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Localized magnetic states in biased bilayer and trilayer graphene

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Abstract
We study the localized magnetic states of an impurity in biased bilayer and trilayer graphene. It is found that the magnetic boundary for bilayer and trilayer graphene shows mixed features of Dirac and conventional fermions. For zero gate bias, as the impurity energy approaches the Dirac point, the impurity magnetization region diminishes for bilayer and trilayer graphene. When a gate bias is applied, the dependence of impurity magnetic states on the impurity energy exhibits a different behavior for bilayer and trilayer graphene due to the opening of a gap between the valence and the conduction band in the bilayer graphene with an applied gate bias. The magnetic moment and the corresponding magnetic transition of the impurity in bilayer graphene are also investigated.

1. Introduction

The intense research currently devoted to graphene, a two-dimensional carbon honeycomb lattice, has uncovered a wealth of fascinating properties such as the anomalous quantized Hall effect, the absence of weak localization and the existence of minimal conductivity [1–5]. Graphene has a high mobility and its carrier density is controllable by an applied gate voltage [2] and a spin–orbit interaction [6–10].

Graphene structures have been the focus of much interest [12–20, 26]. In particular, adatoms may be positioned on graphene by current nanotechnology [21], rendering the study and manipulation of local electronic properties. Ab initio calculations for transition metal adatoms [22] show a tendency to the formation of local magnetic moments. Recently Uchoa et al [23] examined the condition for the emergence of localized magnetic moments on adatoms with inner shell electrons on a single-layer graphene. It is found that the impurity magnetization boundary exhibits anomalous characteristics. In contrast to the case of an impurity in an ordinary metal, the impurity can magnetize for any small charging energy due to the low density of state (DOS) at the Dirac point. On the other hand, detailed experimental studies [26] on multi-layer graphene showed a marked modification of the electronic structure with the number of layers. Hence, we expect [11] a qualitative difference in the magnetic properties of the adatoms on multi-layer graphene; an issue which we address here by inspecting the localized magnetic state of an impurity in a biased bilayer and trilayer graphene. We find that the size of the magnetic region decreases rapidly compared with that in monolayer graphene, the impurity can magnetize even when the energy of the doubly occupied state is below the Fermi level. The impurity magnetization region is asymmetric due to the special nature of the quasiparticles having mixed features of Dirac and conventional fermions. When a gate bias is applied, the dependence of the impurity magnetic states on the impurity energy for a bilayer graphene exhibits a different behavior from that for a trilayer graphene due to the opening of a gate-induced gap between the valence and the conduction band in the bilayer graphene. Calculating the occupation of the impurity level and the susceptibility in the bilayer graphene we show that the magnetic moment decreases with increasing the inter-layer coupling.

2. Bilayer graphene

Figure 1 shows the lattice structure of the bilayer graphene with the adatom. The inter-layer stacking is assumed to be the...
Bernal order where the top layer has its A sublattice atop the sublattice B of the bottom layer. The bias voltage $V$ is applied across the layers. The system Hamiltonian

$$H = H_{TB} + H_i + H_f$$

contains the graphene bilayer term $H_{TB}$, which in a tight-binding approximation reads

$$H_{TB} = 2 \sum_{l=1} \left[ H_l + H_T + H_V \right],$$

with

$$H_l = -t \sum_{\langle i,j \rangle \sigma} \left[ a_{i \sigma}^{\dagger}(\mathbf{R}) b_{j \sigma}(\mathbf{R}) + b_{j \sigma}^{\dagger}(\mathbf{R}) a_{i \sigma}(\mathbf{R}) \right],$$

$$H_T = -t_p \sum_{i,\sigma} \left[ a_{i \sigma}^{\dagger}(\mathbf{R}) b_{2\sigma}(\mathbf{R}) + b_{2\sigma}^{\dagger}(\mathbf{R}) a_{i \sigma}(\mathbf{R}) \right],$$

$$H_V = \frac{V}{2} \sum_{i,\sigma} \left[ a_{i \sigma}^{\dagger}(\mathbf{R}) a_{i \sigma}(\mathbf{R}) + b_{i \sigma}^{\dagger}(\mathbf{R}) b_{i \sigma}(\mathbf{R}) - a_{2\sigma}^{\dagger}(\mathbf{R}) a_{2\sigma}(\mathbf{R}) - b_{2\sigma}^{\dagger}(\mathbf{R}) b_{2\sigma}(\mathbf{R}) \right].$$

