First Principles Study of Irradiation-Induced Magnetism in Graphite

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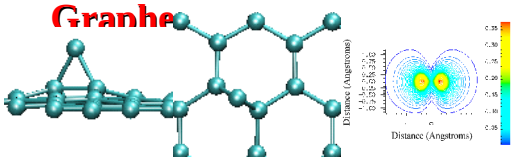
Motivation

• Studying intrinsic magnetic properties of pure carbon systems

• Experimental observation of magnetism in hydrogen and helium irradiated graphite samples by P. Esquinazi *et. al.* In Phys. Rev. Lett. 91, 227201 (2003)

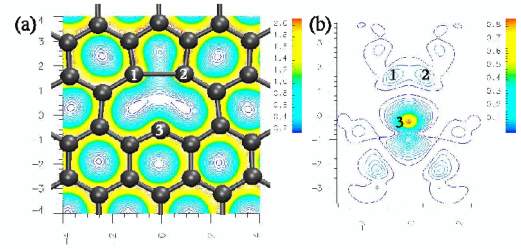
Adatom on Graphe



• Ground state of an adatom in bridge-like structure

•Ground state magnetic with **moment 0.45 μ_B**

Vacancy in Graphene



a) Equilibrium structure of a vacancy in graphene
- weak bond between ions 1 and 2 making the pentagon complete
- A small Jahn-Teller distortion

b) Ion 3 has sp^2- dangling bond contributing the calculated total **magnetic moment of 1.1 μ_B**

Computational Methods

•Density Functional Theory (DFT)
•Basis Set: Plane Waves
•Supercell Approximation: Periodic Boundaries
•Exchange-Correlation Functional: GGA (PW91)
•PAW Potentials
•Kinetic Energy Cut-off: 400 eV
•Computer Program: VASP

Effect of Helium on Vacancy in Graphene

• Equilibrium position of helium 3.06 Å above the graphene slab with a vacancy

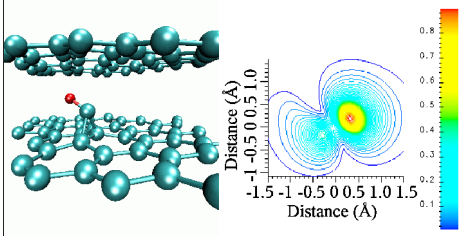
• Adsorption energy: 16 meV

•No effect on observed magnetism; vacancies and adatoms the main contributors

• Possible reason for small magnetic signal observed: recombination with adatoms (interstitals)

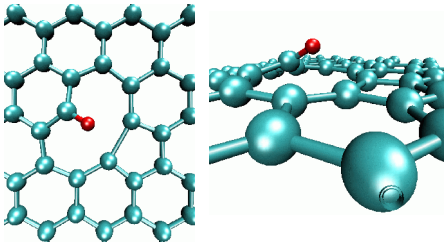
Effect of Hydrogen on Defects in Graphene

• Adatom with a hydrogen magnetic due to change in hybridization; **magnetic moment 1.2 μ_B**



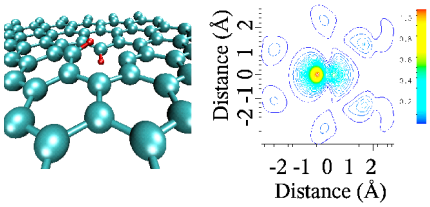
Hydrogen's ground state on graphene with a vacancy

• Hydrogen 1.2 Å above the sheet



• ground state has **not magnetic moment**

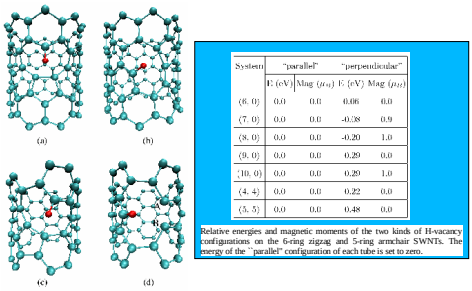
The related references to this work:
• Migration of adatom on graphene:
Phys. Rev. Lett. 91, 017202 (2003)
• Migration of adatom on nanotubes:
Phys. Rev. B 69, 073402 (2004)
• Magnetic Properties of adatom on nanotubes:
Phys. Rev. B 69, 155422 (2004)
• Magnetic properties of a vacancy in graphene and in nanotubes:
New J. Phys. 6, 68
• Irradiation induced magnetism in graphite:
Phys. Rev. Lett. 93, 187202 (2004)



• Vacancy surrounded by two hydrogens magnetic: **Magnetic moment 0.9 μ_B** arises from the dangling bond of the ion neighbour to the vacancy site

• Hydrogen also effective in preventing the recombination of adatoms and vacancies

• In nanotubes, the result depends on the chirality of a tube and the configuration of the defect site



System	"parallel"		"perpendicular"	
	E (eV)	Mag (μ_B)	E (eV)	Mag (μ_B)
(6, 0)	0.0	0.0	0.06	0.0
(7, 0)	0.0	0.0	-0.38	0.9
(8, 0)	0.0	0.0	-0.20	1.0
(9, 0)	0.0	0.0	0.29	0.0
(10, 0)	0.0	0.0	0.29	1.0
(4, 4)	0.0	0.0	0.22	0.0
(5, 5)	0.0	0.0	0.48	0.0

Relative energies and magnetic moments of the two kinds of H-vacancy configurations on the 6-ring zigzag and 5-ring armchair SWNTs. The energy of the "parallel" configuration of each tube is set to zero.

• Magnetic moment does not necessarily mean ferromagnetism
  • Method unable to provide information of coupling
  • H provides long-range perturbations of electronic system (see Phys. Rev. Lett. 84, 4910–4913 (2000))

Summary

• Mobility of adatoms creates difficulties in measuring the magnetic properties of pure Frenkel pairs at room temperature

• Presence of hydrogen slows defects' motion down; recombination more difficult