Controllability of ferromagnetism in graphene

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We systematically study magnetic correlations in graphene within Hubbard model on a honeycomb lattice by using quantum Monte Carlo simulations. In the filling region below the Van Hove singularity, the system shows a short-range ferromagnetic correlation, which is slightly strengthened by the on-site Coulomb interaction and markedly by the next-nearest-neighbor hopping integral. The ferromagnetic properties depend on the electron filling strongly, which may be manipulated by the electric gate. Due to its resultant controllability of ferromagnetism, graphene-based samples may facilitate the development of many applications.

The search for high temperature ferromagnetic semiconductors, which combine the properties of ferromagnetism (FM) and semiconductors and allow for practical applications of spintronics, has evolved into a broad field of materials science\textsuperscript{1,2}. Scientists require a material in which the generation, injection, and detection of spin-polarized electrons is accomplished without strong magnetic fields, with processes effective at or above room temperature\textsuperscript{3,4}. Although some of these requirements have been successfully demonstrated, most semiconductor-based spintronics devices are still at the proposal stage since useful ferromagnetic semiconductors have yet to be developed\textsuperscript{5}. Recently, scientists anticipate that graphene-based electronics may supplement silicon-based technology, which is nearing its limits\textsuperscript{5,6}. Unlike silicon, the single layer graphene is a zero-gap two-dimensional (2D) semiconductor, and the bilayer graphene provides the first semiconductor with a gap that can be tuned externally\textsuperscript{6}. Graphene exhibits gate-voltage controlled carrier conduction\textsuperscript{6,10}, high field-effect mobility, and a small spin-orbit coupling, making it a very promising candidate for spintronics application\textsuperscript{11,12}. In view of these characteristics, the study of the controllability of FM in graphene-based samples is of fundamental and technological importance, since it increases the possibility of using graphene in spintronics and other applications.

On the other hand, the existence of FM in graphene is an unresolved issue. Recent experimental and theoretical results in graphene\textsuperscript{13,14} show that the electron-electron interactions must be taken into account in order to obtain a fully consistent picture of graphene. The honeycomb structure of graphene exhibits Van Hove singularity (VHS) in the density of states (DOS), which may result in strong ferromagnetic fluctuations, as demonstrated by recent quantum Monte Carlo simulations of the Hubbard model on the square and triangular lattices\textsuperscript{15,16}. After taking both electron-electron interaction and lattice structure into consideration, the bidimensional Hubbard model on the honeycomb lattice\textsuperscript{18–20} is a good candidate to study magnetic behaviors in graphene. Early studies of the bidimensional Hubbard model on the honeycomb lattice were based on mean field approximations and the perturbation theory\textsuperscript{20}. However, the results obtained are still actively debated because they are very sensitive to the approximation used. Therefore, we use the determinant quantum Monte Carlo (DQMC) simulation technique\textsuperscript{21,22} to investigate the nature of magnetic correlation in the presence of moderate Coulomb interactions. We are particularly interested on ferromagnetic fluctuations as functions of the electron filling, because the application of local gate techniques enables us to modulate electron filling\textsuperscript{5,10}, which is the first step on the road towards graphene-based electronics.

The structure of graphene can be described in terms of two interpenetrating triangular sublattices, A and B, and its low energy magnetic properties can be well described by the Hubbard model on a honeycomb lattice\textsuperscript{18–20},

\[ H = -t \sum_{i\sigma} a_{i\sigma}^\dagger b_{i+\sigma} + t' \sum_{i\gamma\sigma} (a_{i\sigma}^\dagger a_{i+\gamma\sigma} + b_{i\sigma}^\dagger b_{i+\gamma\sigma}) + h.c. + U \sum_{i} (n_{a|i} n_{a|i} + n_{b|i} n_{b|i}) + \mu \sum_{i\sigma} (n_{a|i\sigma} + n_{b|i\sigma}) \]  (1)

where \( t \) and \( t' \) are the nearest and next-nearest-neighbor (NNN) hopping integrals respectively, \( \mu \) is the chemical potential, and \( U \) is the Hubbard interaction. Here, \( a_{i\sigma} \) (\( a_{i\sigma}^\dagger \)) annihilates (creates) electrons at site \( \mathbf{R}_i \) with spin \( \sigma \) (\( \sigma = \uparrow, \downarrow \)) on sublattice A, \( b_{i\sigma} \) (\( b_{i\sigma}^\dagger \)) annihilates (creates) electrons at the site \( \mathbf{R}_i \) with spin \( \sigma \) (\( \sigma = \uparrow, \downarrow \)) on sublattice B, \( n_{a|i\sigma} = a_{i\sigma}^\dagger a_{i\sigma} \) and \( n_{b|i\sigma} = b_{i\sigma}^\dagger b_{i\sigma} \).

