Symmetry content and spectral properties of charged collective excitations for graphene in strong magnetic fields

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Abstract – We show that graphene in a strong magnetic field with partially filled Landau levels sustains charged collective excitations — bound states of a neutral magnetoplasmon and free particles. In the limit of low density of excess charges, these are bound three-particle complexes. Some of these states are optically bright and may be detected in spectroscopy experiments, providing a direct probe of electron-electron interactions in graphene. The charged excitations can be classified using the geometrical symmetries — non-commutative magnetic translations and generalized rotations — in addition to the dynamical SU\(_4\) symmetry in graphene. From the SU\(_4\) symmetry point of view, such excitations are analogous to bound states of two quarks and one antiquark \(q\bar{q}q\) with four flavors. We establish a flavor optical selection rule to identify the bright states for experimental studies.

Since the isolation of graphene, the unusual behavior of its electrons in a magnetic field has been the subject of intense study [1,2]. Early on, the anomalous integer quantum Hall effect was observed, with plateaus detected at filling factors \(\nu = \pm 2(|n| + 1)\), where \(n\) is the Landau level (LL) index [3,4]. Subsequently, in higher fields, plateaus were seen at \(\nu = 0, \pm 1, \pm 4\), signifying a lift in the fourfold (spin and valley) LL degeneracy, most likely due to the many-body effects [5]. Recently, fabrication of high-quality samples has allowed the fractional quantum Hall effect (FQHE) to be observed at \(\nu = \frac{1}{3}\), indicating again the presence of a strongly correlated electron state [6,7]. Importantly, such signatures of correlated many-body physics emerge already at low magnetic fields, and in higher fields these correlations determine the physics. Here we establish the existence of collective charged excitations which are essentially bound states of a neutral magnetoplasmon binding free particles. In the limit of low density of excess carriers such collective excitations can be thought of as three-particle interacting states. We will demonstrate that in graphene complexes of electrons, akin to \(q\bar{q}q\) quark-triplets in high-energy physics, arise naturally. This is because the spin and valley pseudospin in graphene combine to form four flavors analogous to the four flavors of 1st- and 2nd-generation quarks [8]. Analysis of the symmetry content of these states allows the classification of optically active and dormant states, highlighting the usefulness of high-energy analogues in graphene.

We study the formation of such excitations in pristine graphene in strong magnetic fields with a low density of excess electrons in partially filled LLs. More specifically, we consider inter-LL excitations which, in the presence of excess charges, may bind one additional electron (or hole). Such excitations i) lie above charged topological collective excitations, Skyrmions [9–11], and ii) are directly accessible via optical and tunneling experiments. In particular, we are interested in the behavior of graphene close to the charge neutrality point and thus focus on filling

\(^{1}\)The dynamical interactions, strong for quarks and electromagnetic for electrons, are of course fundamentally different. Besides, electrons in graphene lack the color of true quarks. The latter makes the existence of states analogous to \(q\bar{q}q\) and \(q\bar{q}\bar{q}\) possible in graphene, while such states cannot be colorless and, as such, do not exist in elementary particle physics.

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Fig. 1: (Color online) Predicted absorption spectra in pristine graphene with \( \nu = -2 + \epsilon \) for the bright state at energy \(-0.09 E_0\). The inset schematically shows the relevant optical transitions. Energies are given in units of \( E_0 \) (see text) with zero energy corresponding to the CR energy, \( \hbar \omega_c \).

