Influence of interlayer asymmetry on magneto-spectroscopy of bilayer graphene

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Abstract

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 Ref. [1]. We also analyze the magneto-optical spectra of bilayer flakes in the photon-energy range corresponding to transitions between degenerate and split bands of bilayers.

Key words: A. Graphene, D. Cyclotron resonance, D. Optical properties
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1. Introduction

Bilayer graphene [2, 3, 4, 6, 7, 8, 9] is one of several graphite allotropes [10] 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 [2, 4, 5]. 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 [4, 7, 8, 9], which may play an important role in forming the transport characteristics of such devices [6].

In this paper we analyze 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 quantizing magnetic fields, taking into account the

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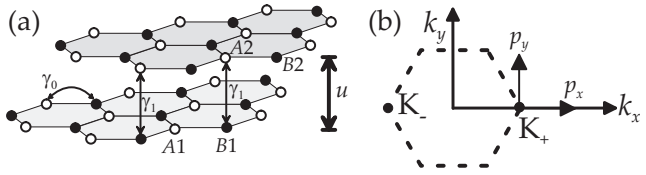

Figure 1: (a) Schematic view of the bilayer graphene lattice containing four atoms in the unit cell: A1 (white circles) and B1 (black) in the bottom layer and A2 (white) and B2 (black) in the top one. Also shown are the hopping parameters γ_0 and γ_1 used in the tight-binding model and interlayer asymmetry u. (b) First Brillouin zone of bilayer graphene with two inequivalent K points: K_+ and K_-. Note the difference between two momentum frames of reference: $\mathbf{k} = \mathbf{K}_\xi + \mathbf{p}$ where $\mathbf{k} = (k_x, k_y)$ and $\mathbf{p} = (p_x, p_y)$.

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 [1]), 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 generalizes the self-consistent analysis [7] 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 magneto-optical spectra of bilayers in the infra-red spectral range that has recently become accessible in optical experiments using cyclotron irradiation sources [12, 13, 14], and where transitions between degenerate and split bands of the bilayer may occur.

2. The Landau level spectrum in charged bilayers: 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 1(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 [2]. The hexagonal Brillouin zone has two inequivalent degeneracy points $K_\xi = (\xi\frac{4\pi}{3a}, 0)$ (where $\xi \in \{+, -\}$ 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:

$$\hat{H}=\xi \begin{pmatrix} -\frac{u}{2} & 0 & 0 & v\pi^{\dagger} \\ 0 & \frac{u}{2} & v\pi & 0 \\ 0 & v\pi^{\dagger} & \frac{u}{2} & \xi\gamma_1 \\ v\pi & 0 & \xi\gamma_1 & -\frac{u}{2} \end{pmatrix}. \quad (1)$$

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_-, and v is related to the nearest-neighbour hopping parameter γ_0 ($v = \frac{a\sqrt{3}}{2\hbar}\gamma_0$) and $\pi = p_x + ip_y$. The interlayer-asymmetry parameter u, 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 (LL). The LL spectrum can be obtained from the Hamiltonian in Eq. (1) using the Landau gauge $\mathbf{A} = (0, -Bx)$ and the fact that operators $\pi = p_x + ip_y - eBx$ and $\pi^{\dagger} = p_x - ip_y + eBx$ coincide with lowering (raising) operators in the basis of Landau functions $\psi_m(x, y) = e^{iq_y y/\hbar}\phi_m(x + q_y\lambda_B^2)$ (where $\phi_m(x)$ are the wave functions of a quantum harmonic oscillator),

$$\pi\psi_m = -i\frac{\hbar}{\lambda_B}\sqrt{2m}\psi_{m-1},\ \pi\psi_0 = 0,\ and\ \pi^{\dagger}\psi_m = i\frac{\hbar}{\lambda_B}\sqrt{2(m+1)}\psi_{m+1}, \quad (2)$$

