Weak ferromagnetism and spiral spin structures in honeycomb Hubbard planes

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Abstract
Within the Hartree Fock RPA analysis, we derive the spin wave spectrum for the weak ferromagnetic phase of the Hubbard model on a honeycomb lattice. Assuming a uniform magnetization, the polar (optical) and acoustic branches of the spin wave excitations are determined. The bipartite lattice geometry produces a \(q\)-dependent phase difference between the spin wave amplitudes on the two sub-lattices. We also find an instability of the uniform weakly magnetized configuration towards a weak antiferromagnetic spiraling spin structure, in the lattice plane, with wavevector \(Q\) along the \(\Gamma-K\) direction, for electron densities \(n > 0.6\). We discuss the effect of diagonal disorder on both the creation of electron bound states and the enhancement of the density of states, and the possible relevance of these effects to disorder-induced ferromagnetism, as observed in proton-irradiated graphite.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
Recent interest in strongly correlated systems in non-square lattices, such as the triangular, honeycomb and kagomé lattices, is justified by the possible realization of exotic metallic [1, 2], magnetic [3, 4] and superconducting states [5] in both inorganic and organic materials. On the organic side, graphite and related carbon allotropes are physical systems where growing evidence for exotic types of ground states has been accumulated over the last few years. In graphite, for example, experimental research put forward evidence for unusual metallic and magnetic properties [2, 6, 7, 4]. In particular, ferromagnetism has been observed at high temperature in graphite [6] which may not be due to magnetic impurities. Also, the observation [2] of magnetic order induced by proton irradiation challenges the theoretical description. Graphite is not alone on the ferromagnetic-order-by-disorder scenario, with
Figure 1. Primitive vectors ($a_1$ and $a_2$) for the honeycomb lattice. The vectors $\delta_1$, $\delta_2$ and $\delta_3$ connect the A site to its three neighbouring B sites. The hexagonal first Brillouin zone corresponds to the reciprocal lattice vectors, $b_1$ and $b_2$.

The inorganic CaRuO$_3$ material also exhibiting disorder-induced ferromagnetism [8]. Recent experimental work [9–11] has produced atomic thin graphite planes where the exciting physics of 2D Dirac fermions may be directly observable. Motivated by these experimental studies and because the microscopic origin of ferromagnetism in these compounds is far from being understood, we decided to study the magnetic properties of a doped Hubbard model on a honeycomb lattice—a single graphite plane. To the best of our knowledge, ferromagnetic spin waves in the honeycomb lattice (as an itinerant electron system) have not been studied in the past. The fact that the honeycomb lattice is a Bravais lattice with a basis immediately presents us with the possibility of observing both polar and acoustic spin waves [12, 13]. We focus our research on the stability of the weak homogeneous ferromagnetic phase found in [14]. The paper is organized as follows: the model is introduced in section 2 and the energy spectrum for the weak homogeneous ferromagnetic system is derived; section 3 is devoted to the description of spin waves in the weak homogeneous ferromagnetic phase; the possibility of spiral spin states is investigated in section 4, where a spiral arrangement is found with lower energy than that of a uniform magnetization; section 5 gives a discussion on the possibility of formation of electronic bound states, due to impurities, and of the possible relevance of disorder to experiments on proton-irradiated graphite.

2. Model Hamiltonian

The Hubbard model is defined as

$$H = - \sum_{i,j,\sigma} (t_{i,j} + \mu \delta_{ij}) c_{i,\sigma}^\dagger c_{j,\sigma} + U \sum_{i} c_{i,\uparrow}^\dagger c_{i,\uparrow} c_{i,\downarrow}^\dagger c_{i,\downarrow},$$

where $c_{i,\sigma}^\dagger$ ($c_{i,\sigma}$) represents a creation (destruction) electron operator with spin $\sigma$ at site $i$, $t_{i,j}$ is the hopping integral between two sites $i$ and $j$, $U$ is the on-site Coulomb repulsion, and $\mu$ is the chemical potential. In the honeycomb lattice we identify two sub-lattices, A and B (see figure 1), where the primitive vectors of the underlying triangular lattice are denoted by $a_1$ and $a_2$. For later use we also define the vector $a_3 = (a_1 - a_2)$. The reciprocal lattice vectors are $b_1$ and $b_2$ and they define a hexagonally shaped first Brillouin zone. Also shown in figure 1 are the vectors connecting any A atom to its nearest neighbours, denoted as $\delta_1$, $\delta_2$ and $\delta_3$. The electrons living on each sub-lattice will be denoted by field operators $a$ and $b$, respectively. The Fourier transformation between real and momentum spaces is given by

