Collective excitations and the nature of Mott transition in undoped gapped graphene

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Abstract

The particle–hole continuum (PHC) for massive Dirac fermions provides an unprecedented opportunity for the formation of two collective split-off states, one in the singlet and the other in the triplet (spin-1) channel, when the short-range interactions are added to the undoped system. Both states are close in energy and are separated from the continuum of free particle–hole excitations by an energy scale of the order of the gap parameter $\Delta$. They both disperse linearly with two different velocities, reminiscent of spin–charge separation in Luttinger liquids. When the strength of Hubbard interactions is stronger than a critical value, the velocity of singlet excitation, which we interpret as a charge composite boson, becomes zero and renders the system a Mott insulator. Beyond this critical point the low-energy sector is left with a linearly dispersing triplet mode—a characteristic of a Mott insulator. The velocity of the triplet mode at the Mott criticality is twice the velocity of the underlying Dirac fermions. The phase transition line in the space of $U$ and $\Delta$ is in qualitative agreement with our previous dynamical mean field theory calculations.

(Some figures may appear in colour only in the online journal)

1. Introduction

Graphene is a huge network of sp$^2$ bonds formed between carbon atoms on a two-dimensional (2D) sheet [1]. The low-energy effective model for 2p$_z$ electrons in this system is the 2 + 1-dimensional Dirac theory which contains an energy scale proportional to $v_F$—the Fermi velocity of electrons [2]. Effects of interactions and/or disorder can be taken into account on top of this non-interacting fixed point [2]. Breaking the sub-lattice symmetry gives rise to a gap in the single-particle excitations spectrum [3], which introduces the gap parameter $\Delta$ as another energy scale. The gapped graphene can be modeled with 2 + 1-dimensional massive Dirac fermions [4]. Gapped graphene supports an interesting new class of excitations in the spectrum, such as domain walls [5]. Moreover, the single-particle gap (mass) gives rise to suppression of Coulomb fields at nanometer length scales [6]. The gap could also give rise to a universal nonlinear optical response [7]. When the many-body interactions of various form are added to the massive Dirac theory, the nature of many-body excitations becomes even more interesting. Recent ab initio estimates of the strength of the Coulomb repulsion in various forms of graphene suggests that the Hubbard parameter in graphene can be quite remarkable [8], which introduces a third energy scale, $U$. Therefore, an interesting question here would be the effects of local Hubbard-type interactions on the excitation spectrum of gapped graphene and the nature of transition to the Mott insulating phase in gapped graphene. Our recent full-fledged dynamical mean field theory (DMFT) investigation of the so called ionic-Hubbard model on the honeycomb lattice suggested three phases [9]: (i) a band insulating phase for $\Delta \gg U$, (ii) a Mott insulating phase for $\Delta \ll U$, and (iii) a semi-metallic phase for $U \sim \Delta$. The above study addresses...
the nature of the ground state. In order to study the connection of excited states and the ground state one notes that, deep in the Mott insulating phase, the energy scale $U$ is expected to be dominant over $\Delta$ such that the low-energy excitations are expected to be spin fluctuations arising from super-exchange interaction induced by the large $U$. We would like to focus on the evolution of the collective spin and charge dynamics in this system as a function of the Hubbard $U$ for a system with a non-zero gap parameter $\Delta$. It turns out that the presence of a non-zero gap parameter facilitates the separation of two collective states, each of which has a different velocity, reminiscent of the spin–charge separation in Luttinger liquids.

2. Formulation of the problem

The Hamiltonian we consider here is the so called ionic-Hubbard model, which is defined by

$$
H = \sum_{\mathbf{k}s} \psi_{\mathbf{k}s}^\dagger [\mathbf{a} \cdot \hbar \mathbf{v}_F \mathbf{k} + \sigma_z \Delta] \psi_{\mathbf{k}s} + U \sum_{\mathbf{q}s} (n_{\mathbf{k}s}^{f} n_{\mathbf{k}s}^{\dagger} + n_{\mathbf{k}s}^{b} n_{\mathbf{k}s}^{\dagger}),
$$

where $n_{\mathbf{k}s}^{f} = f_{\mathbf{k}s} f_{\mathbf{k}s}$, with $f = a, b$ corresponding to number operator at the $j$th unit cell at sub-lattices A, B, respectively. The spinor notation $\psi_{\mathbf{k}s}^\dagger = (a_{\mathbf{k}s}^{\dagger} b_{\mathbf{k}s}^{\dagger})$ has been used. Here $s = \uparrow, \downarrow$ denotes the $z$ component of the physical spin, $\mathbf{k} = (k_x, k_y)$ is a two-dimensional momentum vector and $\mathbf{a}$ stands for Pauli matrices. $U$ is the strength of the on-site Coulomb interaction, known as Hubbard parameter, and $\Delta$ is the mass parameter. The quadratic part of this Hamiltonian can be diagonalized by a simple unitary transformation to the basis of conduction ($+$) and valence ($-$) states and the corresponding eigenvalues are given by

