

Charge $2e$ Skyrmions in Bilayer Graphene

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Quantum Hall states that result from interaction induced lifting of the eightfold degeneracy of the zeroth Landau level in bilayer graphene are considered. We show that at even filling factors electric charge is injected into the system in the form of charge $2e$ Skyrmions. This is a rare example of binding of charges in a system with purely repulsive interactions. We calculate the Skyrmion energy and size as a function of the effective Zeeman interaction and discuss the signatures of the charge $2e$ Skyrmions in the scanning probe experiments.

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The fourfold valley and spin degeneracy of Landau levels (LLs) in monolayer and bilayer graphene, the recently discovered two-dimensional semimetals [1–3], give rise to interesting phenomena at high magnetic fields, where the Coulomb interactions between the electrons become important. In the monolayer, the Coulomb interactions lift the LL degeneracy, giving rise to new spin- and/or valley-polarized incompressible quantum Hall (QH) states [4–6]. The effective Hamiltonian that describes the Coulomb interaction within a single LL is approximately $SU(4)$ symmetric [7,8] with respect to the rotations in the combined spin-valley space. The splitting of the LLs thus corresponds to the spontaneous symmetry breaking of the $SU(4)$ -symmetric quantum Hall ferromagnet (QHFM). The precise order in which spin and valley degeneracy get lifted is determined by the interplay between the Zeeman interaction and valley anisotropy [9,10], both of which are much smaller than the Coulomb interaction. The spin- and valley-polarized QH states in the monolayer were predicted to feature unusual edge states [11,12], as well as spin and valley Skyrmions [13], which are smooth topologically nontrivial textures of the ferromagnetic order parameter that carry the electron charge e [14].

Bilayer graphene features a LL at zero energy, which has a twofold orbital degeneracy: In each valley there are two zero-energy states ($a = 0, 1$), with wave functions corresponding to the ground state and the first excited state of the magnetic oscillator [15]. Taking into account valley and spin degeneracies, the zeroth LL in the bilayer is eightfold degenerate. Coulomb interactions are expected to lift the eightfold degeneracy [16]. In this Letter, we consider the interaction induced QH states at even filling factors and analyze their new properties arising due to the orbital isospin. We shall see that these QH states exhibit interesting collective and topological excitations. In particular, we predict that pairs of charge e excitations bind into Skyrmions that carry charge $2e$. Such a binding of charges is surprising, because the Coulomb interactions between electrons are purely repulsive. Another example

of such a binding was predicted to occur in the spin QHFM with a small Zeeman interaction [17,18]. The weak pairing of Skyrmions considered in Refs. [17,18], however, can occur only when the Zeeman interaction is extremely small; in contrast, charge $2e$ Skyrmions in bilayer graphene can be thought of as robust tightly bound pairs, which exist in a wide range of the effective Zeeman interaction.

Below, we analyze the dependence of Skyrmion energy and size on the effective valley Zeeman interaction, which can be tuned [19,20] by creating a potential difference between the top and bottom layers: $\Delta\nu = eEd$ [see Fig. 1(a)]. Furthermore, we find that slightly away from even filling factors, $|\Delta\nu| = |\nu - 2M| \ll 1$, there is a finite density of charge- $2e$ Skyrmions in the system. At a small density, the Skyrmions form a triangular lattice, while above a critical density $\Delta\nu_*$ they form a bipartite square lattice [21,22].

The zeroth LL states in different valleys reside solely in the opposite layers. Therefore, any valley texture of the order parameter directly translates into a spatial modulation of the charge density in both layers. This results in a spatial modulation of the local density of states (LDOS), which can be probed directly by scanning tunneling microscopy (STM), owing to the fact that the surface of bilayer graphene is exposed. Thus STM provides a way to study the properties of a single charge $2e$ Skyrmion, as well as the properties of a lattice of such Skyrmions. The STM technique has already been employed to study electronic properties of the monolayer graphene with atomic spatial resolution [23,24]. The STM technique enables resolution of the LDOS features on the scale of ~ 1 meV, which should be sufficient to observe the LDOS modulation in the QHFM, whose energy scale is set by the Coulomb interaction, of the order of 10 meV at $B = 10$ T [11].

