Excitation spectrum of correlated Dirac fermions

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Abstract. Motivated by the puzzling optical conductivity measurements in graphene, we speculate on the possible role of strong electronic correlations on the two-dimensional Dirac fermions. In this work we employ the slave-particle method to study the excitations of the Hubbard model on honeycomb lattice, away from half-filling. Since the ratio \( \frac{U}{t} \approx 3.3 \) in graphene is not infinite, double occupancy is not entirely prohibited and hence a finite density of doublons can be generated. We therefore extend the Ioffe-Larkin composition rule to include a finite density of doublons. We then investigate the role played by each of these auxiliary particles in the optical absorption of strongly correlated Dirac fermions.

1. Introduction

Strong correlation has remarkable manifestations in properties of metals. Recent emergence of the so called Dirac materials in solids has provided a new playground for condensed matter physicists. One of the most important two-dimensional examples of Dirac materials is graphene the low energy theory of which is described by 2+1 dimensional Dirac Hamiltonian. As far as the non-interacting Dirac theory is concerned, the single-particle eigen-states of this Hamiltonian are chiral which protects them from complete back scattering. Many other amazing properties of graphene follows from the underlying Dirac nature of its free theory. On top of any free theory an interesting theoretical question would be, what happens if some forms of interactions are turned on. In this respect one of the most important forms of interactions are local (on-site) interactions parameterized by the so called Hubbard \( U \). This type of local correlations in ordinary metals leads to interesting spin liquid followed by Mott insulating behavior when the parent metal is at half-filling [1]. Upon doping away from half-filling the on-site Coulomb repulsion would also lead to possible high temperature superconductivity. Given such a background for the short-range Coulomb interactions in two dimensions, it is interesting to ask similar questions about the Dirac matter: i.e. what would be the consequences of strong electronic correlations on the physical properties of Dirac solids?

In the present work, we are interested in examining the role of strong correlations on the physical properties of Dirac electrons. One of the important problems of condensed matter systems is the optical conductivity that couples to the particle-hole channel of excitations, hence providing information not accessible in the single-particle measurements such as angular resolved photo emission spectroscopy (ARPES) or tunneling experiments. Recent \textit{ab-initio} estimates of the Hubbard \( U \) in graphene shows that it is remarkably strong on the scale of \( \sim 10 \) eV, i.e.
more than 3 times larger than the hopping amplitude $t$ of electrons between the $2p_z$ orbitals of neighboring carbon atoms \[2\]. Therefore it is timely to approach the problem from strong coupling side, and study the manifestations of strong correlation. In this work we employ slave-boson method to calculate the optical conductivity of Dirac electrons of graphene at zero temperature.

2. Slave Boson Method and Hubbard Model

Hubbard model is the canonical model to describe strong correlations in solids. This model consists of two terms, kinetic term that in the case of graphene has a low-energy Dirac nature; and an on-site Coulomb repulsion term of strength $U$. Hamiltonian of this model is shown by \[1\] where $t$ is the hopping amplitude and $(i,j)$ implies that sites $i,j$ are nearest neighbors.

\[
H = -t \sum_{\langle i,j \rangle,\sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow} \tag{1}
\]

The hopping takes place on the honeycomb lattice between two different sub-lattices ($A, B$). In general the Hilbert space of Hubbard model at each site has four states: empty state, two single occupancy states with opposite spin and double occupancy. When $U$ is infinite, the double occupancy is forbidden and the doubly occupied configurations are prohibited by infinitely large $U$. In the case of graphene, the Hubbard $U \approx 3.3t$ \[2\] and hence is not infinitely large. Therefore all four states are allowed. A well established technique to study the Hubbard model in this regime is the so called slave-particle method. In this work we use the slave boson formalism to study Hubbard model in the intermediate range of $U$. Instead of using physical electron operators $c_{i,\sigma}^\dagger$ one introduces auxiliary particles $f_{i,\sigma}^\dagger$, $h_i^\dagger$ and $d_i^\dagger$. The electron creation operator is represented by,

\[
c_{i,\sigma}^\dagger = f_{i,\sigma}^\dagger h_i + \sigma d_i^\dagger f_{i,-\sigma} . \tag{2}
\]

