Electronic Supplementary Information

Embedding epitaxial (blue) phosphorene in between device-compatible functional layers

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Tab. S1 Intensity ratio of the P 2*p* and Au 4*f* core levels depending on the deposition time

Sample	Deposition time [min]	P2*p*/Au4*f* ratio [a.u.]
1[a)]	2.5	0.32
2	1	0.30
3	5	0.34
4	7	0.29
5	10	0.33

[a)]Standard ad-atom rich structure with coverage as shown in Figure S2b and c

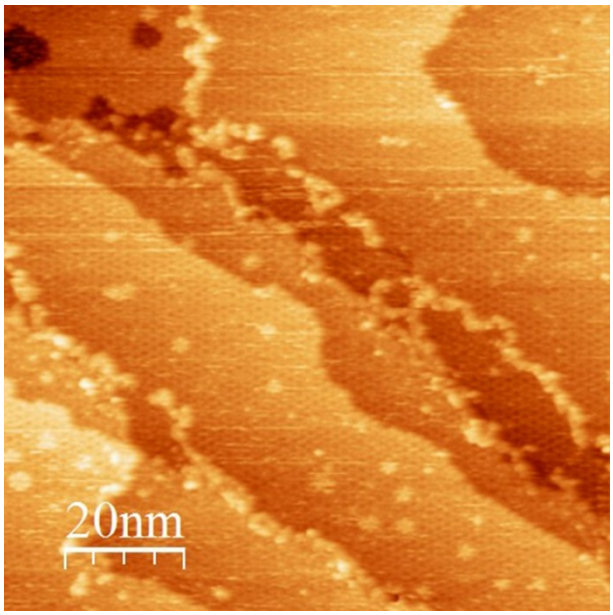

Fig. S1 STM image (100×100 nm^2) of 5 min long (namely twice the time needed to achieve standard ad-atom rich structure, see Table S1) phosphorus deposition showing the formation of phosphorus clusters on top of epitaxial phosphorene on Au(111) ruling out the formation of additional phosphorene layers because of the self-limited growth mechanism.

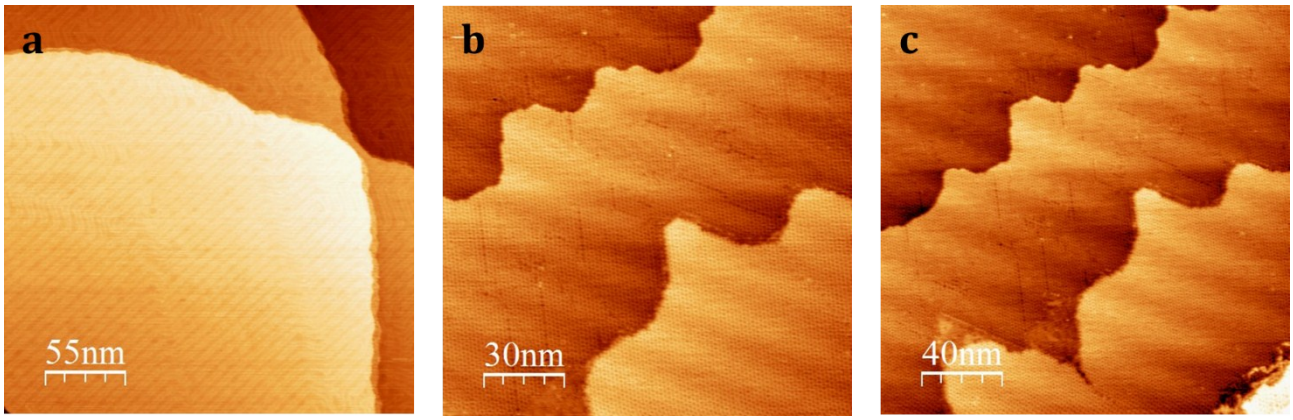

Fig. S2 STM images of bare Au(111) (275×275 nm^2) (a), phosphorene on Au(111) surface 150×150 nm^2 (b) and 200×200 nm^2 (c) showing large-scale epitaxy with uniform and continuous layer covering the gold terraces.