The effective Coulomb interaction Hamiltonian for the zeroth LL in the bilayer is approximately $SU(4)$ symmetric in the valley-spin space; however, the symmetry in the orbital isospin space is broken due to the different orbital

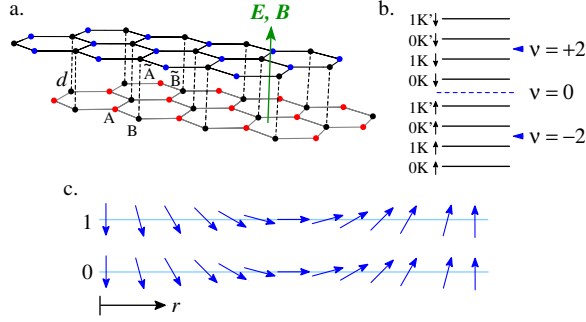


FIG. 1 (color online). (a) Bilayer graphene lattice. Perpendicular electric field E generates effective valley Zeeman interaction $\Delta_v = eEd$, where $d = 0.34$ nm is the separation between the layers. (b) The order of the zeroth LL splitting, assuming that effective valley Zeeman interaction Δ_v favors K valley states and $\Delta_v < E_z$. (c) Texture corresponding to the charge $2e$ Skyrmion at $\nu = \pm 2$. Vectors illustrate the rotation of the order parameter in the valley space.

wave functions of the two states [16]. This results in the following picture of the zeroth LL splitting: At even filling factors ($\nu = 2M$ filled sublevels) M pairs of orbital states with the same valley and spin are filled, while the states at odd filling factors $\nu = 2M + 1$ are obtained from the $\nu = 2M$ QH state by filling one of the remaining states with orbital isospin $a = 0$. This order of the zeroth LL splitting is due to two facts: (i) Exchange energy within the LL with isospin $a = 0$ is higher than that for the LL with isospin $a = 1$; (ii) there is exchange energy between filled $a = 0$ and $a = 1$ LLs with the same spin and valley, which makes the energy of the state where $a = 0, 1$ LLs with the same spin and valley are filled (e.g., $0K \uparrow$ and $1K \uparrow$) lower than the energy of a state polarized in the orbital space along the $a = 0$ direction (e.g., $0K \uparrow, 0K' \uparrow$ LLs are filled). The order in which valley and spin degeneracies get lifted is determined by the competition between the symmetry-breaking terms: the Zeeman interaction $E_z = g\mu_B B$ and the effective valley Zeeman interaction Δ_v . In the experiment Δ_v is typically small, and it can be tuned by gates [20]. We assume that Δ_v is tuned to be smaller than E_z . Furthermore, we assume that Δ_v is nonzero and favors the K valley [25]. This leads to the splitting picture illustrated in Fig. 1(b). In the following, we shall be especially interested in the states at filling factors $\nu = -2, +2$, marked by arrows in Fig. 1(b). Since these two states are related by the particle-hole symmetry, we shall focus on the $\nu = -2$ state.

We start with recalling the Landau level spectrum in the bilayer graphene [15]. The low-energy excitations near the K, K' point are described by the Schrödinger equation $\varepsilon \psi_{K,K'} = H_{K,K'} \psi_{K,K'}$, with the Hamiltonian given by

$$H_{K,K'} = -\frac{1}{2m} \begin{bmatrix} 0 & \pi^{\dagger 2} \\ \pi^2 & 0 \end{bmatrix} + \hat{h}_w, \quad \hat{h}_w = \xi v_3 \begin{bmatrix} 0 & \pi \\ \pi^\dagger & 0 \end{bmatrix}, \quad (1)$$

where $\pi = p_x - ip_y$. For the K valley, the upper (lower)