The operators $f_{i,\sigma}, h_i, d_i$ are associated with spinons and holons and doublons, respectively. Spinons are particles with spin but no charge. Holons and doublons are particles without spin but they have positive and negative charges, respectively. There is an equivalent between new (unphysical) Hilbert space and old (physical) Hilbert space of real electrons:

\[
h_i^\dagger |\text{vac}\rangle = |0\rangle_i \quad f_{i,\sigma}^\dagger |\text{vac}\rangle = |\uparrow\rangle_i , \quad f_{i,-\sigma}^\dagger = |\downarrow\rangle_i \quad d_i^\dagger |\text{vac}\rangle = |\uparrow\downarrow\rangle_i
\]

Since each lattice site is either empty, singly occupied or doubly occupied, the number of above auxiliary fields satisfies a local constraint is represented by:

\[
y_i = \sum_{\sigma} N_{i,\sigma}^f + N_{i}^d + N_{i}^h - 1 = 0. \tag{3}
\]

Substituting the slave-boson representation of electron creation operators in the Hubbard model we obtain the following representation of the Hubbard Hamiltonian in terms of auxiliary particles \[3\]:

\[
H = -t \sum_{\langle i,j \rangle} (\hat{\chi}_{i,j}^f + \hat{\Delta}_{i,j}^f + \hat{\Delta}_{i,j}^b + h.c.) + U \sum_i d_i^\dagger d_i \tag{4}
\]

where bosonic and fermionic bi-particle operators $\hat{\chi}$ and $\hat{\Delta}$ are define as:

\[
\hat{\chi}_{i,j}^f = \sum_{\sigma} f_{i,\sigma}^\dagger f_{j,\sigma} , \quad \hat{\chi}_{i,j}^b = h_i^\dagger h_j - d_i^\dagger d_j , \quad \hat{\Delta}_{i,j}^f = \sum_{\sigma} f_{i,-\sigma} f_{j,\sigma} , \quad \hat{\Delta}_{i,j}^b = d_i h_j + h_i d_j
\]
We decouple the above Hamiltonian into bosonic and fermionic hopping \((\hat{\chi})\) and pairing \((\hat{\Delta})\) channels by using Hubbard-Stratonovic transformation and write the partition function as:

\[
Z = \int D\hat{f}D\hat{h}D\hat{d}D\chi D\Delta D\lambda e^{-\int dt \mathcal{L}}
\]

\[
\mathcal{L} = \sum_{i,\sigma} f_{i,\sigma}^\dagger \frac{\partial}{\partial \tau} f_{i,\sigma} + \sum_i h_i^\dagger \frac{\partial}{\partial \tau} h_i + \sum_i d_i^\dagger \frac{\partial}{\partial \tau} d_i + H - \sum_i \lambda_i y_i.
\]

The field \(\lambda_i\) is Lagrange multiplier that enforces the local constraint Eq. (3) and in saddle point approximation we replace it by site-independent (mean field value) \(\lambda\). As can be seen in Eq. (2), in the enlarged Hilbert space of auxiliary particles, the physical electron is invariant under a local \(U(1)\) gauge transformation. This internal gauge field is the glue that binds the auxiliary particles back together to form the physical electrons. Therefore it is important to consider the \(U(1)\) phase fluctuation of the corresponding order parameters. Within the slave-boson mean field, the order parameters are replaced by their mean field values as \(\Delta_{i,j} \rightarrow \Delta\) and \(\hat{\chi}_{i,j} \rightarrow \chi\). Of course they may have non-trivial dependence on the direction of the bond connecting sites \(i\) and \(j\). Such non-s-wave order parameters can be important in highly doped graphene when it is doped to the M-point of the Brillouin zone [4]. But since in the present work we are concerned with the normal state where superconducting pairing amplitudes will be assumed to be zero, we are not concerned with these possibilities. Ignoring the superconducting order, the normal state Hamiltonian in the slave-boson mean field becomes,

\[
H_{\text{eff}} = H_f + H_h + H_d + E_0
\]

\[
H_f = -\lambda \sum_{i,\sigma} f_{i,\sigma}^\dagger f_{i,\sigma} - t\chi^b \sum_{<i,j>,\sigma} f_{i,\sigma}^\dagger f_{j,\sigma} + h.c.
\]

\[
H_h = - (\lambda - \mu_e) \sum_i h_i^\dagger h_i - t\chi^f \sum_{i,j} h_i^\dagger h_j + h.c.
\]

\[
H_d = - (-U + \lambda + \mu_e) \sum_i d_i^\dagger d_i + t\chi^f \sum_{i,j} d_i^\dagger d_j
\]

\[
E_0 = 6tN\chi^f \chi^b + (\lambda - \mu_e) N + \mu_e N_e
\]

