Landau level spectroscopy of graphene (Raman scattering and far-infrared absorption)

Electron-phonon and electron-electron interactions

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Probing Electronic Excitations in Mono- to Pentalayer Graphene by Micro Magneto-Raman Spectroscopy

Stéphane Berciaud,*† Marek Potemski,‡ and Clément Faugeras*‡

How Perfect Can Graphene Be?

P. Neugebauer,1 M. Orlita,1,2,3,* C. Faugeras,1 A.-L. Barra,1 and M. Potemski1

Magneto-Raman Scattering of Graphene on Graphite: Electronic and Phonon Excitations

Phys. Rev. Lett. 107, 036807 – Published 14 July 2011


"The ZOO of magneto-phonon resonances in graphene"
D.M. Basko, P. Leszczynski, C. Faugeras… et al., to be published

Landau level spectroscopy of electron-electron interactions in graphene

PRL 114, 126804, (2015)

C. Faugeras,1 S. Berciaud,2 P. Leszczynski,1 Y. Henni,1 K. Nogajewski,1 M. Orlita,1 T. Taniguchi,3 K. Watanabe,3 C. Forsythe,4 P. Kim,4 R. Jalil,5 A.K. Geim,5 D.M. Basko,6,* and M. Potemski1,†
Why? Graphene: a truly two-dimensional crystal of sp²-bonded carbon

Properties & Applications of Graphene

New Areas: active plasmonics
Industries: Electronics Photonics Telecommunications

New Areas: extremely efficient and ultra-thin thermal distributors
Industries: Electronics Photonics

Transparent electronics
- LCD displays
- Touchscreens
- Solar cells
- Lasers
- Ultra-fast photodetectors

Optical properties
- Composite materials
- Conductive inks
- Flexible electronics

Mechanical properties
- Aircraft industry
- Automotive industry
- Electronics

Electronic properties
- Transistor applications
- Integrated circuits
- Telecommunications

2D-based heterostructures
- Transistor applications
- Integrated circuits
- Photovoltaic applications

Other 2D crystals
- Medical and pharmaceutical applications
- Transistor applications
- Photovoltaic applications

2D material
- Industries: Medical and Pharmaceutical
- Transistor applications
- Photovoltaic applications

Graphene-Based Revolutions in ICT And Beyond

This talk: fundamental properties studied with magnetic fields (spectroscopy)
Dispersion relations and corresponding Landau level ladders

Electronic states, generic (quasi) 2D structure of \( sp^2 \) carbon (Bernal stacking)

\( E = E(\vec{k}) \)

\( E = E_{n_i}(B) \)

\( B = 0 \)

\( B > 0 \)

\( \text{~ graphene + (effective) bilayers} \)
Dispersion relations and corresponding Landau level ladders

Electronic states, generic (quasi) 2D structure of $sp^2$ carbon (Bernal stacking)

$\sim$ graphene + (effective) bilayers

$E = E(\vec{k})$

$B = 0$

$E = E_{n_i}(B)$

$B > 0$
Landau level spectroscopy

Probing inter Landau level excitations: $L_i \rightarrow L_j$
Absorption/transmission

$$T = T(h\nu, B)$$

resonant when

$$h\nu = E_{\text{exc}}$$

$$\sigma^+, \sigma^-$$

<table>
<thead>
<tr>
<th>Selection rules</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta n = \pm 1$</td>
</tr>
<tr>
<td>$\Delta n = +1 : \sigma^+$</td>
</tr>
<tr>
<td>$\Delta n = -1 : \sigma^-$</td>
</tr>
<tr>
<td>and</td>
</tr>
<tr>
<td>$\Delta n = 2, 4, 5, 7, 8$</td>
</tr>
<tr>
<td>if trigonal warping</td>
</tr>
</tbody>
</table>

V.P. Gusynin & S.G. Sharapov, PRB, 2006
M. Koshino & T. Ando, PRB, 2008
M. Mucha-Kruczynski et al., J. Phys., 2009
M.L. Sadowski et al., SSC, 2007
Raman scattering

\[ E_{\text{exc}} = h \nu' - h \nu \]

\[ \sigma^+ / \sigma^- \]

Selection rules

\[ \Delta n = 0 \]
strong
\[ \sigma^+ / \sigma^-, \sigma^- / \sigma^- \]

\[ \Delta n = \pm 2 \]
weaker
\[ \sigma^+ / \sigma^-, \sigma^- / \sigma^+ \]

\[ \Delta n = \pm 1 \]
if trigonal warping or coupled to phonon

O. Kashuba & V.I. Falko PRB, 2009; M. Mucha-Kruczynski et al., PRB, 2010
What can be learned from magneto-optics?

Band structure  √
Scattering: efficiency (mechanism)?
Scattering ?