$$
e^\pm(\mathbf{k}) = \pm \sqrt{\hbar v_F k^2 + \Delta^2}, \quad \Delta \neq 0,
$$

where $\hbar v_F = \sqrt{3}a/2$ and $a$ is the C–C bond length. The hopping amplitude between the nearest neighboring carbon atoms is $t \sim 2.8$ eV. Due to the paramagnetic nature of the non-interacting system (i.e. $U = 0$), the occupation numbers and energies of the conduction and valence bands are independent of spin orientation. We consider the undoped system with precisely one $2p_e$ electron per carbon atom and assume the temperature to be zero. Then we extend the collective mode analysis of [11] to the case with $\Delta \neq 0$, where the single-particle spectrum is given by equation (2). This change in the single-particle spectrum, changes the borders of the particle–hole continuum. However as will be show here, at the border corresponding to massless spectrum, interesting collective quanta can be formed when short-range Coulomb interactions are turned on.

Using the equation of motion (EOM) for a peculiar type of inter-band fluctuations and employing the Hartree–Fock factorization in the higher rank correlation functions generated by the EOM [11], one finds that the eigenvalue equation for the collective excitations in the triplet and singlet channels for the Dirac fermions in the presence of the short-range Coulomb interactions are given by

$$1 \pm U \chi^0(\mathbf{q}, \omega) = 0,
$$

where the $-$ ($+$) sign corresponds to triplet (singlet) channel [12] and the sign difference can be traced back to the fermion anti-commutation relation. The approximations employed in obtaining the above equation are: (i) the Hartree–Fock factorization of the EOM and (ii) among the terms generated by the short range interactions, only processes that involve two valence band operators and two conduction band operators are retained [11]. Note that, although $\chi^0$ in the above expression is the particle–hole (polarization) bubble, the (approximate) bosonic operators which satisfy the above equation are of a peculiar form which is different from the usual particle–hole fluctuation [11]. The non-interacting susceptibility $\chi^0$ employed in the above equation is defined by

$$
\chi^0(\mathbf{q}, \omega) = \frac{1}{N} \sum_{\mathbf{k}} \frac{\bar{n}_{\mathbf{k}+\mathbf{q}} - \bar{n}_{\mathbf{k}}}{\omega + i\eta - (\varepsilon_{\mathbf{k}+\mathbf{q}} - \varepsilon_{\mathbf{k}})}.
$$

For the dispersion relation equation (2), the susceptibility has been calculated by many authors, the analytic form of which reads [13, 14]:

$$
\chi^{(0)}(\mathbf{q}, \omega) = \frac{-|\mathbf{q}|^2}{\pi \hbar (v_F^2 |\mathbf{q}|^2 - \omega^2)} \times \left\{ \frac{2\Delta}{\hbar} + \frac{\varepsilon_{\mathbf{k}}^2 |\mathbf{q}|^2 - \omega^2 - 4\Delta^2 / \hbar^2}{\sqrt{\varepsilon_{\mathbf{k}}^2 |\mathbf{q}|^2 - \omega^2}} \right\} \times \arctan \left( \frac{\sqrt{\varepsilon_{\mathbf{k}}^2 |\mathbf{q}|^2 - \omega^2}}{2\Delta / \hbar} \right).
$$

To understand the structure of this function, in figure 1 we have plotted the real and imaginary part of this function versus $\omega$ for some representative value of $\mathbf{q}$. As can be seen,
the imaginary part is non-zero for $\omega > \omega_\Delta(q)$, where

$$\omega_\Delta(q) = \sqrt{v_F^2|q|^2 + 4\Delta^2/h^2}$$

(6)
corresponds to the lower boundary of the PHC of massive Dirac fermions. The singularity of the above inter-band response at $\omega = \omega_\Delta(q)$ is of a weak logarithmic form, which could give rise to a solution for equation (3) only in the triplet channel and for extremely large values of $U$, which is unphysical. The $\chi^0$, however, possesses another much more interesting simple pole structure at a lower energy scale, $\omega = \omega_0(q)$, where

$$\omega_0(q) = v_F|q|.$$  

(7)

Note that this equation defines a line in the plane of $\omega$ and $|q|$, which corresponds to the lower boundary of the PHC of massless ($\Delta = 0$) Dirac fermions and is determined by the characteristic velocity $v_F$ of the underlying Dirac fermions. This could be considered as a property of the 'parent' massless Dirac system, which manifests itself as a simple pole structure when a mass parameter $\Delta$ is turned on in the non-interacting part of the Hamiltonian. At the gapless point, where $\Delta = 0$, the two energy scales at $\omega_0(q)$ and $\omega_\Delta(q)$ merge and the resulting singularity of $\chi^0$ will be of inverse-square-root form [10].