In above relation \(N\) is the total number of lattice sites half of which are on sub-lattice A, and the other half on sub-lattice B giving total \(N/2\) unit cells. The quantity \(N_e\) is the total number of electrons. All hopping terms between the nearest neighbors and hence from sub-lattice A to B and vice versa. The chemical potential \(\mu_e\) controls the total number of electrons with respect to empty lattice, while \(\lambda\) forces the constraint of Eq. (3). The above equation parameterizes the chemical potential \(\mu_f = \lambda, \mu_h = \lambda - \mu_e, \mu_d = \lambda + \mu_e - U\) in terms of two independent Lagrange multipliers \(\mu_e\) and \(\lambda\).

3. Extension of Ioffe-Larkin composition rule in presence of doublons

Now that the Hamiltonian in the mean field approximation is decomposed into pieces containing various auxiliary particles, we need a rule that combines the conductivity (i.e. current-current correlation function) in different sectors and gives the conductivity of physical electrons. To this end, we consider phase fluctuation of order parameters around the mean field saddle point values and look at the \(U(1)\) gauge theory of the normal state. In this situation we have two order parameters, the phase fluctuations of which are given by \(a_{ij}\) link fields as follows,

\[
\chi_{ij}^{f,b} = e^{ia_{ij}} \chi_{ij}^{f,b}
\]
In terms of these phase variables the Lagrangian becomes,

\[ L = L_f + L_h + L_d \quad (9) \]

\[ L_f = \sum_{i,\sigma} f_i^\dagger (\frac{\partial}{\partial \tau} - \mu_f) f_i,\sigma - t\chi \sum_{(i,j),\sigma} e^{ia_{ij}} f_i^\dagger f_j,\sigma + \text{h.c.} \]

\[ L_h = \sum_i h_i^\dagger (\frac{\partial}{\partial \tau} - \mu_h) h_i - t\chi \sum_{i,j} e^{ia_{ij}} h_i^\dagger h_j + \text{h.c.} \]

\[ L_d = \sum_i d_i^\dagger (\frac{\partial}{\partial \tau} - \mu_d) d_i + t\chi \sum_{i,j} e^{ia_{ij}} d_i^\dagger d_j + \text{h.c.} \]

in above equation \( a_{i,j} \) is defined as \([5, 6]\):

\[ (\vec{r}_i - \vec{r}_j).\vec{a}[(r_i + r_j)/2] = \int \vec{a}.d\vec{r}. \]

We are interested to study low energy physics on the Honeycomb lattice where the low energy states is described by massless Dirac fermions \([7]\):

\[ H^\pm = v_F \gamma^0 \vec{\gamma}.\vec{p} \quad (10) \]

In above Hamiltonian \( \tau_z = \pm \) is associated with two Dirac points \( (K, K') \) and \( v_F \) is Fermi velocity of electrons and equals \( \sqrt{3}\alpha a/2 \) and \( \vec{p} \) are two dimensional vectors. The matrices \( \vec{\gamma} \) are given by:

\[ \gamma^0 = -\sigma_z \quad , \quad \gamma^1 = i\sigma_y \quad , \quad \gamma^2 = \pm i\sigma_x \equiv i\tau_z \sigma_x. \quad (11) \]

