The Role of Electron-electron Interaction on Anti-Ferromagnetism in Graphene-on-Substrate: A Tight Binding Approach

H S GOUDA[1], S SAHU[1] and G C ROUT[2]

[1]Nano sensor Lab, School of Applied Sciences, KIIT University, Odisha, India

[2]Condensed Matter Physics Group ,Physics Enclave, Plot No.- 664/4825, Lane -4A,Shree Vihar, C. S. Pur, PO- Patia, Bhubaneswar- 751031, Odisha, India.
Email : gcr@iopb.res.in

Abstract. We report here a tight-binding theoretical study of anti-ferromagnetism in graphene taking electron hopping up to third nearest neighbors. The graphene-on-substrate introduces in equivalence in the two sub-lattices of the honey-comb unit cell. The Hubbard type of repulsive Coulomb interaction within mean-field approximation introduces anti-ferromagnetic(AFM) order in both the lattices. The AFM sub-lattice magnetizations are calculated using Zubarev's Green's function technique and computed self-consistently for different ranges of electron-electron interaction.

Keywords. Graphene , anti-ferromagnetism , Coulomb Potential , substrate effect.

1. Introduction

Graphene is a novel material that was successfully fabricated as monolayer [1]. Recent progress in graphene research has shown different remarkable properties i.e. high strength and high crystalline quality, high electron mobility, lack of band gap, thermal conductivity, electrical conductivity and optical transparency etc. The above mentioned properties are very interesting for various applications of graphene such as piezoelectric devices, super capacitors, photo detectors, nano-electronics and field effect transistors (FET) [2-4]. Theoretically, ab-initio calculations along with density functional theory (DFT) are widely used to study the electronic band structure and magnetic properties of graphene such as anti-ferromagnetism, ferromagnetism, diamagnetism and paramagnetism in

graphene [5]. There are also different theoretical models used to study the magnetism in nano-graphene by using Monte Carlo simulations [6]. Gouda et.al. have reported the tight binding model calculation of the substrate effect on the AFM sub-lattice magnetization [7]. In the present communication, we report the interplay between temperature and Coulomb interaction; and substrate induced gap and Coulomb interaction.

2. Formalism

The tight-binding model Hamiltonian for anti-ferromagnetic order is

$$H_0 = \sum_{k,\sigma} (\varepsilon_{a,k} a_{k,\sigma}^\dagger a_{k,\sigma} + \varepsilon_{b,k} b_{k,\sigma}^\dagger b_{k,\sigma}) + \sum_{k,\sigma} (\varepsilon_{13k} a_{k,\sigma}^\dagger b_{k,\sigma} + \varepsilon_{13k}^* b_{k,\sigma}^\dagger a_{k,\sigma}) \quad (1)$$

where $\varepsilon_{a,k} = \varepsilon_a - \mu + x_a V + \Delta + \varepsilon_{2k}$ and $\varepsilon_{b,k} = \varepsilon_b - \mu + x_b V - \Delta + \varepsilon_{2k}$ where $a_{k,\sigma}^\dagger(a_{k,\sigma})$ and $b_{k,\sigma}^\dagger(b_{k,\sigma})$ are the creation (annihilation) operators with spins ($\sigma = \uparrow, \downarrow$) on A and B sites respectively. Here ε_a and ε_b are the site energies at A and B sites. The site energies ε_a and ε_b at A and B sites are modified by energy, $\varepsilon_a + \Delta$ and $\varepsilon_b - \Delta$ due to substrate induced gap Δ. Here x_a and x_b are impurity concentrations at sub-lattice sites respectively with corresponding attractive potential V. Further the hopping energy are for first neighbors, $\varepsilon_{1k} = -t_1|\gamma_1(k)|$, second neighbors, $\varepsilon_{2k} = -t_2|\gamma_2(k)|$ and third-nearest-neighbors, $\varepsilon_{3k} = -t_3|\gamma_3(k)|$, where t_1, t_2, t_3 represent the respective hopping integrals. The dispersion $|\gamma_1(k)|$ for electron hoppings between nearest carbon atoms at Γ point is

$$|\gamma_1(k)| = \sqrt{1 + 4\cos^2\left(\frac{k_y a}{2}\right) + 4\cos\left(\frac{\sqrt{3}}{2} k_x a\right) \cos\left(\frac{k_y a}{2}\right)} \quad (2)$$

The repulsive Coulomb interactions at both the sub-lattices are given by

$$H_U = U \sum_i [\; n_{i,\uparrow}^a n_{i,\downarrow}^a + n_{i,\uparrow}^b n_{i,\downarrow}^b \;] \quad (3)$$

where $n_{i,\sigma}^a$ and $n_{i,\sigma}^b$ are the electron occupancy number operators for two sub-lattices. Using Hartree-Fock mean-field approximation, the Coulomb correlations are written as

$$n_{i,\uparrow}^a n_{i,\downarrow}^a \approx\; < n_{i,\uparrow}^a > n_{i,\downarrow}^a + < n_{i,\downarrow}^a > n_{i,\uparrow}^a - < n_{i,\uparrow}^a > < n_{i,\downarrow}^a >$$

$$n_{i,\uparrow}^b n_{i,\downarrow}^b \approx\; < n_{i,\uparrow}^b > n_{i,\downarrow}^b + < n_{i,\downarrow}^b > n_{i,\uparrow}^b - < n_{i,\uparrow}^b > < n_{i,\downarrow}^b >.$$

