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The Theoretical Study of the Effects of Coulomb Interaction and Substrate Interaction on Charge gap in Paramagnetic State of Graphene

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Abstract. We address here a tight binding model Hamiltonian to describe the evolution of charge gap in paramagnetic state of graphene on substrate. The Hamiltonian consists of nearest neighbor electron hoppings between two sublattices and symmetry breaking effect arising due to graphene on different substrates. The Coulomb interaction in graphene is considered here within mean-field approximation in the paramagnetic limit. The temperature dependent charge gap is calculated by Zubarev's Green's function technique and is computed numerically. It is observed that the charge gap gradually increases with Coulomb interaction in paramagnetic state up to a critical Coulomb energy $U_c \approx 2.35\, t_1$ where t_1 is the nearest neighbor electron hopping. The charge gap is enhanced with enhancement of the substrate interaction.

Keywords. Graphene, Paramagnetism, Coulomb Interaction
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1. Introduction

The magnetism in carbon allotropes has indeed been fundamental and also a controversial problem for long time [1,2]. Very recently the magnetic behavior of graphene has become the main focus of intensive studies because of its possible potential applications in spintronics [3,4]. While room temperature

ferromagnetism in graphene samples obtained from graphene oxide was observed by Wang et al. [5], the diamagnetism at room temperature and paramagnetism at low temperature are reported recently by Sepioni et al [6]. The paramagnetic orbital susceptibilities are theoretically predicted for electrons in doped graphene, when many particle Coulomb interactions [7] and lattice effects [8] are taken into consideration for calculation. Graphene on h-BN substrates (gra/h-BN) has electronic qualities comparable to those of suspended graphene. Substrate induced asymmetry appears in electron densities in two sub-lattices at low temperatures. Sahu et. al. have reported a tight binding model study of ferromagnetic and anti-ferromagnetic properties of graphene on substrate[9,10]. In this paper, we propose a theoretical tight-binding model to investigate the temperature dependent charge gap in paramagnetic limit of Coulomb interaction in graphene by varying different physical parameters such as substrate induced gap and impurities. Using this tight binding model, we have reported the effect of band filling on the temperature dependent charge gap [11]. In the present communication, we investigate the effect of substrate induced gap on the temperature dependent electron densities at the sub-lattices and finally on the charge gap in the paramagnetic state of the graphene-on-substrate.

2. Formalism and Calculation of charge gap

The tight binding Hamiltonian for electron in graphene can be written as

$$\mathbf{H_0}=\sum_{\mathbf{k},\boldsymbol{\sigma}}(\boldsymbol{\epsilon}_{\mathbf{a}}\,\mathbf{a}^{\dagger}_{\mathbf{k},\boldsymbol{\sigma}}\,\mathbf{a}_{\mathbf{k},\boldsymbol{\sigma}}+\boldsymbol{\epsilon}_{\mathbf{b}}\,\mathbf{b}^{\dagger}_{\mathbf{k},\boldsymbol{\sigma}}\,\mathbf{b}_{\mathbf{k},\boldsymbol{\sigma}}) + \sum_{\mathbf{k},\boldsymbol{\sigma}}\boldsymbol{\epsilon}_{\mathbf{1k}}\left(\mathbf{a}^{\dagger}_{\mathbf{k},\boldsymbol{\sigma}}\,\mathbf{b}_{\mathbf{k},\boldsymbol{\sigma}}+\mathbf{b}^{\dagger}_{\mathbf{k},\boldsymbol{\sigma}}\,\mathbf{a}_{\mathbf{k},\boldsymbol{\sigma}}\right) \qquad (\mathbf{1})$$

