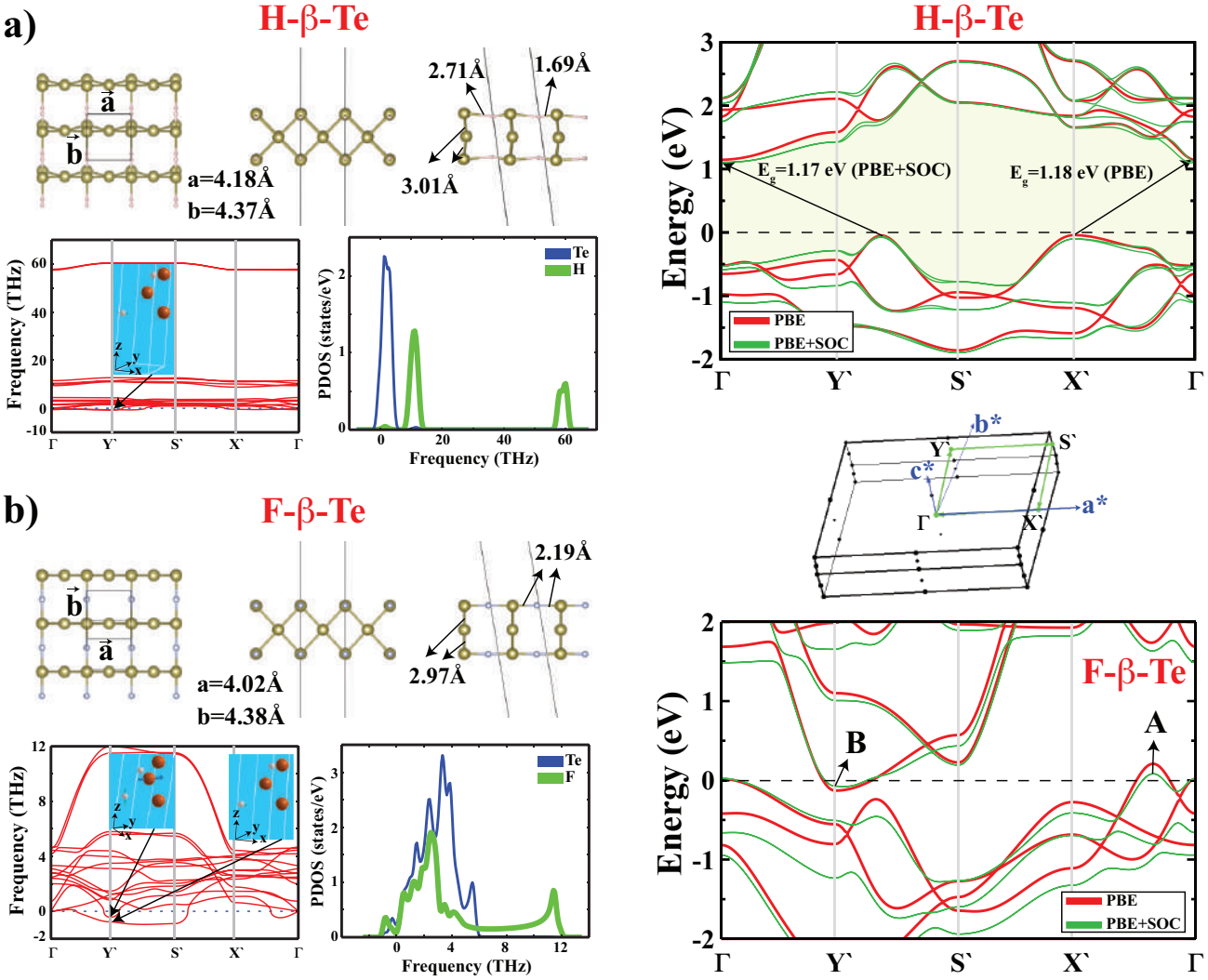

Figure 6: The geometric structure, phonon dispersion curve and phonon projected density of states (PDOS) of an alternate atomic configuration (similar to O-β-Te) of a) H-β-Te and F-β-Te. Our results indicate that this alternate configuration of F-β-Te has a fewer number of negative phonon frequencies than the other F-β-Te structure depicted in Fig. S3. This alternate initial configuration of H-β-Te has relaxed to similar local minimum H-β-Te structure, thus having similar phonon dispersion curves and electronic properties. The electronic band structure of H-β-Te and F-β-Te with and without SOC effects is also given in addition to a diagram of the corresponding Brillouin zone with the blue arrows depicting reciprocal lattice vectors.