Since AFM order is assumed to be present in graphene - on- substrate, the magnetic spin orientation at A sub-lattice becomes opposite to that of B site. Here

the electron occupancies at A and B sites are written as $< n_\sigma^a >= \frac{n}{2} + \frac{\sigma m_a}{2}$ and $< n_\sigma^b >= \frac{n}{2} - \frac{\sigma m_b}{2}$. Here n, m_a and m_b are the respective electron occupancy and sub-lattice magnetizations. The m_a and m_b are oppositely directed. The total Hamiltonian becomes $H_T = H_0 + H_U$.

3. Calculation of AFM magnetization

The total Hamiltonian is solved by Zubarev's Green's function method [8]. The electron correlation functions associated with A and B sub-lattices for electron momentum $\vec{k}$ and spin σ are defined as $< n_{k,\sigma}^a >=< a_{k,\sigma}^\dagger a_{k,\sigma} >$ and $< n_{k,\sigma}^b >=< b_{k,\sigma}^\dagger b_{k,\sigma} >$ .They are calculated from the corresponding Green's functions and are written as

$$< n_{k,\sigma}^a > = \frac{1}{(E_{1k\sigma}-E_{2k\sigma})}\left[\left(E_{1k\sigma} - \varepsilon_{b,\sigma}(k)\right)f(\beta E_{1k\sigma}) - \left(E_{2k\sigma} - \varepsilon_{b,\sigma}(k)\right)f(\beta E_{2k\sigma})\right] \qquad (4)$$

$$< n_{k,\sigma}^b > = \frac{1}{(E_{1k\sigma}-E_{2k\sigma})}\left[\left(E_{1k\sigma} - \varepsilon_{a,\sigma}(k)\right)f(\beta E_{1k\sigma}) - \left(E_{2k\sigma} - \varepsilon_{a,\sigma}(k)\right)f(\beta E_{2k\sigma})\right] \qquad (5)$$

where $f(\beta E_{\alpha k\sigma})$ stands for Fermi distribution function with quasiparticle energy $E_{\alpha,k,\sigma}$ with $\alpha = 1,2$. The sub-lattice magnetizations at A and B sites are defined as

$$m_a = \sum_k [< a_{k,\uparrow}^\dagger a_{k,\uparrow} > - < a_{k,\downarrow}^\dagger a_{k,\downarrow} >] \; ; m_b = \sum_k [< b_{k,\uparrow}^\dagger b_{k,\uparrow} > - < b_{k,\downarrow}^\dagger b_{k,\downarrow} >] \qquad (6)$$

Using correlation functions given in equations (4) and (5), the sub-lattice magnetizations i.e. m_a and m_b are calculated from equation (6).

4. Results and Discussion

The magnetizations m_a and m_b at A and B sub-lattices for different spin orientations are computed numerically and self-consistently for total Brillouin zone taking 100x100 grid points for electron momentum in graphene plane. To facilitate numerical computation, 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 dimensionless form. The scaled parameters are $\tilde{t}_1 = -1$, $\tilde{t}_2 = \frac{t_2}{t_1}$, $\tilde{t}_3 = \frac{t_3}{t_1}$, attractive impurity potential $v = \frac{V}{t_1}$, repulsive Coulomb potential $u = \frac{U}{t_1}$,

substrate induced gap $d_1 = \frac{\Delta}{t_1}$, site energy at A-site $ea = \frac{\varepsilon_a}{t_1}$, site energy at B-site $eb = \frac{\varepsilon_b}{t_1}$ and chemical potential $um = \frac{\mu}{t_1}$. The results are discussed in figures 1-2.

