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Interaction between the localized states in graphene

Angsula Ghosh [a,b,*], H.O. Frota [a]

[a] Department of Physics, Federal University of Amazonas, 69077-000 Manaus, AM, Brazil
[b] São Paulo State University, Institute for Theoretical Physics, São Paulo, SP, Brazil

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ABSTRACT

The formation of the localized magnetic moments is studied due to the presence of two-impurities in the two sublattices of a single-layer graphene sheet. The interaction between two similar magnetic impurities and also the hybridizations are decisive in determining the boundary between the magnetic and the non-magnetic states. A strong chemical potential dependence of the above phase boundary is evident. An anomalous scaling of the boundary separating the above regions is more pronounced in the two-impurity case when compared to that of the single impurity.

1. Introduction

The pioneering work of Andre Geim and Kostya Novoselov [1] has triggered a huge interest in the scientific community to study graphene, mainly motivated by their unusual electronic properties, such as the behavior of non-massive chiral Dirac fermion at low excitation energies. The high-electron mobility in graphene and its planar structure make it suitable for applications in nanoscience and nanotechnology leading to a new era of carbon-based electronics.

Graphene, a two-dimensional allotrope of carbon with the sp^2 hybridization state is distributed in a hexagonal lattice formed by two interpenetrating triangular sublattices, A and B. In the lattice plane, the s and p orbitals of the carbon atom form covalent bonds and thus provide a high mechanical strength to graphene. The remaining p orbitals of the carbon atoms in each sublattice, in the direction perpendicular to the plane of the lattice, hybridize, forming a conduction and a valence energy bands, known as π^* and π bands, respectively. The hexagonal distribution results in two bands that touch each other at two high-symmetry points in the Brillouin zone, K and K', known as Dirac points, thereby leading to a zero gap semiconductor. Around these points the dispersion relation is given by $E = \pm\hbar v_F \mid \vec{k} \mid$, with the energy E varying linearly as a function of the moment $\vec{k}$, where $\hbar$ is the reduced Planck constant and v_F is the Fermi velocity. Hence, it is similar to the dispersion relation of the photon, with the speed of light c playing the role of the velocity of the electrons in the Fermi level of graphene. As a two-dimensional Dirac fermion system, graphene presents unconventional and interesting electronic behavior. For instance, graphene shows a minimum conductivity of about e^2/h, an anomalous quantum Hall effect and a nonzero cyclotron mass m_c described by $E = m_c v_F^2$, although from the linear spectrum of fermion in graphene it would be zero. This similarity with Dirac fermions enables the prediction of the properties of the charge carriers in graphene from the relativistic Dirac equation, such as tunneling through a potential barrier without any reflection, which is known as Klein's paradox.

Impurity states are regarded as important contributors to the unusual and singular properties of graphene [2,3]. In the last few years there has been an increased attention to study the effect of magnetic adatom in a pristine graphene due to its potential use in spintronics. Doping with magnetic impurities could envisage the creation of local spins in graphene including the possibility of opening a gap. Recent progress in scanning tunneling microscopy made it possible to position adatoms in graphene and image the impurity states with high spatial resolution [4]. Various studies have been performed to characterize the necessary conditions under which the transition metal adatom on graphene can form localized magnetic moment. A systematic first-principles study of transition metals from Sc to Zn, including nonmagnetic adatoms Cu and Au, embedded in graphene has also been performed [5]. Isolated hydrogen atoms absorbed on graphene are predicted to induce magnetic moments [6]. Recently, two identical impurities on a zigzag nanoribbon has been studied to demonstrate that the

* Corresponding author at: Department of Physics, Federal University of Amazonas, 69077-000 Manaus, AM, Brazil.
E-mail address: angsula@ufam.edu.br (A. Ghosh).

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chemical potential and the spin-orbit coupling could drive the transition of the local-spin exchange from ferromagnetism to antiferromagnetism [7]. The impurity interaction control via the adjustment of the chemical potential has also been considered to observe that a weak repulsion is observed when the two atoms reside on the same sublattice and a stronger attraction when they are on different sublattices [8]. Double impurities have also been considered for local density of states calculations [9].