where $a^{\dagger}_{k\sigma}$ $(a_{k\sigma})$ and $b^{\dagger}_{k\sigma}$ $(b_{k\sigma})$ are the creation (annihilation) electron operators with spin s ($\sigma =\uparrow,\downarrow$) on sublattices A and B respectively. Here ϵ_a and ϵ_b are the sight energies at A and B sublattices. Here $t_1(= 2.5\ eV - 3.0\ eV)$ is the nearest-neighbor-hopping energy. In case of a graphene-on-substrate system, the electron interaction with the static potential induced by the substrate comes into play. As a result, a modulated potential, where A site has energy +Δ and B site with energy –Δ , leads to the breaking of the symmetry between A and B sites and gives rise to a gap. Such a symmetry breaking Hamiltonian is written as $\mathrm{H_{sub}} = \Delta\sum_{\mathrm{i},\sigma}\mathrm{a}^{\dagger}_{\mathrm{i},\sigma}\,\mathrm{a}_{\mathrm{i},\sigma} - \Delta\sum_{\mathrm{i},\sigma}\mathrm{b}^{\dagger}_{\mathrm{i},\sigma}\,\mathrm{b}_{\mathrm{i},\sigma}$. To stop both the electrons occupying the same site, the effect of Coulomb repulsion is introduced. The Hamiltonian

describing the Coulomb interaction with an effective Coulomb energy U is written as

$$H_U = U\sum_i\left[n^a_{i\uparrow}n^a_{i\downarrow} + n^b_{i\uparrow}n^b_{i\downarrow}\right] \quad (2)$$

where $n^a_{i\uparrow}(n^a_{i\downarrow})$ represent the occupation number of electron of up(down) spin at A sublattice. For weak coupling, the Hamiltonian can be decoupled by Hartree–Fock mean-field decoupling scheme, i.e. $Un^a_{i\uparrow}n^a_{i\downarrow} \approx U\langle n^a_{i\uparrow}\rangle n^a_{i\downarrow} + U\langle n^a_{i\downarrow}\rangle n^a_{i\uparrow} - U\langle n^a_{i\uparrow}\rangle\langle n^a_{i\downarrow}\rangle$. In the paramagnetic limit, the position and spin independent average electron densities at A and B sub-lattices are written as $\langle n^a\rangle = \langle n^a_{i\uparrow}\rangle = \langle n^a_{i\downarrow}\rangle$ and $\langle n^b\rangle = \langle n^b_{i\uparrow}\rangle = \langle n^b_{i\downarrow}\rangle$. After Fourier transformation in momentum space, the total Hamiltonian is given by $H = H_0 + H_{sub} + H_U$. Applying Zubarev's Green's function technique, the electron densities $\langle n^a\rangle = 1/N\sum_k\langle a^\dagger_k a_k\rangle$ and $\langle n^b\rangle = 1/N\sum_k\langle b^\dagger_k b_k\rangle$ are calculated for A and B sub-lattices respectively and written as

$$\langle n^a\rangle = \sum_k\left[\frac{(\omega_{1k}-\bar{\epsilon}_b)f(\beta\omega_{1k})-(\omega_{2k}-\bar{\epsilon}_b)f(\beta\omega_{2k})}{(\omega_{1k}-\omega_{2k})}\right] \quad (3)$$

$$\langle n^b\rangle = \sum_k\left[\frac{(\omega_{1k}-\bar{\epsilon}_a)f(\beta\omega_{1k})-(\omega_{2k}-\bar{\epsilon}_a)f(\beta\omega_{2k})}{(\omega_{1k}-\omega_{2k})}\right] \quad (4)$$

Here $f(\beta\omega_{1k})$ is the Fermi distribution function with $\beta = (kBT)^{-1}$

The quasi particle bands ω_{1_k} and ω_{2_k} are written as $\omega_{1_k} = \frac{P+\sqrt{P^2-4Q}}{2}$ and $\omega_{2_k} = \frac{P-\sqrt{P^2-4Q}}{2}$, where $P = \bar{\epsilon}_a + \bar{\epsilon}_b$, $Q = \bar{\epsilon}_a\bar{\epsilon}_b - \epsilon_1(k)\epsilon_1^*(k)$ with $\bar{\epsilon}_a = \epsilon_a + \Delta + U\langle n^a\rangle$ and $\bar{\epsilon}_b = \epsilon_b - \Delta + U\langle n^b\rangle$ and the nearest neighbor hopping energy is $\epsilon_{1k} = -t_1|\gamma_1(k)|$

The physical quantities are scaled by the nearest neighbor hopping integral t1. The parameters are the substrate induced gap $d_1 = \frac{\Delta}{t_1}$, repulsive Coulomb energy $u = \frac{U}{t_1}$ and temperature $t = \frac{k_BT}{t_1}$.

