Landau level degeneracy and quantum Hall effect in a graphite bilayer

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We derive an effective two-dimensional Hamiltonian to describe the low energy electronic excitations of a graphite bilayer, which correspond to chiral quasiparticles with a parabolic dispersion exhibiting Berry phase $2\pi$. Its high-magnetic-field Landau level spectrum consists of almost equidistant groups of four-fold degenerate states at finite energy and eight zero-energy states. This can be translated into the Hall conductivity dependence on carrier density, $\sigma_{xy}(N)$, which exhibits plateaus at integer values of $4e^2/h$ and has a “double” $8e^2/h$ step between the hole and electron gases across zero density, in contrast to $(4n+2)e^2/h$ sequencing in a monolayer.

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For many decades, the electronic properties of a graphite monolayer have attracted theoretical interest due to a Dirac-type spectrum of charge carriers \( \mathbf{1, 2} \). Recently Novoselov \textit{et al.} \( \mathbf{3} \) fabricated ultra-thin graphitic devices including monolayer structures. This was followed by further observations \( \mathbf{4, 10, 11} \) of the classical and quantum Hall effects (QHE) in such systems confirming the expectations \( \mathbf{3} \) of an unusual phase of Shubnikov de Haas oscillations and QHE plateaus sequencing, as manifestations of a peculiar magneto-spectrum of chiral Dirac-type quasiparticles containing a Landau level at zero energy \( \mathbf{1} \).

In this Letter we show that quasiparticles in a graphite bilayer display even more intriguing properties including a peculiar Landau level (LL) spectrum: these are chiral quasiparticles exhibiting Berry phase $2\pi$, with a dominantly parabolic dispersion and a double-degenerate zero-energy LL incorporating two different orbital states with the same energy. Taking into account spin and valley degeneracies, the zero-energy LL in a bilayer is 8-fold degenerate, as compared to the 4-fold degeneracy of other bilayer states and the 4-fold degeneracy of all LLs in a monolayer. The structure and degeneracies of the Landau level spectrum in a bilayer determine a specific sequencing of plateaus in the density dependence of the QHE conductivity $\sigma_{xy}(N)$ which is distinguishably different from that of Dirac-type quasiparticles in a graphite monolayer and of non-chiral carriers in conventional semiconductor structures.

We model a graphite bilayer as two coupled hexagonal lattices including inequivalent sites $A, B$ and $\tilde{A}, \tilde{B}$ in this gapless semiconductor \( \mathbf{7} \). \( \mathbf{8} \) The low energy states of electrons are described by

$$\hat{H}_2 = -\frac{1}{2m} \left( \begin{array}{cc} 0 & (\pi \gamma^\dagger)^2 \\ (\pi \gamma)^2 & 0 \end{array} \right) + \hat{h}_w + \hat{h}_a;$$

$$\hat{h}_w = \xi \nu_3 \left( \begin{array}{cc} 0 & \pi \\ \pi \dagger & 0 \end{array} \right), \quad \text{where} \quad \pi = p_x + ip_y;$$

$$\hat{h}_a = \xi u \left[ \frac{1}{2} \left( \begin{array}{cc} 1 & 0 \\ 0 & -1 \end{array} \right) - \frac{v^2}{\gamma_1^2} \left( \begin{array}{cc} \pi \dagger \pi & 0 \\ 0 & -\pi \dagger \pi \end{array} \right) \right].$$