$$a_{i,\sigma}^\dagger = \frac{1}{\sqrt{N_A}} \sum_k e^{i k \cdot R_i} a_{k,\sigma}^\dagger, \quad b_{i,\sigma}^\dagger = \frac{1}{\sqrt{N_B}} \sum_k e^{i k \cdot R_i} b_{k,\sigma}^\dagger,$$
and we take $N_A = N_B = N$ as the number of unit cells. In the calculations below, we shall consider first and second neighbour hopping integrals, $t$ and $t'$, respectively. The Hubbard model then takes the form

$$H = \sum_{k,\sigma} \left( D(k) - \mu \right)(a_{k,\sigma}^\dagger a_{k,\sigma} + b_{k,\sigma}^\dagger b_{k,\sigma}) + \sum_{k,\sigma} \left[ \phi(k)a_{k,\sigma}^\dagger b_{k,\sigma} + \phi^*(k)b_{k,\sigma}^\dagger a_{k,\sigma} \right] + H_U, \quad (3)$$

with

$$D(k) = -2t' \sum_{i=1}^{3} \cos(\alpha_i \cdot k),$$

$$\phi(k) = -t \sum_{i=1}^{3} e^{i\delta \cdot \delta}.$$  

(4)

In the ferromagnetic ground state, the average occupancy of lattice sites is given by

$$\langle a_{i,\sigma}^\dagger a_{i,\sigma} \rangle = \frac{n}{2} + \sigma \frac{m}{2}, \quad \langle b_{i,\sigma}^\dagger b_{i,\sigma} \rangle = \frac{n}{2} + \sigma \frac{m}{2},$$

(5)

with the spin index $\sigma = \pm 1$. This may also be generalized to describe antiferromagnetic ordering if we replace $m$ with $-m$ in one of the equations (5) [14]. A Hartree–Fock treatment of the Hubbard term, $H_U$, taking into account equation (5), yields a set of quasi-particle bands given by

$$E_{\sigma}^{\alpha}(k) = D(k) + \frac{U}{2} (n - \sigma m) + \alpha |\phi_k|, \quad (6)$$

where $\alpha = \pm$ is a band index. In the ferromagnetic phase the single particle Green’s functions can be written, in momentum space, as

$$G_{\sigma}^{aa}(i\omega_n, k) = \sum_{j=\pm} \frac{1/2}{i\omega_n - E_{\sigma}^{j}(k)}$$

$$G_{\sigma}^{ab}(i\omega_n, k) = \sum_{j=\pm} \frac{j e^{i\delta \cdot k}/2}{i\omega_n - E_{\sigma}^{j}(k)}$$

$$G_{\sigma}^{ba}(i\omega_n, k) = \sum_{j=\pm} \frac{j e^{-i\delta \cdot k}/2}{i\omega_n - E_{\sigma}^{j}(k)}$$

$$G_{\sigma}^{bb}(i\omega_n, k) = G_{\sigma}^{aa}(i\omega_n, k), \quad (10)$$

where we have defined $e^{i\delta \cdot k} = \phi(k)/|\phi(k)|$. The Hartree–Fock magnetization is given by

$$m = (1/2N) \sum_{k,\sigma,\sigma'} \sigma f[E_{\sigma}^{\alpha}(k)].$$

Since we are mainly concerned with the ferromagnetic phase, the calculations in sections 3 and 4 are performed for an electronic density smaller than half filling. Therefore, electrons at the Fermi level will not be treated as massless Dirac fermions. Such treatment is usually appropriate for electrons in graphite planes at half filling (or close to half filling) [15].

