Classical to quantum crossover of the cyclotron resonance in graphene: a study of the strength of intraband absorption

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Abstract. We report on absolute magneto-transmission experiments on highly doped quasi-free-standing epitaxial graphene targeting the classical-to-quantum crossover of the cyclotron resonance. This study allows us to directly extract the carrier density and also other relevant quantities such as the quasiparticle velocity and the Drude weight, which is precisely measured from the strength of the cyclotron resonance. We find that the Drude weight is renormalized with respect to its non-interacting (or random phase approximation) value and that the renormalization is tied to the quasiparticle velocity enhancement. This finding is in agreement with recent theoretical predictions, which attribute the

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renormalization of the Drude weight in graphene to the interplay between broken Galilean invariance and electron–electron interactions.

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1. Introduction

The low-frequency dynamical conductivity, $\sigma(\omega)$, a fundamental property of electrical conductors, is also central for the physics of graphene and for possible applications of this material in opto-electronics [1–4]. A tempting description of $\sigma(\omega)$ in graphene refers to a simple model of non-interacting Dirac-fermion quasiparticles with the characteristic linear dispersion $E(k) = \pm v_F \hbar |k|$, which at zero temperature occupy the electronic states up to the Fermi energy $|E_F| = \hbar v_F \sqrt{\pi |n|}$, where $v_F$ stands for the (Fermi) velocity parameter and $n$ is the free-carrier concentration. Such an effective single-particle (ESP) picture correctly reproduces the measured strength of interband-absorption processes with the characteristic onset at $2|E_F|$ and the dispersionless universal amplitude $\sigma_{\text{inter}} = \sigma_{\text{uni}} = e^2/(4\hbar)$ [5–11]. Further simple reasoning in terms of the Drude approach yields the well-known expression for the integrated strength $D = \int_0^\infty \sigma_{\text{intra}}(\omega) \, d\omega$ of intraband processes (the so-called Drude weight):

$$D = \frac{e^2}{2\hbar} v_F \sqrt{\pi |n|} = \frac{\pi |n| e^2}{2m_c} = \frac{2\sigma_{\text{uni}} |E_F|}{\hbar},$$

in which $m_c = |E_F|/v_F^2$ accounts for the effective cyclotron mass of carriers at the Fermi energy.

The experimental measurements available so far [12, 13] are in conflict with this simple prediction. They suggest a suppression of the Drude weight up to 40% (notably, in graphenes with high carrier concentrations) and therefore raise the issue of the applicability of the ESP picture to describe the dynamical conductivity of graphene. These doubts are additionally fostered by a startling report on non-vanishing absorption below the $2|E_F|$ threshold for interband processes [11].

Formally, the Drude approach follows from the commonly used random phase approximation (RPA) [14], which indeed grants equation (1), but, with the velocity parameter fixed at its bare value [15] $v_{F,0} = 0.85 \times 10^6 \text{ m s}^{-1}$—the value of $v_{F,0}$ can be accurately estimated, for example, from calculations based on density-functional theory at the local density approximation (LDA) level [16–18]. Markedly, $v_{F,0}$ is smaller than the apparent (renormalized) Fermi velocity, $v_F = (1.0–1.1) \times 10^6 \text{ m s}^{-1}$, which is derived from most of the spectroscopy studies on graphene-based systems [19–27] (and from beyond-LDA theoretical approaches).
In fact, the Drude weight determined by the bare velocity, $D_0 = (v_{F,0}/v_F)D$, could partially explain the experimental claims [12, 13] of a suppressed $D$. However, the RPA approach leading to equation (1) with $v_F$ replaced by $v_{F,0}$ misses some important physics [15]. A more adequate formalism, based on many-body diagrammatic perturbation theory [15], provides a more complex scenario in which self-energy (i.e. velocity enhancement) and vertex (i.e. excitonic) corrections compete with each other. Nevertheless, when the carrier concentration is sufficiently high, the approximate validity of the ESP model of non-interacting Dirac-fermion quasiparticles is expected to be recovered and equation (1), with the renormalized (apparent) velocity parameter, may be justified. Thus the issue of the strength of the Drude weight in graphene, one of its very basic electronic properties, remains a matter of controversy.

