Carrier Scattering from Dynamical Magnetoconductivity in Quasineutral Epitaxial Graphene

M. Orlita, C. Faugeras, R. Grill, A. Wysmolek, W. Strupinski, C. Berger, W. A. de Heer, G. Martinez, and M. Potemski

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Carrier scattering from dynamical magneto-conductivity in quasi-neutral epitaxial graphene

M. Orlita,[1,2,*] C. Faugeras,[1] R. Grill,[2] A. Wysmolek,[3] W. Strupinski,[4]
C. Berger,[5,6] W. A. de Heer,[5] G. Martinez,[1] and M. Potemski[1]

[1] *Laboratoire National des Champs Magnétiques Intenses, CNRS-UJF-UPS-INSA, 25, avenue des Martyrs, 38042 Grenoble, France*
[2] *Institute of Physics, Charles University in Prague, Czech Republic*
[3] *Institute of Experimental Physics, University of Warsaw, Hoża 69, PL 00-681 Warsaw, Poland*
[4] *Institute of Electronic Materials Technology, PL 01-919 Warsaw, Poland*
[5] *School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332, USA*
[6] *Institut Néel/CNRS-UJF BP 166, F-38042 Grenoble Cedex 9, France*

The energy-dependence of the electronic scattering time is probed by Landau level spectroscopy in quasi neutral multilayer epitaxial graphene. From the broadening of overlapping Landau levels we find that the scattering rate $1/\tau$ increases linearly with energy ϵ. This implies a surprising property of the Landau level spectrum in graphene – the number of resolved Landau levels remains constant with the applied magnetic field. Insights are given about possible scattering mechanisms and carrier mobilities in the graphene system investigated.

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Understanding carrier scattering in graphene [1] is one of the fundamental but still unclear issues in the physics of electronic properties of this noble material. Initial, model prediction for graphene [2, 3] done for short- and long-range scatterers independent of carrier density assumed the relaxation rate $1/\tau$ that is directly proportional to carrier energy ϵ. This is a simple consequence of the linear density of states with energy, $D(\epsilon) \sim |\epsilon|$, in graphene which displays the Dirac band structure with a constant Fermi velocity v_F. The $1/\tau \sim |\epsilon|$ relation implies that the low temperature (Boltzmann) conductivity $\sigma \sim v_F^2 \tau(E_F) D(E_F)$ of graphene is insensitive to changes of carrier concentration n and/or position of the Fermi energy $E_F \sim \sqrt{|n|}$ [2, 3]. Surprisingly, experiments on gated graphene flakes show different behavior, the conductivity varies linearly with carrier density: $\sigma(n) \sim |n|$ [4, 5]. The simple $\tau(\epsilon) \sim |\epsilon|^{-1}$ rule may not apply if dominant scatterers are of the specific resonant type or ripples type [6, 7], as well as for charge impurities [8–11]. Nevertheless, the unexpected, strong dependence of $\sigma(n)$ on carrier concentration remains puzzling [6, 12] and it is not fully clarified whether this behavior is intrinsic to graphene or has a well-defined extrinsic origin.

In this Letter we present a positive test of the $1/\tau \sim |\epsilon|$ rule, which we have performed on quasi-neutral layers of epitaxial graphene [13–16]. To probe this graphene system with a fixed Fermi energy we profit of the dynamical magneto-conductivity method and follow the energy dependence of the spectral broadening Γ of inter Landau level (LL) transitions measured in our infrared transmission experiments. Notably, the optical conductivity response of graphene involves interband excitations across the Dirac point and allows us to to determine the parameter Γ over a broad energy range. We have found that in the limit of strongly overlapping LLs, which corresponds to the quasi-classical limit in graphene and in which the simple relation $\Gamma = \hbar/\tau(\epsilon)$ is justified, this spectral broadening becomes $\Gamma = \alpha \cdot |\epsilon|$. This linear dependence on energy is valid at all magnetic fields and for all transition indexes ($\alpha = 0.026$ in the sample investigated). This experimentally determined scattering rate $1/\tau(\epsilon) \sim |\epsilon|$ dependence rules out specific extrinsic scattering mechanisms, such as resonant scatterers or charge impurities, in the epitaxial graphene layers which are protected from the environment. A straightforward but intriguing consequence of our finding is that the number of resolved LLs is constant, independent of the magnetic field. Possible consequences of the $1/\tau \sim |\epsilon|$ relation on the carrier mobility in extrinsically unperturbed graphene are discussed.