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

For a symmetric bilayer, $u = 0$, energies ϵ_m of the Landau levels are described by [2, 15, 16, 17, 18]:

$$\begin{aligned} \epsilon_0 &= 0; \\ \epsilon_{m\beta}^{c(s)} &= \frac{\beta}{\sqrt{2}}\left(\gamma_1^2 + \Gamma(2m-1) \pm \sqrt{\gamma_1^4 + 2\Gamma\gamma_1^2(2m-1) + \Gamma^2}\right)^{\frac{1}{2}}, m \geq 1; \end{aligned} \quad (3)$$

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

Nonzero asymmetry u, caused by a possible difference in electric potential energy between the layers, modifies the LL spectrum [2, 19, 20, 21, 22]. To model this effect, we employ a self-consistent theory of the charging of bilayer graphene. In order to reproduce the conditions of recent experiments [1] where the transition energy between low-energy LLs was measured as a function of a varying magnetic field but for a fixed filling factor, ν, we extend the self-consistent analysis of Ref. [7] from the zero-magnetic field regime into the regime of quantizing 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 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 external magnetic field B, a total excess density, $n = \nu\frac{eB}{h}$, must be induced

using the gate in order to keep the filling factor ν fixed while changing B. The density n is shared between the two layers: $n = n_1 + n_2$ where n_1 (n_2) 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 en_1 on the first layer alone and can be related to the unscreened density n_2,

$$u(\nu, B) = w + \frac{e^2 c_0 n_2(\nu, B)}{\epsilon_0 \epsilon_r}. \tag{4}$$

Here ϵ_r is the effective dielectric constant determined by the SiO_2 substrate, and w takes into account finite asymmetry of a neutral structure (internal electric field due to, for example, initial non-intentional doping of the flake by 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 ν we choose a starting u, and diagonalize Hamiltonian Eq. (1) to find the LL spectrum and the eigenstates with $m \lesssim M_{max}$ where $M_{max} \sim 300$. 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 valley 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 $M_{max} \sim 300$, the results were independent of M_{max}.

The self-consistently calculated values of u obtained for several values of the filling factor ν are shown in Fig. 2(a) and 2(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 $+\nu$ to $-\nu$ corresponds to reversing the applied electric field and inducing excess densities $-n$,$-n_1$ and $-n_2$ and thus reversing the sign of u. Also, with decreasing B all curves tend towards $u = 0$ and $u \approx -60$meV in Fig. 2(a) and 2(b), respectively. These values are the results of the self-consistent calculation with corresponding values of w in the absence of a magnetic field [7]. Examples of the low-energy LL spectrum for $\nu = +4$, $w = 0$ and for $\nu = +12$, $w = -100$meV are shown in Fig. 2(c) and 2(d). To list the LLs in Fig. 2, we use three symbols: $sm\xi$, 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 those levels are degenerate when $u = 0$ [2, 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

