Orissa Journal of Physics
ISSN 0974-8202

Vol. 23, No.2
August 2016
pp. 147-152

Theoretical Study of Impurity Effect on Anti-Ferromagnetic (AFM) order in Graphene-on-Substrate

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Received: 2.6.2016 ; *Revised : 3.7.2016* ; *Accepted : 1.8.2016*

Abstract. We report here a tight binding study of the effect of impurity concentration on anti-ferromagnetic (AFM) order in graphene-on-substrate. The onsite Coulomb interactions at two sub-lattices of graphene are treated within mean-field approximation and the electron occupancies at two sub-lattices include spin moments in opposite directions giving rise to antiferromagnetism in graphene. The sub-lattice magnetizations are calculated by Green's function method and are solved self-consistently for different impurity concentrations.

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Keywords. Graphene , anti-ferromagnetism , impurity concentration.
PACS No- 71.10.Fd, 75.10.Lp, 75.30.Ds

1. Introduction

The magnetic ordering in graphene has potential applications in spintronic applications [1]. It is found that the spins of same sib-lattices are ordered ferromagnetically and spins of different sub-lattices are ordered anti-ferromagnetically. Magnetic ordering of graphene in general is governed by Lieb's theorem [2]. It states that the ground state spin of a bipartite lattice with repulsive electron-electron interaction as described Hubbard model equals half of the difference of electron occupancies. The substrate induced gap leads to difference in spins at different sub-lattices. A recent tight-binding calculation predicts the occurrence of ferromagnetism (FM) and anti-ferromagnetism (AFM) in the graphene [3]. The repulsive electron-electron interaction in the bipartite

lattice of graphene will produce imbalances in sub-lattice atoms leading to spin polarization of the ground state of the system . In theory, magnetic ordering has been demonstrated in nano ribbons and vacancies in bulk graphene [4]. Monte-Carlo calculations of Sorella et al [5] have shown that the Hubbard model in half filled honey-comb lattice would exhibit a Mott-Hubbard transition at a finite U.

2. Formalism and calculations Greens Function

In order to investigate the AFM order present in the monolayer graphene, The tight- binding model of Hamiltonian is proposed as

$$H_{TB} = H_0 + H_1 + H_2 + H_3 + H_s + H_I + H_U \quad (1)$$

$$H_0 = \sum_{i,\sigma} \epsilon_a \; a^{\dagger}_{i,\sigma} a_{i,\sigma} \; + \sum_{j,\sigma} \epsilon_b \; b^{\dagger}_{j,\sigma} b_{j,\sigma} \quad (2)$$

$$H_1 = -t_1 \sum_{<i,j,\sigma>} (\gamma_1(k) \; a^{\dagger}_{i,\sigma} b_{j,\sigma} + \gamma_1^{*}(k) \; b^{\dagger}_{j,\sigma} \; a_{i,\sigma}) \quad (3)$$

$$H_2 = -t_2 \sum_{<<i,j,\sigma>>} \gamma_2(k) \left[a^{\dagger}_{i,\sigma} \, a_{j,\sigma} + \; b^{\dagger}_{j,\sigma} \, b_{i,\sigma} \right] \quad (4)$$

$$H_3 = -t_3 \sum_{<<<i,j,\sigma>>>} (\gamma_3(k) \; a^{\dagger}_{i,\sigma} b_{j,\sigma} + \; \gamma_3^{*}(k) \; b^{\dagger}_{j,\sigma} \, a_{i,\sigma} \;) \quad (5)$$

$$H_s = \Delta \sum_{i,j,\sigma} (a^{\dagger}_{i,\sigma} a_{i,\sigma} - \; b^{\dagger}_{j,\sigma} \, b_{j,\sigma}) \quad (6)$$

$$H_I = V_0 \sum_{i,j,\sigma} (x_a \, a^{\dagger}_{i,\sigma} a_{i,\sigma} + x_b \, b^{\dagger}_{j,\sigma} \, b_{j,\sigma}) \quad (7)$$

where $a^{\dagger}_{k,\sigma}(a_{k,\sigma})$ and $b^{\dagger}_{k,\sigma}(b_{k,\sigma})$ are the creation (annihilation) operators with spins $(\sigma = \uparrow, \downarrow)$ on sub-lattices A and B respectively. H_0 represents the Hamiltonian for onsite energies with ϵ_a and ϵ_b as the site energies at A and B sub-lattices. Since graphene is placed on substrate, the Hamiltonian for the substrate is given by equation (6) and the site energies ϵ_a and ϵ_b at A and B sites are modified by energy, $\epsilon_a + \Delta$ and $\epsilon_b - \Delta$ due to substrate induced gap Δ. H_I represents the impurity interaction in the pristine graphene with impurities x_a and x_b at A and B sites with impurity potential (V_0). Further H_1, H_2 and H_3 represent first, second, third nearest-neighbor electron hopping interactions with energy dispersions $\epsilon_{1k} = -t_1|\gamma_1(k)|$, $\epsilon_{2k} = -t_2|\gamma_2(k)|$ and $\epsilon_{3k} = -t_3|\gamma_3(k)|$, where