The presence of a magnetic adatom in a metal has been successfully studied using the Anderson model [10,11], which recently has been applied also to study magnetic moment formation in graphene [7,12,13]. Depending on the relation between the constitutive parameters of this model, the adatom orbital can be empty, single or doubly occupied. In particular, for temperatures higher than the Kondo temperature [14], there is a formation of local magnetic moment when the adatom and the conduction electrons are weakly hybridized and the Coulomb interaction between the electrons in the adatom orbital is greater than their binding energy. Moreover, for hybridization energy higher than the electron binding energy the orbital presents the valence fluctuation regime [14]. It has been observed that the coupling of a adatom to a graphene sublattice results in a much easier formation of magnetic moment due to the anomalous broadening of the electronic levels of the adatom [14]. In the present work we study the formation of the local magnetic moments due to the presence of two similar magnetic impurities in pristine graphene. The formation of the magnetic states in the single-layer graphene can be seen to depend on the interaction between the two impurities as observed in metals [11]. Moreover, the anomalous scaling of the magnetic boundary separating the magnetic and non-magnetic states alike the single-impurity in graphene continues to exist. The chemical-potential driven phase transition is also considered.

2. The model

The model Hamiltonian of graphene with two impurities hybridized with two sublattices of a single-layer graphene as shown in Fig. 1 is written as

$$H = H_{TB} + H_f + H_V \tag{1}$$

where H_{TB} is the tight binding Hamiltonian of the graphene, H_f is the impurity Hamiltonian, H_V is the hybridization of the adatom localized states with the graphene conduction electrons.

The tight binding Hamiltonian is given by

$$H_{TB} = -t\sum_{\langle i,j\rangle\sigma,l}\left[a^{\dagger}_{\sigma}(R_i)b_{\sigma}(R_j) + H.c.\right] \tag{2}$$

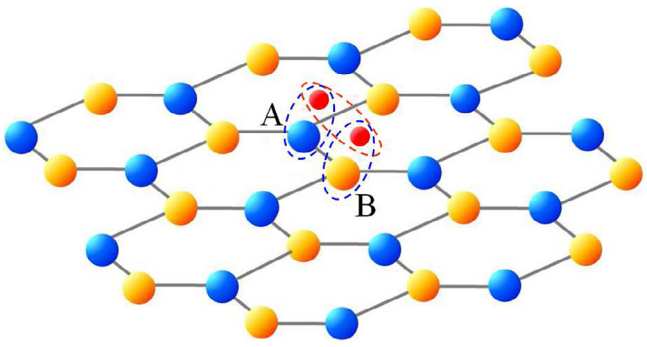

Fig. 1. Schematic diagram of the lattice structure of single layer graphene with two impurity atoms, each one coupled to the lattices A and B, respectively, and to each other.

where the operator $a_{\sigma}(R_i)$ ($b_{\sigma}(R_j)$) annihilates a state with spin σ at the position $R_i(R_j)$ on the sublattice A(B), $\langle i,j\rangle$ stands for summation over the nearest neighbors and the parameter t is the nearest neighbor hopping energy. In momentum space, we have

$$H_{TB} = -t\sum_{\mathbf{k},\sigma}\left[\phi(\mathbf{k})a^{\dagger}_{\mathbf{k},\sigma}b_{\mathbf{k},\sigma} + \phi(\mathbf{k})b^{\dagger}_{\mathbf{k},\sigma}a_{\mathbf{k},\sigma}\right] \tag{3}$$

where $\phi(\mathbf{k}) = \sum_{\delta}e^{i\mathbf{k}.\bar{\delta}_i}$ with $\bar{\delta}_1 = a(\hat{x}/2 + \sqrt{3}/2\hat{y})$, $\bar{\delta}_2 = a(\hat{x}/2 - \sqrt{3}/2\hat{y})$ and $\bar{\delta}_3 = -a\hat{x}$ as the nearest neighbor vectors. a is the lattice parameter. Diagonalizing the Hamiltonian (3) one generates two bands $\epsilon_{\pm}(\mathbf{k}) = \pm t|\phi(\mathbf{k})|$, which can be linearized around the Dirac points $\mathbf{K}$ at the corners of the Brillouin zone: $\epsilon_{\pm}(\mathbf{K}+\mathbf{q}) \sim \pm v_F|\mathbf{q}|$, where $v_F = 3ta/2$ is the Fermi velocity of the Dirac electrons.