Classical condition for observation of cyclotron resonance (Landau quantization)

\[ \tau_{\text{scattering}} > T_{\text{cyclotron}} \]
\[ \tau_s > 1/\omega_c \]
\[ \mu > 1/B_{\text{min}} \] rough estimate of carrier mobility

More general:

Spectral broadening \[ \Gamma \leftarrow 1/\tau_{\text{scat}} \]

Scattering mechanisms \[ \Gamma = \Gamma(B, E) \]
What can be learned?

Band structure  √

Scattering: efficiency (and mechanism)  √

Interactions (?):

  electron-phonon

  electron-electron
Interactions?

Tuning the excitations in resonance

\[ E_n \rightarrow E_m \]
Interactions?

resonant electron-phonon coupling?

\[ E_{\text{phonon}} \]

\[ L_n \to L_m \]

strength of interaction \( \delta \)

\[ E_{\text{ph}} = E_{1,3} \]

other magneto-phonon" resonance

+ more than this!!
Electron-electron interactions and inter Landau level transitions

Parabolic dispersions
equidistant LLs

Restoring single electron spectrum
of excitations at \( k \sim 0 \) (Kohn theorem)

\[ E_{\text{opt}} = E_{\text{single particle}} = \hbar \omega_c \]

\[ k_{\text{opt}} \sim 1/\lambda \ll k_{\text{coll}} \sim 1/l_B \]

Optics is useless to study
the many-body effects !?

Electron-electron interactions and inter Landau level transitions

Linear dispersions
non-equidistant spacing

$E_{\text{opt}} = E_{\text{single particle}} + \Delta_{\text{corr}}$

$k \sim l_{e-h}/l_B^2$

$E_C \neq E_{\text{exch}}$

$\Delta_{\text{corr}} = \delta \div \infty \; ?$

Expectations:

Rather large deviations from effective single electron model?

$\Delta_{\text{corr}} \sim \gamma_{nm} \sqrt{B} \; ?$

R. Roldan *et al.*, PRB, 2010
J. Sari, C. Toke, PRB, 2013
Graphene: Electron-electron interactions at B=0

Dirac cones reshaped by interaction effects in suspended graphene

D. C. Elias¹, R. V. Gorbachev¹, A. S. Mayorov¹, S. V. Morozov², A. A. Zhukov³, P. Blake³,
L. A. Ponomarenko¹, I. V. Grigorieva¹, K. S. Novoselov¹, F. Guinea⁴* and A. K. Geim¹,³

In graphene, electron-electron interactions are expected to play a significant role, as the screening length diverges at the charge neutrality point and the conventional Landau theory that enables us to map a strongly interacting electronic liquid into a gas of non-interacting fermions is no longer applicable¹,². This should result in considerable changes in graphene’s linear spectrum, and even more dramatic scenarios, including the opening of an energy gap, have also been proposed³-⁵. Experimental evidence for such spectral changes is scarce, such that the strongest is probably a 20% difference between the Fermi velocities $v_F$ found in graphene and carbon nanotubes⁶. Here we report measurements of the cyclotron mass in suspended graphene for carrier concentrations $n$ varying over three orders of magnitude. In contrast to the single-particle picture, the real spectrum of graphene is profoundly nonlinear near the neutrality point, and $v_F$ describing its slope increases by a factor of more than two and can reach $\approx 3 \times 10^6$ m s$^{-1}$ at $n < 10^{10}$ cm$^{-2}$. No gap is found at energies even as close to the Dirac point as $\sim 0.1$ meV. The observed spectral changes are well described by the renormalization group approach, which yields corrections logarithmic in $n$.


$\nu = \nu_0 - \frac{\alpha c}{4\varepsilon} \ln \left| \frac{E}{W} \right|$
OUTLINE

Band structure
  mono to pentalayer graphene

Scattering efficiency
  graphene on graphite: the best ever seen graphene

Electron-phonon interaction
  the ZOO of magneto-phonon resonances

Electron-electron interaction

Conclusions
What can be learned from magneto-optics?
Band structure!

Probing Electronic Excitations in Mono- to Pentalayer Graphene by Micro Magneto-Raman Spectroscopy

Stéphane Berciaud,*† Marek Potemski,*‡ and Clément Faugeras*‡

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‡Laboratoire National des Champs Magnétiques Intenses, CNRS/UJF/UPS/INSA, Grenoble F-38042, France
What can be learned from magneto-optics?
Band structure!
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Band structure!
What can be learned from magneto-optics?
Band structure!
What can be learned from magneto-optics?
Scattering: efficiency (mechanism)!
Graphene on graphite: best ever seen graphene!!

