Resonant impurity band induced by point defects in graphene

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Abstract – It is pointed out that due to direct couplings of electronic states, point defects on graphene are strongly correlated and cannot be treated as independent scatterers. In particular, for large on-site defect potential and finite quasi-particle lifetime, we show that defects induce an impurity band with density of state characterized by the Wigner semi-circle law. By including long-range Coulomb interaction, we show that depending on quasi-particle lifetime and defect density, the impurity band may support ferromagnetism. Furthermore, the impurity band can enhance the conductivity of graphene to the order of \( 4e^2/h \), in agreement with experimental observations.

Recent experimental realization of single-layer graphene [1] has raised much interest in studying 2D Dirac fermions in the context of condensed-matter physics. One of graphene’s peculiar properties is the anomalous electronic properties associated with defects and disorders. The notable example is the observed ferromagnetic state induced by bombarding graphene with protons [2]. Here the induced magnetism results from \( \pi \)-electrons [3]. Another example is the finite conductivity at the Dirac point [4], indicating that there is finite density of states at the Dirac point. These observations appear to deviate from what is expected for ideal and clean graphene. Since real graphene must involve disorders, it indicates that disorders may play an important role in these phenomena.

Experimentally, there are many possible forms of disorders found in graphene [5]. Previous studies indicate that localized states would appear near the zig-zag edge [6] of graphene and cause magnetic behavior in carbon nanoribbons [7]. For large defects such as cracks, they tend to contain zig-zag edges. In this case, magnetic moments arise from localized states and interact via RKKY interaction, which tends to make graphene antiferromagnetic [8]. Hence the remaining candidates for the observed ferromagnetism in graphene are defects of small sizes or simply point defects. Here the simplest point defects are single-atom vacancies or hydrogen chemisorption defects. These defects generally create complicated disturbances in graphene and may even form ordered structures [9]. However, for low density of quenched defects, they can be simulated by a large potential \( u \) on a lattice point with distortion of nearby lattice point [10]. Indeed, the magnetic moment induced by single point defects in graphene widely gets support by calculations based on density functional theory (DFT) [11]. Furthermore, theoretical calculations indicate that it is sufficient to simulate a single-atom vacancy or a hydrogen chemisorption defect solely by an on-site potential \( u \). It is shown that these defects play a crucial role in inducing magnetism in \( \pi \)-electrons [10]. For finite density of defects, based on Lieb’s theorem [12], the Hubbard model predicts opposite magnetization for two sublattices on graphene. Thus the short-range Coulomb interaction does not favor ferromagnetism and this calls for consideration of the long-range Coulomb interaction. Nonetheless, even though calculations [10,11] by DFT do support the conclusion of the Hubbard model, the results obtained appear to be sensitive to sizes and boundary conditions [10]. On the other hand, even though weak antilocalization is observed in agreement with the theory [13], theoretical work on conductivity itself shows varying results [14,15] while experimental data consistently indicate that the conductivity is larger than the value \( 4e^2/\pi h \) obtained by many calculations [15]. It is therefore important to have a more reliable method for analyzing disordered graphene.

Theoretically, it is known that the electronic state near a point defect on graphene is semi-localized with amplitude decaying as \( 1/r \) when the distance \( r \) of the electron to

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the defect is large [16,17]. The semi-localized behavior is responsible for the observed long-range \((\sqrt{3} \times \sqrt{3})R30^\circ\) superstructure in STM images [18] and also explains why numerical results may depend on the sizes and boundary conditions of simulated graphene. Furthermore, it implies that semi-localized electrons interact strongly and calls for an appropriate method to take the semi-localization into consideration.

In this letter, we show that for finite quasi-particle lifetime, the semi-localization nature of defect states enables a resonant impurity band to form near zero energy. The existence of the impurity band explains finite conductivity at the Dirac point. Furthermore, by taking the long-range Coulomb interaction into account, we show that the impurity band may support ferromagnetism for an infinite graphene; while for graphene of small sizes, our results are consistent with calculations of DFT. Specifically, we shall investigate an infinite graphene with randomly distributed point defects, characterized by an on-site potential \(u\). In the large \(u\) limit, it is shown that the electronic spectrum due to randomly distributed point defects can be mapped into that of a random matrix. As a result, an impurity band with density of states characterized by the Wigner semi-circle law [19] forms. This impurity band appears to be observed in previous numerical results [20]. Due to the appearance of anomalous density of states near the Dirac point, we show that the conductivity is enhanced beyond \(4e^2/\pi \hbar\). By further including the long-range Coulomb interaction, it is shown that the impurity band supports ferromagnetism with the induced magnetic moment depending on the quasi-particle lifetime and defect density.