The impurity Hamiltonian is described by

$$H_f = \sum_{\sigma}\epsilon_f(f^{\dagger}_{a\sigma}f_{a\sigma} + f^{\dagger}_{b\sigma}f_{b\sigma}) + U(n_{a\uparrow}n_{a\downarrow} + n_{b\uparrow}n_{b\downarrow}) + V_{ab}(f^{\dagger}_{a\sigma}f_{b\sigma} + f^{\dagger}_{b\sigma}f_{a\sigma}), \tag{4}$$

where $f^{\dagger}_{a\sigma}$ ($f^{\dagger}_{b\sigma}$) is the creation operator of a state with a spin $\sigma = \uparrow,\downarrow$ at the impurity of the sublattice A(B), $n_{a\sigma} = f^{\dagger}_{a\sigma}f_{a\sigma}$ and $n_{b\sigma} = f^{\dagger}_{b\sigma}f_{b\sigma}$ are the occupation number operators for the impurities in the sublattices A and B respectively. ϵ_f is the energy of the adatom electron, and U is the Coulomb interaction due to the double occupancy of an energy level in the adatom. The impurities interact with each other via the potential V_{ab}. For simplicity we adopt a mean-field approximation to the electronic correlations of the impurities, $Un_{\alpha\uparrow}n_{\alpha\downarrow} = U\sum_{\sigma}\langle n_{\alpha-\sigma}\rangle f^{\dagger}_{\alpha\sigma}f_{\alpha\sigma} - U\langle n_{\alpha\uparrow}\rangle\langle n_{\alpha\downarrow}\rangle$, where $\alpha = a, b$. Hence, the impurity Hamiltonian can be rewritten as

$$H_f = \sum_{\sigma}(\epsilon_{a\sigma}f^{\dagger}_{a\sigma}f_{a\sigma} + \epsilon_{b\sigma}f^{\dagger}_{b\sigma}f_{b\sigma}) + V_{ab}f^{\dagger}_{a\sigma}f_{b\sigma} + V^{\dagger}_{ab}f^{\dagger}_{b\sigma}f_{a\sigma}$$

where $\epsilon_{a\sigma} = \epsilon_f + U\langle n_{a-\sigma}\rangle$ and $\epsilon_{b\sigma} = \epsilon_f + U\langle n_{b-\sigma}\rangle$. The impurity orbital of the sublattice B is sited at the origin of the sublattice B and that of the sublattice A is at $\mathbf{r}_a = a\hat{r}$.

The hybridization of the impurity orbitals is given by

$$H_V = \frac{V_a}{\sqrt{N_a}}\sum_{\mathbf{k}}e^{ik_x a}a^{\dagger}_{k\sigma}f_{a\sigma} + \frac{V_b}{\sqrt{N_b}}\sum_{\mathbf{k}}b^{\dagger}_{k\sigma}f_{b\sigma} + H.c, \tag{5}$$

where N_a (N_b) denotes the number of atoms in the sublattice a (b), V_a and the V_b are the hybridization interactions of the impurities in the sublattices A and B respectively.

3. The formalism

The formation of a magnetic moment depends on the occupation of the two spin states of the impurities. The localized moment is formed when $n_{\alpha\uparrow} \neq n_{\alpha\downarrow}$. The interaction between the impurities V_{ab} is important to understand the relation between the localized states of the single-layer graphene. The self-consistent calculations of the density of states in the presence of the hybridization and V_{ab} is performed for the determination of the occupation of the impurities. The occupation of the impurity level can be determined by

$$n_{\alpha\sigma} = \frac{1}{2\pi}\int_{-\infty}^{\mu}d\omega A_{\alpha f\sigma}(\omega) \tag{6}$$

where the spectral function is given by

$$A_{\alpha f\sigma}(\omega) = -2\Im G^{R}_{\alpha f\sigma}(\omega). \tag{7}$$

The single particle retarded Green's function [15–18] of the f electrons is $G^{R}_{\alpha f\sigma}(t) = -i\theta(t)[f_{\alpha\sigma}(t), f_{\alpha\sigma}(t)]$ and the Fourier transform of that of the sublattice b can be written as