3. Results and Discussion

The A and B sub-lattices in the honeycomb lattice become asymmetric due to the substrate on which the graphene is placed. The electron density $\langle \boldsymbol{n^a}\rangle$ at A site and the electron density $\langle \boldsymbol{n^b}\rangle$ at B site become different leading to the condition,

$\langle \boldsymbol{n^a} \rangle > \langle \boldsymbol{n^b} \rangle$. Since the total charge at a site is proportional to the electron density, there occurs at a charge gap in the graphene-on-substrate due to Coulomb correlation energy $(\boldsymbol{U})$ in the paramagnetic phase. The magnitude of the charge gap is defined as $\Delta_{\boldsymbol{c}} = \boldsymbol{U}\left[\langle \boldsymbol{n^a} \rangle - \langle \boldsymbol{n^b} \rangle\right]$. Hence the temperature dependent electron densities $\langle \boldsymbol{n^a} \rangle \boldsymbol{and} \langle \boldsymbol{n^b} \rangle$ given in equations (3) and (4) are computed self-consistently for different values of substrate induced gap $(\boldsymbol{d_1})$ and finally the charge gap is computed $(\boldsymbol{i.e.}\, \boldsymbol{z_1} = \frac{\Delta_c}{\boldsymbol{T_1}})$ taking $\boldsymbol{110 \times 110}$ grid points of the electron momentum. The results are shown in figures 1, 2 and 3.

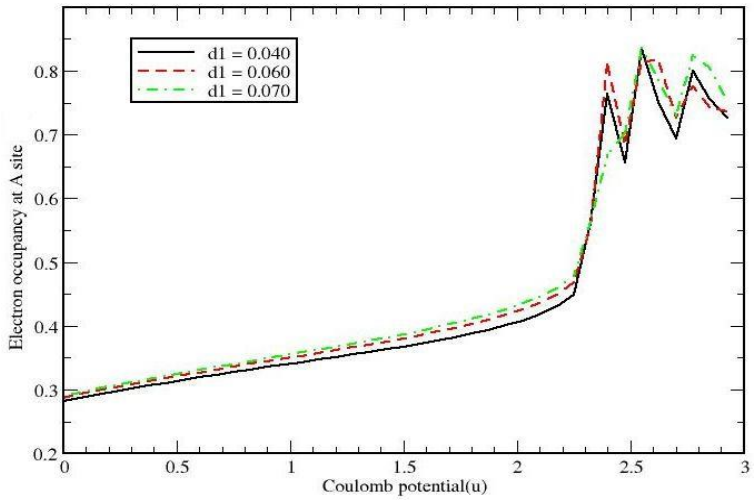

Fig. 1. The variation of A site electron occupancy $\langle n^a \rangle$ with Coulomb potential (u) for different substrate induced gap $d_1 = 0.04, 0.06, 0.08$ for a fixed temperature $t = 0.04$ and electron band filling $n = 1$.

The figure 1 shows the plot of electron occupancy versus Coulomb potential(u) for different substrate induced gap($d_1 = 0.040, 0.060, 0.070$). For given $d_1 = 0.040$ the A-site electron occupancy linearly increases upto critical Coulomb potential $U_c \approx 2.35t_1 \approx 6.533eV$ and then suddenly rises to higher value and then becomes unstable for $> u_c \approx 2.35$. The electron occupancy increases with increase of substrate induced gap for all values of coulomb potential up to the critical value u_c in the paramagnetic phase.