The operator $a_{i \sigma}(\mathbf{R})$ ($b_{i \sigma}(\mathbf{R})$) annihilates a state with a spin $\sigma$ at the position $\mathbf{R}$ on the sublattice A (B) of the $l$ plane. $t$ is the nearest neighbour in-plane hopping energy, $t_p$ is the interlayer hopping energy. For the hybridization with the localized impurity states we write

$$H_i = V_f \sum_{\sigma} \left[ f_{\sigma}^{\dagger} b_{1\sigma}(0) + b_{\sigma}^{\dagger} f_{\sigma}(0) \right],$$

where $f_{\sigma}$ ($f_{\sigma}^{\dagger}$) is the annihilation (creation) operator of a state with a spin $\sigma$ at the impurity, and $V_f$ is the hybridization strength. In the momentum space we have

$$H_l = -t \sum_{k,\sigma} \left[ \phi(\mathbf{k}) a_{1\sigma}^{\dagger} b_{k\sigma} + \phi^*(\mathbf{k}) b_{1\sigma}^{\dagger} a_{k\sigma} \right],$$

$$H_T = t_p \sum_{k,\sigma} \left[ a_{1\sigma}^{\dagger} b_{2\sigma} + b_{2\sigma}^{\dagger} a_{1\sigma} \right],$$

$$H_V = \frac{V}{2} \sum_{k,\sigma} \left[ a_{1\sigma}^{\dagger} a_{1\sigma} + b_{1\sigma}^{\dagger} b_{1\sigma} - a_{2\sigma}^{\dagger} a_{2\sigma} - b_{2\sigma}^{\dagger} b_{2\sigma} \right],$$

$$H_i = \frac{V_f}{\sqrt{N}} \sum_{k,\sigma} \left[ f_{\sigma}^{\dagger} b_{1\sigma} + b_{1\sigma}^{\dagger} f_{\sigma} \right].$$

where $\phi(\mathbf{q}) = -t \sum_{i=1}^{3} e^{iq_i}$ with $\delta_1 = \frac{q}{2}(1, \sqrt{3}, 0)$, $\delta_2 = \frac{q}{2}(1, -\sqrt{3}, 0)$, $\delta_3 = a(1, 0, 0)$ (here $a$ is the lattice spacing), and $N$ is the number of sites on sublattice B of plane 1. Diagonalizing $H_{TB}$ we find the spectrum

$$E_{\pm}(\mathbf{k}) = \pm \left[ \epsilon_k^2 + \frac{t_p^2}{2} + V^2 \pm \sqrt{\frac{t_p^4}{4} + (t_p^2 + V^2)\epsilon_k^4} \right],$$

where $\epsilon_k = \pm |\phi(\mathbf{k})|$ is the Bloch zone by $\epsilon_k = \pm v_F |k|$ where $v_F$ is the Fermi velocity. The impurity is described by the Hamiltonian $H_f$ with

$$H_f = \sum_{\sigma} \varepsilon_0 f_{\sigma}^{\dagger} f_{\sigma} + U n_{\uparrow} n_{\downarrow},$$

where $n_{\sigma} = f_{\sigma}^{\dagger} f_{\sigma}$ is the occupation number operator, $\varepsilon_0$ is the single electron energy at the impurity. The Coulomb interaction is included as a finite Anderson term $U$. For simplicity, we adopt a mean field approximation to the electronic correlations at the impurity, $U n_{\uparrow} n_{\downarrow} \approx U \sum_{\sigma} \varepsilon_\sigma f_{\sigma}^{\dagger} f_{\sigma} - U (n_{\uparrow}) (n_{\downarrow})$. The impurity Hamiltonian is rewritten as $H_f = \sum_{\sigma} \varepsilon_\sigma f_{\sigma}^{\dagger} f_{\sigma}$ with $\varepsilon_\sigma = \varepsilon_0 + U n_{\sigma}$. To investigate the localized magnetic states, we calculate the occupation number of the electrons of a given spin $\sigma$ at the impurity. At low temperatures all the states below the Fermi level $\mu$ are completely occupied and the occupation of the impurity is determined by

$$\langle n_{\sigma} \rangle = \int_{-\infty}^{\mu} d\omega \rho_f^{\sigma}(\omega),$$

where $\rho_f^{\sigma}(\omega)$ is the DOS at the impurity level. We infer this from the retarded Green’s function

$$G_f^{\sigma \sigma}(t - t’) = -i \theta(t - t’)(\langle f_{\sigma}(t), f_{\sigma}^{\dagger}(t’) \rangle).$$

