The influence of interlayer asymmetry on the magnetospectroscopy of bilayer graphene

M. Mucha-Kruczyński, E. McCann*, V.I. Fal’ko
Department of Physics, Lancaster University, Lancaster, LA1 4YR, UK

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A B S T R A C T
We present a self-consistent calculation of the interlayer asymmetry in bilayer graphene caused by an applied electric field in magnetic fields. We show how this asymmetry influences the Landau level spectrum in bilayer graphene and the observable inter-Landau level transitions when they are studied as a function of high magnetic field at fixed filling factor as measured experimentally in [E.A. Henriksen, Z. Jiang, L.-C. Tung, M.E. Schwartz, M. Takita, Y.-J. Wang, P. Kim, and H.L. Stormer, Phys. Rev. Lett. 100 (2008), 087403]. We also analyse the magneto–optical spectra of bilayer flakes in the photon-energy range corresponding to transitions between degenerate and split bands of bilayers.

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1. Introduction

Bilayer graphene [1–7] is one of several graphite allotropes [8] that display unique physical properties determined by the hexagonal symmetry of their two-dimensional crystalline structure. The electronic band structure of bilayer graphene close to the Fermi energy consists of two degenerate bands touching each other at the charge neutrality point (the position of the Fermi energy in a neutral system) and two bands split by the interlayer coupling [1,3,9]. In bilayer-based field-effect transistors a band gap in the electronic structure can be opened using a transverse electric field that breaks the symmetry between the layers [3,5–7], which may play an important role in forming the transport characteristics of such devices [4].

In this paper we analyse self-consistently the interlayer asymmetry parameter for bilayer graphene field-effect transistors, using the tight-binding approximation. The focus of this study is the manifestation of interlayer asymmetry in the magnetospectroscopy of bilayers, and we perform a self-consistent analysis for bilayer flakes subjected to quantising magnetic fields, taking into account the possibility that there is finite asymmetry in a neutral structure. When the material is kept at constant filling factor upon the variation of magnetic field (the measurement scheme employed in recent experiment [10]), the necessity to vary the charge density on the layers causes significant asymmetry and the Landau level (LL) spectrum in a strong magnetic field is altered considerably. Our calculation generalises the self-consistent analysis [5] developed for bilayers at zero magnetic field, and the analysis presented here improves the rigour of Ref. [11] where the variation of interlayer asymmetry on density and its influence on Landau level transition energies was estimated by neglecting screening effects. Also, we calculate the magneto–optical spectra of bilayers in the infrared spectral range that has recently become accessible in optical experiments using cyclotron irradiation sources [12–14], and where transitions between degenerate and split bands of the bilayer may occur.

2. The Landau level spectrum in charged bilayers: A self-consistent analysis

A schematic view of bilayer graphene (marked with the hopping integrals considered throughout this paper) and the Brillouin zone of bilayer graphene are shown in Fig. 1(a) and (b), respectively. Bilayer graphene consists of two coupled hexagonal lattices with inequivalent sites A1, B1 and A2, B2 in the first and second graphene sheets, respectively, which are arranged according to Bernal (A2–B1) stacking [1]. The hexagonal Brillouin zone has two inequivalent degeneracy points Kp = (ξ \frac{4π}{3a}, 0) (where ξ ∈ {±, −} and a is the lattice constant). Here, we take into account only the nearest-neighbour in-plane and interlayer (A2–B1) coupling and
the lowest-order terms in the electron band energy expansion in electron momentum (determined as the deviation of the electron wave number from the corner $K_+$ or $K_-$ of the hexagonal Brillouin zone that is identified below as the centre of the valley), which corresponds to the Hamiltonian

$$
\hat{H} = \xi \begin{pmatrix}
-u & 0 & 0 & v \pi \gamma_1 \\
0 & u & v \pi & 0 \\
v \pi & 0 & u & \xi \gamma_1 \\
v \pi & u & \xi \gamma_1 & -u
\end{pmatrix}.
$$

(1)

It is written in a basis of sublattice Bloch states $\psi_+ = (\psi_{A1}, \psi_{B2}, \psi_{A2}, \psi_{B1})^T$ in valley $K_+$ and $\psi_- = (\psi_{B2}, \psi_{A1}, \psi_{B1}, \psi_{A2})^T$ in valley $K_-$. $u$ and $v$ is related to the nearest-neighbour hopping parameter $y_0 (v = \frac{\pi n}{2\xi})$ and $\pi = p_x + ip_y$. The interlayer-asymmetry parameter $v$, which will be at the heart of this study, describes the on-site energy difference between different layers in the bilayer.