component of the wave function corresponds to the amplitude on the sublattice A (\tilde{B}) [see Fig. 1(a)], which belongs to the bottom (top) layer; for the K' valley, the order of components is reversed. The effective mass m can be expressed in terms of the $\tilde{A}\tilde{B}$ interlayer hopping amplitude $\gamma_1 \approx 0.39$ eV and the Fermi velocity in the monolayer $v_F \approx 10^6$ m/s, $m = \gamma_1/2v_F^2$; $\xi = +1$ (-1) for the K' (K) valley. The trigonal warping term \hat{h}_w originates from the weak direct $\tilde{A}\tilde{B}$ hopping $\gamma_3 \ll \gamma_1$ [15], with the effective velocity given by $v_3 = (\sqrt{3}/2)\gamma_3 a$, $a = 0.142$ nm.

To analyze the Landau level spectrum, we choose the Landau gauge $A_y = Bx$, $A_x = 0$, for which the eigenstates can be classified according to the value of the wave vector k_y : $\psi_{K,K'}(x, y) = e^{ik_y y} \psi_{K,K'}(x)$. The wave vector k_y translates into the guiding center position $X = k_y \ell_B^2$, where $\ell_B = \sqrt{\hbar c/eB}$ is the magnetic length. Below, for simplicity we shall choose units where $\ell_B = 1$. First let us ignore the small trigonal warping term. Then the effective 1D Hamiltonian for $\psi_{K,K'}(x)$ takes the following form:

$$H_{K,K'} = -\hbar\omega_c \begin{bmatrix} 0 & a_X^2 \\ a_X^{\dagger 2} & 0 \end{bmatrix}, \quad a_X = i[\partial_x + (x - X)], \quad (2)$$

where $\omega_c = eB/mc$ is the cyclotron energy. The Hamiltonian (2) has two zero modes with the following wave functions: $\psi_{K,K'}^a(x, y) = e^{ixy} \varphi_{a,X}(x)$, $a = 0, 1$, where $\varphi_{a,X}(x)$ denotes the a th excited level of the magnetic oscillator. Below, we shall denote the annihilation operators of the zero modes by $c_{a,\kappa,X}$, $\kappa = K, K'$. Including the trigonal warping term \hat{h}_w does not change the energies of the zero modes [15]. The corrections to the wave functions of the zero modes due to \hat{h}_w are proportional to $(v_3/v_F) \times \sqrt{\gamma_1/\hbar\omega_c}$ [22]; for experimentally relevant field values $B \sim 20$ T, the square root term is of the order of one, so the corrections are negligible due to the smallness of the ratio $v_3/v_F \ll 1$.

Now we proceed to the analysis of the zeroth LL splitting. We neglect the LL mixing, which allows us to project the Coulomb Hamiltonian onto the zeroth LL:

$$H_{\text{int}} = \frac{1}{2S} \sum_{\mathbf{q}\kappa\kappa'} V(\mathbf{q}) \rho_\kappa(\mathbf{q}) \rho_{\kappa'}(-\mathbf{q}), \quad (3)$$

where $\rho(\mathbf{q})$ are the density operators restricted to the zeroth LL, $S = L_x L_y$ is the sample volume, κ and κ' are valley indices, and the matrix element is given by $V(\mathbf{q}) = \frac{2\pi e^2}{\varepsilon q}$. The projected density components are given by

$$\rho_\kappa(\mathbf{q}) = \sum_{a,b} F_{ab}(\mathbf{q}) \bar{\rho}_\kappa^{ab}, \quad (4)$$

$$\bar{\rho}_\kappa^{ab}(\mathbf{q}) = \sum_{\bar{X}} \exp(iq_x \bar{X}) c_{a,\kappa,\bar{X}}^\dagger c_{b,\kappa,\bar{X}},$$

where $X_\pm = \bar{X} \pm \frac{q_y}{2}$, $F_{00}(\mathbf{q}) = e^{-q^2/4}$ and $F_{11}(\mathbf{q}) = (1 - q^2/2)e^{-q^2/4}$ are the usual form factors for the lowest LL and the first excited LL, respectively, and $F_{01}(\mathbf{q}) = -[(q_y + iq_x)/\sqrt{2}]e^{-q^2/4}$ and $F_{10}(\mathbf{q}) = [(q_y - iq_x)/\sqrt{2}]e^{-q^2/4}$ are the

form factors corresponding to the density components which mix the two orbital states.