The studied sample is a standard multilayer epitaxial graphene (MEG) grown on the C-terminated surface of silicon carbide (4H-SiC[000$\bar{1}$]) [13] with intentional thickness of 50 layers. A significant part of layers displays an electronic band structure as that of an isolated graphene monolayer, which results from the characteristic rotational stacking of these graphene sheets [17]. This is confirmed by micro-Raman spectra measured on our sample, which show a single-component 2D band [18, 19], with some Bernal-stacked residuals on selected locations. The sheets studied in this experiment are quasi-neutral [20], only several layers close to the interface and on the surface of MEG become significantly doped (up to 10^{13} cm^{-2}) [21].

To measure the infrared transmittance, the sample 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 transmitted light was detected by a composite bolometer which operated at $T = 2$ K and which was placed directly below the sam-

ple. Measurements were done in a Faraday configuration, using a superconducting magnet. The sample is fully opaque in the energy range 85-120 meV and also displays a relatively weak transmission around 200 meV. Both absorption bands are related to phonon-related absorption in the SiC substrate. The spectra presented here are either conventional magneto-transmission spectra normalized by the zero field transmission $T(\omega,B)/T(\omega,0)$ or differential spectra $T(\omega,B+\Delta B)/T(\omega,B-\Delta B)$. The differential technique is more precise and helps to correct for possible magnetic field induced changes in the response of the bolometer.

A representative magneto-transmission spectrum at $B = 4$ T (normalized to the response at $B = 0$) of our MEG sample is shown in Fig. 1(a),(b). This spectrum is composed of a series of absorption lines which, accordingly to previous studies [14, 20, 22–24], can be easily identified as due to optically active transitions between Landau levels $L_{\pm n}$ of massless Dirac fermions, with the characteristic energies: $E_{\pm n} = \pm v_F\sqrt{2e\hbar B|n|}$, $n = 0, 1, 2\ldots$ These are the interband transitions: $L_{-(n+1)}\rightarrow L_n$ and $L_{-n}\rightarrow L_{n+1}$, which are reflected in the spectra of our quasi-neutral graphene sheets. From the characteristic field dependence of these transitions, $\hbar\omega = v_F\sqrt{2e\hbar B}(\sqrt{n}+\sqrt{n+1})$, we find $v_F = (1.025 \pm 0.005)\times 10^6$ m.s^{-1}, in agreement with previous estimations [14, 20]. The $L_{-1(0)}\rightarrow L_{0(1)}$ transition disappeared from spectra (due to occupation effect at LL filling factor $\nu \approx 6$) at fields of 100 mT which implies density $n \approx 10^{10}$ cm^{-2} in the studied layers.

For a more quantitative analysis of the measured magneto-transmission $T(\omega,B)$, we first note that the absorption of light by our graphene sheets is relatively weak and therefore:

$$T(\omega,B) = 1 - C\cdot \mathrm{Re}[\sigma_{xx}(\omega,B)]/(\epsilon_0 c) \qquad (1)$$

where the proportionality factor $C = N_{\mathrm{eff}}\cdot\frac{1}{2}(\kappa^2+3)/(\kappa^2+1)$ accounts for the effective number N_{eff} of graphene sheets in our sample including the effect of possible partial coverage of the SiC surface, and for multi-reflections of light from the SiC substrate with the refractive index $\kappa = 2.6$ [14]. $\mathrm{Re}[\sigma_{xx}(\omega,B)]$ is the real part of the dynamical longitudinal conductivity of a single graphene sheet expressed in Kubo-Greenwood formalism [25]:

$$\mathrm{Re}[\sigma_{xx}(\omega,B)] = \frac{e^3}{2\hbar}\frac{B}{\omega}\sum_{r,s} M_{r,s}(f_s - f_r)\delta_\Gamma(E_r - E_s - \hbar\omega), \qquad (2)$$

where $0 \le f_r \le 1$ stands for the occupation factor of the r^{th} LL and $\delta_\Gamma(E)$ for a Lorentzian of width Γ. $M_{r,s} = v_F^2\gamma\delta_{|r|,|s|\pm1}$ are velocity operator matrix elements in which $\gamma = 2$ if $r = 0$ or $s = 0$ and otherwise $\gamma = 1$.