In this paper, we report on testing the ESP model against the results of cyclotron-resonance (CR) absorption experiments, performed on highly doped graphene specimens. Consistently following the ESP picture, we note that the Drude weight is fully transformed into the strength of the CR absorption in the classical limit and, with a good precision, into the strength of inter-Landau-level (LL) transitions in the quantum limit, i.e. into a quantity which is accurately determined from our absolute magneto-transmission measurements. The carrier concentration is deduced from the analysis of the oscillatory broadening of the CR response with LL filling factor, whereas the effective mass (and, in turn, the apparent Fermi velocity) is directly read from the dispersion of the CR with magnetic field. The Drude weight, as measured from the strength of the CR absorption, is found to be consistent with the estimate given by equation (1) in which the velocity parameter has the meaning of its apparent (spectroscopically measured or theoretically renormalized) value.

2. Experimental results

The investigated graphene sheet was prepared by thermal decomposition of the Si-terminated substrate of 6H–SiC. Subsequent hydrogenation of the ‘zero layer’ yields a quasi-free-standing epitaxial graphene monolayer [28]. Such specimens are typically highly p-doped with the Fermi energy $E_F \approx -0.3$ eV and carrier mobility $\mu \approx (2–3) \times 10^3$ cm$^2$ (V s)$^{-1}$ [29]. In this paper, we present data from one particular specimen; however, consistent results have been obtained on another similar sample.

To measure the far-infrared transmittance, a macroscopic area of the sample ($\approx 4$ mm$^2$) was exposed to the radiation of a globar, which was analyzed by a Fourier transform spectrometer and delivered to the sample via light-pipe optics. The light was detected by a composite bolometer placed directly below the sample, kept at $T = 1.8$ K. From the substrate-normalized transmission $T(B)$, the real part of the optical conductivity, $\Re [\sigma_{xx}(\omega, B)]$, was obtained for each separate magnetic field. The extraction of the conductivity from the transmission is described in detail in the appendix.

Experimental data are presented in figures 1 and 2 in the form of the conductivity spectra and as a false-color plot of $\Re [\sigma_{xx}(\omega, B)]$, respectively. In agreement with previous studies on highly doped graphene [29, 30], the magneto-optical response corresponds to the quasi-classical regime of the CR [31–33], since the condition $\mu B \sim 1$ applies. The CR line is approximately a single Lorentzian, its position in the spectra increasing linearly with $B$, see figure 2. At low magnetic fields (in particular below $B = 10$ T), a departure from linearity is visible, see figure 3(a). In this regime the optical response is dominated by plasmons confined on the scale...
Figure 1. The real part of the longitudinal conductivity $\sigma_{xx}(\omega, B)$ (in units of $\sigma_{uni}$) as extracted from the absolute magneto-transmission $T(B)$ using the procedure described in the appendix. Individual curves are plotted every 1 T. Thick curves are plotted every 4 T.

of a few microns due to graphene grain boundaries and/or substrate terraces. This regime has been discussed in detail elsewhere [34].

In high magnetic fields, the CR absorption still follows an overall linear-in-$B$ evolution, consistent with the moderate mobility of the sample. However, at the same time, it first shows clear indications of the approaching quantum regime, in which CR absorption corresponds to transitions between well-defined adjacent LLs. A number of CR studies of graphene in this regime have already been performed; see, e.g., [19–27]. A typical response in the quantum regime closely reflects the properties of the LL spectrum:

$$E_i = \text{sign}(i) v_F \sqrt{2e\hbar |i| B}, \quad i = 0, \pm 1, \pm 2 \ldots$$

Inter-LL excitations follow a clear $\sqrt{B}$-dependence, and the CR absorption gains a multi-mode character [35] due to the non-equidistant level spacing. In the low-temperature limit we always get at least two CR absorption modes unless the Fermi level is placed precisely between two LLs.