We now analyze the order in which the eightfold degeneracy of the LL gets lifted. The split QH states with filling factor $|\nu| \leq 3$ ($\nu + 4$ filled sublevels) correspond to the following wave functions:

$$|\Psi_\nu\rangle = \prod_{i=1}^{\nu+4} \prod_X d_{i,X}^\dagger |\Omega\rangle, \quad d_{i,X}^\dagger = \sum_{a,\kappa,s} \bar{U}_{a,\kappa,s}^i c_{a,\kappa,s,X}^\dagger, \quad (5)$$

where \bar{U} is a unitary matrix, s is the electron spin, and Ω is an empty zeroth LL.

To find the ground state for $\nu = -2$, we compare the energies of two states: (i) Two $a = 0$ LLs with different valley and/or spin indices are filled; for example, $d_1^\dagger = c_{0,K,\uparrow}^\dagger$, $d_2^\dagger = c_{0,K',\uparrow}^\dagger$, and (ii) $a = 0$ and $a = 1$ LLs with the same valley and spin indices are filled; $d_1^\dagger = c_{0,K,\uparrow}^\dagger$, $d_2^\dagger = c_{1,K,\uparrow}^\dagger$. The energy of the first state is twice the exchange energy of a nondegenerate lowest LL:

$$\langle H_{\text{int}} \rangle_1 = -2N\Delta_0, \quad \Delta_0 = \frac{1}{2} \sqrt{\frac{\pi}{2}} \frac{e^2}{\epsilon \ell_B}, \quad (6)$$

where N is the total number of states in one nondegenerate LL. Averaging the Coulomb interaction over the second state, we obtain $\langle H_{\text{int}} \rangle_2 = -\frac{11}{4} N\Delta_0$. Thus the energy of the second state is lower than the energy (6) of the first state, and the spin- and valley-polarized state $|\psi_0\rangle$ is the ground state at $\nu = -2$. The state at $\nu = +2$ can be obtained from $|\psi_0\rangle$ by charge conjugation.

Now we proceed to the excitations of the $\nu = -2$ QH state. The lowest-energy electron-hole pair at $\nu = -2$ is obtained by moving an electron with orbital isospin $a = 1$ from the filled LL into one of the empty LLs. The energy of such an excitation, $E_{\text{eh}} = \frac{7}{2} \Delta_0$, is lower than the energy $E_{\text{eh}}' = 4\Delta_0$ of a particle-hole excitation that is obtained by removing an electron with isospin $a = 0$.

In some QHFM, the lowest-energy charge excitations are Skyrmions, which are topologically nontrivial smooth textures of the order parameter [14]. On the qualitative level, the textures carry charge because the charge and spin or valley dynamics in the QHFM are entangled [14].

Can Skyrmions exist in bilayer graphene? Skyrmions of charge e are energetically unfavorable because they involve flipping valley isospin (or spin) for either $a = 0$ or $a = 1$ states in some region, and in that region the filled $a = 0$ and $a = 1$ states would have different valley isospin (spin), which leads to a loss of the exchange energy Δ_0 per flipped valley isospin.

Another possibility is Skyrmions of charge $2e$, which can be created by making two identical valley textures for $a = 0$ and $a = 1$ orbital states. Such textures are described by a unit vector \mathbf{n} , with $n_z = -1$ (+1) corresponding to filling K (K') states. On the intuitive level, we expect such textures to be energetically favorable: Since $a = 0$ and $a = 1$ orbital states rotate simultaneously, no exchange between 0 and 1 states is lost. Below, we find the energy

of the $2e$ Skyrmion and, by comparing it to the energies of the single-particle excitations, establish that such Skyrmions are indeed energetically favorable.