The effective Hamiltonian $\hat{H}_2$ operates in the space of two-component wave functions $\Phi$ describing electronic amplitudes on $A$ and $B$ sites and it is applicable within the energy range $|\epsilon| < \frac{1}{2} \gamma_1$. In the valley $K$, $\xi = +1$, we determine $\Phi_{\xi=+1} = (\phi(A), \phi(B))$, whereas in the valley $\tilde{K}$, $\xi = -1$ and the order of components is reversed, $\Phi_{\xi=-1} = (\phi(B), \phi(A))$. Here, we take into account two possible ways of $A \Rightarrow B$ hopping: via the dimer state (the main part) or due to a weak direct $AB$ coupling, $\gamma_{AB} \equiv \gamma_3 \ll \gamma_{AB}$ (the term $\hat{h}_a$). They determine the mass $m = \gamma_1/2\nu_3^2$ and velocity $v_3 = (\sqrt{3}/2) a \gamma_{AB}/\hbar$. Other weaker tunneling processes \( \mathbf{12} \) are neglected. The term $\hat{h}_a$ takes into account a possible asymmetry between top and bottom layers (thus opening a mini-gap $\sim u$).
For comparison, the monolayer Hamiltonian \( \hat{H}_1 \),
\[
\hat{H}_1 = \xi v \left( \begin{array}{cc} 0 & \pi \hat{1} \\ \pi & 0 \end{array} \right) \equiv \xi v (\sigma_x p_x + \sigma_y p_y),
\]
is dominated by nearest neighbor intralayer hopping \( \gamma_{AB} = \gamma_{BA} \equiv \gamma_0 \gg \gamma_1 \)
(i.e., in an electron gas with a small density at a very low magnetic field) whereas the energy spectrum within the interval \( \frac{1}{2} \gamma_1 (v_3/v)^2 < |\epsilon| < \frac{1}{2} \gamma_1 \)
is dominated by nearest neighbor intralayer hopping. For equivalent parameters in bulk graphite \( \gamma_0 \), \( v_3 \ll v \) in Eq. (\ref{eq:magnetic_field}). Thus, the linear term \( \hat{h}_{\text{nr}} \), which is similar to \( \hat{H}_1 \), is relevant only for very small electron momenta and thus the Fermi energy in a 2D gas with density \( N \), \( m_e = p/(\partial \epsilon_1 / \partial p) \equiv (\gamma_1/2m)^2 \sqrt{1 + 4\pi^2 e^2 N/\gamma_1^2} \). The relation in Eq. (\ref{eq:magnetic_field}) interpolates between a linear spectrum \( \epsilon_1 \approx vp \) at high momenta and a quadratic spectrum \( \epsilon_1 \approx p^2/2m \), where \( m = \gamma_1/2v \). Such a crossover happens at \( p \approx \gamma_1/2v \), which corresponds to the carrier density \( N^* \approx \gamma_1^2/(4\pi^2 e^2 v^2) \). The experimental graphite values \( \gamma_1 \), \( 10 \)
give \( N^* \approx 4.36 \times 10^{12} \text{cm}^{-2} \), whereas the dimer band \( \epsilon_2 \) becomes occupied only if the carrier density exceeds \( N^*(2) \approx 2\gamma_2^2/(4\pi^2 e^2 v^2) \approx 8\pi^2 \approx 3.39 \times 10^{13} \text{cm}^{-2} \). The estimated effective mass \( m \) is light: \( m \approx 0.05m_e \) using the bulk graphite values \( \gamma_1 \), \( 10 \).

The \( 4 \times 4 \) Hamiltonian \( \hat{H} \) contains information about the higher energy band \( \epsilon_2 \), and, therefore, is not convenient for the analysis of transport properties of a bilayer which are formed by carriers in the low energy band \( \epsilon_1 \). We separate \( \hat{H} \) into \( 2 \times 2 \) blocks, where the upper left diagonal block is \( \hat{H}_{11} \equiv \xi \left( \begin{array}{cc} p_x & \epsilon_1 \gamma_0 \\ \epsilon_1 \gamma_0 & p_y \end{array} \right), \)
the lower right diagonal block is \( \hat{H}_{22} = -\epsilon_1 \gamma_0 \), and the off-diagonal blocks are \( \hat{H}_{12} = \hat{H}_{21} = \epsilon_1 \gamma_0 \). Then, we take the \( 4 \times 4 \) Green function determined by \( \hat{H}_1 \), evaluate the block \( G_{11} \) related to the lower-band states, and use it to identify the effective low-energy bilayer Hamiltonian \( \hat{H}_2 \). Using \( G_{0a} = (H_{\alpha\alpha} - \epsilon)^{-1} \), we write
\[
G = \left( \begin{array}{cc} G_{11} & G_{12} \\ G_{21} & G_{22} \end{array} \right) = \left( \begin{array}{cc} G_{11}^{-1} & H_{12} \\ H_{21} & G_{22}^{-1} \end{array} \right)^{-1} \equiv (\hat{H} - \epsilon)^{-1}.
\]