A closer inspection of the data in figures 1 and 2 (upper panel) reveals broadening of the CR line that appears at magnetic fields of $B_m \approx 27, 20, 16$ and 13.5 T and possibly also at 11.5 T. This broadening is best manifested as a decrease in the peak intensity, see figures 1 or 2, whereas the total peak area remains fairly constant, see figure 3(b). This sequence of field values matches the simple rule $mB_m = \text{const}$ (with $m = 3, 4, 5, 6$ and 7) and allows us to identify the filling factors $\nu = -4m$, which correspond to the half-filled last occupied LL. The broadening at fields $B_m$ thus reflects two CR modes, namely the transitions $L_{-m} \rightarrow L_{-m+1}$ and $L_{-m-1} \rightarrow L_{-m}$, with nearly equal intensities. It is worth noting that the CR width oscillating with the filling...
Figure 2. Upper panel: LL fan chart with schematically shown CR transitions in the quantum regime. Lower panel: a color plot of the real part of the experimentally determined longitudinal optical conductivity $\sigma_{xx}(\omega, B)$. The dashed lines correspond to transitions between adjacent (hole) LLs in graphene, $L_{-m} \rightarrow L_{-m+1}$. In both panels, a Fermi velocity $v_F = 0.99 \times 10^6$ m s$^{-1}$ was considered.

factor might also be reminiscent of another phenomenon—the difference in screening properties of a full or half-filled last-occupied LL in a two-dimensional (2D) parabolic-band electron gas [36–39]. Apart from the discussed weak oscillations in $\nu$, the CR width does not exhibit any clear field dependence, which would facilitate the identification of the main source of scattering in the sample, based on predictions of available theories, see, e.g., [40–42].
Figure 3. (a) Maximum of the CR absorption as a function of the magnetic field. The dashed line denotes the theoretical position of CR for the effective mass of $m = 0.058m_0$. The departure of the CR position from the linear-in-$B$ behavior at low fields is consistent with the appearance of confined magneto-plasmons [34, 43]. (b) Drude weight (area) extracted using a Drude–Lorentzian formula (see [34, 43]) and the directly read-out amplitude of $\text{Re} [\sigma_{xx}(\omega, B)]$. The horizontal dotted lines show the expected ESP and ‘bare’ Drude weights for a specimen with a 95% graphene coverage (see the main text), taking into account the apparent (renormalized) $v_F = 0.99 \times 10^6 \text{m s}^{-1}$ and bare $v_{F,0} = 0.85 \times 10^6 \text{m s}^{-1}$ Fermi velocities, respectively.

The sequence of the CR line broadening $m B_m = \text{const}$ is compatible with two possible scenarios. Either the Fermi level in graphene is pinned by electronic states in the SiC substrate, i.e. $E_F = \text{const} = -v_F\sqrt{2e\hbar m B_m}$, or the carrier density $n$ simply remains constant with varying $B$. In the latter case, the density is given by $|n| = \text{const} = N_f m \xi(B)$, where $\xi(B)$ is the LL degeneracy $eB/\hbar$ and $N_f = 4$ is the number of fermion flavors in graphene (i.e. a degeneracy factor associated with spin and valley degrees of freedom). Even though charge transfer between the substrate and graphene cannot be excluded ($E_F = \text{const}$), this effect is negligible as testified by the CR weight being nearly constant with $B$, see figure 3(b). Anyway, in both cases, the carrier density at $B = 0$ and also at each $B_m$ is $|n| = 4em B_m/\hbar = (7.9 \pm 0.2) \times 10^{12} \text{cm}^{-2}$. 

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Importantly, this value of carrier density is extracted entirely from the LL occupation, i.e. independently of \( v_F \) and \( E_F \). This represents a significant advantage over previous studies of graphene in the quasi-classical regime [29, 30]. We also note that the observation of individual filling factors in data, revealed as well-defined steps in the position of the CR line in figure 2, serves as an indication of high homogeneity of the carrier density on the macroscopic scale.