We shall start by modeling a point defect in graphene. Although point defects may induce more complicated disturbance on graphene or may even form ordered structure [9], as indicated by ab initio calculations, for low density of quenched defects, it is sufficient to model a point defect by an on-site potential \(u\) in the tight-binding model as long as the low energy near \(E = 0\) for \(\pi\)-electrons is concerned [10]. Let the tight-binding Hamiltonian for electrons in the \(\pi\)-band of an infinite graphene be \(H_0\). If the defect is located at \(\vec{r} = 0\), the wave function \(\psi\) for an electron then satisfies

\[
(H_0 + U\delta_{\vec{r},0}) \psi(\vec{r}) = E\psi(\vec{r}).
\]

(1)

Here and in the following \(\vec{r}\) is restricted to points on the honeycomb lattice. \(H_0\psi(\vec{r}) = -t\sum_{\vec{r}_i} \psi(\vec{r}_i)\) with \(t\) being the hopping amplitude and \(\vec{r}_i\) being the position of nearest neighbors to \(\vec{r}\). The solution \(\psi\) can be found by resorting to Huygens’ principle which implies the existence of a particular solution propagating outwards from the defect to infinity. This solution represents a state localized near the defect. Obviously, the solution that represents Huygens’ solution is the Green’s function, \(G(\vec{r}, \vec{r}', E)\), which describes the amplitude for the electron to propagate from \(\vec{r}'\) to \(\vec{r}\) and satisfies

\[
(E - H_0) G(\vec{r}, \vec{r}', E) = \delta_{\vec{r}, \vec{r}'}.
\]

(2)
the localized state hybridizes with extended states at the same energy. The degree of hybridization is determined by the boundary condition. In the case when \( \psi \rightarrow \phi_k \) as \( r \rightarrow \infty \) with \( \phi_k \) being the plane wave function with \( E_k = E_0 \), by appropriate superposition of \( \phi_k \) and \( \text{Im}[G(\vec{r}, 0, E)] \), one obtains \( \psi(\vec{r}) = \phi_k(\vec{r}) + uG(\vec{r}, 0, E_0)\psi(0) \), which reproduces the usual scattering solution obtained by the Lippmann-Schwinger equation.

As indicated in the above, since \( \psi_p(\vec{r}) \) goes as \( 1/r \), it implies that defects would couple strongly when many defects are in present. The strong coupling implies that point defects cannot be treated perturbatively. Clearly, the above construction can be generalized to the case with \( N_I \) impurities located at \( \vec{r}_i \) with \( i = 1, 2, 3, \ldots, N_I \). In this case, we have the obvious solution \( \psi_p(\vec{r}) = \sum_{i=1}^{N_I} A^*_i \text{Re}[G(\vec{r}, \vec{r}_i, E)] \). As a generalization of eq. (4), the energy of semi-localized states satisfies

\[
\begin{vmatrix}
1/u - g_{11} & -g_{12} & \cdots & -g_{1N_I} \\
-g_{12} & 1/u - g_{22} & \cdots & -g_{2N_I} \\
\vdots & \vdots & \ddots & \vdots \\
-g_{N_I1} & -g_{N_I2} & \cdots & 1/u - g_{N_I N_I}
\end{vmatrix} = 0, \tag{6}
\]

where \( g_{ij} = \{\text{Re}[G(\vec{r}_i, \vec{r}_j, E)] \} = g_{ji} \). For low density of defects, one expects that energies of semi-localized states are near 0. Since \( \text{Re}[G(0, 0, E)] \approx -\gamma E \) for \( E \sim 0 \), in the limit of \( u \rightarrow \infty \), eq. (6) reduces to

\[
\begin{vmatrix}
\gamma E & -g_{12}^0 & \cdots & -g_{1N_I}^0 \\
-g_{12}^0 & \gamma E & \cdots & -g_{2N_I}^0 \\
\vdots & \vdots & \ddots & \vdots \\
-g_{N_I1}^0 & -g_{N_I2}^0 & \cdots & \gamma E
\end{vmatrix} = 0, \tag{7}
\]