3. Magnetic collective excitations

We obtain the magnetic collective excitations from the poles of the transverse spin susceptibility calculated in the RPA approximation. Because there are two sub-lattices, the susceptibility is actually a second-order tensor given by the expression

$$\chi_{+/-}^{ij}(q, i\omega_n) = \int_0^{1/T} dr e^{i\omega_n r} \langle T_S^+(q, r) S_j^-(-q, 0) \rangle$$

(11)
magnetic excitation, namely the usual acoustic mode $\omega_{\Gamma_1}$ along the $-X$ direction (see figure 2), for example, and for $|q| > 0.25\Gamma X$ only the optical branch remains. On the other hand, in the $\Gamma - K$ direction (see figure 3) it is the optical branch that vanishes at $|q| \approx 0.2\Gamma X$ while the acoustic mode survives. The vanishing frequency of the acoustic (or optical) modes at finite momentum is associated with an instability of the homogeneous weak ferromagnetic phase towards a state exhibiting possibly weak ferromagnetic order in the $z$ direction and spiral order in the $xy$ plane, which will be analysed in section 4 below.
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Figure 2. Left panel: spin waves (line with circles for $\omega_{opt}$ and continuous line for $\omega_{ac}$) and Stoner continuum border (thin dashed–dotted line) along the $\Gamma$–X direction of the Brillouin zone. Right: band energies along the $\Gamma$–X direction (the band energies are measured relatively to the chemical potential). The parameters are $U = 4$, $t = 1$, $t' = -0.2$, $n = 0.75$, and the magnetization and chemical potential are $m = 0.25$ and $\mu = 0.36$. The energy through all this paper is in units of $t$. (Subscripts $u$ and $d$ in the right panel denote spin projections $\uparrow$ and $\downarrow$, respectively.)

Figure 3. Left panel: spin waves (line with circles for $\omega_{opt}$ and continuous line for $\omega_{ac}$) and Stoner continuum border (thin dashed–dotted line) along the $\Gamma$–K direction of the Brillouin zone. Right panel: quasi-particle band structure along the $\Gamma$–K direction (band energies measured relative to chemical potential). The parameters are the same as in figure 2. (Subscripts $u$ and $d$ in the right panel denote spin projections $\uparrow$ and $\downarrow$, respectively.)

The eigenvector of the matrix $\chi^{(0)}(q,\omega(q))$ that is associated with the eigenvalue $N/U$ gives the spin wave amplitudes over the A and B sub-lattices. We note that equation (13) is
equivalent to the eigenproblem

\[
\frac{1}{N} K^0(q, \omega + i0^+) \begin{bmatrix} \langle S_A^+ \rangle \\ \langle S_B^+ \rangle \end{bmatrix} = \frac{1}{U} \begin{bmatrix} \langle S_A^+ \rangle \\ \langle S_B^+ \rangle \end{bmatrix},
\]

where \( \langle S_A^+ \rangle, \langle S_B^+ \rangle \) denote the spin wave amplitudes over the two sub-lattices [16]. In our case, \( \chi^{(0),aa} = \chi^{(0),bb} \) and \( \chi^{(0)ab} = (\chi^{(0)ba})^* \). The equations for the eigenvalue \( \lambda \) and the corresponding eigenvector are

\[
\lambda = \chi^{(0),aa} \pm |\chi^{(0)ab}| \tag{18}
\]

\[
\langle S_A^+ \rangle = \pm \frac{\chi^{(0)ab}}{|\chi^{(0)ab}|} \langle S_B^+ \rangle \tag{19}
\]

where \( \lambda = N/U \) is the relevant eigenvalue, as can be seen from equation (17). Equation (19) shows that the spin wave amplitudes are related by a phase factor. Therefore, the phase of the complex matrix element \( \chi^{(0)ab} \) determines the angle between the transverse spin components \( \langle S_A^+ \rangle \) and \( \langle S_B^+ \rangle \). The optical mode in the \( \Gamma-X \) direction (shown in figure 2) starts off with \( \langle S_A^+ \rangle = -\langle S_B^+ \rangle \) for small \( q \), as expected of an optical mode, but the angle between \( \langle S_A^+ \rangle \) and \( \langle S_B^+ \rangle \) decreases monotonically from \( \pi \), upon increasing wavevector, and equals \( \pi/2 \) when \( \omega_{\text{opt}} \) attains its minimum. At that point, \( \chi^{(0)ab} \) is pure imaginary. On the other hand, in the acoustic mode the angle increases from zero to \( \pi/2 \), when \( \omega_{\text{ac}} \) is maximum (the angle between the spins is illustrated in figure 4). As the wavevector further increases, the acoustic mode frequency rapidly decreases and vanishes at \( q \approx 0.25\Gamma X \).

