Journal of Magnetism and Magnetic Materials ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Current Perspectives

Tight-binding model study of substrate induced pseudo-spin polarization and magnetism in mono-layer graphene

Sivabrata Sahu [a], G.C. Rout [b,*]

[a] School of Applied Sciences (Physics), KIIT University, Bhubaneswar 751024, Odisha, India
[b] Condensed Matter Physics Group, Physics Enclave, Plot No. – 664/4825, Lane-4A, Shree Vihar, Chandrasekharpur, Po-Patia, Bhubaneswar 751031, Odisha, India

ARTICLE INFO

Article history:
Received 19 April 2015
Received in revised form
25 December 2015
Accepted 20 January 2016

Keywords:
Graphene
Coulomb interaction
Pseudo-spin polarization
FM Magnetization
AFM Magnetization

ABSTRACT

We present here a tight-binding model study of generation of magnetism and pseudo-spin polarization in monolayer graphene arising due to substrate, impurity and Coulomb correlation effects. The model Hamiltonian contains the first-, second- and third-nearest-neighbor hopping integrals for π electrons of graphene besides substrate induced gap, impurity interactions and Coulomb correlation of electrons. The Hubbard type Coulomb interactions present in both the sub-lattices A and B are treated within the mean-field approximation. The electronic Green's functions are calculated by using Zubarev's technique and hence the electron occupancies of both sub-lattices are calculated for up and down spins separately. These four temperature dependent occupancies are calculated numerically and self-consistently. Then we have calculated the temperature dependent pseudo-spin polarization, ferromagnetic and anti-ferromagnetic magnetizations. We observe that there exists pseudo-spin polarization for lower Coulomb energy, $u < 2.2t_1$ and pseudo-spin polarization is enhanced with substrate induced gap and impurity effect. For larger Coulomb energy $u > 2.5t_1$, there exists pseudo-spin polarization (p); while ferromagnetic (m) and antiferromagnetic (p_m) magnetizations exhibit oscillatory behavior. With increase of the substrate induced gap, the ferromagnetic and antiferromagnetic transition temperatures are enhanced with increase of the substrate induced gap; while polarization (p) is enhanced in magnitude only.

1. Introduction

Ferromagnetic instability due to exchange interaction in the three dimensional (3D) electron gas has been studied in great detail [1,2]. Similar studies have suggested the existence of ferromagnetic phase in the diluted two-dimensional (2D) electron gas with transition from paramagnetic to ferromagnetic phase [3]. There has been strong experimental indications on the existence of ferromagnetism in highly disordered graphites [4,5]. Though a number of different mechanisms have been proposed [6,7], the origin of the magnetic phase is still unclear. The recent experiments in true 2D-graphene systems [8,9] show that electron-electron interaction and disorder effect have to be taken in order effect to obtain a fully consistent picture of magnetism in graphene [10].

The electronic structure of perfect graphene plane exhibits the linear relationship between momentum and electronic energy. This singular dispersion relation in graphene is a direct consequence of its honey-comb lattice structure. The effective mass of electron in graphene is zero at the Dirac point similar to the universal electrodynamics. In perfect graphene sheet, the chemical potential crosses the Dirac point and electronic density of states vanishes at the Fermi-energy. The vanishing of the effective mass or density of states has profound consequences. The Coulomb interaction remains unscreened and gives rise to an inverse quasi-particle life time that increases linearly with energy or temperature.

It is well known that direct exchange interaction between electrons can lead to a ferromagnetic instability in dilute electron gas [1,11]. Similar exchange instability of electron gas can be generalized for pure and doped 2D-graphene. It has been reported by Peres et al. [12] that the extended defects such as dislocation, disinclination edges and micro cracks can leads to self-doping where charge is transformed to/from defect to graphene. The extended defects can also lead to self-doping and hence localized disorders such as vacancies and adatoms are introduced. Thus Peres et al. [12] have considered the influence of disorder by introducing Coulomb interaction for the generation of ferromagnetism. The possibilities of other instabilities in a graphene plane reflecting the Coulomb interaction have been studied in the

* Corresponding author.
E-mail addresses: siva1987@iopb.res.in (S. Sahu), gcr@iopb.res.in (G.C. Rout).

http://dx.doi.org/10.1016/j.jmmm.2016.01.062
0304-8853/

literature [13,14]. In spite of this, the study of instability due to Coulomb interaction in graphene is extensive. It is worth mentioning that the simple analysis of Stoner criterion for ferromagnetism fails in graphene, as the density of states of un-doped graphene vanishes at the Fermi level.