Then, in the Fermi energy range, \( \gamma_1 \), \( \epsilon \), \( \gamma_1 \)
the spectrum determined by \( \hat{H}_2 \) in Eq. (\ref{eq:magnetic_field}) agrees with \( \epsilon_1 (p) \)
found using the \( 4 \times 4 \) Hamiltonian \( \hat{H}_1 \). Similarly to bulk graphite \( \gamma_1 \), \( 10 \)
the effect of \( \hat{h}_{\text{nr}} \) consists of trigonal warping, which deforms the isoenergetic lines along the directions \( \varphi = \varphi_0 \). For the valley \( K \), \( \varphi_0 = 0 \), \( \varphi_0 \), \( \frac{\pi}{3} \), \( \varphi_0 \), whereas for \( K \), \( \varphi_0 = \pi \), \( \frac{4\pi}{3} \), \( \varphi_0 \). At the lowest energies \( |\epsilon| < \frac{1}{2} \gamma_1 (v_3/v)^2 \), trigonal warping breaks the isoenergetic line into four pockets, which can be referred to as one “central” and three “leg” parts. The central part and leg parts have minimum \( \epsilon \) at \( p = 0 \) and at \( |p| = \gamma_1 v_3/v^2 \), angle \( \varphi_0 \), respectively. For \( v_3 \approx 0.1v \), we find (using the data in Ref. \( \gamma_1 \)) that the separation of a 2D Fermi line into four pockets would take place for very small carrier densities

\[
\epsilon_2 \approx \pm \frac{1}{2} \gamma_1 \left[ 1 + 4v^2 p^2/\gamma_1^2 - 1 \right].
\]
N < N_c = 2(v_3/v)^2 N^* \sim 1 \times 10^{11} \text{ cm}^{-2}$. For $N < N_c$, the central part of the Fermi surface is approximately circular with area $A_c = \pi \varepsilon_c^2/(\hbar v_3)^2$, and each leg part is elliptical with area $A_l \approx \frac{4}{3} A_c$. This determines the following sequencing of the first few LL’s in a low magnetic field, $B \ll B_c \approx \hbar N_c/4e \sim 1T$. Every third Landau level from the central part has the same energy as levels from each of the leg pockets, resulting in groups of four degenerate states. These groups of four would be separated by two non-degenerate LLs arising from the central pocket.

In structures with densities $N > N_c$ or for strong magnetic fields $B > B_c$, the above described LL spectrum evolves into an almost equidistant staircase of levels. We derive such a spectrum numerically from Eq. (1) using the Landau gauge $A = (0, Bx)$, in which operators $\pi^\dagger$ and $\pi$ coincide with raising and lowering operators in the basis of Landau functions $e^{i k_B \phi_n(x)}$, such that $\pi^\dagger \phi_n = i(\hbar/\lambda_B) \sqrt{2(n+1)} \phi_{n+1}$, $\pi \phi_n = -i(\hbar/\lambda_B) \sqrt{2n} \phi_{n-1}$, and $\pi \phi_0 = 0$, where $\lambda_B = \sqrt{\hbar/(eB)}$. In this we followed an approach applied earlier to bulk graphite by using the bulk parameters for intralayer $v$ and interlayer $\gamma_1$, varying the value of the least known parameter $v_3$. The spectrum for the valley $K$ ($\xi = 1$) is shown in Fig. 3 as a function of the ratio $v_3/v$ for two different fields. Fig. 3(a) shows the evolution of the twenty lowest levels for $B = 0.17T$ as a function of $v_3$, illustrating the above-mentioned crossover from an equidistant ladder at $v_3 = 0$ to groups of pocket-related levels.

