ELECTRONIC PROPERTIES OF NITROGEN DOPED GRAPHENE NANOWIGGLES.

Anderson Soares da Costa Azevêdo (Mestrando da Pós Graduação em Ciência e Engenharia de Materiais - UFPI);
Eduardo Costa Girão (Departamento de Física – UFPI);
anderson4r2@gmail.com.

Introduction

Early researchers demonstrated considerably interest in investigating nanoscale carbon materials. This is due to their remarkable properties and promising potential applications in novel nanoelectronic devices (Son et al., 2006). These materials include: fullerenes, carbon nanotubes and graphene nanoflakes. In this work, we investigate the electronic properties and emergence of magnetic states in periodic nitrogen-doped graphene structures of complex geometry inspired into nanostructures fabricated and reported in the literature (Cai et al., 2010).

Methods

The electronic structure calculations were performed within the framework of density functional theory (DFT) as implemented in the SIESTA code (Soler et al., 2002), which solves Kohn-Sham equations self-consistently. We used a double ξ basis plus a polarization function to expand the electronic wave functions and the generalized gradient approximation (GGA) to account for exchange and correlation effects. A 400 Ry cutoff was used for the grid integration when representing the charge density. All systems were relaxed until Hellman-Feynman forces and stresses reached values no larger than 0.01 eV/Ang and 0.1 GPa, respectively.

Studied structures

The basic building block for the systems studied in this work are graphene nanowiggles characterized by a periodic repetition of hexagonal graphene nanoflakes junctions. We consider the pristine structure formed only by a periodic junction of a hexagonal graphene nanoflake with chemical formula $C_{42}H_{18}$. It is the one depicted in Fig. 1a. which we labelled as Flake-0. Such a flake can be viewed as a central hexagonal ring surrounded by other six similar rings, resulting in a system with an armchair edge geometry. We consider a set of nitrogen-doped versions of this flake, where substitutional doping is set at different sites of the molecule. We consider six possibilities, named as Flake-i, with $i = 1,2,\ldots,6$. These are illustrated in Figs. 1b-g, where we represented carbon, hydrogen and nitrogen atoms as black, blue and green spheres, respectively. In addition, we consider variations of the Flake-i ($i = 2,4,5,6$) systems for which we remove the hydrogens from nitrogen edge atoms. These systems, named as Flake-i', are represented in Figs. 1h-k. Figure 2 shows the periodic cells of the pristine structure, W00 (composed of two Flake-0), and four configurations for combinations of Flakes-1. We adopt the notation parallel(P) and oblique(O) to identify a set of wiggles formed by a periodic junction of Flakes-1 (Fig. 1b.).

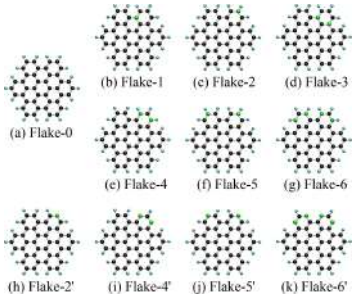

Figure 1: Graphene nanoflakes structures.

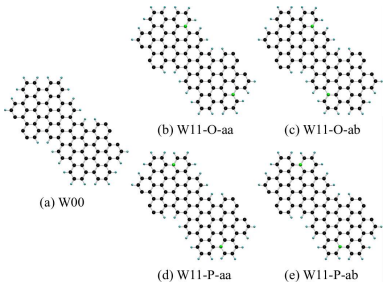

Figure 2: Graphene nanowiggles structures.

Results and discussion

This section is dedicated for the description of electronic properties of the investigated structures. Table 1 shows the HOMO-LUMO Gap and Total energy calculated for: a non-polarized (NP) and ferromagnetic (FM) states of the eleven Flakes configurations.

Table 1: Comparative table for Flakes properties.

STRUCTURE	TOTAL ENERGY $[eV]$	HOMO-LUMO GAP $[eV]$
FLAKE 0 NP	-6787.3085	2.4570
FLAKE 1 NP	-6901.5867	0.0000
FLAKE 1 FM	-6901.7050	0.6159
FLAKE 2 NP	-6901.6651	0.0000
FLAKE 2 FM	-6901.7660	0.3503
FLAKE 3 NP	-7015.8102	0.0733
FLAKE 3 FM	-7015.9876	0.7704
FLAKE 4 NP	-7016.8465	0.2169
FLAKE 5 NP	-7016.2960	0.2523
FLAKE 6 NP	-7246.5282	0.3409
FLAKE 2' NP	-6886.2864	2.4297
FLAKE 4' NP	-6985.8192	2.2785
FLAKE 5' NP	-6985.2408	2.4305
FLAKE 6' NP	-7183.7923	2.1776

Table 2 shows the total energy and magnetic moment calculated for: a non-polarized (NP), ferromagnetic (FM) and antiferromagnetic (AFM) states of the eleven Wiggles simulations.