where \( g_{ij}^0 = \{\text{Re}[G(\vec{r}_i, \vec{r}_j, 0)] \} \). For randomly distributed positions of defects, eq. (7) implies that except for the scaling factor \( \gamma \), energies are exactly the eigenvalues of a symmetric random matrix. In the limit \( N_I \rightarrow \infty \), it is known that the density of \( \gamma E \) follows the Wigner semi-circle law [19]. Converting the Wigner semi-circle law to the density of electronic states, \( D(E) \), we find

\[
D(E) = \frac{n_I \gamma}{2\pi \Delta^2} \sqrt{4\Delta^2 - \gamma^2 E^2}, \tag{8}
\]

where \( n_I \) is the density of defects and \( \Delta^2 = N_I \langle (g_{ij}^0)^2 \rangle \) with \( \langle \cdots \rangle \) denoting the average over positions of defects. Thus the electronic states due to defects form an impurity band with the width \( w \) being determined by \( \Delta \) via the relation \( w = 4\Delta/\gamma \). This impurity band appears to be observed in a previous numerical simulation [16]. We note in passing that eq. (6) can be also obtained by summing the Dyson series for \( g_{ij} \). However, the existence of the impurity band is a non-perturbative result which cannot be obtained by summing each averaged term (averaged over the impurity potential) in the Dyson series.

For randomly distributed defects on graphene with \( N \) lattice points, the probability for a defect being located at \( \vec{r}_i \) is \( 1/N \). Hence \( \langle (g_{ij}^0)^2 \rangle = \sum_{ij} (g_{ij}^0)^2/N^2 \), which shows that \( \langle (g_{ij}^0)^2 \rangle \) is nothing but the Fourier transformation of \( \langle (g_{ij}^0)^2 \rangle \) at \( k = 0 \). We find \( \Delta^2 = \gamma N_I/2 \) and hence \( w = \sqrt{8\gamma N_I/\gamma} \), which decreases when \( \delta \) decreases. Note that because \( g_{ij}^0 \) does not vanish only when \( i \) and \( j \) belong to different sublattices, the impurity band results from the bipartite nature of graphene.

We first examine the magnetic property. For this purpose, we include the Coulomb interaction. Since screening is not important at the two-dimensional Dirac point, it is necessary to use the long-range Coulomb potential [1]

\[
H_C = \frac{e^2}{8\pi\epsilon_0} \sum_{i,j,\sigma,\sigma'} C_{i\sigma}^\dagger C_{j\sigma'} \frac{1}{|\vec{r}_i - \vec{r}_j|} C_{j\sigma'}^\dagger C_{i\sigma}. \tag{9}
\]

The magnetism can be examined by considering the case of two defects, in which semi-localized states are filled with two electrons. In this case, solutions to eq. (7) split into \( E_+ \) and \( E_- \) with the splitting, \( E_+ - E_- = 2|g^0(\vec{r}_i - \vec{r}_j)| \). Hence the splitting oscillates in the same way as \( g^0 \) does. Obviously, two electrons can be filled in \( E_\pm \) separately, or filled in the same states with the spin state being singlet. If Hund’s rule dominates, two electrons fill in \( E_\pm \) separately with their spins being in the triplet state. The total energies for two electrons for various possible spin configurations are \( E_+ + E_- + C_+ - J_+ \) (spin-triplet), \( E_+ + E_- + C_+ + J_- \) (spin-singlet), \( 2E_+ + C_+ + J_- \) (spin-singlet), and \( 2E_- + C_- \) (spin-singlet). Here \( C_{ab} \) and \( J_{ab} \) are Coulomb and exchange integrals given by

\[
C_{ab} = \frac{e^2}{8\pi\epsilon_0} \sum_{\vec{r}_1, \vec{r}_2} \frac{|\Psi_a(\vec{r}_1)|^2|\Psi_b(\vec{r}_2)|^2}{|\vec{r}_1 - \vec{r}_2|}, \tag{10}
\]