By going to the continuum limit and considering phase fluctuations of order parameters as internal gauge field, we can write:

\[ L_f = \int d\vec{r} \quad \bar{f}(\vec{r},\sigma) \gamma^0 (\frac{\partial}{\partial \tau} - \mu_f) f(\vec{r},\sigma) + \hbar v_f \sum_{\sigma} \int d\vec{r} \quad \bar{f}(\vec{r},\sigma) \vec{\gamma}.(\frac{\vec{\nabla}}{i} + \vec{a}) f(\vec{r},\sigma) \quad (12) \]

\[ L_h = \int d\vec{r} \quad \bar{h}(\vec{r},\sigma) \gamma^0 (\frac{\partial}{\partial \tau} - \mu_h) h(\vec{r},\sigma) + \hbar v_h \sum_{\sigma} \int d\vec{r} \quad \bar{h}(\vec{r},\sigma) \vec{\gamma}.(\frac{\vec{\nabla}}{i} + \vec{a}) h(\vec{r},\sigma) \quad (13) \]

\[ L_d = \int d\vec{r} \quad \bar{d}(\vec{r},\sigma) \gamma^0 (\frac{\partial}{\partial \tau} - \mu_d) d(\vec{r},\sigma) + \hbar v_d \sum_{\sigma} \int d\vec{r} \quad \bar{d}(\vec{r},\sigma) \vec{\gamma}.(\frac{\vec{\nabla}}{i} + \vec{a}) d(\vec{r},\sigma). \quad (14) \]

Note that \( \vec{f} \) is defined as \( f^\dagger \gamma^0 \) and \( f = (f_A, f_B) \) is a spinor encoding the two sub-lattice structure of the honeycomb lattice. Similar definition holds for holons and doublons. Finally the velocity of three auxiliary particles are given by:

\[ v_f = \frac{\sqrt{3} t\alpha \chi^b}{2} \quad , \quad v_h = \frac{\sqrt{3} t\alpha \chi^f}{2} \quad , \quad v_d = -\frac{\sqrt{3} t\alpha \chi^f}{2}. \]

The coupling between the auxiliary particles and the internal gauge field is of the following form,

\[ L_{int} = \vec{J}_f.\vec{a} + \vec{J}_h.\vec{a} + \vec{J}_d.\vec{a}. \quad (15) \]
Minimization of the above above term with respect to the internal gauge field \( \vec{a} \) gives

\[
\vec{J}_f + \vec{J}_h + \vec{J}_h = 0.
\]

This relation is indeed very plausible. At every given site the total number of auxiliary particles is conserved. Therefore the at a given site, there is only one of the above auxiliary particles available. Likewise at a neighboring site to which a hopping is going to happen, another particle is available. Therefore always a third particle (among, \( f \), \( d \) and \( h \) particles) is missing from the nearest neighbor hopping. Hence only two of the above auxiliary particles produce currents which by the very constraint forces the currents to satisfy the above relation.

Now let us couple an external electro-magnetic (EM) gauge field \( \vec{A} \). Assuming that spinons have no EM charge, while charge of holons and doublons are +1 and \(-1\), respectively, these bosons interact with EM field via the following minimal substitution:

\[
(\vec{\nabla} + \vec{a}) \rightarrow (\vec{\nabla} + \vec{a} \pm \vec{A}).
\]

Having introduced the external EM gauge field \( \vec{A} \), expanding up to second order in \( \vec{A} \), and performing the integration over auxiliary fields we obtain

\[
S_{\text{eff}}(a, A) = \sum_q \Pi_{\mu,\nu}(q)a_\mu(q)a_\nu(-q) + \Pi^h_{\mu,\nu}(q)[a(q) - A(q)]_\mu[a(-q) - A(-q)]_\nu
+ \Pi^d_{\mu,\nu}[a(q) + A(q)]_\mu[a(-q) + A(-q)]_\nu
\]

where \( \Pi_{\mu,\nu} \) is current-current correlation function and is defined by:

\[
\Pi^\alpha_{\mu,\nu} = \text{tr}\langle J^\alpha_{\mu}(q)J^\alpha_{\nu}(-q)\rangle.
\]

Here \( \alpha = f, h, d \) and \( q \equiv (\vec{q}, i\omega_n) \). If we integrate out internal gauge field \( a \), effective action in terms of \( A \) becomes,

\[
S_{\text{eff}}(A) = \sum_q \Pi_{\mu,\nu}A_\mu A_\nu
\]

where

\[
\Pi = \frac{4\Pi^d\Pi^h + \Pi^d\Pi^f + \Pi^h\Pi^f}{\Pi^d + \Pi^h + \Pi^f}.
\]

This relation arises from the fact that fermions and holons and doublons are coupled to each others via internal gauge fields and hence they can not move independently. Although we employed the linearized dispersion of underlying electrons on a honeycomb lattice to emphasize Dirac nature of the electrons, but the above relation relies on the constraint imposed by the internal gauge field and therefore it also holds when a tight-binding band picture valid over the entire hexagonal Brillouin zone of graphene is used.