Equation (2) is a simplified version of a more general Kubo formula (see e.g. Refs. 26 and 27) which includes initial and final spectral functions. Our simplified formula can be strictly derived for energy-independent and purely imaginary self-energies of the initial and final LLs, $i\Gamma_s$ and $i\Gamma_r$, and in the limit of relatively narrow levels/transitions [25], i.e., $\Gamma_{r,s} \ll \hbar\omega$ and $\Gamma_{r,s} \ll |E_{r,s}|$. On the other hand, since we deal with interband excitations only, the condition $\Gamma_{r,s} \ll |E_{r,s} - E_{r+1,s+1}|$ does not have to be fulfilled and adjacent LLs thus may overlap. With these approximations, the broadening Γ in Eq. 2 corresponds to $\Gamma = \Gamma_r + \Gamma_s$, and in a view of remarkable electron-hole symmetry of bands in graphene, also corresponds to $\Gamma \approx 2\Gamma_{r,s}$. At a given magnetic field, the broadening Γ can be viewed as a function of the resonance energy/frequency $\Gamma = \Gamma(\hbar\omega_{s\rightarrow r})$, where $\hbar\omega_{s\rightarrow r} = |E_r| + |E_s| \approx 2|E_{r,s}|$. In further, we also accept the possibility that $\Gamma = \Gamma(\hbar\omega)$ is a continues function of the energy/frequency. As a matter of fact, the conductivity (2) is practically identical if $\Gamma(\hbar\omega_{r\rightarrow s})$ is fixed for each transition or $\Gamma(\hbar\omega)$ is a slowly varying function over the interval $\hbar\omega_{r\rightarrow s} \pm \Gamma(\hbar\omega_{r\rightarrow s})$. This latter condition as well as $\Gamma_{r,s} \ll \hbar\omega$ and $\Gamma_{r,s} \ll |E_{r,s}|$ are a posteriori fulfilled in our experimental data.

Since we probe here quasi-neutral graphene layers at low temperatures, we assume that the Fermi energy is pinned to the 0^{th} Landau level and therefore $f_s = 1$ and

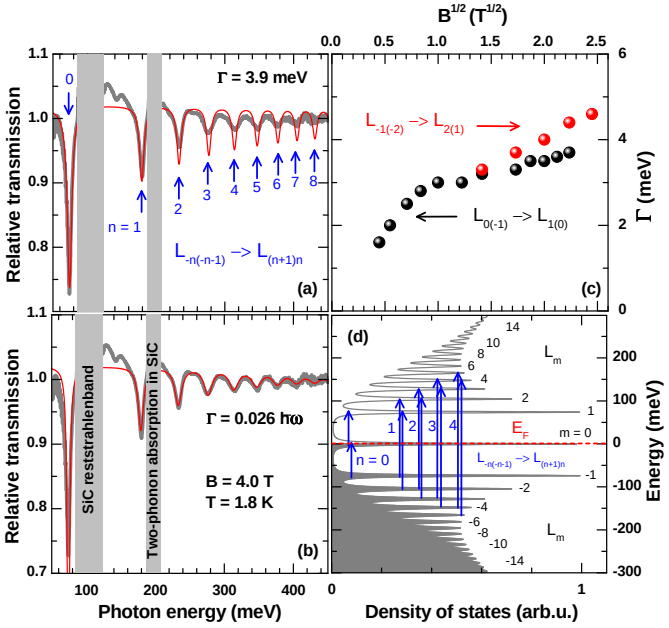

FIG. 1: (color online) Part (a) and (b): Experimentally obtained transmission spectrum of multilayer epitaxial graphene compared to theoretically expected curves. (c) Broadening Γ as derived from our data for $L_{0(-1)}\rightarrow L_{1(0)}$ and $L_{-1(2)}\rightarrow L_{2(1)}$ transitions (d) DOS in graphene at $B = 4.0$ T as calculated for the energy-dependent broadening $\Gamma = 0.026E$. The broadening of states around zero energy has been set constant to $0.026E_1$. Several observed absorption lines are schematically marked by vertical arrows.