t_1, t_2, t_3 represent the respective hopping integrals. The repulsive Coulomb interactions at both the sub-lattices are given by

$$H_U = \frac{U}{2}\sum_{i,\sigma}\left(n^a_{i,\sigma}n^a_{i,-\sigma} + n^b_{i,\sigma}\, n^b_{i,-\sigma}\right) \tag{8}$$

here $n^{\beta}_{i,\sigma}$ represents electron occupation number at two sub-lattices i.e. $\beta = A, B$ with repulsive Coulomb energy U.

3. Calculation of Green's functions and AFM magnetization

The Hamiltonian H_U is considered within Hartree-Fock mean-field approximation where the average electron occupancies at A and B sub-lattices are written as $< n^a_{i,\sigma} >= \left(\frac{n}{2} + \frac{\sigma ma}{2}\right)$ and $\left(< n^b_{i,\sigma} >= \frac{n}{2} - \frac{\sigma mb}{2}\right)$ indicating that A and B site magnetic moments are oppositely oriented for anti-ferromagnetism and n represents the total electron occupancy. The total Hamiltonian H_{TB} in equation (1) is solved by Zubarev' s Green's function technique [6]. The A site and B site electron occupancies are calculated from the correlation functions derived from the Green's functions. The A site and B-site magnetization are calculated from the given relations. Finally m_a and m_b are computed self- consistently.

$m_a = \sum_k[< a^{\dagger}_{k,\uparrow}a_{k,\uparrow} > - < a^{\dagger}_{k,\downarrow}a_{k,\downarrow} >], m_b = \sum_k[< b^{\dagger}_{k,\uparrow}b_{k,\uparrow} > - < b^{\dagger}_{k,\downarrow}b_{k,\downarrow} >]$

4. Results and Discussion

All the physical parameters are scaled by first-nearest-neighbor hopping integral ($t_1 = 2.78$ eV). The temperature appears as $t = \frac{k_B T}{t_1}$ in unit less form . The scaled quantities are $\tilde{t}_1 = -1$, $\tilde{t}_2 = \frac{t_2}{t_1}$, $\tilde{t}_3 = \frac{t_3}{t_1}$, attractive impurity energy $v = \frac{V_0}{t_1}$, repulsive Coulomb energy $u = \frac{U}{t_1}$, substrate induced gap $d_1 = \frac{\Delta}{t_1}$, energy at A-site $ea = \frac{\varepsilon_a}{t_1}$, energy at B-site $eb = \frac{\varepsilon_b}{t_1}$ and chemical potential $um = \frac{\mu}{t_1}$. The temperature dependent magnetizations(m_a, m_b) are shown in figures 1-2 for different concentrations (x_a and x_b) to A and B sub-lattices.

