Orissa Journal of Physics
ISSN 0974-8202

Vol. 23, No.1
February 2016
pp. 79-84

The frequency dependent neutron scattering spectra of FM-ordered graphene: A tight-binding study

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Received : *11.12.2015* ; *Accepted : 14.01.2016*

Abstract. We report here a microscopic tight-binding study of frequency dependent neutron scattering spectra forferromagneticordering in the graphene systems. The tight-binding Hamiltonian consists of electron hopping up to third -nearest- neighbors, substrate and impurity effects in presence of Coulomb interaction of electrons separately at two inequivalent A and B sublattices of graphene. We calculate the two particle electron Green's functions by using Zubarev's Green's function technique. The frequency dependent scattering intensity of the system is computed numerically. Thespectra displays a sharp peak at the neutron momentum transfer energy at low energies and another higher energy peak appearing at substrate induced gap.

Keywords. Graphene, Ferromagnetic spin-susceptibility, neutron scattering

PACS Nos. 81.05ue, 73.22.-f

1. Introduction

Graphene is a truly two dimensional zero band gap honey-comb lattice with one electron per orbital. Graphene as a possible new magnetic material has stimulated renewed research interest for application in spintronic devices [1]. The pristine graphene does not contain magnetism due to equivalence of the two sublattices of carbon atoms. Many theoretical works [2, 3, 4, 5] have predicted defected induced magnetism in graphene by simulating in-equivalence in two

sublattices of carbon atoms. Further the impurity effect, substrate induced gaps, defects and Coulomb correlation effect are expected to generate magnetism in graphene. The origin of magnetism in graphene is unclear till yet. The imbalance of electronic states in sublattice atoms due to substrate effect gives rise to electron-electron interaction leading to spin polarization [6]. It is to note that the spins on the same sublattices are found to exhibit ferromagnetic order and the spins of different sublattices are found to be exhibit antiferromagnetic order. We assume here that the graphene-on-substrate creates in equivalence in A and B sublattices leading to magnetism in graphene. We introduce Coulomb interaction in both the sublattices in the form of Hubbard model [7, 8]. We calculate the dynamic ferromagnetic spin susceptibility by using Zubarev's technique of two particle Green's function. The imaginary part of the ferromagnetic susceptibility describes the neutron scattering intensity for different temperature and frequency.

2. Formalism and calculation of susceptibility

Based upon our earlier reports [7, 8], the tight-binding model Hamiltonian for monolayer graphene is written below taking into account electron hopping upto third-nearest neighbors.

$$H_1 = \sum_{k,\sigma}\left(\varepsilon_a\, a^{\dagger}_{k,\sigma}a_{k,\sigma} + \varepsilon_b b^{\dagger}_{k,\sigma}b_{k,\sigma}\right) \tag{1}$$

$$H_2 = \sum_{k,\sigma} V\,(x_a\, a^{\dagger}_{k,\sigma}a_{k,\sigma} + x_b b^{\dagger}_{k,\sigma}b_{k,\sigma}) \tag{2}$$

$$H_3 = \sum_{k,\sigma} \Delta\,(\, a^{\dagger}_{k,\sigma}a_{k,\sigma} - \, b^{\dagger}_{k,\sigma}b_{k,\sigma}) \tag{3}$$

$$H_4 = -\sum_{\beta=1,3}\sum_{k,\sigma} t_\beta\left(\gamma_\beta(k)\, a^{\dagger}_{k,\sigma}b_{k,\sigma} + \gamma^{*}_{\beta}\,(k)\, b^{\dagger}_{k,\sigma}a_{k,\sigma}\right) \tag{4}$$

$$H_5 = -t_2\sum_{k,\sigma}\left(\gamma_2(k)\, a^{\dagger}_{k,\sigma}a_{k,\sigma} + \gamma^{*}_{2}\,(k)\, b^{\dagger}_{k,\sigma}b_{k,\sigma}\right) \tag{5}$$

Here H_1 represents the on-site hopping of electrons with site energies ϵ_a and $\epsilon_b$at A and B sites respectively. The Hamiltonian H_2 represents the external doping effect with doping concentrations x_a and x_b respectively at A and B sub- lattices with same impurity potential$V$. The Hamiltonian H_3 represents the substrate effect on graphene which raises the A site energy by Δ and lowers the B site energy by Δ, there by introducing a substrate induced gap 2Δ. The Hamiltonian H_4represents the electron hopping between nearest-neighbors and the third-nearest- neighbors represented by $\beta = 1$ and 3 and the corresponding electron dispersions are $\epsilon_1(k) = -t_1\gamma_1(k)$ and $\epsilon_3(k) = -t_3\gamma_3(k)$ respectively. The

