# Supplementary information:

**Table S1** Lattice parameters, bond lengths and bond angles of four phosphorene allotropes and four graphane conformers.

| | | α-P | | β-P | | γ-P | | δ-P | |
|---|---|---|---|---|---|---|---|---|---|
| | | This work | Ref. | This work | Ref. | This work | Ref. | This work | Ref. |
| a/Å | | 3.30 | 3.28[1] | 3.30 | 3.28[2] | 3.41 | 3.41[3] | … | |
| b/Å | | … | | 3.30 | 3.28[2] | 5.34 | 5.34[3] | 5.72 | 5.56[3] |
| c/Å | | 4.59 | 4.54[1] | … | | … | | 5.49 | 5.46[3] |
| α/° | | 90 | 90[1] | 90 | 90[4] | 90 | 90[3] | 90 | 90[3] |
| β/° | | 90 | 90[1] | 90 | 90[4] | 90 | 90[3] | 90 | 90[3] |
| γ/° | | 90 | 90[1] | 120 | 120[4] | 90 | 90[3] | 90 | 90[3] |
| bond lengths/Å | | 2.221 | 2.23[5] | 2.270 | | 2.279 | | 2.275 | |
| | | 2.269 | 2.26[5] | | | 2.280 | | 2.226 2.265 | |
| bond angles/° | | 96.0 | | 93.3 | | 96.8 | | 104.6 | |
| | | 103.8 | | | | 99.9 | | 102.2 101.9 | |
| | | α-G | | β-G | | γ-G | | δ-G | |
| | | This work | Ref. | This work | Ref. | This work | Ref. | This work | Ref. |
| a/Å | | 2.46 | 2.46[6] | 2.46 | 2.46[6] | 2.46 | 2.46[6] | … | |
| b/Å | | … | | 2.46 | 2.46[6] | 4.26 | 4.26[6] | 4.59 | 3.83[7] |
| c/Å | | 4.26 | 4.26[6] | … | | … | | 4.27 | 3.83[7] |
| α/° | | 90 | | 90 | | 90 | | 90 | |
| β/° | | 90 | | 90 | | 90 | | 90 | |
| γ/° | | 90 | | 120 | | 90 | | 90 | |
| bond lengths/Å | C-H | 1.106 | 1.10[6] | 1.112 | 1.11[8] | 1.103 | 1.10[8] | 1.110 | 1.108[7] |
| | C-C | 1.541 | 1.54[6] | 1.526 | 1.52[8] | 1.516 | 1.52[8] | 1.564 | 1.539[7] |
| | | 1.601 | | | | 1.564 | 1.56[8] | 1.558 1.554 | 1.541[7] |
| bond angles/° | HCC | 105.3 | 107.3[6] | 108.3 | 107.4[6] | 108.4 | 107.16[6] | 106.8 | |
| | | 105.5 | | | | 107.6 | | 107.7 107.1 | |
| | CCC | 105.9 | 111.2[6] | 110.6 | 111.5[6] | 108.5 | 110.7[6] | 111.5 | |
| | | 116.9 | 111.4[6] | | | 111.9 | 112.3[6] | 111.9 111.6 | |

The basic structural data of these eight monolayers are listed in Table S1 and the results are consistent with previous studies.

