

Supplemental Material

To extend our work to other system containing localized p electrons, we studied the magnetism of graphene with single vacancy based on the DFT+ U method.

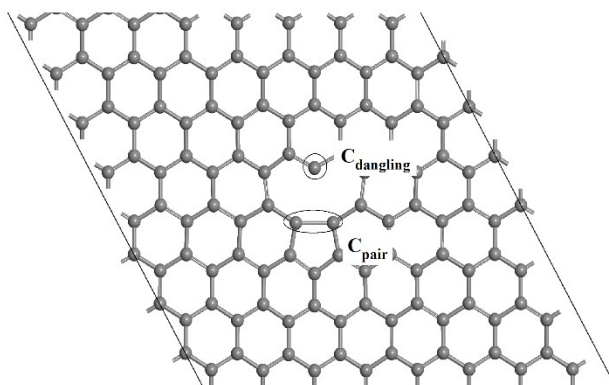


Figure S1 Optimized geometric structure of graphene with single vacancy. The carbons nearby the vacancy are denoted as C_{pair} and C_{dangling} , respectively.

The optimized geometric structure is obtained based on DFT with PBE type of GGA functionals, as shown in Figure S1. As discussed in Ref. 1, nearby the vacancy, there is one dangling bond (C_{dangling}) and two localized p_z electrons which form a localized π bond (C_{pair}). Both contribute strongly to the magnetism of the system.

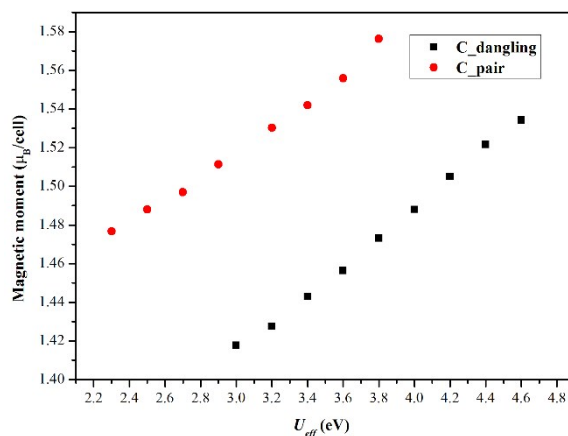


Figure S2 Magnetic moment of the system as a function of U_{eff} at sites C_{dangling} and C_{pair} , respectively.

To study the U -dependent magnetism of the system, we calculated the magnetic moment of the system using the DFT+ U method with a series of values of U_{eff} on the sites C_{dangling} and C_{pair} , respectively. As shown in Figure 2S, the magnetic moment increases almost linearly with the increasing of U_{eff} at the C_{dangling} site and the C_{pair} site, indicating that Coulomb on-site repulsion cannot be ignored between localized p electrons in this system.

To estimate the contribution of the localized density of states (DOS) in this system, we calculated the PDOS of p_z electrons of carbon atoms at GNR and ELD. As shown in Figure S3, most of the localized DOS near the Fermi level is contributed by the carbon atoms at edge-A and C_2 sites.

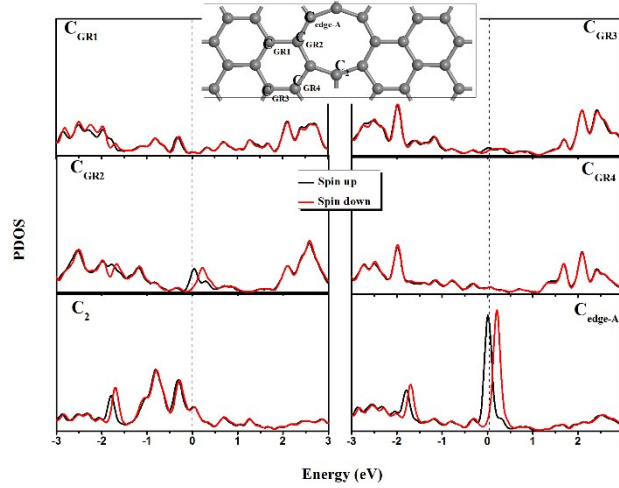


Figure S3 PDOS of p_z electrons of carbon atoms at different sites. C_{GR1-4} are carbon atoms at the GNR; C_2 and C_{edge-A} are carbon atoms at the center and edges of the ELD, respectively, as shown in the insert. The Fermi energy is set to be zero.

Reference

- [1] O. V. Yazyev and L. Helm, *Phys. Rev. B*, **75**, 125408 (2007).