Electronic Supplementary Material (ESI) for Nanoscale

Structural and Electronic Properties of Bilayer and Trilayer Graphdiyne

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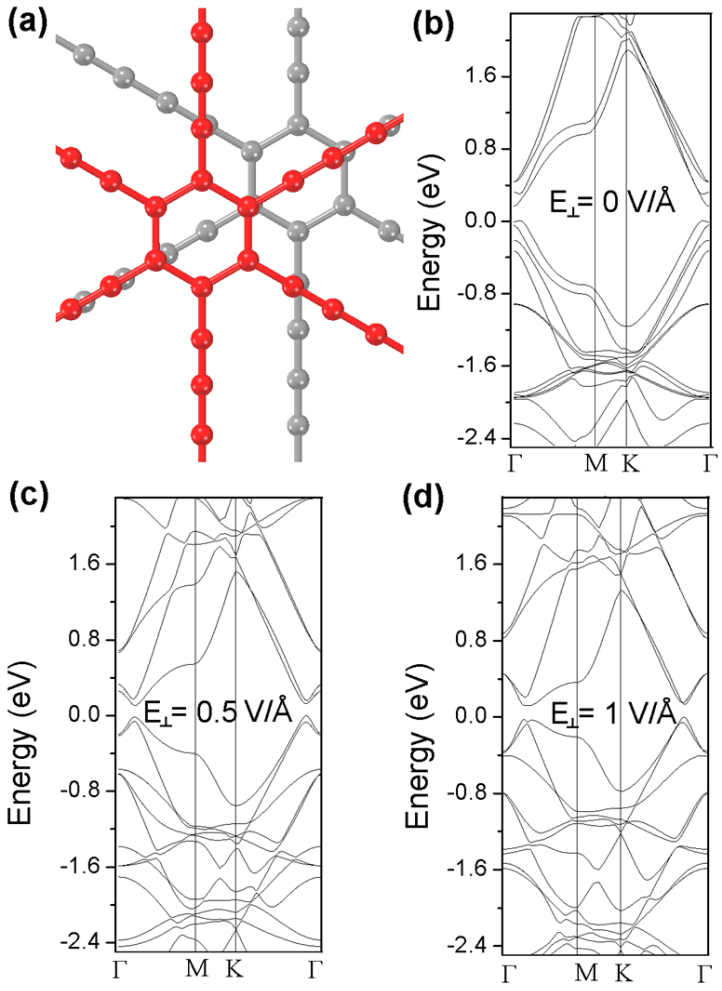

Figure S1. (a) Bilayer graphdiyne with the acetylenic bond of the top layer (red ball) stacked over the benzoic ring of the bottom layer (gray ball), where one of the carbon belonging to the benzoic ring of the top layer located right above that of the bottom layer. Since its interlayer binding energy is 28.9 meV/atom, which is 0.6 meV/atom less than the most stable configuration, this configuration may also exist experimentally under certain circumstances. (b-d) Band structures of this configuration under three different electric field strengths. The bandgaps are 0.16, 0.12, and 0.11 eV when the external vertical electric field are 0, 0.5, and 1 V/ Å respectively, which also supports the conclusion about the electric field effect derived in the main article.

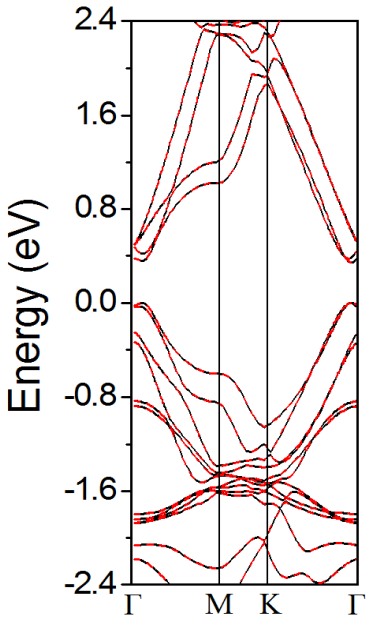

Figure S2. Comparison of the electronic structure of bilayer graphdiyen stacked in the AB(β1) manner calculated with vdW correction (black solid line) and without (red dashed line). These two bands are almost identical. There is also no appreciable difference in the band structure between the method with and without vdW correction for monolayer graphdiyne and bilayer graphdiyne in other configurations. Therefore it is safe to conclude that the vdW correction derived from the work of Ortmann *et al*[1] causes negligable influence on bandstructure for a given configuration.

Reference

1. F. Ortmann, F. Bechstedt and W. G. Schmidt, *Physical Review B*, 2006, **73**, 205101.