Model study of temperature dependent dynamic anti-ferromagnetic spin susceptibility in graphene

Sivabrata Sahu 1,a), H. S. Gouda 1,b), S. K. S. Parashar 1,c) and G. C. Rout 2,*

1 School of Applied Sciences (Physics), Campus-3, 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
a) siva1987@iopb.res.in, b) himanshugouda@yahoo.in, c) sksparashar@yahoo.com
*Corresponding author: gcr@iopb.res.in

Abstract. We report here a microscopic theoretical study of dynamic anti-ferromagnetic spin susceptibility of electrons for graphene systems, which deal with a tight-binding model Hamiltonian consisting of the hopping of electrons up to third nearest-neighbours, impurity and substrate effects besides Coulomb interaction of electrons at A-and B- sub-lattices. The spin susceptibility involves four two-particle Green’s functions, which are calculated by Zubarev’s Green’s function technique. The up and down spin electron occupancies at A and B sub-lattices are computed numerically and self-consistently. The temperature dependent susceptibility becomes smooth for $U = u_c t_2$ with critical Coulomb potential $u_c = 2.2$, whereas the susceptibility shows more suppression at low temperatures in anti-ferromagnetic phase for $u < u_c$. The susceptibility is suppressed due to the increase of external periodic frequency imposed on the system. The A site doping enhances the susceptibility, while the B site doping suppresses it. The increase of the substrate induced gap enhances the susceptibility. The energy dependent real part of susceptibility shows that susceptibility minima shift to higher energies with increase of substrate induced gap.

Keywords: Graphene, dynamic spin susceptibility, Coulomb interaction, Anti-ferromagnetism

1. Introduction

The tremendous recent progress in graphene research is driven by its remarkable properties including magnetism. It has been predicted that the bipartite graphene structures could exhibit magnetic ordering which has potential application for spintronics devices [1, 2]. The repulsive electron-electron interaction in the bipartite lattice of graphene will produce imbalances of sub-lattice atoms leading to spin polarization of the ground state of the system [1]. The spins on same sub lattice are found to exhibit ferromagnetic coupling, whereas spins on different sub lattices couple anti-ferromagnetically. In theory, magnetic ordering has been demonstrated in nano ribbons [3] and vacancies in bulk graphene [4]. The experimental reports on the detection of ferromagnetic and...
anti-ferromagnetic orderings [5, 6] and measurement of defect induced para-magnetism [7, 8] are rare. Several theoretical studies offered explanations for the diversity of phenomena related to magnetic orderings [9, 10]. Monte-Carlo calculations of Sorella et al [11] have found that the Hubbard model in half filled honeycomb lattice would exhibit a Mott-Hubbard transition at a finite U. The minimal on-site strong Hubbard interaction is believed to produce the anti-ferromagnetic ground state [12]. Recent field theoretical calculations suggest that a direct transition from the Dirac semi-metal to the Neel state occurs [13, 14]. A recent tight-binding calculation predicts the occurrence of ferromagnetism (FM) and anti-ferromagnetism (AFM) in the graphene [15]. Applying this model Hamiltonian, we have calculated and reported [16] magnetic spin susceptibility for ferromagnetic and anti ferromagnetic ordering in graphene. Sahu et.al. [17,18,19] have reported the opening of band gap in AA-stacked and AB-stacked bi-layer graphene. Here we report the tight-binding theoretical model calculation of dynamic anti-ferromagnetic spin susceptibility.

2. Theoretical Model

Based upon our earlier calculation [15, 20, 21, 22], the following tight-binding model Hamiltonian describes the mono layer graphene system on-substrate is given by

\[ H = \sum_{k,\sigma} \left[ \varepsilon_{a,\sigma}(k) a_{k,\sigma}^\dagger a_{k,\sigma} + \varepsilon_{b,\sigma}(k) b_{k,\sigma}^\dagger b_{k,\sigma} \right] + \sum_{k,\sigma} \left[ \varepsilon_{13}(k) a_{a,k,\sigma}^\dagger b_{a,k,\sigma} + \varepsilon_{13}(k) b_{a,k,\sigma}^\dagger a_{a,k,\sigma} \right] \]