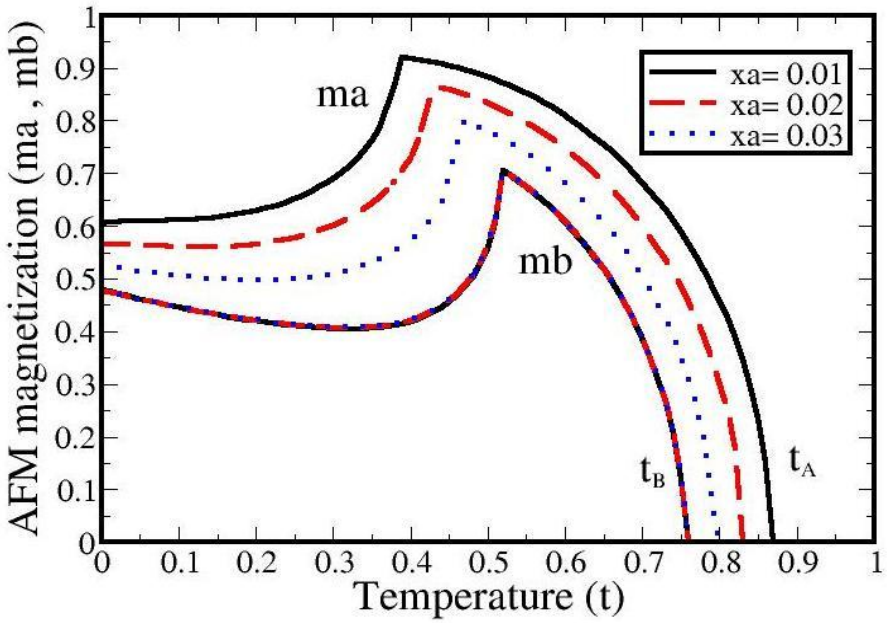

Fig. 1. The plot of magnetizations (m_a, m_b) vs. temperature (t) for fixed Coulomb potential u = 3.3, impurity potentials $v = -4.5$ and $x_a = 0.01, 0.02, 0.03$ and $x_b = 0$ at substrate induced gap $d_1 = 0.090$ and band filling n = 0.75.

Figure.1 shows the effect of impurities ($x_a = 0.01, 0.02, 0.03$) at A-site on magnetization m_a. For given impurity concentration, the magnetization is suppressed at low temperature and they exhibit mean-field behavior at higher temperature. A-site magnetization (m_a) becomes higher than that of B-site magnetization (m_b) with corresponding Neel temperature $t_A = 0.86$ and $t_B = 0.76$. With increase of A-site impurity concentration, the A-site magnetization is suppressed considerably throughout the temperature range. However A-site impurity has no effect on B-site magnetization. It is to note further that sub-lattice magnetizations(m_a, m_b) of graphene-on-substrate are suppressed as at lower temperatures due to the presence of impurities.

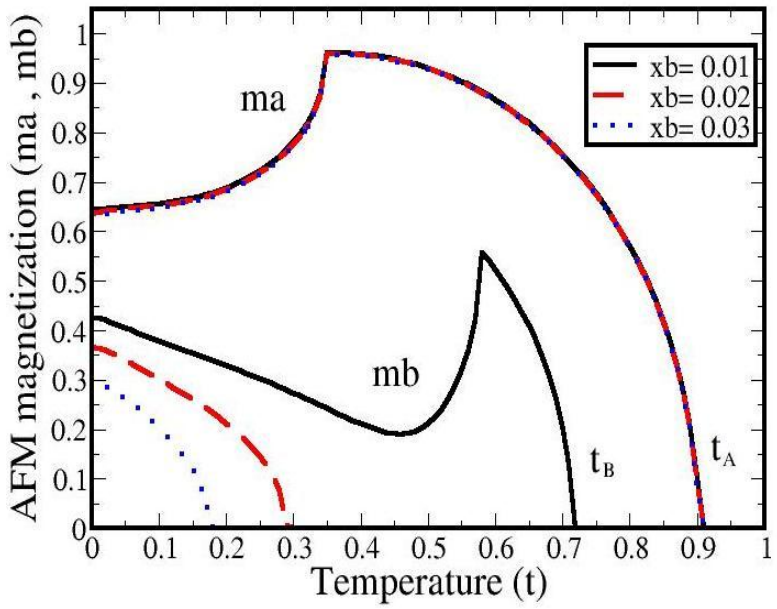

Fig. 2. The plot of magnetizations (m_a, m_b) vs. temperature (t) for impurity potentials $v = -4.5$, $x_a = 0$ and $x_b = 0.01, 0.02, 0.03$ for same parameters given in figure 1.

Figure. 2 shows that the effect of impurities $x_b = 0.01, 0.02, 0.03$ at B sub-lattice on magnetization(m_b). With increase of impurity, the B-site magnetization(m_b) is suppressed appreciably with large suppression of Neel temperature and the magnetization (m_b) vanishes for the impurity $x_b = 0.05$. However the A-site magnetization (m_a) remains unaffected by B-site impurities. On the other hand, the graphene-on-substrate exhibits for the magnetization for impurity $x_b = 0.05$ only due to the magnetization of A-site atoms.

5. Conclusions

We have reported here the impurity effect on AFM sub-lattice magnetizations below the anti-ferromagnetic ordering temperature (Neel temperature). It is observed that A-site magnetization is suppressed by A-site impurity concentration, while B-site magnetization remains unaffected. Similar results are obtained for B-site impurities. In present case, we have considered nitrogen impurity at carbon sites with attractive impurity potential $\boldsymbol{V = -4.5} *$

$\mathbf{t_1 = -12.51eV}$ for fixed band filling $\mathbf{n = 0.75}$ and substrate induced gap $\mathbf{d_1 = 0.090}$. Recently, we have reported effect of band filling effect on antiferromagnetically ordered magnetization in graphene [7].

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