As \( \nu = \mu \pm \epsilon \) for \( \mu = 0, \pm 1, \pm 2 \) and \( \epsilon \ll 1 \). By way of illustration, let us consider the filling factor \( \nu = -2 + \epsilon \), so that the \( n = 0 \) LL is almost empty (see fig. 1, inset). A neutral collective excitation, or magnetoplasmon, is formed when an electron, excited to an empty state in the \( n = 0 \) LL, becomes bound to the hole formed in the \( n = 1 \) LL. On their own, such neutral excitations form a continuum of extended states, with the lower continuum edge corresponding to the cyclotron resonance (CR) mode at energy \( \hbar \omega_c = \nu \sqrt{\frac{2eB}{c}} \) [12]. In addition, they may bind one of the few excess electrons in the \( n = 0 \) LL, thus forming a negatively charged trion \((\text{e}eh)\) complex, \( X^- \). These discrete bound states are stable when located below the magnetoplasmon continuum. We demonstrate the existence of such states, and determine their spectral properties. We find that the symmetry of graphene severely restricts optical transitions so that most collective states are dark. The bright bound states give rise to additional optical absorption peaks below the CR as shown in fig. 1 and whose observation is within experimental reach. The binding energies of the extra electron in the \( X^- \) are \( \sim 0.1 E_0 \), similar to large FQHE gaps expected in pristine graphene [13,14] \((E_0 \simeq 40 \text{ meV at } B = 10 \text{ T})\). Here 

\[
E_0 = \sqrt{\frac{\epsilon^2}{2 \mu_B^2}}
\]

is the characteristic energy of Coulomb interactions in strong fields, \( E_0 \ll \hbar \omega_c \); \( \mu_B = (\hbar c/eB)^{1/2} \) is the magnetic length and \( \epsilon \) the effective dielectric constant. In this regime, virtual transitions between LLs are suppressed as powers of \( E_0/\hbar \omega_c \) and are neglected, constituting the high-magnetic-field approximation [11,13,15–17].

Due to the particle-hole symmetry in graphene, positively charged trions \( X^+ \), or \((\text{e}hh)\) states, exist as \( X^- \) counterparts. This occurs for the complementary filling factors \( -\nu \), such as, \( -\nu = 2 - \epsilon \), when the \( n = 0 \) LL is almost completely filled but contains a few holes. The \( X^+ \) states at \( -\nu \) have the same energies and optical strengths as their \( X^- \) counterparts at \( \nu \), but are active in the opposite circular polarization.

With the underlying dynamical SU\(_4\) symmetry of graphene, resulting from two possible spin \((\uparrow, \downarrow)\) and two valley pseudospin \((\uparrow, \downarrow)\) projections, the trion states are in fact completely analogous, from the symmetry point of view, to bound states of two quarks and one antiquark with four flavors, \( qq\bar{q} \) for the \( X^- \) and \( q\bar{q}q \) for the \( X^+ \) (see footnote 1). The corresponding multiplets are shown in fig. 2; LL splittings in fig. 2(a) are shown for clarity. In particular, the state designated in fig. 1 as \( X^- \) realizes the fundamental representation of the SU\(_4\) group, fig. 2(b). The elementary particle physics classification of states and the techniques of the SU\(_4\) group [8] turn out to be very useful in graphene. They allow us to perform a complete group-theoretical analysis of the bound states and to determine the multiplicities of the emerging groups of excitations. Importantly, these enable us to establish a flavor optical selection rule, explaining which of the states are dark and which are bright for experimental studies.

In the following, we focus on the \( X^- \) states, which may occur for fillings \( \nu = \mu + \epsilon \), where \( \epsilon \ll 1 \) and \( \mu \) is an integer; the \( X^+ \) case proceeds analogously. The operator creating a charged collective excitation is

\[
R^I_{k,j,N} = \sum_{m_1,m_2=0}^{\infty} A_{k,j,N}(m_1,m_2) c_{N_1,m_1}^\dagger c_{N_2,m_2}^\dagger d_{N_m}^j, \tag{1}
\]

where the hole representation \( d_{Nm}^j \equiv c_{Nm} \) is used for all completely filled levels. The composite index \( N = \{n, s_z, \sigma_z\} \) carries the LL index \( n \), the spin \( s_z = \pm \frac{1}{2} \) and the pseudospin \( \sigma_z = \pm \frac{1}{2} \) (labeling valleys \( K \) and \( K' \)), while \( N = \{N_1, N_2, N_3\} \).