(1)

where \( \varepsilon_{a}(k) = \varepsilon_a - t_2 \gamma_2(k) + V_0 x_a + \Delta, \varepsilon_{b}(k) = \varepsilon_b - t_2 \gamma_2(k) + V_0 x_b - \Delta \) and \( \varepsilon_{13}(k) = -t_3 \gamma_3(k) - t_2 \gamma_2(k) \). Here \( \varepsilon_a \) and \( \varepsilon_b \) are the site energies of A- and B- sites respectively. Here \( t_j \) and \( \gamma_j(k) \) with \( j = 1-3 \) represent the hopping integrals and dispersions respectively up to third-nearest-neighbour hopping. Further \( x_a \) and \( x_b \) are the impurities at A and B sites with impurity potential \( V_0 \). The graphene substrate introduces energy gap with excess energy at \( +\Delta \) at A site and lower energy \( -\Delta \) at B site and thereby forming a substrate induced gap of \( 2\Delta \). The Hubbard type Coulomb interaction is introduced as \( H_U = \sum_{a,i} n_{a,i}^a n_{a,i}^a + \sum_{b,i} n_{b,i}^b n_{b,i}^b \).

The Coulomb interaction is treated here within Hartree-Fock approximation, where the mean occupation \( < n_{a,\sigma}^> \) of electron at A and B sites for up and down spins are computed numerically and self-consistently. This Hamiltonian introduces anti-ferromagnetism in graphene System.

3. Calculation Of Dynamic Longitudinal anti-ferromagnetic Susceptibility

The Fourier transformed dynamic longitudinal magnetic susceptibility \( \chi^{zz}(q, \omega) \) for antiferromagnetic states in monolayer graphene is defined as

\[ \chi^{zz}(q, \omega) = \ll (m_A(q,t) - m_B(q,t)) ; (m_A(-q,0) - m_B(-q,0)) \gg_{\omega} \]

(2)

where the \( z \) component of magnetizations are defined as \( m_A(q,t) = \sum_k (a_{k+q,1}^+ a_{k,1} - a_{k+q,1}^+ a_{k,1}) \) at A site and \( m_B(q,t) = \sum_k (b_{k+q,1}^+ b_{k,1} - b_{k+q,1}^+ b_{k,1}) \) at B site. Then the equation (2) can be re-written in terms of the two particle Green’s functions as
The two particle Green’s functions are written as \( x_1 = \langle \alpha_1 | \beta_1 \rangle \), \( x_2 = \langle \alpha_2 | \beta_1 \rangle \), \( x_3 = \langle \alpha_3 | \beta_2 \rangle \), and \( x_4 = \langle \alpha_2 | \beta_2 \rangle \). Here \( a_{k,\sigma} \) and \( b_{k,\sigma} \) are respectively the creation (annihilation) operators of electrons at A- and B- sub-lattices. The above two particle Green’s functions are calculated by using Zubarev’s technique [7]. During the calculation for each Green’s function, we solve the four coupled Green’s function equations. Finally the temperature and frequency dependent susceptibility is calculated in terms of the Fermi-Dirac distribution functions.

4. Result and Discussion

The impurity and substrate effects produce in-equivalence in A and B site atoms. In presence of Coulomb interaction the anti-ferromagnetism is produced in the Graphene system. We calculate and compute numerically and Self-consistently the electron occupancies of A and B sites i.e. \( n_a \) and \( n_b \) respectively. The temperature dependent dynamic anti-ferromagnetic susceptibility is computed. The physical parameters involved in the calculation are made dimensionless with respect to nearest-neighbour hopping integral \( (t_1) \). The parameters are \( t_1 = -1, t_2 = t_3, t_2 = t_4, \) impurity potential \( (\nu) = V_0/t_1 \), Coulomb potential \( (u) = U/t_1 \), temperature \( (t) = k_B T/t_1 \), applied external frequency \( (c1) = \omega/t_1 \), and substrate induced gap \( (d1) = \Delta/t_1 \).