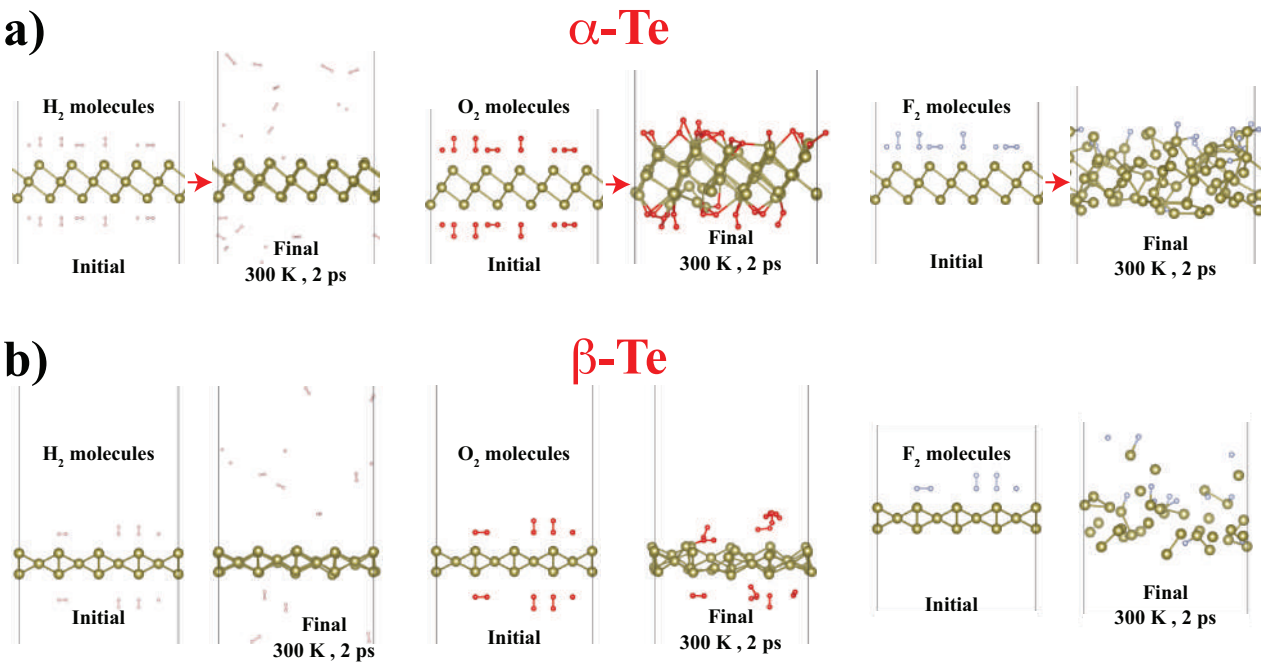

Figure 7: Initial and final (after 2 ps) snapshots of MD calculations at 300 K for high coverage of H_2, O_2, and F_2 molecules above the surface of a) α-Te and b) β-Te.