The oscillator quantum number, \( m \), represents in the symmetric gauge \( A = \frac{1}{2} B \times r \) the guiding center for a single particle, yielding the LL degeneracy. As for the neutral case [17], several of the \( R_{k,j,N} \) states having same \( k \), \( J_z \) and equal or close energies (but different \( N \)'s) are strongly mixed by Coulomb interactions. As a result, the true excitation is given by a linear combination of these (see fig. 2).

The geometrical symmetries of the system allow the classification of the composite excitations by two orbital quantum numbers, half integer \( J_z \) and integer \( k = 0, \frac{1}{2}, \ldots \). The former is due to the symmetry under generalized rotations in graphene. It corresponds to the total \( \hat{J}_z = \sum_j \hat{J}_{1z} \), where a single-particle operator \( \hat{J}_{1z} = \hat{I}_{1z} - \frac{1}{2} \hat{I}_z \otimes \tau_z \) involves the orbital \( \hat{I}_{1z} \) and the sublattice isospin \( \tau_z \) angular-momentum projections [17,18]. The oscillator quantum number, \( k \), is due to magnetic translations whose generator \( \hat{K} = \sum_i \hat{K}_i \) commutes with the Hamiltonian \( \hat{H} \). The single-particle operators are \( \hat{K}_i = \pi_i - \frac{\hbar}{2} \hat{r}_i \times \hat{B} \), where \( \pi_i \) is the kinematic momentum operator and \( \epsilon_i \) is the particle’s charge. For charged complexes, the components do not commute: \([K_\sigma, K_\rho] = \theta \hbar B/c \) for the \( X^- \). However, using the mutual commutativity of \( \hat{H}, \hat{J}_z, \) and \( \hat{K}^2 \), the composite states can be chosen to be
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Fig. 2: (Color online) (a) Filled/empty circles (●/○) denote electrons/holes; hatched regions, the Dirac sea. States in the boxes are resonantly mixed by the Coulomb interactions. (b) Quartet $|x\rangle$ and (c) $X_{\pm}$ states. Shading denotes the different levels of charm. The neutral (e-h) state in (b) $Q_0 = \frac{\hbar}{2} (d\bar{d} + u\bar{u} + s\bar{s} + c\bar{c})$ is the SU4 singlet. The electron in the $n = 1$ LL shown in (a) is resonantly admixed to the $X^-$ from the $[4]$-multiplet with $J_e = \frac{1}{2}$.

...with $N_i = N'_i = \{0, s_i, \sigma_i\}$ and $i = 1, 2$. The graphene Coulomb interaction vertex $W_{N_1 N'_1, N_2 N'_2, N_3 N'_3} \sim \delta_{s_1, s'_1} \delta_{s_2, s'_2} \delta_{s_3, s'_3}$ conserves spin and pseudospin and may be expressed in terms of the vertices calculated using the standard 2D electron wave functions in LLs [17]. The Hamiltonian of the e-h interactions consists of two parts, $H_{eh} = H_{eh}^I + H_{eh}^{II}$, where

$$H_{eh}^I = \sum_{m_1, m_2} \sum_{s_1, s_2, \sigma_1} \sum_{s'_1, s'_2, \sigma'_1} W^{N'_1 \delta_{N_1 N'_1}}_{N_1 N'_1, N_2 N'_2, N_3 N'_3} \left( \begin{array}{c} d_{N_1}^{\dagger} \bar{d}_{N_2} \bar{d}_{N_3} \\ \bar{d}_{N_1} d_{N_2} d_{N_3} \end{array} \right)$$

with $N_1 = N'_1 = \{0, s_1, \sigma_1\}$ and $N_2 = N'_2 = \{1, s_2, \sigma_2\}$.