Using the ESP expression for the cyclotron frequency, \( \omega_c = eB/m_e \), we can extract the cyclotron mass at the Fermi level from the slope of the CR line directly: we find that \( m_c = (0.058 \pm 0.01) m_e \), where \( m_e \) is the electron mass in vacuum. Using the ‘Einstein relation’, \( |E_F| = m_e v^2_F \), together with \( |E_F| = h v_F \sqrt{|n|} \), we obtain \( v_F \) and \( E_F \). We find that \( v_F = (0.99 \pm 0.02) \times 10^6 \text{ m s}^{-1} \), which is sensibly larger than the ‘bare’ value \( v_{F,0} \), and \( |E_F| = (325 \pm 5) \text{ meV} \), which is in excellent agreement with the position of onset of the interband absorption (not discussed in this paper), which provides \( |E_F| = (320 \pm 10) \text{ meV} \). According to the ESP model (1), the deduced Fermi level implies the expected Drude weight \( Dh/\sigma_{\text{uni}} = 2|E_F| = 650 \text{ meV} \).

On the other hand, the Drude weight can be directly extracted from our data by estimating the area under the \( \Re \varepsilon \) \( \sigma_{xx}(\omega, B) \), which bypasses the use of equation (1). To account for the partial transfer of the Drude weight into the lower magneto-plasmon branch [44–46], which is significant at lower magnetic fields, the Drude weight has been extracted by fitting with a standard Drude–Lorentz formula, see [34, 43], assuming the confined-plasmon frequency of \( \omega_0 = 6.5 \text{ meV} \) [34]. This approach provides at higher field results identical to the fitting with a simple Lorentzian curve. From this fitting, we obtained the Drude weight close to \( Dh/\sigma_{\text{uni}} \approx 600 \text{ meV} \), see figure 3(b).

To compare both estimates of the Drude weight, the following corrections should be taken into account: (i) the coverage of the substrate by graphene is not full (see, e.g., AFM measurements on similarly prepared samples [34]), (ii) the hydrogenization process is not always complete—a small part of the sample remains covered only by the non-graphene ‘zero layer’ [47], (iii) bilayer graphene may also appear at selected locations [48] and (iv) the area below the \( \Re \varepsilon \) \( \sigma_{xx}(\omega, B) \), extracted using the simplified equation (A.4), is suppressed with respect to the Drude weight by a few per cent, see the Appendix. All these corrections tend to decrease the Drude weight deduced from the area under the \( \Re \varepsilon \) \( \sigma_{xx}(\omega, B) \) by approximately 5–10%.

To sum up, we find that our two independent estimates of the Drude weight agree with each other with a precision better than 10% and conclude that we do not observe any significant deviation from the validity of the ESP model (1). Our findings thus do not support recent transmission studies [12, 13] in which a significant suppression of the Drude weight (in comparison with the ESP expectation) has been reported. The reason for this discrepancy remains unclear at the moment, although we believe that (at least a part of) the suppression of the Drude weight found in [12] might stem from the normalization procedure used, in which the Drude-type absorption of graphene in the regime of electron–hole puddles (when the sample is neutral on average) is neglected.

3. Discussion

The validity of the ESP model (1) for the description of the strength of intraband absorption reminds us of Landau’s theory of normal Fermi liquids, which establishes a ‘mapping’ between a system of interacting fermions and a gas of weakly interacting quasiparticles with renormalized parameters [14]. The renormalized quasiparticle velocity \( v_F \) is precisely one of...
these parameters, which is completely controlled by the real part of the quasiparticle self-energy evaluated at the Fermi level $E_F$. Quasiparticles, however, do interact among each other and their interactions, which are described by the dimensionless Landau parameters $F_{\ell}^{s,a}$, modify physical observables such as the compressibility and the spin susceptibility or, more generally, the macroscopic response of the interacting electron liquid to external fields [14]. Our experimental test of the validity of the ESP model (1) tells us that interactions among quasiparticles in graphene, which diagrammatically are encoded in ‘vertex’ or ‘excitonic’ corrections, are weak, at least at large carrier densities.