Table 2: Comparative table for Wiggles properties.

STRUCTURE	TOTAL ENERGY $[eV]$	TOTAL MAGNETIC MOMENT $[\mu B]$
W00 NP	13385.8294	-
W11O-aa NP	13614.6744	-
W11O-aa AFM	13614.7282	-0.000001
W11O-ab NP	13614.6909	-
W11O-ab AFM	13614.7166	-0.000000
W11P-aa NP	13614.6119	-
W11P-aa FM	13614.7336	1.976741
W11P-aa AFM	13614.7438	-0.000049
W11P-ab NP	13614.6445	-
W11P-ab FM	13614.7221	1.942044
W11P-ab AFM	13614.7543	-0.000034

Figures 3 - 6 presents the electronic bands and spin density of non-polarized and polarized configurations for different sets of oblique and parallel Wiggles composed of Flakes 1. We observed only the emergence of an antiferromagnetic for oblique configurations, while for parallels sets both states (FM and AFM) were obtained.

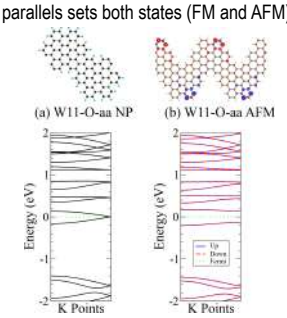

Figure 3: Spin density and energy bands of NP and AFM W11O-aa.

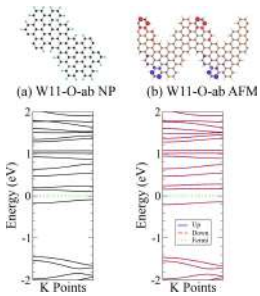

Figure 4: Spin density and energy bands of NP and AFM W11O-ab.

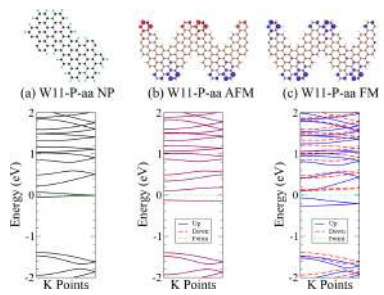

Figure 5: Spin density and energy bands of NP, FM and AFM W11P-aa.

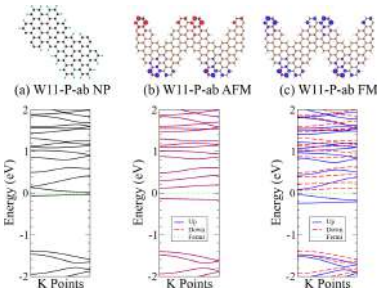

Figure 6: Spin density and energy bands of NP, FM and AFM W11P-ab.

Conclusion

We have investigated the electronic properties and emergence of magnetic states of eleven graphene nanoflakes. We observed that the ferromagnetic state emergence is related with the presence of levels close to Fermi energy in non-polarized structure or presence of an odd number of electrons, due to the nitrogen dopant position. Finally, we observed the emergence of six different magnetic states for a periodic structure composed only of Flakes 1 accordingly to the position of Nitrogen dopant atom.

Acknowledgement

This work was supported by PPGCM of Universidade Federal do Piauí - UFPI. We acknowledge the financial support from Brazilian agency CAPES and FAPEPI.

References

CAI, Jinming et al. Atomically precise bottom-up fabrication of graphene nanoribbons. **Nature**, v. 466, p. 470 – 473, 2010.
SON, Young-Woo et al. Energy gaps in graphene nanoribbons. **Physical Review Letters**, v. 97, n. 216803, p. 1-4, 2006.
SOLER, Jose et al. The Siesta method for ab initio order-n materials simulation. **Journal of Physics: Condensed Matter**, v. 14, n. 3, p. 2745-2779, 2002.