Considering now the spin waves in the \( \Gamma-K \) direction, the angle between the precessing spins is always zero or \( \pi \) in the acoustic and optical modes, respectively. The optical mode
frequency vanishes shortly after the interception with the acoustic branch and only the latter survives for increasing $q$.

The phase difference between spin wave amplitudes just described for the $\Gamma$–$X$ direction is a manifestation of the complex coherence factors appearing in the single-particle Green functions (8) and (9), resulting from the honeycomb lattice geometry.

4. Spiral spin states

The disappearance of the acoustic mode, at wavevector $q \approx 0.25\Gamma X$, and of the optical mode, at wavevector $q \approx 0.20\Gamma X$, suggests an instability to a spiral spin state [17]. In such a state, the spiral spin configuration is characterized by non-zero transverse magnetization at site $j$,

$$\langle S_j^z \rangle = \langle S_j^+ \rangle e^{iQ \cdot R_j},$$

in addition to a uniform alignment in the $z$ direction, $\langle S_j^z \rangle$. The amplitudes of the spiral, $\langle S_{\alpha(B)}^+ \rangle$, at sub-lattices A and B are given by

$$\langle S_A^+ \rangle = \frac{1}{N_e} \sum_k \langle a_{k,\uparrow}^\dagger a_{k+Q,\downarrow} \rangle$$

$$\langle S_B^+ \rangle = \frac{1}{N_e} \sum_k \langle b_{k,\uparrow}^\dagger b_{k+Q,\downarrow} \rangle. \quad (20)$$

In general, there will be a nonzero angle $\theta$ between the transverse sub-lattice magnetizations $\langle S_A^+ \rangle$ and $\langle S_B^+ \rangle$, so that $\langle S_0^+ \rangle = e^{i\theta} \langle S_A^+ \rangle$.

Following [17], the mean field equations are obtained from the minimization of the ground state energy with respect to the order parameters $\langle S_{\alpha(B)}^+ \rangle$ and $\langle S_j^z \rangle$. Each Bloch $k$-state, $\gamma_k$, representing an elementary excitation, is a linear superposition of the fields $a_{k,\uparrow}$, $a_{k+Q,\downarrow}$, $b_{k,\uparrow}$ and $b_{k+Q,\downarrow}$. Conversely, we can rewrite each of the fields as a combination of Bloch states, and recast the expectation value of the kinetic term in (3) as well as the order parameters (20) in terms of $\gamma_k$ operators. Using Wick’s theorem, the expectation value of the Hubbard term in the Hamiltonian (3) can be expressed as

$$\langle H_U \rangle = U \left( \frac{n^2}{2} - \langle S_A^+ \rangle^2 - \langle S_B^+ \rangle^2 - \langle S_A^+ \rangle \langle S_B^+ \rangle - \langle S_B^+ \rangle \langle S_A^+ \rangle \right). \quad (21)$$

By minimizing $\langle H \rangle$, as given in equations (3) and (21), we find that the Bloch $k$-state, $\gamma_k$, diagonalizes an effective $4 \times 4$ Hamiltonian matrix, $H_{\text{eff}}(k)$, which can be expressed in the basis $\langle a_{k,\uparrow}, a_{k+Q,\downarrow}, b_{k,\uparrow}, b_{k+Q,\downarrow} \rangle$, as

$$H_{\text{eff}}(k) = \begin{bmatrix} D_A(k) & C^+(k) \\ C(k) & D_B(k) \end{bmatrix} \quad (22)$$

where the $2 \times 2$ matrices $D_\alpha$ (with $\alpha = A, B$) and $C$ are given by

$$D_\alpha(k) = \begin{bmatrix} D(k) - U \langle S_A^+ \rangle - \mu & -U \langle S_A^+ \rangle \\ -U \langle S_A^+ \rangle & D(k + Q) + U \langle S_A^+ \rangle - \mu \end{bmatrix}, \quad (23)$$

and

$$C(k) = \begin{bmatrix} \phi(k) & 0 \\ 0 & \phi(k + Q) \end{bmatrix}. \quad (24)$$

The mean field equations only determine the phase difference between $\langle S_A^+ \rangle$ and $\langle S_B^+ \rangle$, so we choose $\langle S_A^+ \rangle$ to be real. At each point in the weak ferromagnetic region of the phase diagram we have to choose the spiral wavevector $Q$ that minimizes the ground state energy. This vector should lie along one of the high symmetry directions in the Brillouin zone.