Hamiltonian H_5 represents the electron hopping among the second- nearest -neighbors with dispersion energy$\epsilon_2(k) = -t_2\gamma_2(k)$. Here $t_\beta$and $\gamma_\beta(k)$ with $\beta = 1, 3$ represent the hopping integrals and band dispersions respectively up to third- nearest- neighbor. The Hubbard type Coulomb interaction is introduced as $H_u = \sum_{\alpha,i} n^\alpha_{i,\uparrow} n^\alpha_{i,\downarrow}$, where $\alpha = A$ and B sites. The Coulomb interaction is treated here within Hartree -Fock approximation, where the mean occupancies $< n^\alpha_{-\sigma} >$ of electron at A and B sites for up and down spins are computed numerically and self -consistently. This Hamiltonian introduces ferromagnetism in graphene system.

3. Calculation of Green's functions

The longitudinal ferromagnetic susceptibility $\chi^{zz}(q,\omega)$ is written as

$$\chi^{zz}(q,\omega) = \ll m^z(q,t); m^z(-q,0) \gg_\omega \tag{6}$$

where the z component of magnetization due **to** A and B site electrons is written as

$m^z = m_A(q,t) + m_B(q,t)$which is given by

$$m_A(q,t) = \sum_k (a^\dagger_{k+q,\uparrow} a_{k,\uparrow} - a^\dagger_{k+q,\downarrow} a_{k,\downarrow}) \; ; m_B(q,t)$$
$$= \sum_k (b^\dagger_{k+q,\uparrow} b_{k,\uparrow} - b^\dagger_{k+q,\downarrow} b_{k,\downarrow})$$

The susceptibility defined in equation (6) is rewritten in terms of two particle Green's functions as

$$\chi^{zz}(q,\omega) = \sum_{k,\sigma} \sum_{k',q',\sigma'} \sigma\sigma' [\chi_1 + \chi_2 + \chi_3 + \chi_4] \tag{7}$$

where $\sigma(\sigma')$ is +1(-1) corresponding to up (down) spin orientation of the electrons and the two particle Green's Functions are written as $\chi_1 = \ll \alpha_1; \beta_1 \gg_\omega$, $\chi_2 = \ll \alpha_2; \beta_1 \gg_\omega$, $\chi_3 = \ll \alpha_1; \beta_2 \gg_\omega$, $\chi_4 = \ll \alpha_2; \beta_2 \gg_\omega$, where $\alpha_1 = a^\dagger_{k+q} a_{k,\sigma}$, $\alpha_2 = b^\dagger_{k+q} b_{k,\sigma}$, $\beta_1 = a^\dagger_{k'-q',\sigma'} a_{k',\sigma'}$and $\beta_2 = b^\dagger_{k'-q',\sigma'} b_{k',\sigma'}$. The above two particle Green's functions χ_j (j =1 to 4) are calculated by using Zubarev's technique[9]. During the calculation for each Green's function, we solve the four coupled Green's function equations. Finally

the temperature dependent susceptibility is calculated in terms of the Fermi-Dirac distribution functions. The neutron scattering intensity is directly proportional to the imaginary part of the ferromagnetic susceptibility. The scattering intensity is computed numerically taking 100×100 grid point in momentum space during integration. The neutron scattering intensity is written as $I = -2\pi \sum_{k,\sigma} Im\{\chi^{zz}(q,\omega)\}$.The physical parameters involved in the calculation are made dimensionless with respect to nearest neighbor hopping integral (t_1). The parameters are$\tilde{t}_1 = -1$, $\tilde{t}_2 = \frac{t_2}{t_1}$, $\tilde{t}_3 = \frac{t_3}{t_1}$, impurity potential $v = \frac{V}{2}$, Coulomb potential $u = \frac{U}{t_1}$, temperature $t = \frac{k_B T}{t_1}$, applied external frequency $c_1 = \frac{\omega}{t_1}$ and substrate induced gap $d_1 = \frac{\Delta}{t_1}$.

4. Results and Discussion

The asymmetry at A and B sub lattice atoms is further enhanced by introducing the external impurity and substrate effects in the system. The Coulomb interaction among electrons generates ferromagnetism in graphene system. We calculate and compute numerically and self-consistently the electron occupancies $n_\sigma^a$and n_σ^b respectively of A and B sites for up and down spin orientations. The frequency dependent longitudinal susceptibility is computed for different values of temperature, substrate induced gap, impurity concentration, Coulomb interaction and different hopping parameters. The results are discussed for the figures 1 to 2.