In the presence of a magnetic field $B$ perpendicular to the flake, the bilayer spectrum splits into Landau levels (LLs). The LL spectrum can be obtained from the Hamiltonian in Eq. (1) using the Landau gauge $A = (0, -Bx)$ and the fact that operators $p_{-} = p_x + ip_y$ and $\pi = p_x - ip_y + eBx$ coincide with lowering (raising) operators in the basis of Landau functions $\psi_m(x, y) = e^{\pm i Bx/\hbar} \phi_m(x + q_p, y)$, where $\phi_m(x)$ are the wave functions of a quantum harmonic oscillator,

$$
\pi \psi_m = -i\hbar \sqrt{2m} \psi_{m-1}, \quad \pi \psi_0 = 0, \quad \text{and}
$$

(2)

$$
\pi \psi_m = i\hbar \sqrt{2(m+1)} \psi_{m+1},
$$

where $\lambda = \sqrt{\hbar/eB}$ stands for magnetic length.

For a symmetric bilayer, $u = 0$, the energies $\varepsilon_m$ of the Landau levels are described by [1, 15–18]

$$
\varepsilon_0 = 0;
$$

$$
\varepsilon_m^{c(s)} = \sqrt{\frac{\beta}{2}} \left( \gamma_1^2 + \Gamma (2m - 1) \right) \left( \pm \sqrt{\gamma_1^4 + 2\Gamma \gamma_1^2 (2m - 1) + \Gamma^2} \right)^{\frac{1}{2}}, \quad m \geq 1;
$$

(3)

where $\Gamma = 2 \frac{B^2 \gamma_1^2}{\hbar^2}$ and $\beta$ denotes conduction band (+) and valence band (−) LLs. Indices $c$ and $s$ stand for low-energy and high-energy (split) bands and correspond to minus and plus signs in front of the square root, respectively.

Nonzero asymmetry $v$, caused by a possible difference in electric potential energy between the layers, modifies the LL spectrum [1, 19–22]. To model this effect, we employ a self-consistent theory of the charging of bilayer graphene. To reproduce the conditions of recent experiments [10] where the transition energy between low-energy LLs was measured as a function of a varying magnetic field but for a fixed filling factor $\nu$, we extend the self-consistent analysis of Ref. [5] from the zero-magnetic-field regime into the regime of quantising magnetic fields, taking into account the possibility that there is finite asymmetry in a neutral structure [see Eq. (4) below]. The analysis presented here improves the rigour of Ref. [11] where the variation of interlayer asymmetry on the density and its influence on Landau level transition energies was estimated by neglecting screening effects.

In particular, we consider a gated bilayer with interlayer separation $c_0$. In an external magnetic field $B$, a total excess density, $n = \frac{e^2}{\pi^2 \hbar^2}$, must be induced using the gate in order to keep the filling factor $\nu$ fixed while changing $B$. The density $n$ is shared between the two layers: $n = n_1 + n_2$, where $n_1$ is the excess density on the layer closest to (furthest from) the gate. The difference in electric potential between the layers is related to an incomplete screening of the gate electric field by the charge $e_1$ on the first layer alone and can be related to the unscreened density $n_2$.

$$
u(v, B) = w + \frac{e^2 c_0 \nu_2(v, B)}{\epsilon_0 \epsilon_r}.
$$

(4)

Here $\epsilon_r$ is the effective dielectric constant determined by the SiO$_2$ substrate, and $w$ takes into account the finite asymmetry of a neutral structure (the internal electric field due to, for example, initial non-intentional doping of deposits/adsorbates).