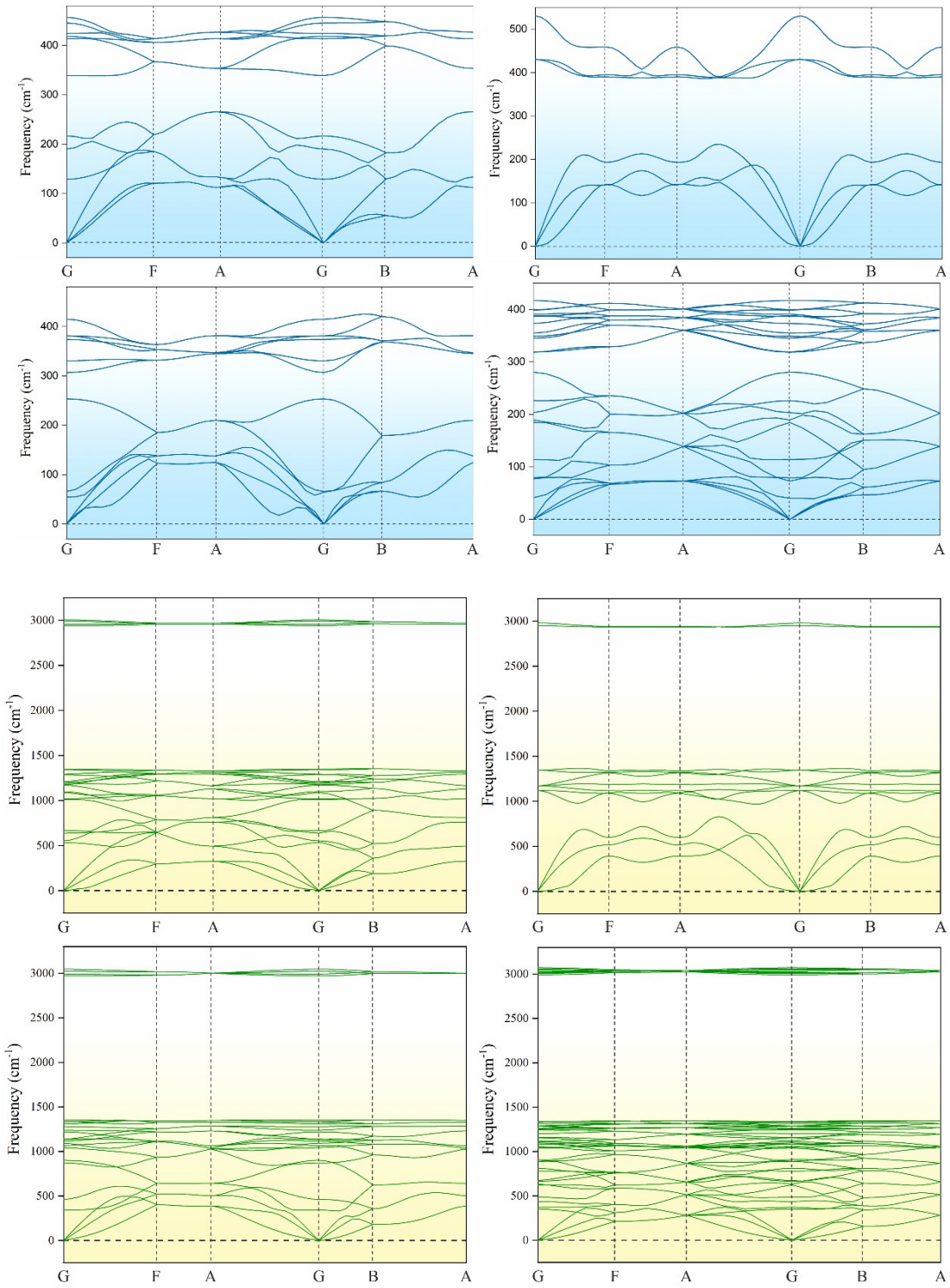


**Fig. S1** Calculated phono dispersions of the eight monolayer polymorphs (a) α-P, (b) β-P, (c) γ-P, (d) δ-P, (e) α-G, (f) β-G, (g) γ-G, (h) δ-G.