The LL spectrum obtained for $B = 1T$, Fig. 3(b) remains independent of $v_3$ over a broad range of its values. Hence, even in the absence of a definite value of $v_3$, we are confident that the LL spectrum in bilayers studied over the field range where $\hbar \lambda_B^{-1} > v_3 m$ can be adequately described by neglecting $v_3$, thus using an approximate Hamiltonian given by the first term in $H_2$, Eq. (1). The resulting spectrum contains almost equidistant energy levels which are weakly split in valleys $K$ and $\bar{K}$.

\[
\varepsilon_n = \pm \hbar \omega_c \sqrt{n(n-1)} - \frac{1}{2} \xi \delta, \quad \text{for } n \geq 2,
\]

\[
\Phi_{n\xi} = C_{n\xi}(\phi_n, D_{n\xi} \phi_{n-2}), \quad \delta = u \hbar \omega_c / \gamma_1.
\]

Here, $\omega_c = eB/m$, $\varepsilon_n^\pm$ and $\varepsilon_n^\mp$ are assigned to electron and hole states, respectively, and $D_{n\xi} = [\varepsilon - \xi u + \xi n \delta]/(\hbar \omega_c \sqrt{n(n-1)})$, $C_{n\xi} = 1/\sqrt{1 + |D_{n\xi}|^2}$. In the limit of valley ($u = 0$) and spin degeneracies, we shall refer to these states as 4-fold degenerate LLs.

The LL spectrum in each valley also contains two levels identified using the fact that $\pi^2 \phi_1 = \pi^2 \phi_0 = 0$,

\[
\left\{ \begin{array}{l}
\varepsilon_0 = \frac{\xi}{2} \xi u; \\
\varepsilon_1 = \frac{\xi}{2} \xi u - \xi \delta;
\end{array} \right.
\]

\[
\Phi_{0\xi} = (\phi_0, 0); \\
\Phi_{1\xi} = (\phi_1, 0).
\]

According to different definitions of two-component $\Phi$ in two valleys, $n = 0, 1$ LL states in the valley $K$ are formed by orbitals predominantly on the $A$ sites from the bottom layer, whereas the corresponding states in the valley $\bar{K}$ are located on $\bar{B}$ sites from the top layer, which is reflected by the splitting $u$ between the lowest LL in the two valleys. In a symmetric bilayer ($u = 0$) levels $\varepsilon_0$ and $\varepsilon_1$ are degenerate and have the same energy in valleys $K$ and $\bar{K}$, thus forming an 8-fold degenerate LL at $\varepsilon = 0$ (here, spin is taken into account). Also, note that the spectrum of high-energy LLs, Eq. (4) is applicable in such fields that $\hbar \lambda_B^{-1} < \gamma_1/2v$. For higher fields the full two-band Hamiltonian $\hat{H}$ has to be used to determine the exact LL spectrum, nevertheless, the 8-fold degeneracy of the zero-energy LL remains unchanged.

The group of 8 states at $|\varepsilon| = 0$ (4 for electrons and 4 for holes, Eq. (11) embedded into the ladder of 4-fold degenerate LL’s with $n \geq 2$, Eq. (3) is specific to the magneto-spectrum of $J = 2$ chiral quasiparticles. It
would be reflected by the Hall conductivity dependence on carrier density, $\sigma_{xy}(N)$ shown in Fig. 4. A solid line sketches the form of the QHE $\sigma_{xy}^{(2)}(N)$ in a bilayer which exhibits plateaus at integer values of $4e^2/h$ and has a “double” $8e^2/h$ step between the hole and electron gases across $N = 0$ that would be accompanied by a maximum in $\sigma_{xx}$. Figure 4 is sketched assuming that temperature and the LL broadening hinder small valley and spin splittings as well as the splitting between $n = 0,1$ electron/hole LL’s in Eqs. 17, so that the percolating states 18 from these levels would not be resolved. To compare, a monolayer has a spectrum containing 4-fold (spin and valley) degenerate LL’s 19, $\varepsilon_0 = 0$ and $\varepsilon_n^{\pm} = \pm\sqrt{2n\hbar v/\lambda_{\parallel}}$ shown on the r.h.s of Fig. 4 which corresponds to Hall conductivity $\sigma_{xy}(N)$ exhibiting plateaus at $(4n+2)e^2/h$ (dotted line 20), as discussed in earlier publications 19.