The sub-lattice magnetizations (m_a, m_b) plotted for Coulomb potential at temperatures $t = 0.04, 0.08, 0.10$ at room temperature and just above it. At a given temperature $t = 0.04$, the magnetization m_a decreases up-to Coulomb potential $u_c = 2.35$ and monotonically increases for higher u. On the other hand AFM magnetization(m_b) gradually increases for higher u, but AFM magnetization vanishes for u less than $U_c = u_c \, t_1 = 2.35 \, t_1 \approx 6.533 \, eV$. It is observed that the sub-lattice magnetizations are in-equivalent i.e. $(m_a > m_b)$ for all values of coulomb potential at any given temperature. The in-equivalence arises due to the substrate induced gap in graphene. The magnetization(m_a) is enhanced for lower Coulomb potential and is suppressed at higher u values showing a turning point in m_a where m_a is greater than a critical magnetization (m_{ac}) and magnetization m_a becomes smaller than critical magnetization (m_{ac}) for this critical Coulomb potential $u_{ca} = 3.58$. On the other hand magnetization(m_b) is enhanced for lower value of Coulomb potential up-to critical Coulomb potential $u_{cb} = 2.51$ and the magnetization is suppressed for Coulomb potential $u > u_{cb}$. The turning point m_a and m_b shows that the corresponding magnetizations are independent of temperature.
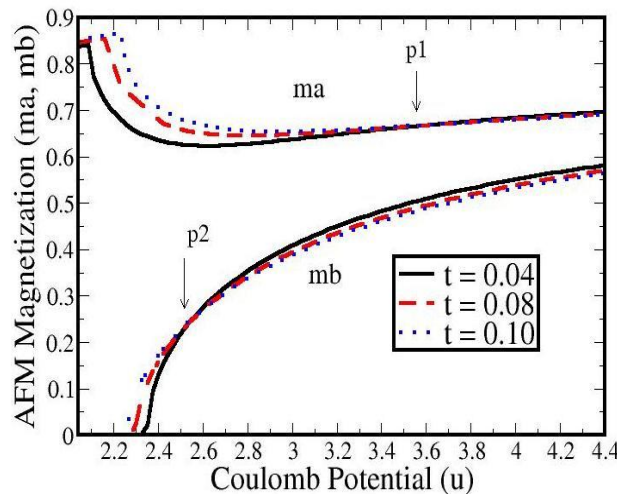
Fig. 1. The plot of magnetizations (m_a, m_b) vs. Coulomb Potential (u) for different temperatures $t = 0.04, 0.08, 0.10$, impurity potentials $v = 0$ and at substrate induced gap $d_1 = 0.090$ and band filling n = 0.75.

Figure. 2 shows that the effect of substrate induced gap d_1 on AFM sub-lattice magnetizations (m_a, m_b). For given $d_1 = 0.10$, the magnetization m_a gradually decreases with Coulomb potential u and finally nearly remain constant with further increase of u. On the other hand AFM magnetization(m_b) is zero for critical Coulomb potential $u_{cb} = 2.4$ for $d_1 = 0.10$. However, the magnetization(m_b) is continuously increases for $u > u_c = 2.4$. With further decrease of substrate induced gap, the magnetization(m_a) is suppressed and magnetization (m_b) is enhanced with decrease of substrate induced gap. Finally the two sub-lattice magnetizations coincide with each other ($m_a = m_b$) for all Coulomb potentials. In the absence of substrate induced gap (i.e. $d_1 < 0.002$), the AFM magnetization attains a flat minimum ($m_{ac} = m_{bc} \approx 0.45$) at critical Coulomb potential ($u_c \approx 2.2$) and increases linearly with further increase of Coulomb potentials.
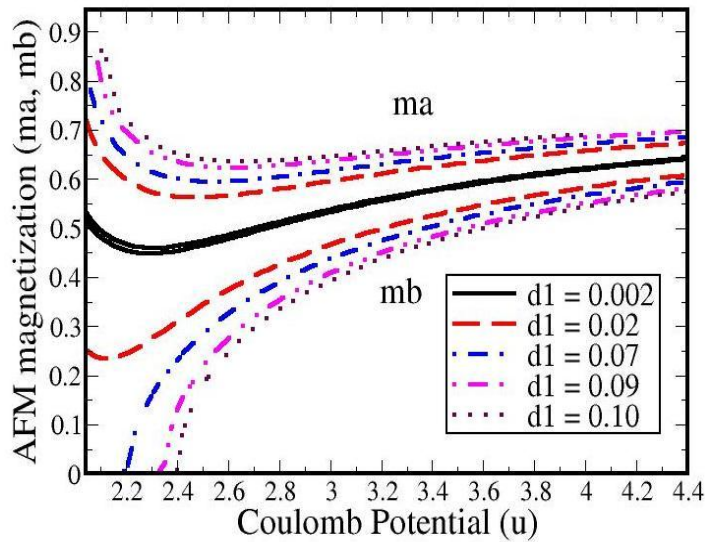
Fig. 2. The plot of magnetizations (m_a, m_b) vs. Coulomb Potential (u) for fixed temperature $t = 0.04$, impurity potentials $v = 0$ and at different substrate induced gap $d_1 = 0.002, 0.02, 0.07, 0.09, 0.10$ and band filling n $= 0.75$.

5.   Conclusions

We have proposed a tight binding model study taking electron hoppings up-to third nearest neighbors for graphene-on-substrate. The on-site Coulomb interaction is treated within mean-field approximation and the average electron density is expressed in terms of electron occupancy and sub-lattice magnetizations for anti-ferromagnetic(AFM) order. The sub-lattice magnetizations are calculated by Green's function technique and are computed numerically. It is observed that the sub-lattice magnetization (m_a) is always greater than the sub-lattice magnetization (m_b). The AFM order at B-site exists above a critical Coulomb potential $u_c \approx 2.4$. The substrate induced gap enhances the magnetization (m_a) and suppresses the magnetization (m_b). However, the two sub-lattice magnetizations coincide with each other for lower substrate induced gap (i.e. $d_1 < 0.002$) for all Coulomb potentials. The present tight binding model calculation displays very strong interplay between the Coulomb interaction and substrate induced gap.

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