By the standard equation of motion, we can derive ($\eta \to 0^+$)

$$G_f^{\sigma \sigma}(\omega) = \frac{\delta_{\sigma \sigma}}{\omega - \varepsilon_\sigma - \Sigma_f^{\sigma \sigma}(\omega) + i\eta},$$

where

$$\Sigma_f^{\sigma \sigma}(\omega) = \frac{V_f^2}{N} \sum_k \left\{ \frac{-(\omega - \frac{V}{2}) v_F |k|^2 + (\omega - \frac{V}{2})(\omega + \frac{V}{2}) - t_p^2 (\omega + \frac{V}{2})}{v_F^2 |k|^4 - 2(\omega^2 + \frac{V^2}{4}) v_F^2 |k|^2 + (\omega^2 - \frac{V^2}{4})^2 - t_p^2 (\omega^2 - \frac{V^2}{4})} \right\}.$$
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### 3. Trilayer graphene

The Hamiltonian for trilayer graphene contains a coupling of the B atom of the second layer to the A atom of the third layer according to the conventional Bernal-type stacking order. Similar to the bilayer graphene case, we find for the impurity Green’s function

\[ G^{f\sigma\sigma'}(\omega) = \frac{\delta_{\sigma\sigma'}}{\omega - \epsilon_{\sigma} - \Sigma' + i\eta}, \]

where

\[ \Sigma' = -\frac{V_f^2}{N} \sum_k \frac{A_1 \psi_\sigma^0 |k|^4 + B_1 \psi_{\sigma'}^0 |k|^2 + C_1}{\epsilon_k^0 |k|^6 + B_2 \psi_{\sigma'}^0 |k|^4 + C_2 \psi_{\sigma'}^0 |k|^2 + D_2} + \frac{\pi V_f^2}{D^2} \left| \frac{\omega}{\sqrt{4V_f^2 + t_{m}^2}} \right|^{2\omega - (\omega - i\eta)} - \frac{\omega - V_f^2/2}{\sqrt{4V_f^2 + t_{m}^2}} \right|^{2\omega - (\omega - i\eta)} \]

\[ \times \left[ \text{sgn} \left( \frac{dx_1}{d\omega} \right) \theta(0 < x_1 < D^2) \right] \]

\[ - \text{sgn} \left( \frac{dx_1}{d\omega} \right) \theta(0 < x_2 < D^2) \]

\[ - \frac{\omega - V_f^2/2}{2} \left[ \text{sgn} \left( \frac{dx_1}{d\omega} \right) \theta(0 < x_1 < D^2) \right] \]

\[ + \text{sgn} \left( \frac{dx_1}{d\omega} \right) \theta(0 < x_2 < D^2) \right], \]

where \( \theta(x) \) is the step function, and

\[ x_{1,2} = \left( \omega^2 + \frac{V_f^2}{4} \right) \pm \frac{1}{2} \sqrt{4(D^2 + t_{m}^2)\omega^2 - t_{m}^2 V_f^2}. \]

The summation over \( k \) in equation (16) is accurate for \( \omega \ll D \) by ensuring the conservation of the total number of states in the Brillouin zone according to the Debye prescription. Substituting \( \Sigma'(\omega) \) into equation (15), the retarded Green’s function \( G_f^{\sigma\sigma}(\omega) \) can be obtained. Note, the determination of \( \langle n_{\sigma} \rangle \) in equation (13) entails a self-consistent calculation of DOS at the impurity level via the relation \( \rho_f^{\sigma}(\omega) = \frac{-1}{\pi} \text{Im} G_f^{\sigma\sigma}(\omega) \). When \( t_{m} = V = 0 \), our present results reduce to those of [23].
behavior as a function of the parameters

The transition curves from the magnetic to the non-magnetic

Substituting equations (22) and (25) in equation (20), we can

From the occupation of the two spin channels on the impurity

4. Numerical analysis

From the occupation of the two spin channels on the impurity we conclude that a localized magnetic moment forms whenever

For a detailed study, conventionally one introduces the dimensionless parameters

\[
x = \frac{D^*}{U} \quad \text{and} \quad y = \frac{(\mu - \epsilon_0)}{U}
\]

with \( \Gamma = \pi V_f^2/D^2 \).