To search for solutions of equation (3), one has to look for the intersection of the real part (green line) with the constant horizontal line $\mp \frac{\pi}{h}$, where the upper (lower) sign corresponds to the singlet (triplet) channel. As can be very clearly seen in the figure, near $\omega_0(q) = v_F|q|$, where the imaginary part of $\chi^0$ is zero, there can be poles both in the singlet and triplet channels. The simple pole structure of the particle–hole propagator near this energy scale implies that poles exist for every value of the Hubbard $U$, no matter how small or how large it is. To see this more clearly, note that the real part of the susceptibility near $\omega_0(q)$ behaves as

$$\text{Re} \chi^0(q, \omega) \approx \frac{|q|\Delta}{\pi v_Fh^2[\omega - \omega_0(q)]}.$$  

(8)

The solutions of the eigenvalue equation (3) for an arbitrary value of $U$ define two collective mode branches in the singlet and triplet channel as

$$\omega_{\text{singlet/triplet}}(q) = v_Fq \left(1 \mp \frac{U\Delta q^2}{\pi h^2v_F^2}\right),$$

(9)

where the upper sign corresponds to the singlet channel and the lower sign corresponds to the triplet channel. These solutions are available even for arbitrarily small $U$. This formula can be interpreted as the spin–charge separation in the sense that the singlet and the spin-1 modes move at different velocities given by

$$v_{c/s} = v_F \left(1 \mp \frac{U\Delta q^2}{\pi h^2v_F^2}\right).$$

(10)

The mere existence and simple pole structure of the $\chi^0$ at the energy scale $\omega_0$ is a unique consequence of the gap opening in the single-particle spectrum of excitations. The presence of a gap in the single-particle spectrum pushes the continuum of free particle–hole pairs to higher energies, and hence the above collective excitations which appear around $\omega_0(q) = v_F|q|$ line are well separated from the boundary of the PHC so that they will be protected from Landau damping to the incoherent background of free particle–hole excitations.

3. Discussion

Although the opening of a single-particle gap in the spectrum of excitations in graphene pushes the lower edge of the particle–hole continuum from the energy scale $\omega_0$ to higher energy scale $\omega_\Delta$, it still leaves behind a signature of underlying massless Dirac fermions in the form of a singular behavior for $\chi^0$ on the line defined by $\omega_0(q) = v_F|q|$ corresponding to the PHC boundary of the underlying massless Dirac fermions. The simple pole left on this line has two consequences: (i) due to the sign change of the susceptibility across $\omega_0$, for arbitrary values of the Hubbard $U$ there will be collective mode solutions at both singlet and triplet channels. The explicit forms of these solutions are given by equation (9). (ii) The different velocities for the two modes is reminiscent of the situation one encounters in one dimension. Albeit the difference is that in 1D the divergences in particle–hole bubbles are of the characteristic inverse-square-root type. In the case of 2D massive Dirac fermions where $\Delta$ is finite, the divergence in the particle–hole propagator is of a simple pole form. When the limit $\Delta \to 0$ is taken, the simple pole merges with the logarithmic singularity at the boundary of the PHC, $\omega_\Delta$, and again gives rise to a inverse-square-root behavior for gapless Dirac fermions in 2D [10]. It is therefore the non-zero value of $\Delta$ which provides a solution in the singlet channel in addition to the known solution of the triplet channel at the $\Delta = 0$ point [10, 11]. In this sense, it appears that the non-zero value of $\Delta$ enhances the separation of the triplet and the singlet modes. Therefore the smallest value of the gap parameter $\Delta$ totally changes the nature of collective excitations in graphene. This
observation may have implications for novel approaches to the bosonization of the 2 + 1-dimensional massive Dirac fermions. Note that if the interaction employed was of a long-range Coulomb form, the solution in the spin-1 channel would be of a linearly dispersing gapped form while the solution in the singlet channel would not exist at all. Therefore the two modes discussed here are a peculiar feature of short-range interactions, which due to the remarkable value of the Hubbard $U$ in graphene may have relevance to physically fabricated samples of gapped graphene.