In our numerical calculations we use $\epsilon_r = 2$.

On the one hand, $u$ influences the LL spectrum via the Hamiltonian in Eq. (1). On the other hand, its value depends on the charge density $n_2$ which can only be obtained with a full knowledge of the LL spectrum and the wave functions corresponding to each LL. Therefore, a calculation of $u$ requires a self-consistent numerical analysis. This calculation consists of the following steps: for each given $B$, $5 < B < 20$ T, and $\nu$ we choose a starting $u$, and diagonalise Hamiltonian Eq. (1) to find the LL spectrum and the eigenstates with $m \leq M_{\text{max}}$, where $M_{\text{max}} \sim 300$. Then, we sum over all filled Landau levels and determine the excess electron densities on each layer. Note that, as a nonzero value of $u$ splits the valleys degeneracy of the LLs [22], care has to be taken, when comparing densities in specific LLs in biased and neutral structures, not to confuse levels in different valleys. Finally, using Eq. (4) we find the asymmetry parameter and, then, iterate the numerical procedure to obtain the self-consistent value of $u$ [23]. Note that, for a sufficiently large cutoff $M_{\text{max}} \sim 300$, the results were independent of $M_{\text{max}}$.

In our calculations we used $v = 10^6$ m/s and $\gamma_1 = 0.4$ eV, corresponding to effective mass $\gamma_1/2u^2 \approx 0.035m_e$. The self-consistently calculated values of $u$ obtained for several values of the filling factor $\nu$ are shown in Fig. 2(a) and (b) for the case of $w = 0$ and a nominal $w = -100$ meV, respectively. In the case when $w = 0$, the induced interlayer asymmetry is antisymmetric with respect to the change of the filling factor from positive to negative. This is because changing the filling factor from $+1$ to $-1$ corresponds to reversing the applied electric field and inducing excess densities $-n_1 - n_2$ and $n_2 - n_1$ and thus reversing the sign of $u$. Also, with decreasing $B$ all curves tend towards $u = 0$ and $u \sim -60$ meV in Fig. 2(a) and (b), respectively. These values are the results of the self-consistent calculation with corresponding values of $w$ in the absence of a magnetic field [5]. Examples of the low-energy LL spectrum for $\nu = +1$, $w = 0$ and for $\nu = +12$, $w = -100$ meV are shown in Fig. 2(c) and (d). To list the LLs in Fig. 2, we use three symbols: $m \xi$, where $s$ attributes the LL to the conduction (+) or valence (−) band, $m$ is the LL index and $\xi \in (+, -)$ identifies the valley ($K_+$ or $K_-$) that the level belongs to, respectively. The Landau levels $m = 0, 1$ have no $s$ index, as

![Fig. 1](image-url)
those levels are degenerate when \( u = 0 \) [1,22]. The sign of the valley splitting of the level \( sm \) depends on the sign of \( u \); for \( u > 0 \), level \( sm^+ \) has higher energy than level \( sm^- \) whereas the opposite is true for \( u < 0 \). Levels \( m = 0, 1 \) behave differently: in this case the energy \( e_{m+(-)} < (> ) \) if \( u > 0 \) and \( e_{m+(-)} > (< ) \) if \( u < 0 \). The size of the valley splitting of the low-energy LLs increases with \( u \) and \( B \) and for \(|u| \approx 0.1 \text{ eV}, B \approx 20 \text{ T} \) [filling factors \( \nu = +12, +16 \) in Fig. 1(a) and \( \nu = -8, -12, -16 \) in Fig. 1(b)] is of the order of 10 meV.