The absence of a $\sigma_{xy} = 0$ plateau in the QHE accompanied by the maximum in $\sigma_{xx}$ in the vicinity of zero density is the result of the existence of the zero-energy LL, which is the fingerprint of a chiral nature of two-dimensional quasiparticles. This contrasts with a gradual freeze-out of both Hall and dissipative conductivities in semiconductor structures upon their depletion. Having compared various types of density dependent Hall conductivity, we suggest that two kinds of chiral (Berry phase $\pi \ell\pi$) quasiparticles specific to monolayer ($J = 1$) and bilayer ($J = 2$) systems can be distinguished on the basis of QHE measurements. It is interesting to note that the recent Hall effect study of ultra-thin films by Novoselov et al. 10 featured both types of $\sigma_{xy}(N)$ dependence shown in Fig. 4.

It is also worth mentioning that the 8-fold degeneracy of the group of $\epsilon = 0$ LL’s in a bilayer, Eqs. 17, is quite unusual in 2D systems. It suggests that e-e interaction in a bilayer may give rise to a variety of strongly correlated QHE states. For structures studied in Ref. 10, with electron/hole densities $N \sim 10^{12}$cm$^{-2}$, such a regime may be realized in fields $B \sim 10T$.

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[1] J.W. McClure, Phys. Rev. 104, 666 (1956).
[2] D. DiVincenzo and E. Mele, Phys. Rev. B 29, 1685 (1984).
[3] F.D.M. Haldane, Phys. Rev. Lett. 61, 2015 (1988).
[4] Y. Zheng and T. Ando, Phys. Rev. B 65, 245420 (2002).
[5] V. Gusynin and S. G. Sharapov, Phys. Rev. Lett. 95, 146801 (2005).
[6] A.H. Castro Neto, F. Guinea, and N. Peres, cond-mat/0509079
[7] N. Peres, F. Guinea, and A.H. Castro Neto, cond-mat/0512011.
[8] C. de C. Chamon, C. Mudry, and X.-G. Wen, Phys. Rev. Lett. 77, 4194 (1996).
[9] D.H. Kim, P.A. Lee, X.G. Wen, Phys. Rev. Lett. 79, 2109 (1997).
[10] Y. Hatsugai and T. Ando, Phys. Rev. B 48, 4204 (1993).
[11] D.V. Khveshenko, Phys. Rev. Lett. 87, 206401 (2001).
[12] T. Stauber, F. Guinea, and M.A.H. Vozmediano, Phys. Rev. B 71, 041405 (2005).
[13] M.S. Dresselhaus and G. Dresselhaus, Adv. Phys. 51, 1 (2002).
[14] R.C. Tatar and S. Rabii, Phys. Rev. B 25, 4126 (1982).
[15] J.C. Charlier, X. Gonze, and J.-P. Michenaud, Phys. Rev. B 43, 4579 (1991).
[16] We quote $\gamma_\parallel = 0.39eV$ and $\gamma_\perp = 0.315eV$, neglect the weakest coupling $\gamma_{4A} = \gamma_{4B} = \gamma_{4C} = 0.044eV$, and use the experimental values $v \approx 8.0 \times 10^4m/s$ from Ref. 17.
[17] K.S. Novoselov et al., Science 306, 666 (2004).
[18] K.S. Novoselov et al., PNAS 102, 10451 (2005).
[19] J.S. Bunch et al., Nano. Lett. 5, 287 (2005).
[20] Note that two parallel equivalent monolayers would display Hall conductivity $2\sigma_{xy}(N)$, thus missing every sec-

![FIG. 4: Landau levels for a bilayer (left) and monolayer (right). Brackets $(n,\xi)$ indicate LL number $n$ and valley index $\xi = \pm 1$. In the center the predicted Hall conductivity $\sigma_{xy}^{(2)}$ as a function of carrier density for bilayer (solid line) is compared to that of a monolayer (dashed line).]
ond plateau, as compared to $\sigma^{(2)}_{xy}(N)$. 