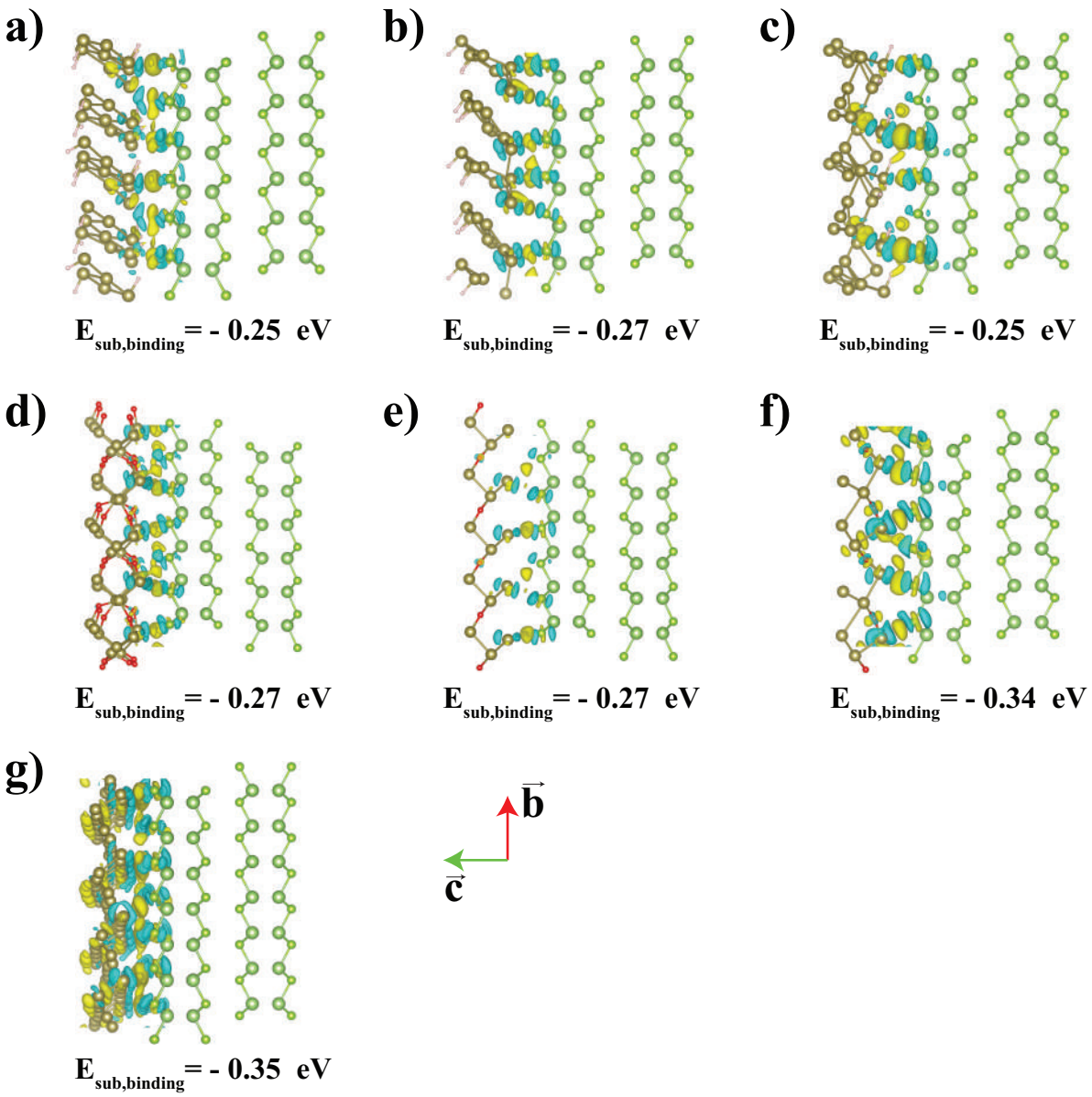

Figure 8: The optimized structures and charge difference of H-β-Te (local min): a) full coverage, b) half coverage (adatoms on opposite side of GaSe), and c) half coverage (adatoms on the same side as GaSe) and O-β-Te: d) full coverage, e) half coverage (adatoms on opposite side of GaSe), and f) half coverage (adatoms on the same side as GaSe). Bare β-Te on GaSe is depicted in g). The isosurface value for the charge difference plots is set to 0.0003 e/Å^3. The yellow isosurface signifies charge accumulation and the blue isosurface signifies charge depletion.

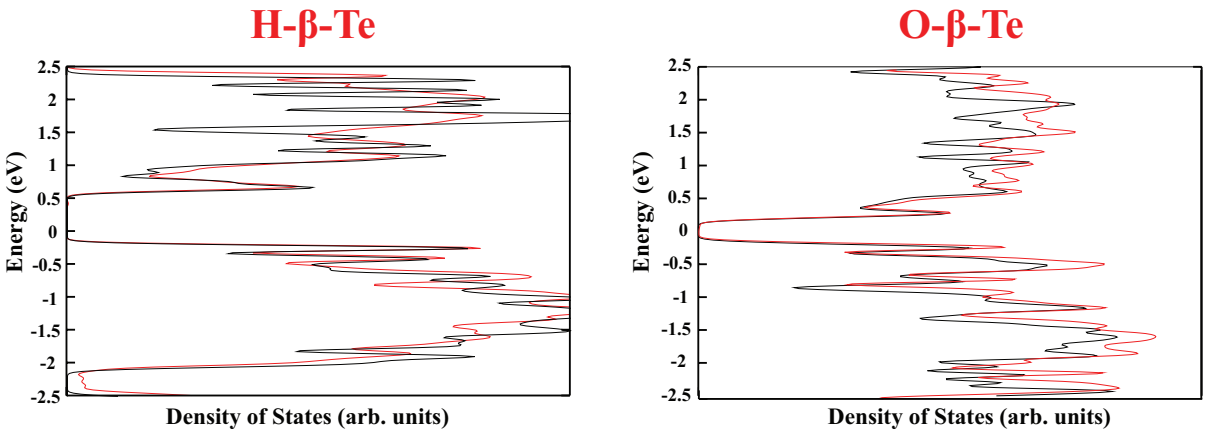

Figure 9: The total (black) density of states of free-standing H-β-Te and O-β-Te compared to the projected (red) density of states (of the respective monolayer) when the structure is on a GaSe substrate.