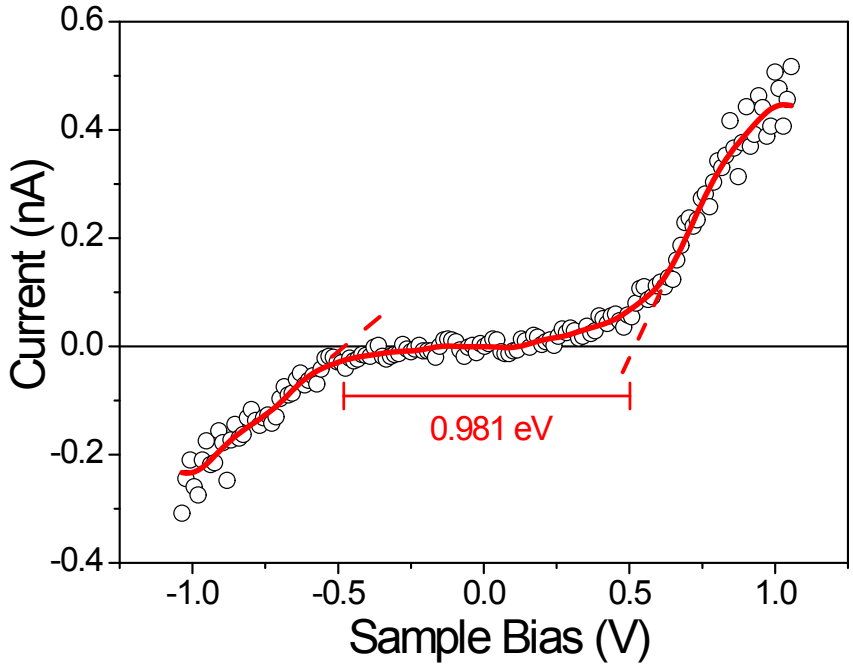

Fig. S3. Average I(V) curve measured by STM from -1 to 1 V on epitaxial phosphorene grown on Au(111) showing a bandgap of 0.981 eV.

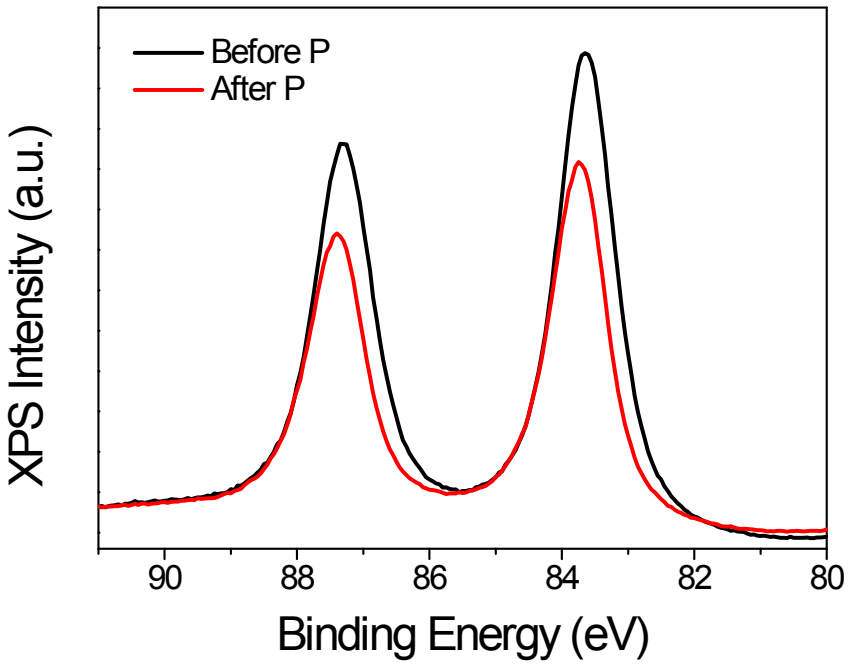

Fig. S4. XPS Au 4*f* core level before (black) and after (red) deposition of phosphorus on Au(111) surface showing a slight shift (<0.1 eV) to higher binding energy after the epitaxial phosphorene growth.