Our experimental findings and the approximate validity of equation (1) can also be interpreted in terms of an enhancement of the Drude weight $D$ with respect to its non-interacting value $D_0$, which is found to be given by $D/D_0 \approx v_F/v_F,0 \approx 1.2$. This result is in excellent agreement with the theoretical predictions of Abedinpour et al [15]. By taking into account screening at the Thomas–Fermi level, the authors of [15] predicted $D/D_0 \approx 1.25$ at a carrier density $|n| = 8 \times 10^{12}$ cm$^{-2}$ and for a dimensionless coupling constant $\alpha_{ee} \approx 0.5$—for a definition of $\alpha_{ee}$ see, e.g., [1]. Note that this is the appropriate value of $\alpha_{ee}$ for our sample, as deduced from a recent ARPES study carried out by Bostwick et al [49] in a nearly identical sample. We stress that the theoretical proof of the approximate validity of equation (1) given in [15] is far from trivial: it is based on a fully microscopic diagrammatic approach in which substantial cancelations between the self-energy and vertex corrections play an essential role. In particular, the authors of [15] demonstrated the smallness of excitonic corrections to the intraband response of a doped graphene sheet, which tend to suppress the Drude weight—see equations (21) and (23) in [15]. Our experimental study corroborates this finding too.

For the sake of completeness, we should note that the Drude weight $D$ is strictly fully transferred into the CR strength only in the quasi-classical limit. In the quantum regime, the total CR weight $D_{CR}$ may differ from $D$. When the Fermi level is placed just in between two LLs ($|v| = 4m + 2, m = 1, 2, 3, \ldots$), we still have $D = D_{CR}$, but for the half-filled last occupied LL ($|v| = 4m, m = 1, 2, 3, \ldots$) we obtain $D_{CR} < D [50]$:  

$$D_{CR} = D \left[ \frac{1}{2} + \frac{1}{4} \left( \sqrt{1 + \frac{1}{m}} + \frac{1}{m} \right) \right].$$

(3)

However, one can easily verify that this suppression, which is related to the transfer of the oscillator strength between intra- and inter-band absorptions, remains very small for $m \geq 2$ (below 2%). The approximation $D = D_{CR}$ is thus very applicable as long as the zero LL is not involved in the CR absorption.

### 4. Conclusions

To conclude, an absolute magneto-transmission experiment on highly doped quasi-free-standing graphene allowed us to follow the oscillator strength of the CR, which exactly matches the zero-field Drude weight. We found that this weight remains very close to $D = 2|E_F|\sigma_{uu}/\hbar = e^2v_F/\pi|n|/(2\hbar) = \pi|n|e^2/(2m_e)$ and that a single-particle-like picture is fully sufficient to account for our data, provided that the renormalized (quasiparticle) parameters $E_F, v_F$ and/or $m_e$ are considered. This implies that the Drude weight in a doped graphene sheet is enhanced with respect to its bare value due to electron–electron interactions and that the enhancement is tied to the Fermi velocity enhancement [51, 52]. Our findings are in excellent agreement with a recent theoretical work [15], which predicted an enhancement of the Drude weight.
in doped graphene stemming from the interplay between broken Galilean invariance and electron–electron interactions.

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Appendix. Extraction of the optical conductivity from transmission spectra

The experimentally obtained substrate-normalized transmission $T(B)$ spectra are used to extract the optical conductivity, $\text{Re}[\sigma_{xx}(\omega, B)]$, of the graphene on the SiC substrate at every field. Figure A.1 shows the complex transmission and (internal) reflection coefficients for our experimental ‘vacuum (v)/graphene (g)/substrate (s)/vacuum (v)’ geometry, which are

$$
\begin{align*}
t_{vs} &= \frac{2}{N_s + 1}, & t_{sv} &= \frac{2N_s}{N_s + 1}, & t_{vgs} &= \frac{2}{N_s + 1 + Z_0\sigma}, \\
N_s &= n_s + i k_s, & d_s &= \text{the thickness of the substrate}, \quad Z_0 = 377 \, \Omega \text{ is the impedance of vacuum and } \sigma \text{ is the 2D optical conductivity of graphene.}
\end{align*}
$$