The figure 1 describes the plot of neutron scattering intensity versus energy (c_1) for different values of neutron momentum transfer energy$q_x = q_y$. We observe a low energy sharp peak (p_1) arising due to neutron momentum transfer energy and another flat peak (p_2) associated with the substrate induced gap energy. With increase of q_x, the peak p_1 shifts to higher energy with enhancement of spectral height and this peak shifts to lower energy with decrease of q_x an ultimately the peak appears at $q_x = 0$. On the other hand, with increase of neutron momentum transfer energy, the peak (p2) height is enhanced and its position is slightly displaced to lower energies indicating that the substrate induced gap is slightly suppressed withq_x. It is to note further that the neutron scattering intensity is enhanced throughout the energy range upto the substrate induced gap and then intensity remains constant beyond this gap energy. The enhancement of intensity with q_x indicates that the magnitude of the ferromagnetic ordering is enhanced within the energy range.

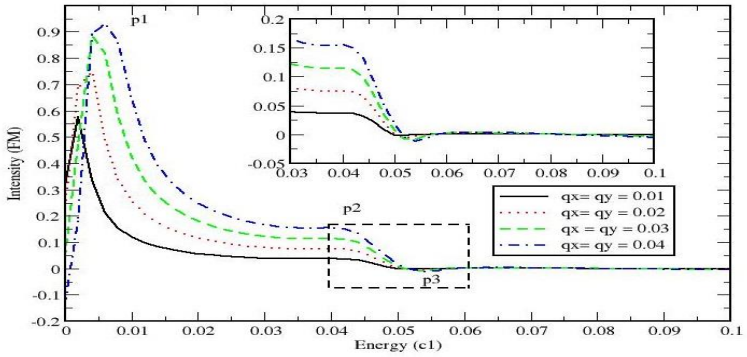

Fig. 1. The plot of intensity (FM) vs. energy (c1) for different values of neutron momentum transfer energy q on substrate Induced gap d1 = 0.04 for Coulomb potential u = 2.2, damping factor e1 = 0.001 at temperature t = 0.025 shown in above figure

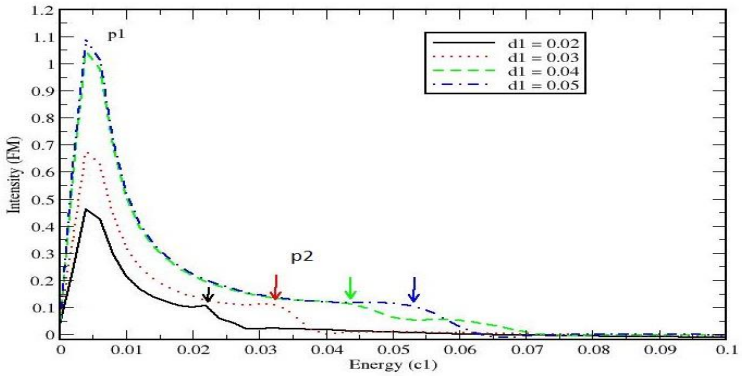

Fig. 2. The plot of intensity (FM) vs energy (c1) for different substrate induced gap d1 = 0.02, 0.0 3, 0.04, 0.05, fixed Coulomb potential u = 2.2, damping factor e1 = 0.001 and temperature t = 0.025 for small wave vector q = 0.03 as shown in above figure.

The figure 2 shows the effect of substrate induced gap on the scattering intensity. The scattering intensity is enhanced throughout the energy range upto the induced gap energy with the enhancement of the substrate effect (indicated by arrow mark). The peak p_1 arising due to neutron momentum transfer energy exhibits enhancement of its peak height with increase of substrate induced gap(d_1). With increase ofd_1, the position of peak p_2 shifts to higher energies showing the enhancement of substrate induced gap. The substrate induced gaps

are observed experimentally with gap values 100 meV for boron nitride [10], 200meV for gold-on-ruthenium [11] and 250 meV for SiC [12]. Tight-binding theoretical calculation by Rout et.al. [13] predicts that the substrate induced gap can be enhanced upto 67% by taking electron-phonon coupling of $\lambda = 0.2\ t_1$ and higher frequency phonon vibration frequency $\omega_0 = 0.6\ t_1$arising due to graphene-on-polarized substrate and superstrate.

5. Conclusions

We present here a tight-binding-binding model calculation of frequency dependent neutron scattering intensity of monolayer graphene. We consider a tight-binding model Hamiltonian incorporating substrate induced gap, coulomb correlation effect. The neutron spectra shows a low energy peak arising due to neutron momentum transfer energy and higher energy flat peak associated with substrate induced gap. The high frequency peak shifts to higher energy with increase of substrate induced gap. The increase of substrate induced gap and neutron momentum transfer energy enhances ferromagnetism ordering in the monolayer graphene within lower energies $(c_1) < d_1$.

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