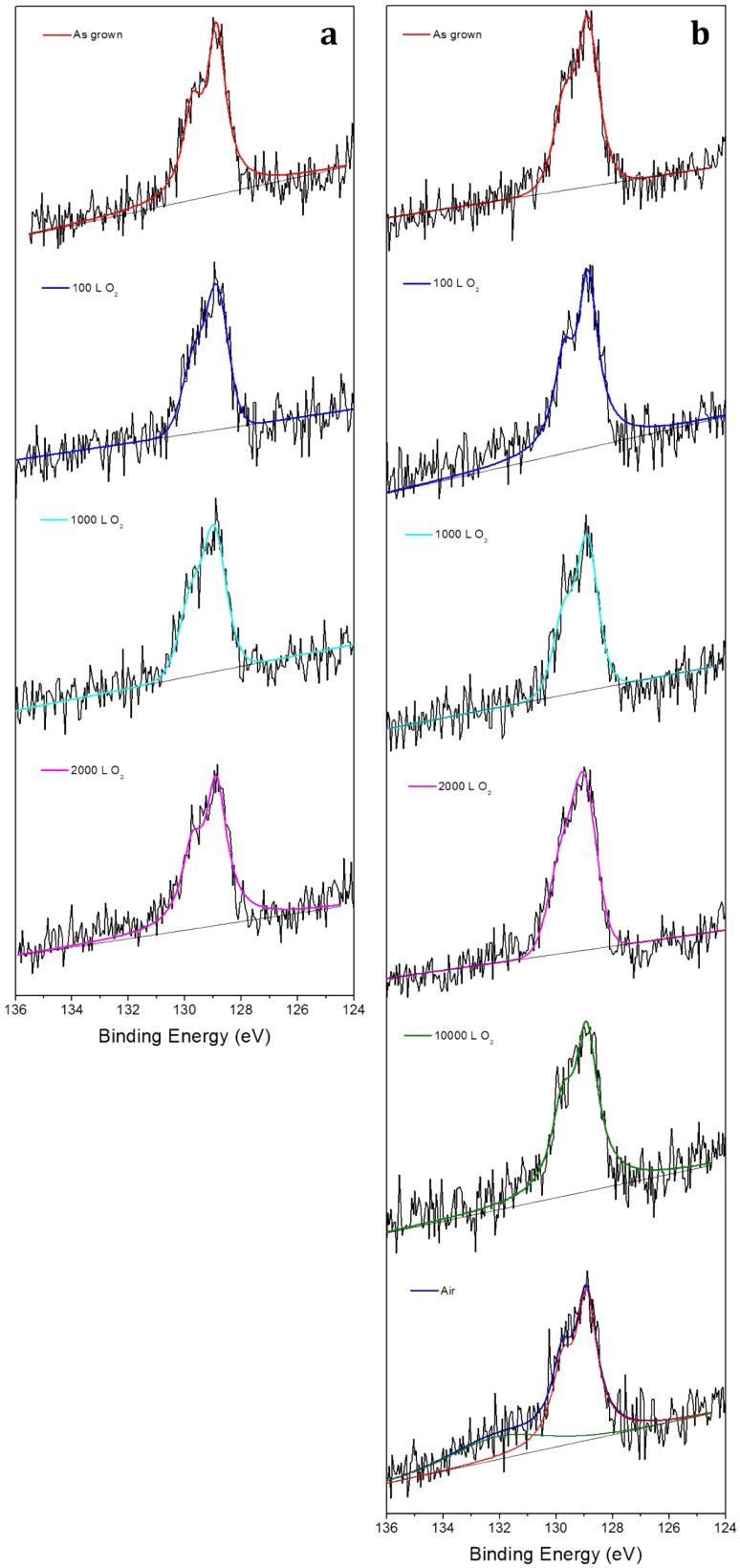

Fig. S5. XPS of the P 2*p* core level when exposed to O_2 doses at partial pressure of 1.3×10^{-7} mbar (a). From top to bottom: as grown (red), 100 L (blue), 1000 L (cyan), 2000 L (magenta). As reported in main text with the O_2 partial pressure of 1.3×10^{-6} mbar (b). From top to bottom: as grown (red), 100 L (blue), 1000 L (cyan), 2000 L (magenta), 10000 L (green), and dry air for 10 min (dark blue).