### 3. Low-energy inter-Landau level transitions and bilayer signature in the far-infrared absorption

Using spectra similar to those shown in Fig. 2(c) and (d), we find the energy of the low-energy inter-Landau level transitions for several filling factors which mimics the experimental conditions of Ref. [10] (the tight-binding approach to this problem has also been adopted in Ref. [11] where the dependence of the interlayer asymmetry on the density \( n \) and its influence on Landau level transition energies were estimated by neglecting screening effects). In that experiment, infrared light of energy \( h \omega \) was incident on the graphene bilayer in a strong external magnetic field and with a constant filling factor in order to excite charge carriers between a prescribed pair of LLs and to probe the energy dispersion. According to the selection rules determined in Ref. [15] (and later extended for \( u \neq 0 \) [22]), only transitions which change the LL index \( m \) by one are allowed. Also, as photons provide a very small momentum transfer, we only consider transitions between levels at the same \( K \) point. Thus, the corresponding low-energy transitions for filling factors \( \nu = +4, +8, +12, +16 \) are \( \xi_1 \rightarrow +2\xi, +2\xi \rightarrow +3\xi, +3\xi \rightarrow +4\xi, +4\xi \rightarrow +5\xi \) and for \( \nu = -4, -8, -12, -16 \) they are \( -2\xi \rightarrow 1\xi, -3\xi \rightarrow -2\xi, -4\xi \rightarrow -3\xi, -5\xi \rightarrow -4\xi \), respectively. However, as transitions between the same levels at different \( K \) points differ too little in energy to have been resolved separately in the above-mentioned experiment (in fact, they can be only be clearly distinguished in Fig. 3 for the case \( \nu = 4 \)), we obtain a single transition energy \( \epsilon_{\text{trans}}^\nu \) for a given filling factor \( \nu \) by comparing the relative intensities of the corresponding transition at each \( K \) point:

\[
\epsilon_{\text{trans}}^\nu = \frac{\epsilon_{\text{trans}}^\nu (K_0^+)\Gamma^\nu (K_0^+) + \epsilon_{\text{trans}}^\nu (K_0^-)\Gamma^\nu (K_0^-)}{\Gamma^\nu (K_0^+) + \Gamma^\nu (K_0^-)}
\]

where \( \epsilon_{\text{trans}}^\nu (K_0^+) \) and \( \Gamma^\nu (K_0^+) \) are the transition energy at the \( K_0^+ \) point and its intensity, respectively. The results obtained for sets of parameters as in Fig. 2(a) and (b) are shown in Fig. 3 \( (w = 0) \) and Fig. 4 \( (w = -100 \text{ meV}) \). We shall first discuss the case \( w = 0 \) presented in Fig. 3. In this case, for a specified value of \( B \), the asymmetry \( u \) changes sign with a change of the sign of the filling factor [Fig. 2(a)], the Landau level spectrum for \( \nu > 0 \) and \( \nu < 0 \) are the same but the \( K \) points have to be exchanged. Therefore, \( \epsilon_{\text{trans}}^\nu (K_0^+) = \epsilon_{\text{trans}}^\nu (K_0^-) \), clearly seen in all four graphs. Moreover, both transitions have the same intensity and contribute equally to \( \epsilon_{\text{trans}}^\nu \) (black solid line). Comparison with the transition energy for a neutral bilayer (grey line) shows that non-zero \( u \) decreases the energy of the transition. The greater \(|u|\) and \( B \), the bigger the difference between the excitation energy in symmetric and biased bilayers. However, this difference decreases with an increase of filling factor.

**Introduction of the parameter \( w \) breaks the symmetry between the conduction and valence band LLs as presented in Fig. 4 for the case of \( w = -100 \text{ meV} \). The valence band excitation has**
Fig. 3. Energy of low-energy inter-LL excitations as a function of magnetic field for \( w = 0 \). The broken lines are the contributions of individual valleys to the transition energy; black dot-dashed and dashed lines denote the transition energy for positive (negative) \( \nu \) at \( K_+ \) (\( K_- \)) and \( K_- \) (\( K_+ \)), respectively. The solid black lines show the contribution of both valleys to the transition energy, calculated according to Eq. (5) (in this case the transition energy is the same for both positive and negative \( \nu \)), whereas solid grey lines depict the transition energy in a neutral \( (u = 0) \) structure. Note that for \( \nu = 8, 12, 16 \) all black lines are very close to each other and difficult to resolve.