Now let us discuss the nature of the phase transition marked by vanishing of the velocity of the singlet mode. As can be seen in equation (10), when the Hubbard $U$ is large enough to satisfy

$$ U_c \Delta = \frac{3\pi}{4} t^2, \quad (11) $$

the velocity of singlet excitations becomes zero and the low-energy sector is exhausted by solely triplet excitations of the form

$$ \omega_{\text{triplet}}(q) = 2v_F q. \quad (12) $$

Thinking in the spirit of slave-boson approach [15], an electron (hole) can be assumed to be composed of its spin part—the spinon, and the charge part—the doublon (holon). Therefore a composite object constructed from an electron and a hole can be thought of as a separate spin-1 composite of two spinons while a charge composite of a doublon and a holon give rise to a spin zero boson. Hence the triplet mode can be interpreted as a triplet bound state of two spinons and a doublon and a holon give rise to a spin zero boson. Therefore the triplet mode can be interpreted as a composite boson constructed from a doublon and a holon, the total charge of which is zero. In this sense the vanishing of the velocity of the singlet mode can be associated with infinite enhancement of the effective mass of charge bosons. This gives rise to localization of charge carriers and hence is a Mott transition [16]. Indeed the decreasing trend $U_c \propto \Delta^{-1}$ as a function of the gap parameter $\Delta$ in equation (11) is in qualitative agreement with our previous DMFT result for the phase boundary of the Mott insulating phase in the ionic-Hubbard model [9]. Note that this agreement holds for very small to moderate values of $\Delta$. For large values of $\Delta$ the phase boundary in DMFT will be given by $U \propto \Delta^{-1}$ [9]. Note that the present analysis is not expected to hold for very large values of $\Delta$. The reason is that when $\Delta$ is large, the (covariant) hyperbolic bands are replaced by simple parabolic bands of ordinary semiconductors. Therefore, instead of using equation (3), one has to do a proper Bethe–Salpeter analysis and the collective states discussed here will be replaced by corresponding excitonic states. However, as far as experimental realization of the gap parameter in graphene is concerned, $\Delta$ will be on the scale of a few tens of meV, which remains quite small in the scale of the hopping term $t \sim 2.8$ eV. Moreover, the fact that the only low-energy excitations left in the system when $U$ goes beyond $U_c$ are spin-1 fluctuations, is further evidence that the phase transition at $U_c$ is to a Mott insulating state.

Because in the Mott insulator, the only possible low-energy modes are spin excitations.

When $U$ is further increased, it can be seen in figure 1 that the real part of $\chi^0$ and $+i\frac{\omega}{v_F}$ (triplet channel) will intersect in another point which is immediately below the edge of the PHC, $\omega_{\Delta}(q)$, and therefore a second triplet mode at slightly higher energy than the first one is expected to form. This solution is anticipated due to a weak logarithmic singularity at $\omega_{\Delta}$. However, this mode is not likely to be realized as it requires quite a large value of $U$. But, since for very large values of $U$ the system already falls in the Mott insulating phase, the ground state maybe totally deformed with respect to the initial starting state of massive Dirac fermions employed in this work.

It is interesting to note that, although the velocity of the underlying Dirac fermions is $v_F$, the velocity of the spin-1 mode is always more than $v_F$. In particular, at the transition to the Mott insulating phase, the velocity of the spin-1 mode will be $2v_F$, i.e. the triplet mode (if we call it tripolon) moves twice as fast as the underlying Dirac electrons. This prediction can be tested not only in gapped graphene systems, but also on a platform based on cold atoms engineered to mimic a massive Dirac theory at nanokelvin energy scales. The existence of a mode whose velocity can be twice the velocity of the underlying Dirac fermions may have interesting implications in teleportation of quantum information, as well as in spin-only forms of transport.

The above two modes discussed here also exist for small values of $U$. Therefore it should be possible to check for their experimental consequences. The good isolation of the energy scale $\omega_{\Delta}$ from the PHC edge $\omega_{\Delta}$ makes the massive Dirac fermions more interesting for performing neutron scattering experiments. In this work, we find that the triplet excitation exists below the Mott transition. Moreover, the Mott insulating phase has its own magnetic excitations. Therefore, in massive Dirac fermion systems such as gapped graphene/graphite or cold atoms arranged on honeycomb lattice, we anticipate a triplet excitation over a large range of values of the Hubbard parameter $U$. Such a low-energy spin-1 branch of excitations will have a characteristic $T/v_s^2$ contribution in the specific heat at constant volume, where $v_s$ is the velocity of spin modes. Below the Mott transition there will be another $T/v_F^2$ contribution coming from singlet charge modes. The influence of these modes on the various properties of massive Dirac fermions remains to be investigated.

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