The e-h vertex

$$W^{N'_1 \delta_{N_1 N'_1}}_{N_1 N'_1, N_2 N'_2, N_3 N'_3} \sim \delta_{s_1, s'_1} \delta_{s_2, s'_2} \delta_{s_3, s'_3}$$

(4)

describes the direct e-h Coulomb interaction and the exchange e-h repulsion. The term $H_{eh}^{II} = \sum_{m_1, m_2} \sum_{s_1, s_2, \sigma_1} W^{N'_1 \delta_{N_1 N'_1}}_{N_1 N'_1, N_2 N'_2, N_3 N'_3} \left( \begin{array}{c} d_{N_1}^{\dagger} \bar{d}_{N_2} \bar{d}_{N_3} \\ \bar{d}_{N_1} d_{N_2} d_{N_3} \end{array} \right)$

(5)

and $N_i = \{1, s_i, \sigma_i\}$, $N'_i = \{0, s_i, \sigma_i\}$, $N_2 = \{1, s_2, \sigma_2\}$, and $N'_2 = \{0, s_2, \sigma_2\}$ does not conserve the number of particles in each of the LLs involved. It describes the resonant conversion between a three-particle trion state and a single electron state in the $n = 1$ LL and contributes $\sim E_0$.

These indicated interaction terms exhaust all Coulomb contributions $\sim E_0$ for the considered charged excitations at $\nu = -2 + \epsilon$. For other excitations and at other filling factors, the effective interaction Hamiltonians are constructed analogously.

The excitations may additionally be characterized by their total spin projection $S_z$ and pseudospin projection (valley index) $\Sigma_z$ [1,2]. However, there exists a larger SU4 group, which includes the direct product of the spin and pseudospin groups SU2 $\otimes$ SU2 $\subset$ SU4 as a subgroup. More
precisely, the Hamiltonian of the Coulomb (and in fact any long-range) inter-particle interactions is $SU_4$ symmetric up to small symmetry-breaking terms [15]. The dynamical $SU_4$ symmetry in graphene is based therefore on the equivalence of the two valleys ($\uparrow, \downarrow$) and the two spin states ($\uparrow, \downarrow$), including the continuous interchange between spins and valleys. The four basis states (flavors) that form a fundamental representation of the $SU_4$ group are $\downarrow\downarrow, \uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow$. We shall label them in analogy with the four flavors of down, up, strange, and charm quarks, by \{ $\downarrow\downarrow, \uparrow\uparrow, \uparrow\downarrow, \downarrow\uparrow$ \} $\equiv \{ d, u, s, c \}$. The generators of the $SU_4$ group can be constructed using the bilinear combinations conserving the number of electrons and holes. We have $C_{ij} = \sum_{N} c_{N}c_{Nj} - \sum_{N} d_{N}d_{Nj}$, where the composite index $N = \{ n, m \}$ and $i, j \in \{ d, u, s, c \}$. The generators satisfy the commutation relations $[C_{ij}, C_{kl}] = \delta_{jk}C_{il} - \delta_{il}C_{kj}$. The operators of total spin $S$ and pseudospin $\Sigma$ can be expressed in terms of the $C_{ij}$. For example, $\Sigma_{z} = \frac{1}{2} (C_{zz} + C_{uu} - C_{ss} - C_{dd})$ and $\Sigma_{+} = \Sigma_{x} + i \Sigma_{y} = C_{ud} + C_{du}$. The degeneracy of the eigenstates follows from the invariance of the Hamiltonian $\hat{H} = U^{\dagger}HU$, where $U = \exp(i \sum_{jk} \Theta_{jk} C_{ij})$, with $\Theta_{jk}$ the $SU_4$ transformation “angles” [8,15].