Before we proceed to the quantitative analysis of charge $2e$ Skyrmions, let us compare excitations at the even and odd filling factors. For simplicity, let us consider the excitations of the state $\nu = -3$, which corresponds to filled $0K\uparrow$ LL. The lowest-energy electron-hole pair is obtained by removing an electron from the $0K\uparrow$ LL and putting it in the $1K\uparrow$ LL; owing to the exchange between $0K\uparrow$ and $1K\uparrow$ states, the energy of such an excitation, $E_{\text{eh}}^{\text{odd}} = \Delta_0$, is lower than the energy $\bar{E}_{\text{eh}}^{\text{odd}} = 2\Delta_0$ of an excitation where the excited electron resides in a LL with a different valley and/or spin index. The existence of orbital Skyrmions at $\nu = -3$ is unlikely, because such Skyrmions correspond to filling $a = 1$ states in some region, which leads to a loss of the exchange energy equal to $\Delta_0/4$ per flipped orbital isospin.

Now we briefly describe the calculation of the energy of a charge $2e$ Skyrmion. As a first step, we derive an effective Hamiltonian describing valley textures of the order parameter:

$$|\psi\rangle = e^{-i\hat{O}} |\psi_0\rangle. \quad (7)$$

In our analysis, we follow the microscopic approach developed in Ref. [26]; however, the dynamics of the order parameter in the bilayer graphene is richer than that in the case of $SU(2)$ - and $SU(4)$ -symmetric QHFM, owing to the presence of the orbital degree of freedom. We parametrize the rotation operator \hat{O} as follows:

$$\hat{O} = \sum_{\mathbf{q},a,b,\mu} \Omega_{ab}^\mu(\mathbf{q}) \hat{S}_{ab}^\mu(-\mathbf{q}), \quad (8)$$

$$\hat{S}_{ab}^\mu(-\mathbf{q}) = \sum_{\bar{X}} e^{iq\bar{X}} \frac{\tau_{\kappa\kappa'}}{2} c_{a,\kappa,X}^\dagger c_{b,\kappa',X_+}, \quad (9)$$

where τ are the Pauli matrices. The rotation (8) is described by four complex parameters:

$$\begin{aligned} u_a &= \Omega_{aa}^x + i\Omega_{aa}^y, & a &= 0, 1, \\ v &= \Omega_{10}^x + i\Omega_{10}^y, & w &= \Omega_{01}^x + i\Omega_{01}^y. \end{aligned} \quad (10)$$

The parameters $u_0(u_1)$ correspond to rotations that involve $0K$ and $0K'$ ($1K$ and $1K'$) states, while v and w parametrize rotations which transform $0K$ into $1K'$, and $1K$ into $0K'$, and vice versa. To simplify calculations, we assume that the rotations are small ($|u_a| \ll 1$, $|v| \ll 1$, $|w| \ll 1$). Then we can expand the texture energy $E = \langle \psi_0 | e^{i\hat{O}^\dagger} H e^{-i\hat{O}} | \psi_0 \rangle - \langle \psi_0 | H | \psi_0 \rangle$ in series in the powers of \hat{O} . This procedure yields an effective action for general textures that involve u_a , v , and w [27]. As a next step, we restrict our attention to the low-energy excitations, where $a = 0$ and $a = 1$ states are rotated simultaneously in the valley space [see Fig. 1(c)], which corresponds to setting $u_0 = u_1 = u$. Integrating out v and w variables [27], we obtain the energy of the low-energy textures. For what follows, we rewrite the resulting

energy functional in terms of the $O(3)$ order parameter $\mathbf{n} = (-u_y, u_x, 0)$:

$$E_{\text{st}} = \frac{\rho_s}{2} \int d^2r (\partial_\mu \mathbf{n})^2, \quad \rho_s = \frac{25}{64\pi} \Delta_0. \quad (11)$$

Although we derived the above equation assuming that \mathbf{n} deviates slightly from $\mathbf{n} = (0, 0, -1)$, due to the rotational invariance in the valley space Eq. (11) is valid for any slowly varying configuration of the order parameter.