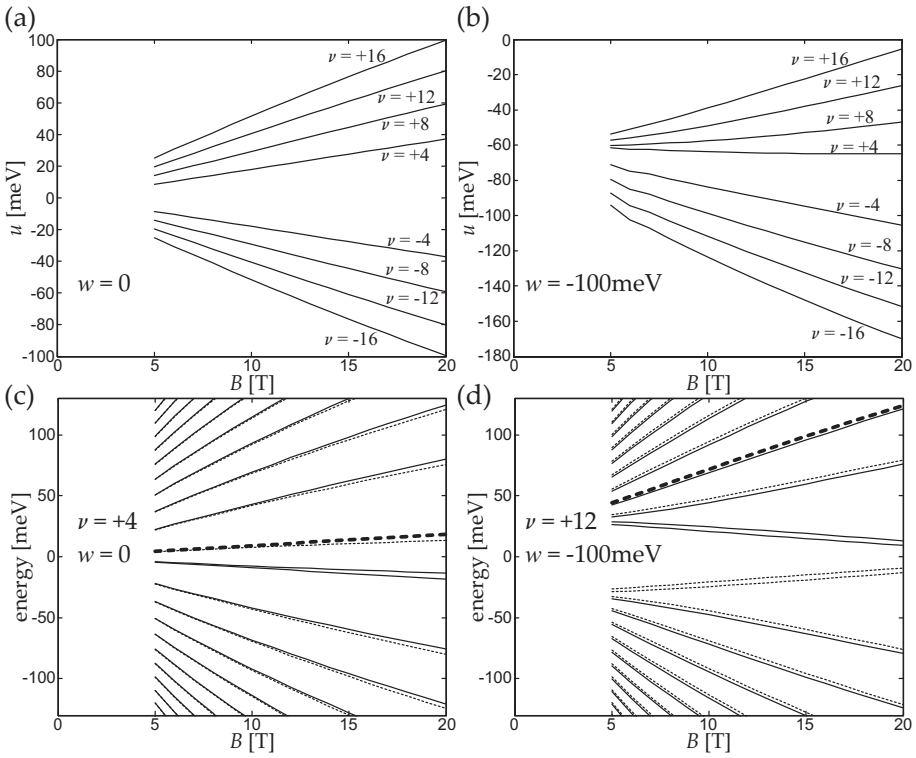

Figure 2: Top row: results of a self-consistent calculation of the interlayer asymmetry u for (a) $w = 0$ and (b) $w = -100$meV. Bottom row: the LL spectrum as a function of applied magnetic field B for constant filling factor and excess density-induced interlayer asymmetry u: (c) $\nu = +4$; $w = 0$, and (d) $\nu = +12$; $w = -100$meV. Solid and dashed lines denote levels belonging to K_+ and K_-, respectively. The line corresponding to the last filled Landau level is shown in bold. In these calculations we used $v = 10^6$m/s and $\gamma_1 = 0.4$eV.

differently: in this case the energy $\epsilon_{m+(-)} < (>)0$ if $u > 0$ and $\epsilon_{m+(-)} > (<)0$ if $u < 0$. The size of the valley splitting of the low-energy LLs increases with u and B and for $|u| \approx 0.1$eV, $B \approx 20$T [filling factors $\nu = +12, +16$ in Fig. 1(a) and $\nu = -8, -12, -16$ in Fig. 1(b)] is of the order of 10meV.

3. Low-energy inter-Landau level transitions and bilayer signature in the FIR absorption

Using spectra similar to those shown in Fig. 2(c) and 2(d), we find the energy of the low-energy inter-Landau level transitions for several filling factors which mimics the experimental conditions of Ref. [1] (the tight-binding approach to this problem has also been adopted in Ref. [11] where the dependence of interlayer asymmetry on density n and its influence on Landau level transition energies was estimated by neglecting screening effects). In that experiment,