$f_r = 0$ in the case of our interband $(r, s \neq 0)$, $L_s \rightarrow L_r$ transitions. As for a pair of degenerate transitions, $L_{-1} \rightarrow L_0$ and $L_0 \rightarrow L_1$, which involve the 0^{th} LL, we can, without loss of generality, set $f_1 = 0$ and $f_0 = f_{-1} = 1$ for any magnetic field, since the sum of intensities of $L_{-1} \rightarrow L_0$ and $L_0 \rightarrow L_1$ transitions is independent of *de facto* partial population of the L_0 level ($|\nu| < 2$) [28]. The phenomenologically introduced broadening parameter Γ is the main focus of this work. In general, this parameter depends on the magnetic field, on the transition index, or, as discussed above on frequency ω. Notably, the physical interpretation of Γ is different in the case of well-overlapped or well-resolved LLs.

In the limit of well separated Landau levels, $\Gamma = \Gamma_r + \Gamma_s$ may correspond to a single-particle quantum level broadening Γ_s of the density of states (DOS) [25]. This regime has been theoretically explored in the case of long-range and short-range disorder, especially for scattering centers insensitive to carrier concentration [2]. For short-range scatterers, Γ_s was found to be independent of the LL index but to increase as $\sqrt{B}$. In the more complex case of long-range scattering, Γ_s may also vary as $\sqrt{B}$ but the broadening of the 0^{th} LL is found to be significantly (by a factor $\sqrt{2}$) larger than that of any other levels. Charged impurities, if present, lead to a narrowing of LLs. The width of the LL is then predicted to oscillate with the filling factor and shows an overall $1/B$ field dependence, $\Gamma \sim 1/D(E_F)$, instead of the $\sqrt{B}$ field dependence [26, 27]. As concluded by the authors of Ref. 26, this excludes charge impurities as the dominant source of scattering in MEG, in which $\sqrt{B}$-type broadening is characteristic of inter-LL transitions [20].

In the limit of densely spaced and therefore strongly overlapping LLs, attained in graphene at sufficiently high energies – see Fig. 1(d) for illustration – the broadening parameter Γ can be interpreted as the inverse of a transport relaxation time, $\Gamma = \hbar/\tau$ within a factor of 2. This interpretation can be justified by the classical Drude approach to describe the carrier motion in a magnetic field [29, 30], in which the magnetic field effects are apparent when the cyclotron frequency ($\omega_c = v_F^2 eB/\epsilon$ for graphene) is comparable to the carrier relaxation rate τ^{-1}. The transport relaxation time is the only parameter which accounts for broadening effects in the dynamical conductivity response of graphene at zero magnetic field [3] and very likely in the magneto-dynamical conductivity response in the classical limit when the density of states is weakly modulated by the magnetic field [31]. Notably, our assumption $\Gamma = \hbar/\tau$ is favored by the electron-hole symmetry in graphene and, therefore, in neutral sheets likely identical relaxation rates for electrons and holes at energies $+\epsilon$ and $-\epsilon$, respectively. The transport relaxation time becomes comparable with the single-particle relaxation time if short-range scattering dominates [32], which is, as shown later on, consistent with our data and which thus facilitates our interpretation.