Our main numerical calculations were performed on a double-48 sites lattice, as sketched in Fig.\textsuperscript{4} where blue circles and yellow circles indicates A and B sublattices, respectively. The structure of the honeycomb lattice leads to the well known massless-Dirac-fermion-like low energy excitations and the two VHS in the DOS

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that $t$ study the ferromagnetic fluctuations within the Hubbard DQMC simulation is a useful tool [22]. The exact value where the mean-field theory does not work well while the ferromagnetic correlation is strengthened, which may be manipulated by the electric gate. Furthermore, the ferromagnetic fluctuations within the Hubbard model on the honeycomb lattice by including $t'$. In the followings, we show that the behaviors of magnetic correlation are qualitatively different in two filling regions separated by the VHS at $<n>=0.75$. In the filling region below the VHS the system shows a short-ranged ferromagnetic correlation and the on-site Coulomb interaction tends to strengthen ferromagnetic fluctuation. The ferromagnetic properties depend on the electron filling, which may be manipulated by the electric gate. Furthermore, the ferromagnetic fluctuation is strengthened markedly as $t'$ increases. Our results highlight the crucial importance of electron filling and the NNN hopping in graphene. The resultant controllability of FM may facilitate the new development of spintronics and quantum modulation.

To study ferromagnetic fluctuations, we define the spin susceptibility in the $z$ direction at zero frequency,

$$\chi(q) = \int_0^\beta d\tau \sum_{\delta,\delta'} \sum_{i,j} e^{i\delta q (i \delta - j \delta')} \langle m_i \cdot m_j (0) \rangle$$

where $m_i(\tau) = e^{iHt_m}(m_i(0))e^{-iH\tau}$ with $m_i = a_i^\dagger a_i - \frac{1}{2} \delta^{i,j} a_i^\dagger a_i$, and $m_i = b_i^\dagger b_i - \frac{1}{2} \delta^{i,j} b_i^\dagger b_i$. Here $\chi$ is measured in unit of $|t|^{-1}$, and $\chi(\Gamma)$ measures ferromagnetic correlation while $\chi(K)$ measures antiferromagnetic correlation.

We first present temperature dependence of the magnetic correlations at $<n>=0.25$ with different $t'$ and $U$. Fig. 2 shows $1/\chi(\Gamma)$ versus temperature at $U=3|t|$, with $t'=t/10$, $t/6$, and $t/5$. Data for $U=5|t|$ as $t'=t/5$ are also shown. In the inset, we present $\chi(q)$ versus momentum $q$ at different temperatures with $t'=0.10|t|$ and $U=3|t|$. Further, we observe that $\chi(\Gamma)$ grows much slower than $\chi(\Gamma)$ with decreasing temperatures. Moreover, $1/\chi(\Gamma)$ exhibits Curie-like behavior as temperature decreases from $|t|$ to about 0.1. Fitting the data as $1/\chi(\Gamma)=\alpha(T-\Theta)$ (solid lines in Fig. 2) shows that $\Theta$ is about 0.02|t|≈580 K at $t'=t/5$, and we also note that both $\Theta$ and $\chi(\Gamma)$ are enhanced slightly as the on-site Coulomb interaction is increased. Positive values of $\Theta$ indicate that the curves of $1/\chi(\Gamma)$ start to bend at some low temperatures and probably converge to zero as $T \to 0$, i.e., $\chi(\Gamma)$ diverges. This demonstrates the existence of ferromagnetic state in graphene.

From Fig. 2 we may also notice that $t'$ plays a remarkable effect on the behavior of $\chi(q)$, and results for

![FIG. 1: (Color online) (a) Sketch of graphene with double-48 sites; (b) First Brillouin zone and the high symmetry direction (red line); (c) DOS (solid dark lines) and fillings $\langle n \rangle$ (dash red lines) as functions of energy with $t'=0.10|t|$; and (d) $t'=0.20|t|.$](image)

![FIG. 2: (Color online) At $<n>=0.25$, inverse of magnetic susceptibility, $1/\chi(\Gamma)$ versus temperature with $U=3|t|$, $t'=t/10$, $t/6$, and $t/5$. Fitted line $1/\chi(\Gamma)=\alpha(T-\Theta)$ are also shown. Inset: Magnetic susceptibility $\chi(q)$ versus $q$ at different temperatures with $t'=0.10|t|$ and $U=3|t|.$](image)

![FIG. 3: (Color online) Magnetic susceptibility $\chi(q)$ versus momentum $q$ at different $t'$, here $U=3|t|$, $<n>=0.25$ and $T=0.10|t|.$](image)
The magnetic correlation is strong around the hall-filling and antiferromagnetic fluctuations. The antiferromagnetic correlation showed strong dependence on the electron filling and the NNN hoping integral. This provides a route to manipulate FM in ferromagnetic graphene-based samples by the electric gate or varying lattice parameters. The resultant controllability of FM in ferromagnetic graphene-based samples may facilitate the development of many applications.

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