Fig. 4. Energy of low-energy inter-LL excitations as a function of magnetic field for \( w = -100 \text{meV} \). Black solid and dashed lines denote the transition energy for positive and negative filling factor, respectively. Grey solid lines depict the transition energy in a neutral \( (u = 0) \) structure.
greater energy than the conduction band excitation at filling factor $\nu = \pm 4$. However, this situation is reversed for higher filling factors $\nu = \pm 8, \pm 12, \pm 16$ (this reversal was not observed in the experiment [10]). For this specific case, $w = -100$ meV, the asymmetry introduced between excitations for filling factors $\nu$ and $-\nu$ is of the size of 3–10 meV. These two effects, the reduction of the transition energy with the increase of $u$ and the breaking of the symmetry between transitions for positive and negative filling factors caused by $w$, partly account for the disagreement between experimental findings and Eq. (3) obtained from a tight-binding model for neutral bilayers as used in Ref. [10] to fit the data.

4. Infrared magneto–optics in bilayer graphene

In this section, we analyse the optical transition spectra corresponding to transitions between LLs in split bands of the bilayers. Here, we use the self-consistently calculated interlayer-asymmetry parameter $u$, and the LL energies and wave functions, and we compute the infrared optical absorption spectra [15,24] of right-handed ($\otimes$) and left-handed (⊗) circularly polarised light $E_\omega = E_{\omega l}^{\otimes,\otimes}$, with $l_\otimes = (l_x - i l_y)/\sqrt{2}$ and $l_\otimes = (l_x + i l_y)/\sqrt{2}$ for bilayer graphene in a strong external magnetic field. The broadening of the Landau levels is modeled using a Lorentzian shape with the same full width at half maximum $\gamma = 60$ meV for all Landau levels. Numerical results for magnetic field $B = 14$ T and filling factors $\nu = 8$ and $\nu = 16$ are shown in Fig. 5. For the case of $w = 0$, the symmetry of the system demands that the intensity of absorption of light with a given polarisation for filling factor $\nu$ and at the $K_x$ point is the same as that of the light with the inverted polarisation at the $K_x$ point for filling factor $-\nu$. This, indeed, is the case for graphs in the left column of Fig. 5, where black solid and dashed lines show absorption of right-handed (left-handed) and left-handed (right-handed) circularly polarised light for the positive (negative) filling factor, respectively. Such a symmetry is broken for the case of $w = -100$ meV, for which the spectra for positive and negative filling factors are shown in the separate
panels, where solid and dashed lines refer to right-handed and left-handed polarization, respectively. In particular, the peak visible for some of the spectra at the radiation energy around 0.4 eV corresponds to electron excitation between the low-energy $m = 0$ LL and one of the two high-energy $m = 1$ LLs. Its position can be used to determine the value of the coupling constant $\gamma_1$, and a small shift in energy of this peak is due to strong magnetic field and asymmetry $u$. Presented curves show a similar shape as those predicted theoretically \cite{15,24} and observed experimentally \cite{12,13} for infrared optical absorption in the biased bilayer.

5. Summary

In this work, we have considered gated bilayer graphene in an external magnetic field. We have shown that keeping the filling factor constant results in a breaking of the symmetry between the graphene layers due to the induced carrier density. We have calculated the interlayer asymmetry $u$ self-consistently and demonstrated its influence on the Landau level spectrum. Using these results, we discussed both low-energy and high-energy inter-LL excitations and compared them to recent experiments concerning optical absorption. In particular, we achieved some improvement over the standard tight-binding model for a neutral bilayer in the explanation of the cyclotron resonance experiment probing low-energy the Landau level spectrum. The adequacy of our Hartree approximation in describing electron–electron interactions in bilayer graphene is uncertain, and comparison of our results with experiments would provide a good test of the reliability of this approximation. Other theoretical investigations \cite{6,25–27} show that corrections to the low-energy spectrum may arise when taking into account exchange interaction.

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References

\cite{23} Neither Landau level broadening nor partial filling factors were considered in this work.