The Coulomb interaction between massless fermions in pristine graphene remains long ranged and unscreened. It is not clear whether this would lead to strongly correlated insulating phase or weakly correlated semi-conducting phase of graphene [15,16]. The local Coulomb interaction is important for the theory to understand the defect induced magnetism [17] and Mott insulator-metal transition in graphene on the surface of Si:X (X=Si, C, Sn, Pb) [18]. The two dimensional graphene [19,20] and polymers [21,22] display strong local as well as non-local Coulomb interactions. The bare on-site Coulomb interaction in benzene was estimated long back to be ~16.93 eV [17]. The weak coupling perturbation theory gives an effective repulsive on-site Coulomb interaction energy of ~10 eV to explain the optical spectroscopy experiments of poly -acetylene [23,24]. Wehling et al. [19] have estimated that the on-site Coulomb interaction is $U \sim 3.3t_1$ and the nearest-neighbor Coulomb interaction is $U \sim 2t_1$ in graphene, where $t_1 = -2.8$ eV is the nearest-neighbor hopping integral. The mean-field Hubbard model calculation for bipartite graphene lattice yields antiferromagnetic ground state for $U_{AFM} \geq 2.2t_1$ [25,26], while the quantum Monte-Carlo and finite size scaling for the Hubbard model give the evidence of a zero temperature transition between non-magnetic semimetal and antiferromagnetic insulator for $U_{AFM} \geq 4.5t_1$ [25–27].

The local magnetic moments can be introduced to graphene in a varieties of forms i.e along the edges of the nano-ribbons [28], around vacancies [29] and adatoms [30]. However, long range ferro-magnetic order in graphene does not occur without exchange coupling between the local moments. By coupling the graphene to an atomically flat Yttrium iron garnet (YIG) [31], ferromagnetic insulator film, ferromagnetism in graphene can be introduced without sacrificing its excellent transport properties. The hybridization between the π orbitals in graphene and the near-by spin polarized d-orbitals in magnetic insulator gives rise to the exchange interaction required for long range ferromagnetic ordering.

It is known that point defects induce localized magnetic moments in graphene [32–34]. Lieb [35] has predicted the presence of bi-bipartite lattice in graphene in case of half filling and repulsive local electron-electron repulsive interaction'U'. The ground state is characterized by the total spin $2S = |N_A - N_B|$, where N_A and N_B are the number of sites in sub-lattices A and B respectively. Point defects like adsorbed atoms or single vacancies unbalance the number of atoms in both the sub-lattices giving rise to localized magnetic moments in graphene. The peculiar characteristics of different magnetic states in graphene have been studied and characterized by model Hamiltonians [36–39]. Many first principle calculations have been performed for hydrogen vacancies by Yazyev [34,40] and others [33,41] and fluorine by Sofo et al. [42]. The recent experiments on fluorinated graphene and irradiated graphene indicate a magnetic behavior competitive with localized spin one-half defects [43–46].

The electron–electron interaction in graphene can lead to other instabilities at low temperatures like ferromagnetic phase [47]. A local on-site repulsive interaction can lead to an anti-ferromagnetic phase, when its value exceeds a critical threshold [25,48]. In the following, we will concentrate on the on-site Coulomb interaction in low and relatively large limits in order to study the pseudo-spin polarization, ferromagnetism (FM) and antiferromagnetism (AFM) in doped graphene on substrate. Earlier we have reported the study of band gap opening in graphene by single impurity taking up to third nearest hoppings in a tight-binding model in absence of Coulomb interaction [49]. We find that Coulomb interaction (U) in bipartite lattice enhances pseudo-spin effect leading to band gap opening [50]. Now we investigate here the charge and spin polarizations leading to oscillatory magnetic phases due to Coulomb interaction in doped graphene on substrate. We present the tight-binding Hamiltonian in Section 2. We calculate the electron Green's functions by Zubarev technique [51] and calculate the electron occupancies of both sub-lattices for two spin orientations in Section 3. The temperature dependent electron occupancies are solved numerically and self-consistently. We present the results and discussion in Section 4 and conclusion in Section 5.