The figure 1 (a) shows the effect of Coulomb interaction on the temperature dependent anti-ferromagnetic dynamic spin susceptibility \( \chi \). The susceptibility decreases with increase of temperature for all values of Coulomb interaction \( u = 1.7 \) to \( 2.5 \). Susceptibility \( \chi \) is very smooth in paramagnetic phase for \( U < u_c t_1 \), whereas \( u_c = 2.2 \) is the critical Coulomb interaction whereas \( \chi \) shows more suppression at low temperatures in anti-ferromagnetic phase for higher Coulomb interaction, \( U > u_c t_1 \). Figure 1(b) shows the effect of external frequency \( c1 = \omega/t_1 \) from 0.2 to 0.4 on the magnetic susceptibility. The increase of external frequency produces extra pressure on the crystal lattice which suppresses the magnetism as well as substrate induced gap. As a result magnetic susceptibility is suppressed with increase of external frequency.
We study here the effect of A site doping as well as B site doping of graphene on the temperature dependent magnetic susceptibility as shown in figure 2. It is observed that with increase of A site doping \( x_A = 0.02 \) to \( 0.010 \), the magnetic susceptibility is enhanced. It is to note further that the Neel temperature shown by the peak position in \( \chi \) (indicated by arrow marks) shifts to higher temperature with increase of doping. Figure 2(b) shows the effect of B site doping on \( \chi \). The increase of B site doping, on the other hand, suppresses the \( \chi \) considerably and the peak positions of \( \chi \) completely disappear with the B site doping.

**FIGURE 1.** The plot of susceptibility (AFM) vs. temperature (t) for different values of Coulomb potential \( u = 1.7, 2.0, 2.2, 2.4, 2.5 \) on substrate induced gap \( d_1 = 0.04 \) at small wave vector \( q_x = 0.03 \) and \( q_y = 0.03 \) (shown in fig. a) and the plot of Susceptibility (AFM) vs. temperature (t) for different energies \( c_1 = 0.2, 0.3, 0.4 \) at fixed Coulomb potential \( u = 2.2 \) and for small wave vector \( q_x = 0.03 \) and \( q_y = 0.03 \).
FIGURE 2. The plot of susceptibility (AFM) vs. temperature (t) for different values of doping concentrations at A site $x_a = 0.02, 0.04, 0.06, 0.08, 0.10$ of impurity potential $v = 1$ for substrate induced gap $d_1 = 0.04$ at small wave vector $q_x = 0.03$ and $q_y = 0.03$ at fixed Coulomb potential $u = 2.2$ (shown in fig. a) and the plot of Susceptibility (AFM) vs. temperature (t) for different values of doping concentrations at B site $x_b = 0.02, 0.04, 0.06, 0.08, 0.10$ of impurity potential $v = 1$ for substrate induced gap $d_1 = 0.04$ at small wave vector $q_x = 0.03$ and $q_y = 0.03$ at fixed Coulomb potential $u = 2.2$ (shown in fig. b)

FIGURE 3. The plot of susceptibility (AFM) vs. temperature (t) for different values of substrate induced gap $d_1 = 0.02, 0.03, 0.04, 0.05$ for fixed Coulomb potential $u = 2.2$ at small wave vector $q_x = 0.03$ and $q_y = 0.03$

FIGURE 4. The plot of susceptibility (AFM) vs. Energy ($c_1$) for different values of substrate induced gap $d_1 = 0.02, 0.03, 0.04, 0.05$, for fixed Coulomb potential $u = 2.2$ at small wave vector $q_x = 0.03$ and $q_y = 0.03$.

The substrate induced gap plays an important role on the magnetic properties of graphene. The figure 3 shows the effect of substrate induced gap $d_1$ on $\chi$. With increase of substrate induced gap, the magnetic susceptibility
is enhanced throughout the temperature range and this effect is clearly observed in the real part of the energy dependent on the anti-ferromagnetic susceptibility as shown in figure 4. The figure 4 shows the energy dependence of real part of AFM susceptibility $(\text{Re } \chi)$. $\text{Re } \chi$ shows a low temperature peak (p1) arising due to neutron momentum transfer energy and another peak (p2) arising due to substrate induced gap. With increase of substrate induced gap, the peak position of peak (p2) is enhanced considerably with its peak position unaltered, while the peak (p1) arising due to substrate induced gap shifts to higher energies (see in inset figure 4).