To determine the multiplicities of the 64 possible $ech$ flavor states, we need to decompose the direct product of the three $SU_4$ multiplets. The electron states form the fundamental $SU_4$ $[4]$-multiplet with four states represented by the Young diagram $\bigotimes$. The hole (antiparticle) transforms as a conjugate multiplet $[\bar{4}]$ and is represented by the Young diagram $\bigotimes$ which is one box short of the unitary singlet $[1]$. The latter transforms as the $SU_4$ vacuum. The decomposition of the direct product of the $SU_4$ $ech$ states gives $[4] \otimes [4] \otimes [4] = [4] \oplus [4] \oplus [20] \oplus [36]$. The corresponding Young diagrams are, respectively,

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Graphically, the resulting multiplets with their flavor contents resolved are shown in figs. 2(b)–(d). There, we use the standard elementary particle physics $SU_4$ representation [8]2. Namely, the three orthogonal axes show: the flavor isospin projection $T_{2} = \frac{1}{2} (C_{uu} - C_{dd})$, the hypercharge $Y = \frac{1}{2} (C_{uu} + C_{dd} + C_{cc}) - \frac{3}{2} C_{ss}$ and the charm $c = C_{cc}$. The Young diagrams (6) show that the states in the $[20]$-multiplet are antisymmetric with respect to permutation of two electrons and symmetric in the $[36]$-multiplet. Notice that the states of the two $[4]$-multiplets describe the combination of an electron in one of the possible $d, u, s, c$ states with an $ch$ symmetric neutral state involving the other electron $Q_0 \equiv \frac{1}{2} (dd + uu + ss + cc)$; the states in the $[20]$-multiplet are conjugate to the baryon $qqq$ [20]-plet of elementary particle physics including the neutron $n$, proton $p$ and $\Lambda, \Sigma$, and $\Xi$ particles.

2The $qqq$ multiplet $[20]$ is conjugate to the baryon $qqq$ [20]-plet of elementary particle physics including the neutron $n$, proton $p$ and $\Lambda, \Sigma$, and $\Xi$ particles.
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were found only for $\mu = \pm 2$. Our results are valid in the presence (as indicated in fig. 2) as well as the absence of spin $\delta \omega s$ and valley $\delta \omega v$ LL splittings, provided $\delta \omega_{s,v} \ll \omega_0$. In very strong magnetic fields $B > 20$ T, the $n = 0$ LL splitting may become of the order of $0.1 E_0$ [5], most probably due to the spontaneous symmetry breaking [1,2]. If this occurs, the SU$_4$ multiplets will split, which requires additional group-theoretical analysis.

The SU$_4$ symmetry allows one to determine, in a very direct and simple manner, the selection rule for trion photocreation $\hbar \omega + e^- \rightarrow (ech)$. Indeed, the interaction of electrons with light of frequency $\omega$ and circular polarizations $\sigma^\pm$ is described by the Hamiltonian

$$\delta \hat{H}_\pm = A \begin{pmatrix} \tau_\pm & 0 \\ 0 & \tau_\pm \end{pmatrix},$$

where $\tau_\pm = \tau_x \pm i \tau_y$ are the isospin Pauli matrices acting in the space of the graphene sublattices, $A = \frac{e \hbar \omega}{2 \omega_{c}}$ and $\mathcal{E}$, the electric-field amplitude [17,22,23]. When expressed in the SU$_4$ terms, $\delta \hat{H}_\pm$ assumes the form $A (dd + uu + ss + cc) \sim Q_0$, i.e., is proportional to the SU$_4$ singlet $Q_0$. This means that the photon is flavorless in the SU$_4$ sense. From this and considerations of the dipole transition matrix elements $\langle (ech)| Q_0 |e^- \rangle$, it immediately follows that the states in the [20] and [36] multiplets are all dark while the states in the [4]-multiplet may be bright, provided they have proper orbital quantum numbers. Let us discuss the latter.