We first look for the most favourable wavevectors lying along the $\Gamma$–$X$ direction. For the same parameters as in figure 2, we find a spiral state with $Q = \frac{1}{2}\Gamma X$. $\langle S_A^+ \rangle = 0.12$
and \( \langle S_x^+ \rangle = 0.036 \). We note that this spiraling state has a smaller \( z \)-component of the magnetization than that in the uniform phase. The angle between transverse magnetizations \( \theta = 0.77\pi \approx 3\pi/4 \). We find, however, that the energy difference between this spiral state and that with uniform magnetization is indeed very small, not exceeding \( 10^{-4}t \) per lattice site. This has been checked for several lattice sizes. We also note that the obtained spin transverse component is not negligible compared to \( \langle S_z \rangle \). The favourable spiral wavevector depends slightly on interaction and density: at \( U = 3.5 \) and \( n = 0.8 \), for instance, \( Q = \frac{19}{80}\Gamma X \) is the most favourable, with \( \langle S_z \rangle = 0.089, \langle S_x^+ \rangle = 0.046 \) and \( \theta = 0.73\pi \).

We now consider states with \( Q \propto \Gamma K \). Overall, the energies of these spiral states are found to be lower than those of the states with \( Q \propto \Gamma X \) considered above. For the same parameters as in figure 2, we find the optimum wavevector to be \( Q = \frac{1}{2}\Gamma K \), where \( \langle S_z \rangle = 0.24 = -\langle S_a^+ \rangle \). The spin configuration is, therefore, planar and the two sub-lattices have opposite magnetizations. The energy (per lattice site) is 0.02 lower than that with uniform magnetization. An approximate representation of this state is shown in figure 5. The length of the most energetically favourable \( Q \) depends significantly on \( U \) and \( n \). If, for instance, \( U = 3.5 \) and \( n = 0.8 \), then \( Q = \frac{1}{2}\Gamma K \) is the most favourable, with \( \langle S_x^+ \rangle = 0.18 = -\langle S_a^+ \rangle \). The energy per lattice site of this state is 0.008 lower than that of the uniform state. In such a planar spin configuration, a small ferromagnetic alignment along \( z \) could still arise in the presence of a weak anisotropy or external magnetic field.

An anisotropic perturbation producing an easy axis (or a small magnetic field) need not exceed an energy of the order \( 10^{-2}t \) per lattice site in order to induce the uniform state, especially at the lower \( U \) values considered. It is still possible that the minimum energy spin structure of the system could be a superposition of spirals with different \( Q \) vectors, as has been found for the \( n = 1 \) Hubbard triangular lattice [18]. The search for such structures, as well as their sensitivity to disorder, is beyond the scope of this work, however. We also find that the spiral states are absent at smaller densities (\( n < 0.6 \)). A schematic representation of our findings is shown in figure 6.

5. Disorder as possible mechanism to ferromagnetism

Ferromagnetic order induced by disorder in proton-irradiated graphite was observed by Kopelovich et al [2]. The disorder induced by proton irradiation can be modelled by diagonal disorder, where the local energy of some sites is modified. The problem of treating disorder and Coulomb interaction together is a hard one in condensed matter physics [19]. Therefore
we start by studying the effect of a finite density of uncorrelated impurities on the electron gas in the honeycomb lattice at half filling, where the low-energy electronic excitations can be described by massless Dirac fermions [15]. The effect of disorder on the properties of Dirac fermions leads to some unexpected results, and was discussed in the context of disordered superconductors by some authors [20, 21]. In our study, we compute the $T$-matrix using the massless Dirac fermion description.