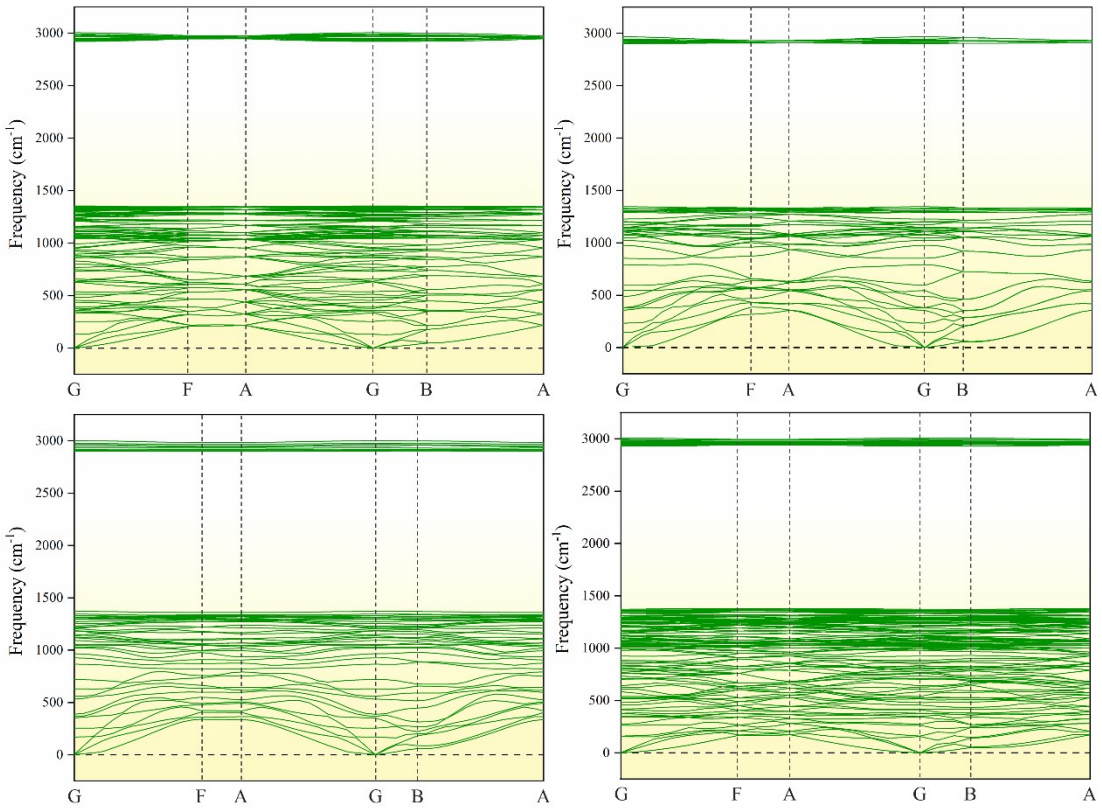


**Fig. S2** Calculated phonon dispersion of four new proposed graphane conformers, (a) γδ-G, (b) αγ-G, (c) βγ-G and (d) αδ-G.

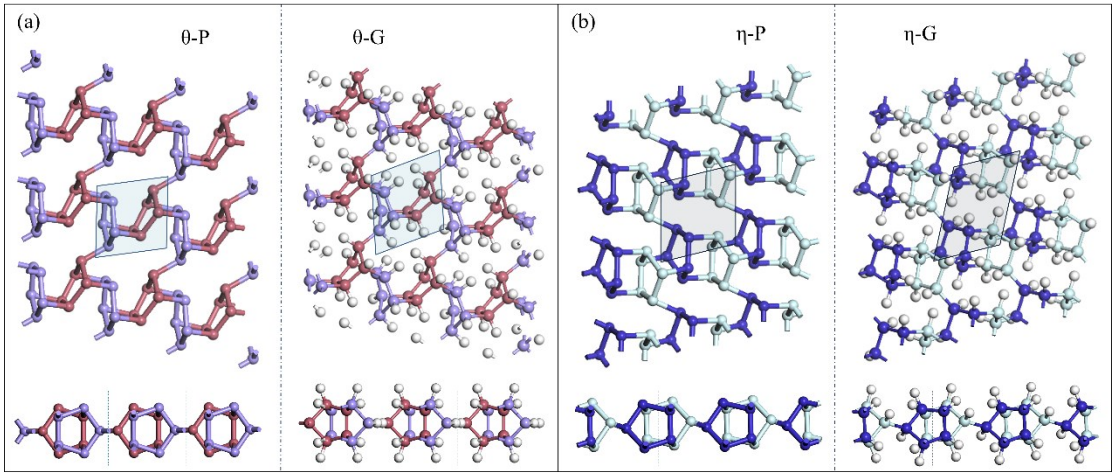


**Fig S3** The top and side view of (a) θ-P and θ-G and (b) η-P and η-G, shaded parts represent original units of these four monolayers.