The optical selection rules for the orbital quantum numbers are $\Delta k = 0$ and $\Delta J_z = \pm 1$ in the $\sigma^\pm$ polarizations. The former reflects the negligible linear momentum of a photon in the dipole approximation and the fact that $k$ is associated with translations (i.e., is the discrete analog of linear momentum for charged states in a magnetic field). Let us consider $X^-$ photocreation $\hbar \omega s + e_{nm} \rightarrow (ech) k_{J_z = -k}$; here and in what follows $J_z$ is that of the seed state. The electron in the initial state in the $n$-th LL has $j_z = |n| - m - \frac{1}{2}$, and $\Delta k = 0$ reads in this case $k = m$. Therefore, only the $X^-$ states from the SU$_4$ [4]-multiplet having $J_z = |n| + \frac{1}{2}$ for $\sigma^+$ and $J_z = |n| - \frac{3}{2}$ for $\sigma^-$ can be photogenerated. We see that the combination of the SU$_4$ and orbital selection rules imposes stringent limitations on optical transitions.

As a result, among all of the bound $X^-$ states found near the charge neutrality point, the only bright states are at $\nu = -2 + \epsilon$ with the binding energy $\sim 0.1 E_0$ ($J_z = \frac{1}{2}$, active in $\sigma^+$), see fig. 3(a). Depending on the broadening, this may give rise to a separate absorption peak below the CR or may show up as a low-energy CR shoulder. This is shown in fig. 1, where the spectra are broadened $\sim 0.05 E_0$, which amounts to 2 meV at $B = 10$ T. We conclude that observation of the bright $X^-$ feature is almost feasible in graphene even for the current broadening/disorder experimental parameters such as mobilities of $\sim 17000$ cm$^2$/Vs [5,14,24]. We may compare the binding energy to that found for trions in the conventional two-dimensional electron gas (2DEG). This is also $\sim 0.1 E_0$ [19], but $E_0$ is smaller than it is in graphene, due to a larger value of the dielectric constant $\epsilon$ for the 2DEG.

The oscillator strength of the bright $X^-$ with $J_z = \frac{1}{2}$ needs some clarification. Indeed, the hole-like collective excitations at $\nu = -2 + \epsilon$ originate in the lower cone (inset in fig. 1). As such, they are optically inactive in the electron-like $\sigma^+$ polarization. This holds, however, only in the lowest order in the Coulomb interactions [17]. In fact, the resonant $e^{-1} \cong X_{00-1}$ processes described by the Hamiltonian $\hat{H}_{\text{ab}}$ and indicated in fig. 4 lead to “dressing” of the optical response. As a result, the oscillator strength of the bright $X^-$ becomes comparable to that of the CR mode per electron. Thus, the intensity of the $X^-$ peak relative to the main CR is proportional to the number of excess electrons $\epsilon$ [25–28]. Notice that because of the $e$-$h$ symmetry, there exists the $X^+$ counterpart at $-\nu = 2 - \epsilon$, which is bright in $\sigma^-$ polarization.

The dark bound $X^-$ and $X^+$ states may in principle be observed by tunneling spectroscopy, which is a sensitive tool for probing discrete states in the spectrum. This can be pursued, e.g., in tunneling experiments involving gate-tunable graphene quantum dots [29]. Also, the various symmetry-breaking terms such as external fields, disorder and lattice defects, ripples, and deformations [1,2] may partially lift the limitations following from the orbital and SU$_4$ selection rules. This can make some of the dark states “gray” and detectable in the absorption spectra as well as possibly in photoluminescence [30].

In conclusion, we considered Coulomb correlations in graphene with a low density of excess electrons or holes in partially filled LLs. We have shown that bound negative $X^-$ and positive $X^+$ trions exist in the spectra. With the SU$_4$ symmetry of graphene these are analogous to quark–antiquark complexes. They are different from their counterparts in the conventional 2DEG systems such as GaAs [25–28]. We expect that the predicted additional absorption peak due to the bright $X^-$ below the CR mode should undergo an evolution with filling factor $\epsilon$ of a partially filled level. This collective excitation can serve as an optical probe as its behavior should reflect correlations.
in the electron system [30,31]. This evolution would be especially interesting to follow and study in the vicinity of ε = \frac{1}{2} and other FQHE fractions.

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Additional remark: After completion of this work we became aware of publication [32], where the observation of a plasmon excitation, a zero-field analogue of the X +, has been reported.

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