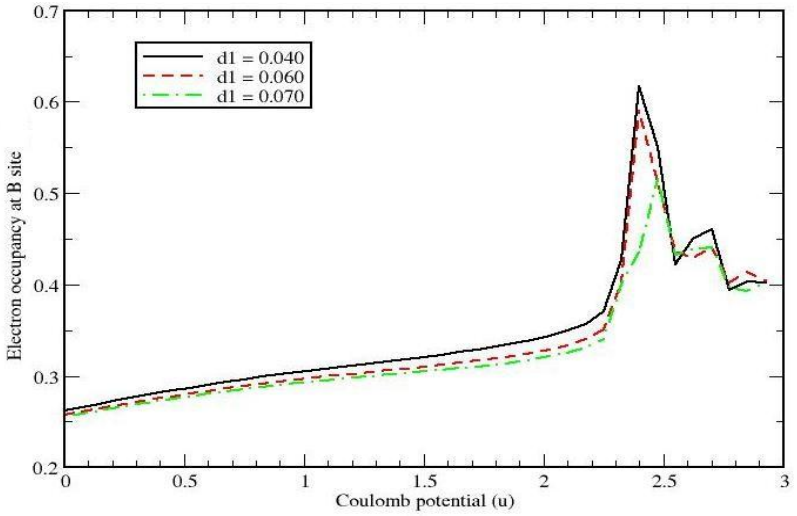

Fig. 2. The variation of B site electron occupancy $\langle n^b \rangle$ with Coulomb potential (u) for different substrate induced gap $d_1 = 0.04, 0.06, 0.08$ for a fixed temperature t=0.04 and electron band filling n =1.

Similarly figure 2 shows the effect of substrate induced gap on the plot of B-site electron occupancy versus Coulomb potential. It is observed that B-site electron occupancy linearly increases up to the critical Coulomb potential $U_c \approx 2.35$. However the B-site electron occupancy is suppressed with increase of substrate induced gap in the paramagnetic phase. The increase of $\langle n^a \rangle$ and decrease of $\langle n^b \rangle$ with increase of substrate induced gap arises due to the in-equivalence in the two sublattices of honeycomb lattice of graphene.

Finally the substrate induced gap is shown in figure 3. The substrate induced gap enhances the charge gap up to the critical Coulomb potential $U_c \approx 2.35$ in the paramagnetic phase of graphene. Further the charge gap is also enhanced with increase of Coulomb potential for any given value of the charge gap. The maximum charge gap is observed to be $z_1 \approx 0.35(\Delta_c = z_1 t_1 \approx 0.973\text{eV})$ for $U_c \approx 2.35$ and substrate induced gap $d_1 = 0.07(\Delta = d_1 t_1 \approx 0.194\, eV)$

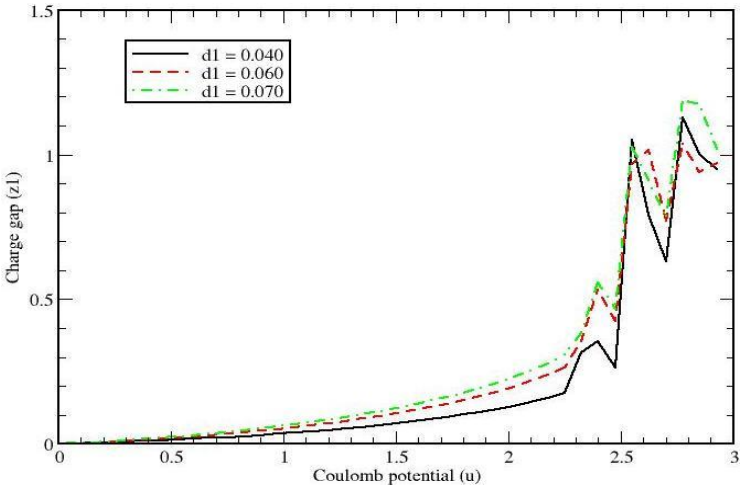

Fig. 3. The variation of charge gap (z_1) with Coulomb potential (u) for different substrate induced gap $d_1 = 0.04, 0.06, 0.08$ for a fixed temperature t=0.04 and electron band filling n=1.

4. Conclusions

We present here a tight binding model taking the neighbor hopping of electron in the honeycomb lattice of graphene-on-substrate. The Coulomb interaction introduces charge gap in graphene at the Dirac point. The charge gap is calculated by Zubarev's Green's function technique. It is observed that the maximum charge gap is found to be $\mathbf{z_1 \approx 0.35}$ for substrate induced gap $\mathbf{d_1 = 0.07}$ and critical Coulomb potential $\mathbf{u_c \approx 2.35 (U_c \approx 6\ eV)}$

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