As our next step, we evaluate the charge density of the texture $\delta\rho = \langle \psi_0 | e^{i\hat{\rho}} \hat{\rho} e^{-i\hat{\rho}} | \psi_0 \rangle - \langle \psi_0 | \hat{\rho} | \psi_0 \rangle$, where $\hat{\rho}$ is the density operator. We find that the charge density is twice the Pontryagin index density:

$$\delta\rho(\mathbf{r}) = 2e p(\mathbf{r}), \quad p(\mathbf{r}) = -\frac{1}{8\pi} \varepsilon_{\mu\nu} (\mathbf{n} [\partial_\mu \mathbf{n} \times \partial_\nu \mathbf{n}]). \quad (12)$$

This relation differs from the usual $SU(2)$ QHFM case [14,26] by a factor of 2, which corresponds to the fact that the texture rotates states in both $a = 0, 1$ LLs.

Apart from the stiffness term (11), there are two other contributions to the texture energy: the valley Zeeman term and the long-range Coulomb interaction:

$$H_z = \Delta_v n_0 \int d^2r n_z, \quad (13)$$

$$H_{\text{coul}} = \frac{1}{2} \int d^2r d^2r' \frac{\delta\rho(\mathbf{r})\delta\rho(\mathbf{r}')}{\varepsilon|\mathbf{r} - \mathbf{r}'|},$$

where $n_0 = 1/2\pi\ell_B^2$ is the LL density of states.

The simplest topologically nontrivial texture of the order parameter \mathbf{n} has topological charge 1 and an electric charge $\pm 2e$. This is to be contrasted with the usual Skyrmions [14], which carry charge $\pm e$. In the limit of vanishing Δ_v , the Coulomb repulsion forces Skyrmions to be infinitely large. Then the Skyrmion energy is determined solely by the stiffness term

$$E_{\text{sk}} = 4\pi\rho_s = \frac{25}{16} \Delta_0. \quad (14)$$

The energy of the Skyrmion–anti-Skyrmion pair, $2E_{\text{sk}} = 25\Delta_0/8$, is lower than the energy of two electron-hole pairs, which equals $7\Delta_0$. Therefore, in the limit $\Delta_v \rightarrow 0$, pairs of electron (hole) excitations bind into charge $2e$ Skyrmions (anti-Skyrmions).

At finite Δ_v the Skyrmion size l_s is determined by the competition between the effective valley Zeeman and Coulomb energies [14]. Optimizing the Skyrmion energy with respect to l_s , we find with logarithmic precision

$$\frac{l_s}{\ell_B} \approx \left(\frac{9\pi^2}{32} \right)^{1/3} \tilde{\Delta}_v^{-1/3} |\log \tilde{\Delta}_v|^{-1/3}, \quad (15)$$

where $\tilde{\Delta}_v = \Delta_v / (e^2 / \varepsilon \ell_B)$. The Skyrmion energy is increased compared to the case $\Delta_v = 0$, $E_{\text{sk}}(\tilde{\Delta}_v) = \frac{25}{16} \Delta_0 + A \Delta_0 \tilde{\Delta}_v^{1/3} |\log \tilde{\Delta}_v|^{1/3}$, where $A = \frac{3^{4/3} \pi^{5/6}}{2^{11/6}}$.

We now briefly address experimental manifestations of the charge $2e$ Skyrmions. STM can be used to study the Skyrmion size (15) as a function of the valley Zeeman interaction, as well as the properties of the Skyrme lattice as a function of filling factor and valley Zeeman interaction. Finally, we note that, in samples with long-range disorder, the charge- $2e$ Skyrmions will lead to an even-odd asymmetry in charging spectra of individual disorder-induced quantum dots, which can be studied in scanning single-electron transistor experiments [28].

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