The above relation is a generalization of what is known as Ioffe-Larkin composition rule to the case where a finite number of doublons is also present. Finite density of doublons arise from the fact that Hubbard \( U \) is not infinitely large. In the following let us repeat the physical argument of Ioffe and Larkin [8] to convince ourselves that the above composition rule in presence of doublons is independent of the details of the band structure of underlying electrons. Assume that the internal gauge fields gives rise to electric field \( \vec{e} \). In the presence of external electric field \( \vec{E} \), effective electric fields for each particles are:

\[
\vec{e}_f = \vec{e}, \quad \vec{e}_h = \vec{e} - \vec{E}, \quad \vec{e}_d = \vec{e} + \vec{E},
\]
as we have assumed that the EM charges belong only to holons and doublons. Current due to each particle is given by the following equation:

\[ \vec{j}_\alpha = \sigma_\alpha \vec{e}_\alpha \]  

where \( \alpha = f, h, d \) and the above relation is seen as a tensorial equation where tensor components are not shown for clarity. Using this relation in the constraints on the currents, Eq. (16) gives the following relation between the internal and external electric fields:

\[ \vec{e} = \frac{\sigma_h - \sigma_d}{\sigma_f + \sigma_h + \sigma_d} \vec{E}. \]  

The physical charge current is defined as difference between doublonic and holonic currents,

\[ \vec{j} = \vec{j}_d - \vec{j}_h = \sigma \vec{E}, \]  

⇒ \[ \sigma = \frac{4\sigma_d \sigma_h + \sigma_d \sigma_f + \sigma_h \sigma_f}{\sigma_d + \sigma_h + \sigma_f}. \]  

This conductivity relation is same as equation that is calculated in Eq. (20) and hence is not dependent on the details of the band dispersion and holds for the low-energy Dirac as well as the full tight-binding kinetic parts. If we consider the situation that there is no doublon in our systems (large limit of \( U/t \)), the doublon channel is blocked and hence the conductivity \( \sigma_d \) of doublons vanishes. Therefore in this limit the composition formula for the conductivity reduces to the relation containing only spinons and holons contributions [9, 10].

4. Normal State

Let us now proceed by rewriting Eq (7) in the Fourier space which leads to the following normal state Hamiltonian:

\[ H_f = \sum_{k, \sigma} \left[ f_{k, \sigma}^A \bar{f}_{k, \sigma}^A \left[ -\mu_f - \chi \eta_k \right] \left[ f_{k, \sigma}^A \right] \right. \]  

\[ H_h = \sum_{k} \left[ h_k^A \bar{h}_k^A \left[ -\mu_h - \chi \eta_k \right] \left[ h_k^A \right] \right. \]  

\[ H_d = \sum_{k} \left[ d_k^A \bar{d}_k^A \left[ -\mu_d - \chi \eta_k \right] \left[ d_k^A \right] \right. \]  

\[ E_0 = (\lambda - \mu_e)N + 6tN \chi \chi^b + \mu_e N_e, \]  

where \( \eta_k = \delta e^{i\vec{k} \cdot \vec{\theta}} \) and summation applies on the nearest neighbor vectors. \( \mu_f, \mu_h, \mu_d \) are defined in previous section. Diagonalizing the above Hamiltonian gives the following energy eigen-values for fermions and holons and doublons:

\[ E_{k, \sigma}^f = -\mu_f \pm \chi \eta_k, \]  

\[ E_{k}^h = -\mu_h \pm \chi \eta_k, \]  

\[ E_{k}^d = -\mu_d \pm \chi \eta_k. \]  

