Linearly doped graphene by charge transfer complex

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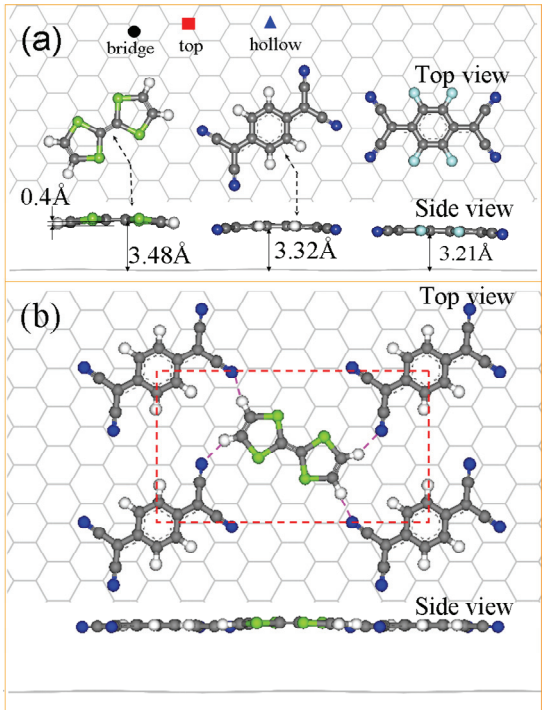
1. Introduction

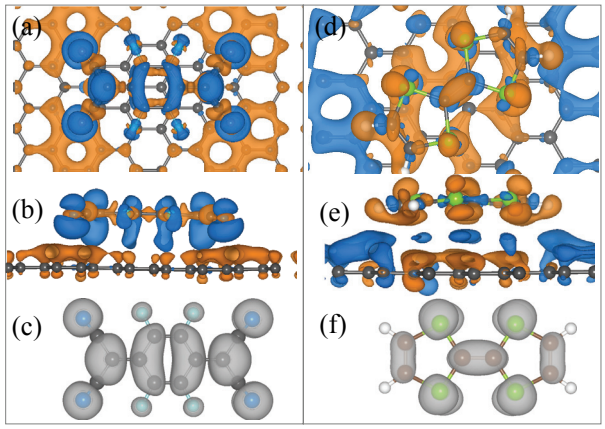
Because of their importance in the emerging field of molecular electronics the electronic properties of graphene have been the focus of intensive research, leading to the concept of a single-electron transistor and a proof of principle of field regulation by external charges. Due to the intrinsic gapless properties, the modification of the electronic structures of graphene gave rise to tremendous theoretical and experimental investigations. However these investigations mainly focus on only one type of controlling the type of the charge carrier of graphene. Here we will show that the type of charge carrier of graphene can be controlled to be *n*-type or *p*-type either by donor or acceptor molecules. Furthermore, the electronic structure of graphene can be tuned by the charge transfer complex consisting of donor and acceptor molecules. Our results suggest that, by tuning the composition of charge transfer complex, the electronic structure of graphene can be engineered.

2. Model setup



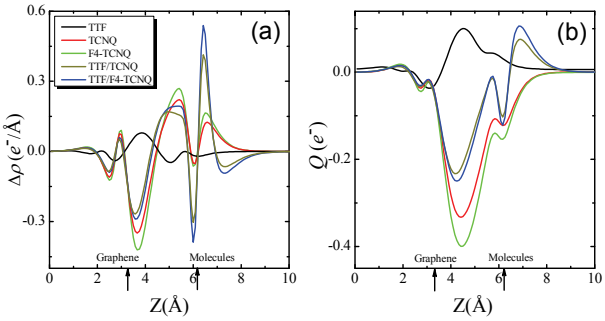
(a) Most stable adsorption structures of single molecule on graphene. (b) The proposed model of charge transfer complex on graphene consisting of TCNQ and TTF. Red dashed line stands for the supercell

3. The differential charge density of molecules on graphene



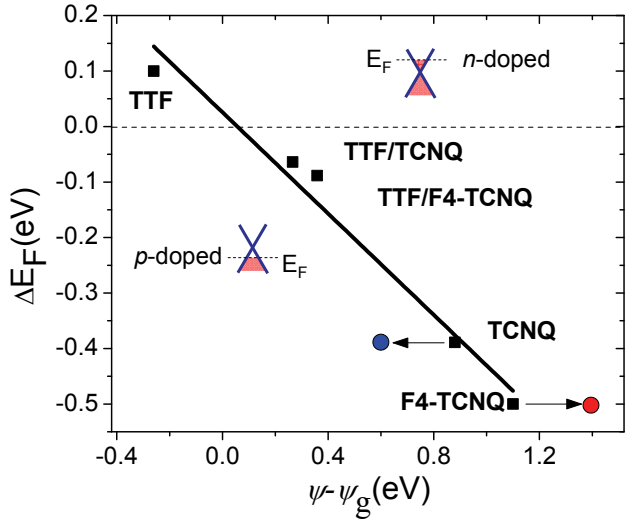
The top view (a,d) and side view (b,e) of the charge rearrangement of F4-TCNQ (left) and TTF (right) on graphene. (c) and (f) show the constant charge density isosurface of F4-TCNQ and TTF adsorbed on graphene around the Fermi level respectively. Blue (orange) colour denotes charge accumulation (depletion) regions.

4. Planar averaged charge density difference



(a) Planar averaged electron density change $\Delta\rho$ induced by single molecule and CTC adsorption on graphene. (b) The integrated charge transfer along the z direction. The average positions of the graphene substrate and molecule plane are indicated by two arrows

5. Potential step and the Fermi level shift



The linear dependence of the relative position of Dirac point and the Fermi level as a function of the potential step.

6. Conclusion

In summary, first-principles calculations and photoemission spectroscopy experiments were carried out to investigate the interfacial electronic structure between graphene and organic molecules. Donor (acceptor) molecules give (attract) electrons to make *n*- (*p*-) type graphene. Theoretical results by DFT calculations are consistent with experimental data. It is found surprisingly that the adsorption of single molecule and formation of charge transfer complex on graphene can change the type and concentration of the charge carriers of graphene linearly. The charge transfer process dominates the interfacial electronic structure. Our results suggest that by tuning the composition of charge transfer complex, the electronic structure of graphene can be engineered.

7. Reference

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