The transition curves from the magnetic to the non-magnetic behavior as a function of the parameters \( x \) and \( y \) for the different hybridization and inter-layer coupling in the bilayer graphene are shown in figure 2. For \( t_p = V = 0 \), our results reduce to those of [23]. The magnetic boundary exhibits an asymmetry around \( y = 0.5 \) and can even cross the line \( y = 1 \). The magnetic region shrinks in the \( x \) direction when the hybridization \( V_f \) is increased; for \( y \) close to 1 (cf equation (28)), the boundary line for magnetic transition shifts away from the \( y \) axis due to the increased influence of graphene on the impurity magnetization with enhanced hybridization. When the inter-layer coupling \( t_p \) is taken into account (see figure 2(b)), the size of the magnetic region diminishes rapidly, and for a large enough \( t_p \), the magnetic boundary shrinks above the line \( y = 0 \). However, the magnetic boundary does not turn symmetric around \( y = 0.5 \) and the above magnetic boundary line crosses the line \( y = 1 \). The origin of this phenomena lies in the peculiar nature of the quasiparticles in the bilayer graphene; they exhibit features akin both to Dirac and to conventional fermions. The contribution of conventional fermions originates from the inter-layer coupling that supports a metallic bilayer graphene and results in effects as for a conventional metallic host on the magnetic properties of the impurity. For large inter-layer coupling we observe, therefore, magnetic boundaries similar to those for an impurity in an ordinary metal. Figure 3 shows for a bilayer graphene the boundary between magnetic and non-magnetic impurity states as a function of the parameters \( x \) and \( y \) (equation (28)) for different impurity energy levels \( \epsilon_0 \). For \( V = 0 \) the size of the magnetic region grows as \( \epsilon_0 \) approaches the energy of the Dirac point. This behavior is reminiscent of the single layer of graphene [23], and originates

\[
\begin{align*}
+ \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_2)} \\
+ \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_2)} \\
- \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_1)} \\
\times \ln \left[ \frac{D^2 - x_3}{x_3} \right] - \frac{\pi V_f^2}{D^2} \left[ \text{sgn} \left( \frac{d x_1}{d \omega} \right) \theta(0 < x_1 < D^2) \right] \\
\times \left[ A_1 + \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_1)} \right] \\
\times \left[ A_1 + \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_1)} \right] \\
\times \left[ A_1 + \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_1)} \right] \\
= \left[ A_1 + \left[ A_1(x_2 + x_3) + B_1 \right] \frac{x_1 - A_1 x_2 x_3 + C_1}{(x_2 - x_1)(x_3 - x_1)} \right],
\end{align*}
\]

where

\[
\begin{align*}
x_1 &= -\frac{B_2}{3} + \frac{2 \sqrt{B_2^2 - 3C_2}}{3} \cos \left( \frac{\arccos T}{3} \right), \\
x_2 &= -\frac{B_2}{3} + \frac{2 \sqrt{B_2^2 - 3C_2}}{3} \cos \left( \frac{2\pi + \arccos T}{3} \right), \\
x_3 &= -\frac{B_2}{3} + \frac{2 \sqrt{B_2^2 - 3C_2}}{3} \cos \left( \frac{4\pi + \arccos T}{3} \right), \\
T &= -\frac{2(B_2^2 - 3C_2)B_2 - 3(B_2C_2 - 3D_2)}{2(B_2^2 - 3C_2)},
\end{align*}
\]

Substituting equations (22) and (25) in equation (20), we can derive self-consistently the occupation on the impurity for the case of a trilayer graphene.

\[
\text{Figure 2. Regions of the magnetic and the non-magnetic phase for the bilayer graphene. The boundary line gives } y \text{ as a function of } x \text{ (defined in equation (28)) at } t_p/D = 0 \text{ for the different } V_f/D \text{ (a) and at } V_f/D = 0.14 \text{ for different } t_p/D \text{ (b). The other parameters are } \epsilon_0/D = 0.029 \text{ and } V/D = 0.
\]
from the suppression of the DOS around the impurity energy level. In contrast, for a nonzero gate bias, when $\varepsilon_0$ is close to the Dirac point from the positive energy side, the size of the region first increases to a maximum, then decreases with decreasing $\varepsilon_0$, as shown in figure 3(b). The explanation for this phenomenon is as follows: the gate bias voltage gives rise to a finite electronic gap between the conduction and the valence band, and induces a large local DOS close to the gap edges [24]. In particular, the DOS may extend into the gap of Dirac fermions in the trilayer graphene. As $\varepsilon_0$ approaches the energy of the Dirac point, the magnetization region of the impurity grows due to the two almost-linear touching bands reminiscent of the bands in monolayer graphene [26]. It is interesting to note that for nonzero gate bias, the impurity magnetization region increases monotonously when $\varepsilon_0$ is close to the Dirac point, which is clearly different from that in the bilayer graphene. This behavior stems from the fact that the gate bias cannot destroy the particle–hole degeneracy in the trilayer graphene [25].