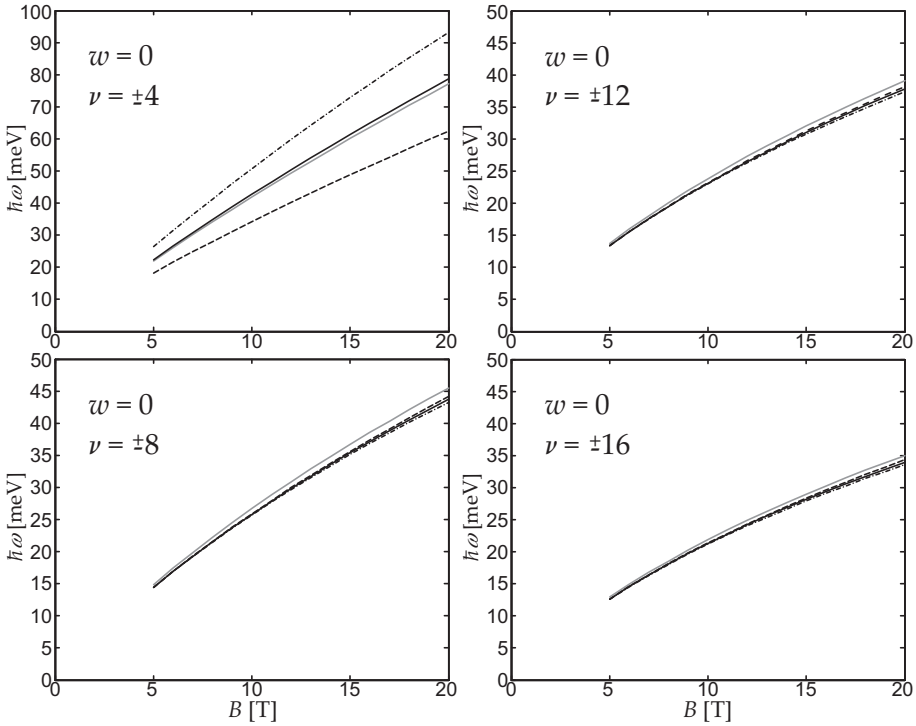

Figure 3: Energy of low-energy inter-LL excitations as a function of magnetic field for $w = 0$. The broken lines are the contributions of individual valleys to the transition energy: black dot-dashed and dashed lines denote the transition energy for positive (negative) ν at K_+ (K_-) and K_- (K_+), 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 ν), 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.

infrared light of energy $\hbar\omega$ 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 $1\xi \to +2\xi$, $+2\xi \to +3\xi$, $+3\xi \to +4\xi$, $+4\xi \to +5\xi$ and for $\nu = -4, -8, -12, -16$ they are $-2\xi \to 1\xi$, $-3\xi \to -2\xi$, $-4\xi \to -3\xi$, $-5\xi \to -4\xi$, respectively. However, as transitions between the same levels at different K points differ too little in energy to have been resolved separately in the abovementioned experiment (in fact, they can be only be clearly distinguished in Fig. 3 for the case $\nu = 4$), we obtain a single

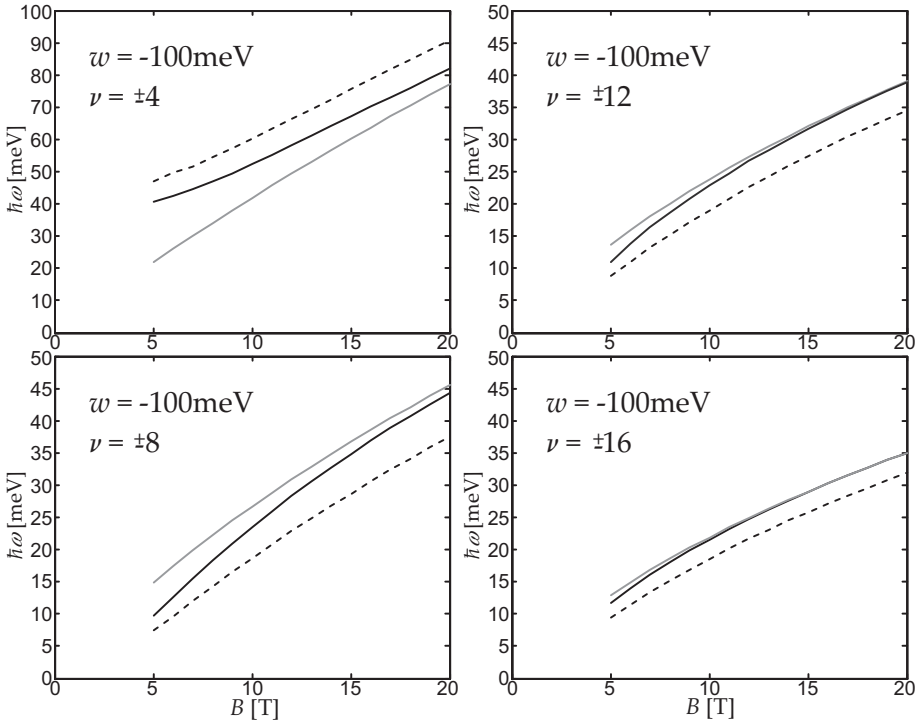

Figure 4: Energy of low-energy inter-LL excitations as a function of magnetic field for $w = -100$meV. Black solid and dashed lines denote the transition energy for positive and negative filling factor, respectively. Grey solid lines depict the transition energy in a neutral ($u = 0$) structure.