5. Conclusion

We present here a microscopic theoretical tight binding model calculation of dynamic anti-ferromagnetic spin susceptibility in mono layer graphene. The model consists of electron hopping up to third nearest-neighbours, substrate induced gap, doping effect and Coulomb interaction at A and B sub lattices. We calculate magnetic spin susceptibility and compute it numerically. We observe that anti-ferromagnetism develops in graphene for critical Coulomb interaction $U > u_c, t_1$ with $u_c = 2.2$. The A site doping enhances and B site doping suppresses the susceptibility. It is to note further that the substrate induced gap enhances the spin susceptibility throughout the temperature and the substrate induced peak observed in real part of AFM susceptibility shifts to higher energy with increasing magnitude of substrate induced gap.

Acknowledgments: The authors gracefully acknowledge the research facilities offered by the Institute of Physics, Bhubaneshwar, India. The author (Sivabrata Sahu) also acknowledges the leave granted by the authorities of Synergy Institute of Engineering and Technology, Dhenkanal, Odisha to continue research work

References

[1] Oleg V Yazyev 2010 Rep. Prog. Phys. 73, 056501
[2] Young-Woo Son, Marvin L. Cohen Steven G. Louie 2006 Nature 444, 347
[3] J. Fernández-Rossier 2008 Phys. Rev. B 77, 075430
[4] J. J. Palacios, J. Fernández-Rossier, and L. Brey 2008 Phys. Rev. B 77, 195428
[5] T. Enoki and K. Takai 2009 Solid state commun. 149, 1144
[6] V. L. Joseph Joly, Manabu Kiguchi et. al. 2010 Phys. Rev. B 81, 245428
[7] Andreas Ney, Pagona Papakonstantinou, Ajay Kumar et.al. 2011 Appl. Phys. Lett. 99,102504
[8] R. R. Nair,M. Sepioni,I-Ling Tsai,O. Lehtinen,J. Keinonen et. al. 2012 Nat. Phys. 8,199
[9] Jens Kunstmann , Cem özdoğan, Alexander Quandt, and Holger Fehske 2011 Phys. Rev. B. 83, 045414
[10] Oleg V. Yazyev and M. I. Katsnelson 2008 Phys. Rev. Lett. 100, 047209
[11] S. Sorella and E. Tosatti 1992 Europhys. Lett. 19, 699
[12] Jun Nagamatsu, Norimasa Nakagawa, Takahiro Muranaka, Yuji Zenitani . 2001 Nature 410, 63
[13] N. M. R. Peres, M. A. N. Arajo, and A. H. Castro Neto 2004 Phys. Rev. Lett. 92, 199701
[14] N. M. R. Peres and M. A. N. Arajo 2002 Phys. Rev. Lett. 65, 132404
[15] Sivabrata Sahu and G.C. Rout, J. Magn. Magn. Mater. 407, 396
[16] Sivabrata Sahu, S.K.S. Parashar and G.C. Rout, 2016 AIP Conf. Proc. 1724, 020111
[17] Sivabrata Sahu, S.K.S. Parashar and G.C. Rout, 2016 Mat. Today: Proc. 3, 39
[18] G.C. Rout, Sivabrata Sahu, S.K. Panda, 2016 AIP Conf. Proc. 1724, 020125
[19] Sivabrata Sahu, S.K.S. Parashar and G.C. Rout, 2016 AIP Conf. Proc. 1728, 020037
[20] Sivabrata Sahu and G. C. Rout, 2015 Physica B 461, 49
[21] Sivabrata Sahu and G.C. Rout, 2014 Adv. Sci. Lett. 20, 834
[22] Sivabrata Sahu and G.C. Rout, 2016 Adv. Sci. Lett. 22, 331