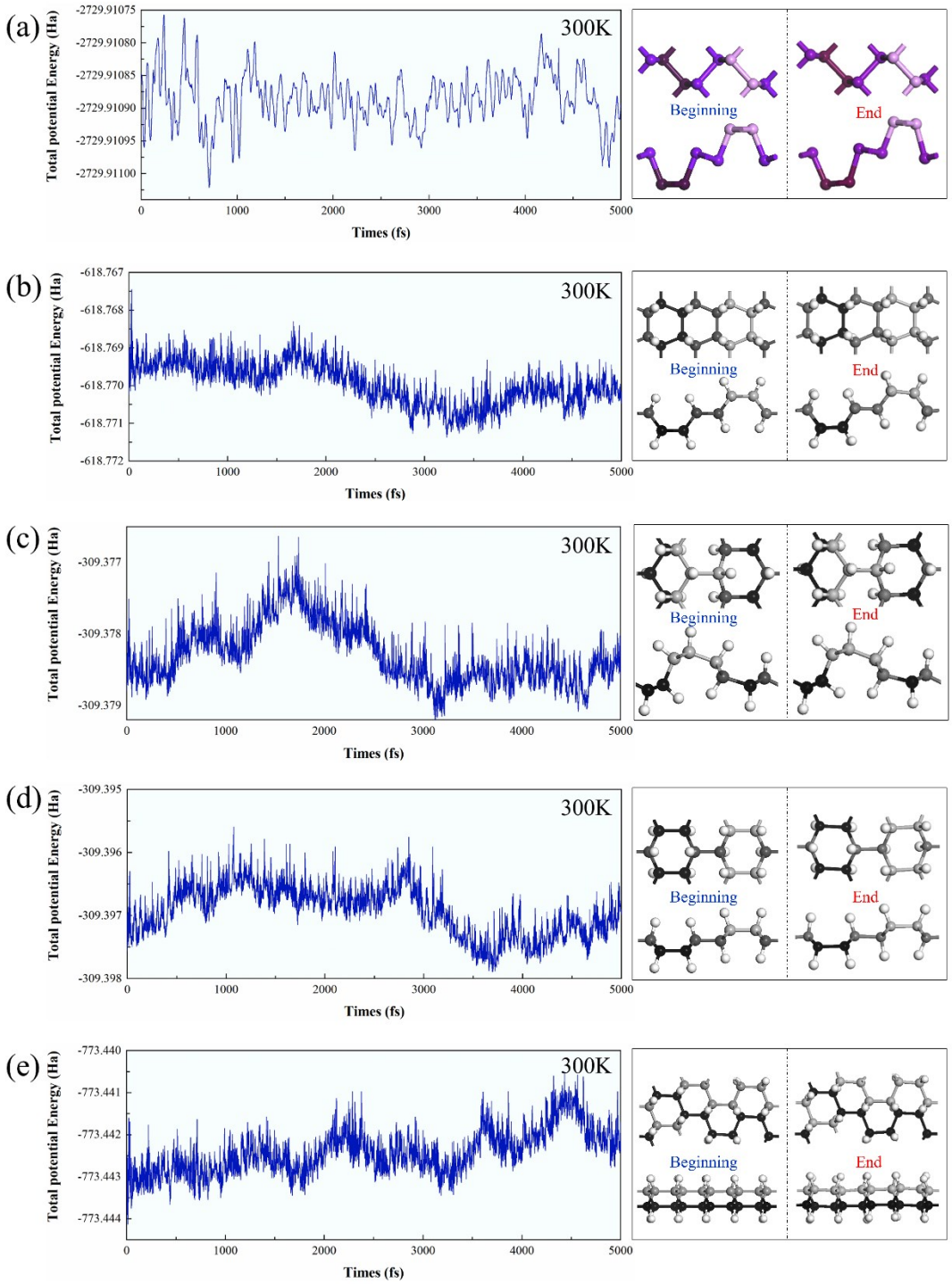


**Fig. S4** The time-dependent potential energy fluctuation in (a) αβ-P, (b) γδ-G, (c) αγ-G, (d) βγ-G and (e) αδ-G.

## References


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