To compare with experiment, we re-consider the (relative) magneto-transmission spectrum measured at $B = 4$ T in Fig. 1. To model this spectrum, we assume that $\sigma_{xx}(\omega, B = 0) = e^2/4\hbar$. This is also the numerical result from Eq. 2 in the limit of strongly overlapping levels. Then following Eqs. 1 and 2 we fit $T(\omega, B)/T(\omega, 0)$ with two adjustable parameters/functions C and $\Gamma(\omega)$. In the first and simplest approach, we set Γ to be constant, independent of the transition energy. As shown in Fig 1(a), we then fail to reproduce the high energy part of the spectrum, but can well reproduce the two lowest, best-separated transitions. We find $C = 1.1$ and $\Gamma = 3.9$ meV by best fitting the shape of the $L_{-1(0)} \rightarrow L_{0(1)}$ transition.

However, most of the spectrum consists of overlapping transitions (in fact with the exception of the two lowest-energy transitions) that can be very well reproduced by assuming $\Gamma = \alpha \cdot \hbar\omega$. The best values are $\alpha = 0.026$ and $C = 1.3$ from the fit shown in Fig. 1(b). This latter value implies a surprisingly low effective number of layers, $N_{\text{eff}} \approx 2$, in the present sample. We related this to a partial coverage of the SiC surface by graphene sheets and to the partial AB-stacking, e.g. into bilayers recently iden-

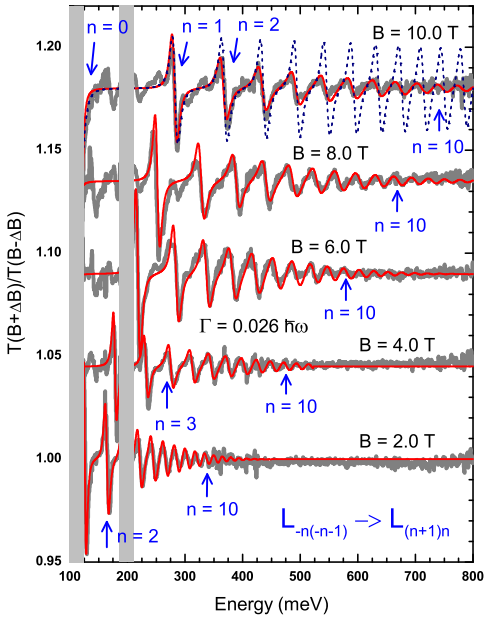

FIG. 2: (color online) Differential transmission spectra $T(B + \Delta B)/T(B - \Delta B)$. The lowest two spectra have been taken for $\Delta B = 0.05$ T, the other with $\Delta B = 0.125$ T. For clarity, the subsequent curves have been shifted by a factor of 0.045. Experimental curves are accompanied by theoretical fits, assuming the broadening parameter $\Gamma = 0.026\hbar\omega$. Let us note that the condition $B/\Delta B > 2n$ must be fulfilled to prevent significant overlap of adjacent transitions that involve n^{th} LL. The dashed line shows the simulated curve for differential transmission at $B = 10$ T and $\Delta B = 0.125$ T for a constant broadening $\Gamma = 8$ meV, which has been set to reproduce the $L_{-1(-2)} \rightarrow L_{2(1)}$ line.

tified as a minor component in MEG [33, 34]. Perhaps another reason could be an increased dielectric screening inside MEG [35], which can reduce the total absorption significantly. Remarkably, the $\Gamma = 0.026\hbar\omega$ rule is universal, in the sense that it applies to spectra measured at any magnetic field within the range of the present measurements. This is illustrated in Fig. 2 with simulations of differential spectra measured in the region of overlapping $n \geq 1$ transitions, at five distinct magnetic fields. The calculated traces, which accurately fit the data, are obtained from Eqs. 1 and 2 with a common set of parameters ($\alpha = 0.026$, $C = 1.3$). Here we note that despite a clear overlap between transitions, each of them is still well-defined in a sense that the above discussed assumption of narrow absorption lines and of slowly varying Γ is indeed well satisfied, $\Gamma = 0.026\hbar\omega \ll \hbar\omega$.