At the mean field level where the above auxiliary particles do not interact, the free energy at a given temperature \( T \) is given by:

\[ F = -\frac{2}{\beta} \sum_{k, \sigma = \pm} \ln(1+e^{-\beta E_{k, \sigma}^f}) + \frac{1}{\beta} \sum_{k, \sigma = \pm} \ln(1+e^{-\beta E_{k, \sigma}^h}) + \frac{1}{\beta} \sum_{k, \sigma = \pm} \ln(1+e^{-\beta E_{k, \sigma}^d}) + (\lambda - \mu_e)N + 6tN \chi \chi^b. \]  

(33)
where $s = \pm$ stands for two branches of energy bands for each auxiliary particle. Minimizing free-energy leads to follow Self-consistency equations:

\[
\frac{\partial F}{\partial \lambda} = 0 \quad \rightarrow \quad 2 \sum_{k,s=\pm} f(E_{k,s}) + \sum_{k,s=\pm} n_B(E_{k,s}^h) + \sum_{k,s=\pm} n_B(E_{k,s}^d) - N = 0, \quad (34)
\]

\[
\frac{\partial F}{\partial \chi^f} = 0 \quad \rightarrow \quad \sum_{k,s=\pm} st|\eta_k|n_B(E_{k,s}^h) + \sum_{k,s=\pm} st|\eta_k|n_B(E_{k,s}^d) + 6tN\chi^b = 0, \quad (35)
\]

\[
\frac{\partial F}{\partial \chi^b} = 0 \quad \rightarrow \quad 2 \sum_{k,s=\pm} st|\eta_k|f(E_{k,s}) + 6tN\chi^f = 0, \quad (36)
\]

where $n_B(E)$ and $f(E)$ are Bose-Einstein and Fermi-Dirac distribution functions, respectively. If temperature is nonzero all of particles are distributed according to their corresponding distribution function among their energy levels and the self-consistent solution of the above equations determines various parameters. However in the limit of $T \rightarrow 0$, Bose particles may have a chance to undergo a Bose condensation that gathers them at the bottom of their energy band. Therefore in zero temperature if holons and doublons have zero energy they could be condensed in the bottom of their energy band that lies at $\vec{k} = 0$. Holons can condense when $\lambda = \mu_e - 3t\chi^f$. In such a case, the minimum energy of doublons becomes $U - 2\mu_e$. Consequently, depending on the value of $\mu_e$ holons or doublons or both of them could be condensed i.e. if $\mu_e = U/2$ we are in half-filling and have particle hole symmetry and both of holon and doublon condense, but for $\mu_e > U/2$ ($\mu_e < U/2$) only doublons (holons) condense. For example if system is doped by holes, following self-consistency equations at $T = 0$ becomes,

\[
\lambda = \mu_e - 3t\chi^f, \quad \chi^b = \frac{1}{2}\delta, \quad \chi^f = \frac{1}{3N} \sum_k |\eta_k|. \quad (37)
\]

The prime in the above summation means that the sum is performed over the occupied fermionic states and $\delta$ is number of holons per site. By substituting $\chi^f$ we obtain $\lambda = \frac{1}{N} \sum_k |\eta_k|$. Away from half-filling where only one of bosons condenses, the boson conductivity will be a strong Dirac delta peak around the accessible energy $\omega_b$ from the condensed momentum. Therefore the boson conductivity will be zero away from $\omega_b$. This makes the total conductivity of physical particles zero away from $\omega_b$ which is not consistent with the experiment of Ref. [11]. Hence as far as the doped graphene is concerned, the bosons will not be in the condensed phase as the light absorption in a wide energy range is non-zero.

5. Conclusions

For the intermediate values of $U$ the presence of doublons are allowed and they are coupled to the fermions and holons by internal gauge field. In the absence of pairing parameters (i.e. in the normal state) we extended the Ioffe-Larkin composition rule to include the contributions of doublons in the physical conductivity. We also re-derived it using a general physical argument. We studied normal state of Hubbard model in zero temperature and showed in this case depending on electronic chemical potential, bosons or doublons or both of them can be condensed. We further concluded that in doped graphene, the bosons (either of holons or doublons) can not be condensed. Therefore one must consider non-zero temperatures in order to understand the optical absorption experiment of Ref. [11].

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