2. Theoretical model

Based upon our earlier model [49,50], the tight-binding model Hamiltonian including π electron hoppings up to third-nearest-neighbor hoppings with hopping integrals t_1, t_2 and t_3 respectively is written as

$$H_0 = \sum_{i,\sigma} \epsilon_a a^{\dagger}_{i,\sigma} a_{i,\sigma} + \sum_{J,\sigma} \epsilon_b b^{\dagger}_{j,\sigma} b_{j,\sigma} - t_1 \sum_{\langle i,j\rangle,\sigma} \left(a^{\dagger}_{i,\sigma} b_{j,\sigma} + b^{\dagger}_{j,\sigma} a_{i,\sigma} \right)$$
$$- t_2 \sum_{\langle\langle i,j\rangle\rangle,\sigma} \left(a^{\dagger}_{i,\sigma} a_{j,\sigma} + b^{\dagger}_{j,\sigma} b_{i,\sigma} + H.C \right)$$
$$- t_3 \sum_{\langle\langle\langle i,j\rangle\rangle\rangle,\sigma} \left(a^{\dagger}_{i,\sigma} b_{j,\sigma} + b^{\dagger}_{j,\sigma} a_{i,\sigma} \right) \quad (1)$$

where $a^{\dagger}_{i,\sigma}$ ($a_{i,\sigma}$) and $b^{\dagger}_{i,\sigma}$ ($b_{i,\sigma}$) create (annihilate) respectively an electron with spin σ on site $\vec{R_i}$ on sub-lattices A and B. The Fourier transformed band dispersions for first-, second- and third-nearest-neighbor hoppings of π electrons are found to be $\epsilon_{1k} = -t_1\gamma_1(k)$, $\epsilon_{2k} = -t_2\gamma_2(k)$ and $\epsilon_{3k} = -t_3\gamma_3(k)$, where $\gamma_1(k)$ for the nearest-neighbor hopping is

$$\gamma_1(k) = e^{ik_x a_0} + 2e^{-i(1/2)k_x a_0}.\cos\frac{\sqrt{3}}{2}k_y a_0 \quad (2)$$

where a_0 is the lattice constant and $\vec{k}(k_x, k_y)$ is the electron momentum. The other dispersions are $\gamma_2(k) = \sum_{\delta_2} e^{i\vec{k}.\vec{\delta_2}}$ and $\gamma_3(k) = \sum_{\delta_3} e^{i\vec{k}.\vec{\delta_3}}$, where $\vec{\delta_2}$ and $\vec{\delta_3}$ are corresponding lattice vectors. Gharekhanl et al. [52] have reported the electronic band structure of patterned graphene taking tight-binding approximation for the π electrons including upto five nearest-neighbor hopping integrals and found opening of gap near Dirac point in graphene by this symmetry breaking. The localized defects such as vacancies and impurities are included in the tight-binding description of the model by adding a local energy term [53]

$$H_{imp} = \sum_{i,\sigma} V_i \left(a^{\dagger}_{i,\sigma} a_{i,\sigma} + b^{\dagger}_{i+\delta,\sigma} b_{i+\delta,\sigma} \right) \quad (3)$$

where V_i is the random potential at sites $\vec{R_i}$. In momentum space the Hamiltonian appears as

$$H_{imp} = \sum_{k,\sigma} V \left(x_a a^{\dagger}_{k,\sigma} a_{k,\sigma} + x_b b^{\dagger}_{k,\sigma} b_{k,\sigma} \right) \quad (4)$$

where V is the interaction potential at impurity sites with impurity concentrations x_a and x_b at sites A and B respectively.

On growing epitaxial graphene on silicon carbide substrates by annealing 4H– and CH–SiC surfaces [54,55], magnetism is

Please cite this article as: S. Sahu, G.C. Rout, Journal of Magnetism and Magnetic Materials (2016), http://dx.doi.org/10.1016/j.jmmm.2016.01.062