\[
J_{ab} = \frac{e^2}{8\pi\epsilon_0} \sum_{\vec{r}_1, \vec{r}_2} \frac{\Psi_a^\dagger(\vec{r}_1)\Psi_b(\vec{r}_1)\Psi_a(\vec{r}_2)^\dagger\Psi_b(\vec{r}_2)}{|\vec{r}_1 - \vec{r}_2|}, \tag{11}
\]

with \( \Psi_\pm \) being the corresponding wave functions to \( E_\pm \). Since \( \Psi_{\pm} \) can be expressed in terms of Green’s functions, both Coulomb and exchange integrals can be calculated in the Fourier space with finite domain of integration so that the preferred spin state for two defects on an infinite graphene can be found. Figure 2 shows our numerical calculations on locations of defects that favor the spin-triplet state for \( u = \infty \). Here one defect is fixed at the center, marked by \( x \), while the other defect is shown by a solid dot only when the spin-triplet is favored. It is seen that for short distances (the central region of fig. 2), our results are in consistent with calculations of DFT and anticipation of Lieb’s theorem for the Hubbard model.
show that the spin-triplet state is favored. For large distances, our results go beyond DFT and differ among different sublattices. This is in agreement with the calculations of DFT. For large distances, our results go beyond DFT and show that the spin triplet state is favored.

spin-triplet states are favored for defects on the same sublattice and spin-singlet states are favored for defects on different sublattices. This is in agreement with the calculations of DFT. For large distances, our results go beyond DFT and show that the spin triplet state is favored. We note in passing that by reducing the strength of the off-diagonal part of long-range Coulomb interaction, our results show that the central region grows which agrees with the anticipation of Lieb’s theorem.

To deal with the finite density of defects, we first note that due to the semi-localization of defect states, defects are strongly coupled. As a result, local spin population is entirely determined by the spin population in the impurity band. This is reflected in the expansion \( C_\sigma = \sum_{E,J} \langle \mu | g(\vec{r}, \vec{r}', E) | C_{E,\sigma} \rangle \). Clearly, one needs to calculate \( A_E^\uparrow A_E^\downarrow \) with \( A \) being the average over defect configuration. Because the normalization \( \sum_\sigma \psi_\sigma^2(\vec{r}) = 1 \), by setting \( E \sim 0 \), we find \( A_E^\uparrow A_E^\downarrow = \delta_{\mu J} / N_I \gamma_\mu \). Hence \( \langle C_\sigma | C_\omega \rangle \) is determined by \( \langle C_{E,\sigma} | C_{E,\omega} \rangle \). By further using the fact \( A_E^\uparrow A_E^\downarrow, A_E^\uparrow A_E^\downarrow, A_E^\uparrow A_E^\downarrow = A_E^\uparrow A_E^\downarrow, A_E^\uparrow A_E^\downarrow, A_E^\uparrow A_E^\downarrow \), we find that the competition between ferromagnetic, antiferromagnetic, and nonmagnetic states is determined by the exchange energy

\[
E_{\text{exchange}} = -\frac{e^2}{8\pi\hbar^2} \sum_{i,j} \frac{1}{|\vec{r}_i - \vec{r}_j|} \left( \frac{N_i}{\sum_k g_{ik} g_{jk}^0} \right)^2 ,
\]

where \( n_\sigma = N_\sigma / N_I \) are fractions of electrons in the spin state \( \sigma \). In the ferromagnetic state, we have \( n_\uparrow \neq n_\downarrow \) with \( E_\sigma \) being the corresponding Fermi energy for the spin state \( \sigma \). The total energy in the impurity band for spin state \( \sigma \) is \( \int E_{E,\sigma} d\mathcal{E}(E) \). For an undoped graphene, \( E_\downarrow = -E_\uparrow \). We find that \( n_\uparrow - n_\downarrow = 2(y\sqrt{1-y^2} + \sin^{-1} y) / \pi \) with \( y = E_\uparrow / 2\Delta \), while the change of the total energy in the impurity band per site is

\[
\Delta K = 2n_I w 3\pi \left[ 1 - (1 - y^2)^{3/2} \right] .
\]