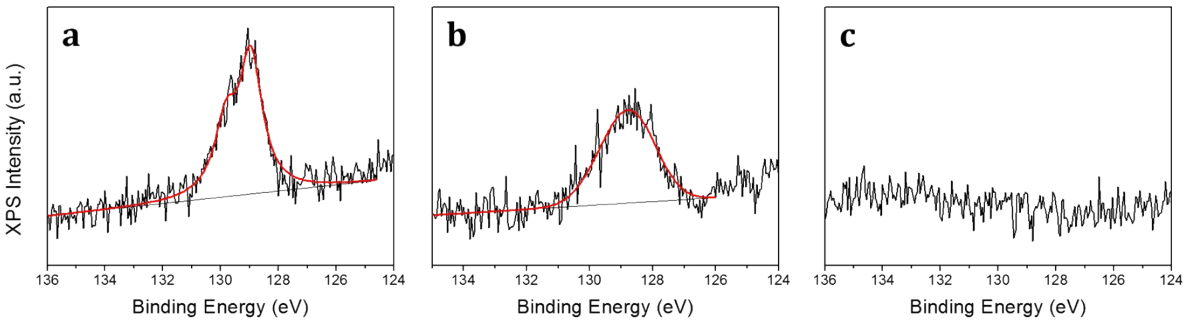

Fig. S6 XPS P 2*p* core level as-grown (a), after 2.5 nm-thick Al_2O_3 capping layer (b), and after air exposure (c) showing that half-thick capping layer (with respect to that discussed in main text) is not enough to appropriately protect the underlying phosphorene. Black and red curves show raw data and fit, respectively.

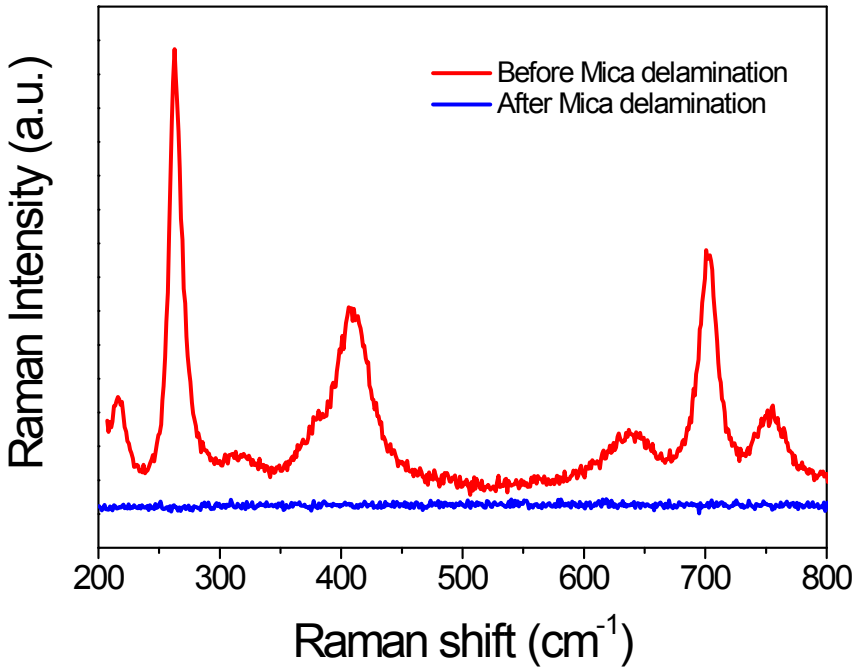

Fig. S7 Raman spectrum of the THF-dipped Al_2O_3-phosphorene-Au/mica sample before (red) and after (blue) mica delamination showing that the THF bath removes the mica.