To sum up, the broadening parameter Γ seems to be constant for transitions between well-separated levels, but tends to increases linearly with energy when levels start to overlap significantly. This behavior remains unchanged up to the highest resolved transitions, i.e., those which originate from practically merging LLs. This is shown in Fig. 1(d), where the DOS has been plotted for the experimentally derived parameters. As discussed above, in the regime of strongly overlapping levels, the broadening of electronic states is governed dominantly by the zero-field relaxation rate and the observed linear in energy increase of Γ indicates a $1/\tau \sim |\epsilon|$ dependence. Interestingly, the same conclusion can be drawn using a simple semi-classical argument: the number of resolved transitions in our spectra is roughly magnetic field independent, i.e., the onset of LL quantization remains always pinned to a certain level ($n_0 \sim 10$, see Fig. 2). The energy of this onset moves to higher energies as $|\epsilon| \sim \sqrt{B}$ and the cyclotron frequency corresponding to this onset thus follows a linear in energy dependence $\omega_c = v_F^2 eB/|\epsilon| \sim |\epsilon|$. If we employ the classical condition for this onset, $\omega_c\tau \sim 1$, we conclude $1/\tau \sim |\epsilon|$.

Having established the $\hbar/\tau = \alpha|\epsilon|$ dependence, let us crudely assume that it is not additionally affected by changes in the carrier concentration. The Fermi energy independent conductivity of "our graphene" is then $\sigma = \frac{2}{\alpha} \cdot \frac{e^2}{h}$ ($\sigma \approx 80e^2/h$ for $\alpha = 0.026$) and we can speculate on a low-temperature carrier mobility, $\mu = ev_F^2\tau/E_F$, in such a model system at various charge densities. As an order of magnitude, we obtain $\mu \approx 10^6$ cm^2/(V.s) for $n = 10^{10}$ cm^{-2}, which is consistent with our recent measurements on quasi-neutral graphene [20]. The mobility falls down to $\mu \approx 10^4$ cm^2/(V.s) for the carrier density as high as $n = 10^{12}$ cm^{-2}, what reflects fairly well typical mobilities measured for charged, single-layer epitaxial graphene on C-face of SiC or interface layer of MEG [21, 36]. Our hypothetical graphene with $n = 10^{13}$ cm^{-2} would have a mobility of $\mu \approx 10^3$ cm^2/(V.s), in striking agreement with typical values measured for high concentration graphene on Si-face of SiC [37].

The $1/\tau(\epsilon)$ dependence reported here is expected for graphene and points towards conventional scattering mechanisms in quasi-neutral epitaxial graphene. Apart from charge impurities, excluded already earlier in MEG due to $\sqrt{B}$-broadening of inter-LL transitions [26], we can also for certain exclude the dominant role of resonant scatterers, which produce the mid-gap states and therefore modify the predicted $1/\tau \sim |\epsilon|$ relation. Those resonant scatters are by definition extrinsic, and maybe important for environment exposed graphene, but are naturally expected to be absent in our protected layers.

To complete our discussion of scattering mechanisms in MEG, we focus on the two lowest energy transitions of Fig. 1(a),(b). These are the two best separated transitions in our spectra, apparently related to nearly non-overlapping LLs ($|n| = 0, 1$ and 2) and therefore, their behavior can be tentatively analyzed in reference to theory in Ref. 2. Figure 1(c) shows that the width of both transitions scales roughly as $\sqrt{B}$. Moreover, the width of the $n = 0$ transitions is roughly the same if not even smaller than the width of the $n = 1$ transition. According to the theoretical analysis of the dynamical magneto-conductivity in the regime of well-separated Landau levels, this observation points towards short- rather than long-range scattering mechanism (both insensitive to carrier energy). Another possibility is the electron-electron scattering which also provides a scattering rate that increases linearly with energy [38].

Concluding, we have investigated the dynamical magneto-conductivity of quasi-neutral epitaxial graphene layers and studied the carrier scattering in these layers. We find that the relaxation rate is linear with the carrier energy. This dependence has been initially expected for graphene, nevertheless, not clearly observed yet. Its interesting implication is that the number of resolved Landau levels is fixed, independent of the magnetic field. The dominant scattering mechanism in the extrinsically unperturbed layers of epitaxial graphene is conventional, very likely due to short-range potentials or due to electron-electron interactions.

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* Electronic address: milan.orlita@lncmi.cnrs.fr

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