Using eq. (12), the dependence of the exchange energy per site on \( n_\uparrow - n_\downarrow \) can be extracted and we find

\[
E_{\text{exchange}} = -\frac{(2 + \sqrt{3})e^2 n_I^2}{4\pi^3 \hbar a} \left( y\sqrt{1 - y^2} + \sin^{-1} y \right)^2 ,
\]

where \( a = 2.46 \text{ Å} \) for graphene. In deriving eq. (14), we have expressed \( g_{ij} \) and \( 1/(|\vec{r}_i - \vec{r}_j|) \) in the momentum space and approximated functions that are smooth in \( q \) by the corresponding values at Dirac points. It is clear that minimizing \( \Delta K + \Delta E_{\text{exchange}} \) with respect to \( y \) determines the induced magnetic moment. We find that the value \( y = 0 \) where the minimum occurs is determined by the intersection of \( y\sqrt{1 - y^2} + \sin^{-1} y \) and \( s y \) with \( s = 0.072 \mu \text{eV} / n_I \). Note that for finite quasi-particle lifetime, because \( \Delta K \) and \( \Delta E_{\text{exchange}} \) have different \( n_I \) dependence: \( \Delta k \sim n_I^{3/2} \) and \( \Delta E_{\text{exchange}} \sim n_I^2 \), their competition leads to a threshold in the defect density to induce ferromagnetism, as shown in fig. 3(a). For \( \Delta \sim O(\text{meV}) \), we find that ferromagnetism can be stabilized for \( n_I \sim O(10^{-2}) \). In fig. 3(b), we show the calculated induced magnetic moment per defect vs. \( \delta \). A clear linear behavior is shown. Since for finite temperature \( T \), \( \delta \sim T \) [21], our result is consistent with the linear temperature behavior of magnetic moment observed in experiments [22].
We now address the issue of the transport property by explicitly calculating the conductivity using the Kubo formula [23]
\[
\sigma = \lim_{\omega \to 0} -\frac{1}{A\hbar\omega} \text{Im} \Pi_{zz}^R(\omega + i\epsilon).
\]  
(15)

Here \(\sigma\) is the dc conductivity, \(A\) is the total area of graphene and
\[
\text{Im} \Pi_{zz}^R(\omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \langle [J_x(t), J_x(0)] \rangle
\]  
(16)

with the total current being given by \(J = \sum_{j, \delta} \delta C_{ij, \delta} C_{j, \delta}^\dagger \) with \(t\) being the hopping amplitude and \(\delta\) connecting nearest neighbors. In zero temperature, by expressing \(C_{ij}\) in terms of \(E_{E\sigma}\), one obtains \(J_{E\sigma} = \sum_{E, \delta} I_{E\sigma} J_{E\delta}^E C_{E\sigma}^E C_{E\delta}^E\) with the coefficient \(I_{E\sigma}\) determined by \(A E_{E\delta}^\dagger \delta\) and \(g_{j, \delta}\). After taking the average over defect configurations, we find the dc conductivity is given by [23]
\[
\sigma = 2\pi n \left| J_{E\sigma}^E \right|^2 D(E) D(E') \bigg|_{E = 0, E' = 0}.
\]  
(17)

By further expressing \(J_{E0}^E\) in the momentum space, we obtain
\[
\sigma = \frac{4e^2 \mu^2 t^2}{h} I,
\]  
(18)

where \(I = \int \frac{d^2k}{(2\pi)^2} \sin^2(k_x/2) |E_k|^4 / (\delta^2 + |E_k|^2)^4\). For small \(\delta\), because \(I \sim 1/(2\pi t^2 \delta^2)\), we find \(\sigma\) is larger than the value \(4e^2/h\) by a factor \(w^2/4\pi \delta^2\) [24]. Since the typical mean free path for graphene is about 30 nanometers [25], \(n_f \sim 10^{-5} - 10^{-6}\). Therefore, for \(\delta/t \sim 10^{-3}\), we find that the enhancement factor is of the order \(O(1)\), in agreement with experimental observations [15].

To summarize, we show that a resonant impurity band with the density of states characterized by the Wigner semi-circle law is induced near zero energy by point defects on graphene. The impurity band may support ferromagnetism with the induced magnetic moment depending on the quasi-particle lifetime and defect density. Furthermore, we find that a threshold in the defect density is required to stabilize ferromagnetism at fixed temperatures. The induced magnetization shows a linear behavior vs. temperature in agreement with experimental observations. Finally, the impurity band enhances the conductivity of graphene to the order of \(4e^2/h\), in agreement with experimental observations.

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