Here $N_s = n_s + i k_s$ is the experimentally known complex refractive index and $d_s$ is the thickness of the substrate, $Z_0 = 377 \, \Omega$ is the impedance of vacuum and $\sigma$ is the 2D optical conductivity of graphene.

In the derivation of the ratio of the transmission coefficient of the sample to that of the bare substrate, all internal reflections in the substrate must be taken into account. The experimental spectral resolution was reduced to $4 \, \text{cm}^{-1}$ in order to suppress the Fabry–Perot interference in the SiC substrate. In this case, the internal reflected rays add incoherently. By plane-wave
counting, the experimental transmission coefficients of the bare substrate and the sample are derived in terms of the complex coefficients:

\[
T_{vsv} = \frac{|t_{vsv} r_{sv}^2 \tau|^2}{1 - |r_{sv}^2 \tau|^2}, \quad T_{gsv} = \frac{|t_{gsv} r_{sv}^2 \tau|^2}{1 - |r_{sv}^2 r_{sv}^2 \tau|^2}.
\] (A.2)

The substrate-normalized transmission follows from equations (A.1) and (A.2):

\[
T = \frac{T_{gsv}}{T_{vsv}} = \frac{|N_s + 1|^4 - |N_s - 1|^4 |\tau|^4}{((N_s + 1)(N_s + 1 + Z_0 \sigma))^2 - ((N_s - 1)(N_s - 1 - Z_0 \sigma))^2 |\tau|^4}.
\] (A.3)

In the experimental spectral range, \( k_s \ll n_s \), therefore \( k_s \) is neglected in all complex coefficients (A.1), except in \( \tau \). We introduce the absorption coefficient of the substrate \( \alpha = |\tau|^2 \) and the substrate–vacuum amplitude reflectivity \( r = (n_s - 1)/(n_s + 1) \) in equation (A.3), and after some algebra the relation between the substrate-normalized transmission and the complex optical conductivity is found:

\[
T = \left[ 1 + \frac{2}{n_s + 1} \frac{1 + \alpha^2 r^3}{1 - \alpha^2 r^4} \text{Re}\{Z_0 \sigma\} + \frac{1}{(n_s + 1)^2} \frac{1 - \alpha^2 r^2}{1 - \alpha^2 r^4} |Z_0 \sigma|^2 \right]^{-1}.
\] (A.4)

Evidently, from the experimental transmission spectra alone, the complex conductivity \( \sigma \) cannot be extracted. However, note that the prefactor in the term depending on \( \text{Re}\{\sigma\} \) is significantly larger than the prefactor in the term depending on \( |\sigma|^2 \); therefore the transmission is determined predominantly by \( \text{Re}\{\sigma\} \). In this case, replacing \( |\sigma|^2 \) with \( |\sigma|^2 \) is a reasonable approximation.

In a magnetic field, equation (A.4) remains valid for each circular polarization separately if \( \sigma \) is substituted with \( \sigma_{\pm} \). For the unpolarized light used in our experiment, the transmission is an average of the two. In order to extract the diagonal conductivity at a finite field, we made a further simplification by ignoring the relatively small effect of \( \sigma_{xy} \) on the transmission and by substituting \( \sigma \) with \( \text{Re}\{\sigma_{xx}\} \) in equation (A.4). To estimate the error introduced by these approximations, we have tested the simplified version of equation (A.4) using the ideal classical CR absorption described by the standard analytical formulae for \( \sigma_{xx} \) and \( \sigma_{xy} \). We found that the area under \( \text{Re}\{\sigma_{xx}\} \), extracted using the above described procedure, is suppressed with respect to the Drude weight by roughly 5%.

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