transition energy ϵ^{ν}_{trans} for a given filling factor ν by comparing the relative intensities of the corresponding transition at each K point:

$$\epsilon^{\nu}_{trans} = \frac{\epsilon^{\nu}_{trans}(K_+)I^{\nu}(K_+) + \epsilon^{\nu}_{trans}(K_-)I^{\nu}(K_-)}{I^{\nu}(K_+) + I^{\nu}(K_-)} \tag{5}$$

where $\epsilon^{\nu}_{trans}(K_\xi)$ and $I^{\nu}(K_\xi)$ are the transition energy at the K_ξ 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 ($w = -100$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 ν and $-\nu$ are the same but the K points have to be exchanged. Therefore, $\epsilon^{\nu}_{trans}(K_+) = \epsilon^{-\nu}_{trans}(K_-)$, clearly seen in all four graphs. Moreover, both transitions have the same intensity and contribute equally to ϵ^{ν}_{trans} (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 excitation

energy in symmetric and biased bilayers. However, this difference decreases with an increase of filling factor.

Introduction of parameter w breaks symmetry between the conduction and valence band LLs as presented in Fig. 4 for the case of $w = -100$meV. The valence band excitation has 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 [1]). For this specific case, $w = -100$meV, the asymmetry introduced between excitations for filling factors ν 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 factor 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. [1] to fit the data. Other investigations [24, 25] show that additional corrections may arise from electron-electron interactions.

4. IR 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, 26] of right ($\oplus$) and left-handed ($\ominus$) circularly polarized light $\mathbf{E}_\omega = E_\omega \mathbf{l}_{\oplus/\ominus}$, with $\mathbf{l}_\oplus = (\mathbf{l}_x - i\mathbf{l}_y)/\sqrt{2}$ and $\mathbf{l}_\ominus = (\mathbf{l}_x + i\mathbf{l}_y)/\sqrt{2}$ 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 ν and at the K_ξ point is the same as that of the light with the inverted polarisation at the $K_{-\xi}$ 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 polarisation, respectively. In particular, the peak visible for some of the spectra at the radiation energy around 0.4eV 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 γ_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 [15, 26] and observed experimentally [12, 13] for IR optical absorption in the biased bilayer.

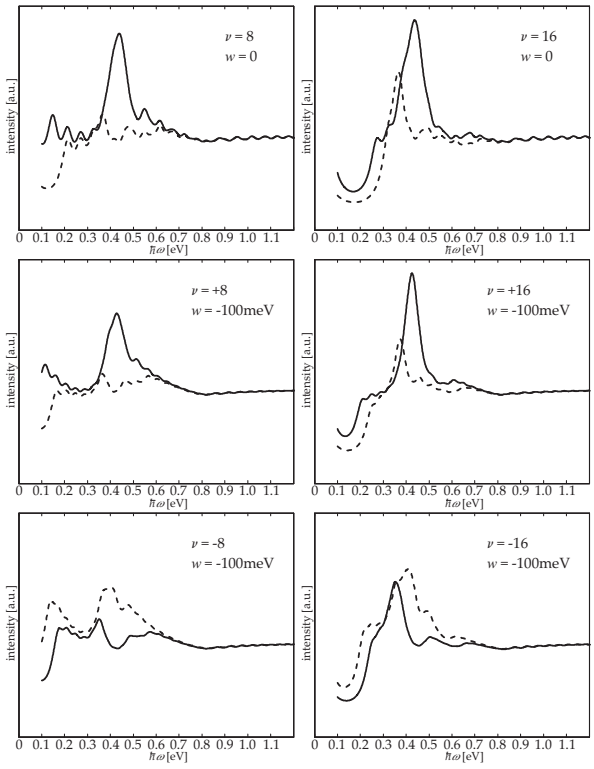

Figure 5: Magneto-optical absorption spectra for bilayer graphene in strong external magnetic field $B = 14$T and for filling factors $\nu = 8$ and $\nu = 16$ (left and right columns, respectively) and the case of $w = 0$ (top row) and $w = -100$meV (middle and bottom row). For the symmetric case of $w = 0$, 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. For the case of $w = -100$meV, solid and dashed lines represent absorption of right and left-handed circularly polarised light, respectively.

5. Summary

In this work, we have considered gated bilayer graphene in 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 neutral bilayer in the explanation of the cyclotron resonance experiment probing low-energy the Landau level spectrum.

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