Figure 4 shows the magnetic transition curve as a function of the parameters $x$ and $y$ (equation (28)) for different $\varepsilon_0$ in the trilayer graphene. For $V_0 = 0$, phenomena such as the asymmetry around the line $y = 0.5$ and the crossing of the line $y = 1$ in the magnetic boundary suggest the existence of Dirac fermions in the trilayer graphene. As $\varepsilon_0$ approaches the energy of the Dirac point, the magnetization region of the impurity grows due to the two almost-linear touching bands reminiscent of the bands in monolayer graphene [26].

To investigate the localized magnetic moment of the impurity in the magnetic region and the magnetic transition we calculate the magnetic susceptibility. The energy of the impurity spin states in a magnetic field $B$ is $\varepsilon_{\sigma} = \varepsilon_0 - \sigma \mu_B B + U_{\sigma} n_{\sigma}$, where $U_{\sigma}$ is the energy of the Dirac point, the magnetization region of the impurity grows due to the two almost-linear touching bands reminiscent of the bands in monolayer graphene [26].

Figure 5 shows the occupation of the impurity spin level and the magnetic susceptibility as a function of $y$ for the different inter-layer coupling in a bilayer graphene. The occupation $\langle n_{\sigma} \rangle$ versus $y$ is a bubble that corresponds to the impurity magnetization. The corresponding susceptibility exhibits two peaks at the magnetization edge indicating the strength of the magnetic transition. For $t_p = 0$, a strong magnetic moment of $\sim 0.7 \mu_B$ forms in almost the whole magnetic region. With increasing the inter-layer coupling $t_p$, the magnetic bubble region diminishes, signaling the decrease of the magnetic moment of the impurity, and the magnetic transition becomes very sharp. There is no localized magnetic moment in the case of a sufficiently strong inter-layer coupling. In this case, the magnetic boundary shrinks below the line $x = 6$ in the $x$ direction (see figure 2(b)).

Figure 6 shows the occupation of the impurity level and the magnetic susceptibility as a function of $y$ for the different impurity energy level $\varepsilon_0$ in the bilayer graphene. The corresponding magnetic boundaries are defined in figures 3(a) and (b) respectively. For $V_0 = 0$, the magnetic bubble shifts towards the $\langle n_{\sigma} \rangle$ axis, and decreases with increasing $\varepsilon_0$. When $\varepsilon_0$ becomes large enough, the bubble...
vanishes, meaning that the impurity loses magnetism in this situation. For large $\varepsilon_0$ the magnetic transition becomes very sharp. Inspecting figures 6(c) and (d), we find that when the gate bias $V$ is applied the magnetic bubble shows a non-monotonic dependence on $\varepsilon_0$, while the magnetic transition becomes very sharp with increasing $\varepsilon_0$. Since the magnetic boundary line shrinks on the left-hand side of the line $x = 4.2$ at $\varepsilon_0/D = 0.082$ (see figure 3(b)), the impurity remains non-magnetic for any $y$, i.e. $n_\uparrow = n_\downarrow$, as shown in figure 6(c).

5. Conclusions

Summarizing, we studied the localized magnetic states of an impurity in biased bilayer and trilayer graphene. We found that the size of the magnetic region decreases rapidly compared with that of a monolayer graphene. The impurity can magnetize even when the energy of the doubly occupied state is below the Fermi level and the impurity magnetization region has a different shape. We can trace this behavior back to the special nature of quasiparticles. When a gate bias is applied, the dependence of the impurity magnetic states on the impurity energy for the bilayer graphene shows a behavior different from that for a trilayer graphene due to the opening of a gap between the valence and the conduction band in the bilayer graphene. Correspondingly, the magnetic moment of the impurity versus the impurity energy in the bilayer graphene is affected strongly by the band gap induced by the gate bias.

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