The Matsubara Green’s functions are determined via the equation-of-motion method. After the usual manipulations [22] we obtain the $2 \times 2$ Green’s function matrix as

$$G(p, i\omega_n) = G^0(p, i\omega_n) + G^0(p, i\omega_n)T(i\omega_n)G^0(p, i\omega_n),$$  \hspace{1cm} (25)$$

with the Matsubara $T$-matrix given by

$$T(i\omega_n) = \frac{V}{N}\left[1 - \frac{V}{N}G^0(i\omega_n)\right]^{-1},$$ \hspace{1cm} (26)$$

where $G^0(p, i\omega_n)$ is the Green’s function matrix with $V = 0$, and

$$G^0(i\omega_n) = \sum_p G^0(p, i\omega_n).$$ \hspace{1cm} (27)$$

For a small density, $n_{\text{imp}}$, of scatterers (but finite in the thermodynamic limit), and after the position of the scatterers has been averaged over ensemble configurations, the Green’s function matrix can be written as

$$G(p, i\omega_n) = \left[[G^0(p, i\omega_n)]^{-1} - \Sigma(i\omega_n)\right]^{-1},$$ \hspace{1cm} (28)$$

where [22]

$$\Sigma(i\omega_n) = Vn_{\text{imp}}\left[1 - \frac{V}{N}G^0(i\omega_n)\right]^{-1}. \hspace{1cm} (29)$$

Once the Green’s function (28) is known, one can proceed to include the effect of correlations into the problem. The electronic bound states are given by the poles of (26). There always is a bound state due to the impurity, below the energy band, independently of the value of $V$. 

Figure 6. Schematic phase diagram of a honeycomb layer, with $t' \neq 0$, showing the paramagnetic (P), antiferromagnetic (AF), weak uniform ferromagnetic (WF) and spiral phases. The Nagaoka phase is a fully polarized ferromagnet. The exact position of the transition lines is not given, except at some special values marked in the axes. The results for $t' = 0$ are not qualitatively different.
The existence of bound states allows for a possible mechanism to the disorder-induced ferromagnetic behaviour in proton-irradiated graphite. Graphite is usually modelled as a half-filled honeycomb plane, in which electrons near the Fermi level have linear dispersion [15]. The sample irradiation produces the displacements of the carbon atoms from their original positions. In this case, even if hydrogen atoms become bonded to some of the carbons, from the lattice point of view a dilution of lattice points is being induced. In this case, we should take the limit $V \to \infty$, even if $n_{\text{imp}}$ is small. This effect leads to a drastic change in the density of states $\rho(\omega)$, where a strong enhancement of $\rho(\omega)$ in the vicinity of $\omega = 0$ is obtained. Such an enhancement can be responsible for a large reduction of the critical $U$ needed for ferromagnetism, as follows from the Stoner criterion. The effect of disorder on the density of states of Dirac fermions is shown in figure 7 for several values of $n_{\text{imp}}$ and $V$. If $V$ is negative there are bound states for the electrons below the bottom of the band. As $V$ increases an enhancement of $\rho(\omega)$ in the vicinity of $\omega = 0$ starts to develop. We have, therefore, two possible routes toward the appearance of magnetic order in graphite, namely bound states and enhancement of the density of states. This type of enhancement of $\rho(\omega)$ is characteristic of acoustic excitations, either fermionic (Dirac fermions) or bosonic (magnons) [23]. This very qualitative view has to be corroborated by more detailed calculations taking into account both disorder and Coulomb interaction. In particular, it is important to compute how the critical lines from the paramagnetic to the magnetic phases change with the amount of disorder. These issues will be the subject of a future publication.

6. Conclusion

In conclusion, we have studied the spin collective excitations of the homogeneous weak ferromagnetic state in the honeycomb lattice and found an instability to spiral spin structures at electron densities above $n \approx 0.6$. Although our calculation is performed for a system with the same type of atoms it is simple to generalize it for honeycomb lattices with different type of atoms as in boron–nitrogen–carbon hexagonal sheets [24]. However, the main differences
will be: (i) the number of optical and acoustic branches increases; (ii) the different site energies due to different atoms will change the form of the spin wave bands. We have also suggested a possible mechanism for ferromagnetism in irradiated graphite: the appearance of bound states due to disorder and the enhancement of the density of states. If it becomes possible in the future to perform neutron scattering experiments on ferromagnetic graphite allotrope the results we present here may have direct experimental importance.

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