G. Li et al., PPRL, 2008
Cyclotron resonance absorption: high temperature but well resolved LLs

LL spacing $> kT$

LL broadening $< LL$ spacing

non-equidistant spacing

multimode

cyclotron resonance absorption
Graphene on graphite

(Very) low field cyclotron resonance absorption

\[ v_F = 1.0 \cdot 10^6 \frac{m}{s} \]

LL broadening: \( \Gamma \approx 35 \mu eV \) (0.4 K)
How perfect can graphene be

\[
\begin{align*}
E_F &\approx 6.5 \text{ meV}, \quad n \approx 3 \cdot 10^9 \text{ cm}^{-2}, \quad m^* = E_F / \nu_F^2 \approx 1.3 \cdot 10^{-3} m_e \\
\end{align*}
\]

Landau level quantization down to \( B_0 = 1 \text{ mT} \)

\[
\mu > \frac{1}{1 \text{ mT}} = 10^7 \frac{\text{cm}^2}{\text{V} \cdot \text{s}}
\]

\[
\gamma = 35 \mu eV \ (0.4 \text{ K}) \quad \rightarrow \quad \tau \approx 20 \text{ ps}, \quad \mu = \frac{e}{m^*} \tau \approx 3 \cdot 10^7 \frac{\text{cm}^2}{\text{V} \cdot \text{s}}, \quad l_F \approx 20 \ \mu m
\]

\[\gamma = E_1 \quad \rightarrow \quad B = 1 \mu T\]

\[B_{Earth} \approx 50 \mu T \quad \rightarrow \quad E_1 \approx 0.25 \text{ meV} = 3 \text{ K} > \gamma = 0.4 \text{ K}\]

Pronounced Landau quantization in the magnetic field of the Earth

Also at 50 K!

P. Neugebauer et. al., PRL, 2009
Graphene on graphite: magneto Raman scattering response phonons + search for a characteristic electronic response
e.g., $L_{-1} \rightarrow L_1$ inter Landau level excitation

![Graphene Raman scattering diagram](image)

**G band**
- $B=10T$
- $\lambda_{\text{exc}}=514.53\text{nm}$
- Temp. = 4K

**2D band**

Distance ($\mu m$)

**Distance ($\mu m$)**

**Distance ($\mu m$)**
Graphene on graphite: magneto-Raman scattering response: an overview
Graphene on graphite: magneto-Raman scattering response: an overview

\[ E_{2g} \text{ phonon} \]

\[ + \text{electronic excitations} \]

C. Faugeras et al., PRL, 2011; M. Kühne et al., PRB, 2012; P. Leszczynski et al., to be published
Graphene on graphite: magneto-Raman scattering response: an overview

C. Faugeras et al., PRL, 2011; M. Kühne et al., PRB, 2012, D. Basko et. al, to be published

"2D" band

E_{2g} phonon

focus on E_{2g} phonon
Interactions?

resonant electron-phonon coupling!

strength of interaction $\delta$
In magnetic fields
Resonant coupling of $E_{2g}$ phonon ("optical") with $\Delta n=\pm 1$ inter Landau level excitations
Theoretical predictions:

\[ \delta \sim \sqrt{2\lambda \cdot E_1(B_{res}) \cdot \sqrt{(1 - f_f) f_i}} \sim \sqrt{\lambda \cdot B_{res} \cdot (1 - f_f) f_i} \]
Magneto-phonon resonance: graphene on graphite

Graphene on graphite: an electronic system of unprecedented quality!

C. Faugeras, et al., PRL, 2011; M. Kühne et al., PRB 2012

J. Yan et al., PRL, 2010
Experiment: magneto-phonon resonance in epitaxial graphene

Neutral graphene
\[ \lambda = 4.5 \cdot 10^{-3} \]

C. Faugeras, et al., PRL, 2009
Magneto-phonon resonance in doped graphene

Graphene flake on Si/SiO$_2$

\[ \delta \sim \sqrt{\lambda \cdot B_{res} \cdot \sqrt{(1 - f_f) f_i}} \]

Magneto-phonon resonances: graphene on h-BN

~ neutral and better electronic quality

Experiment in qualitative agreement with simulations

P. Leszczynski, A. Nicolet, C. Faugeras et al., to be published
Magneto-phonon resonances: bilayer graphene on h-BN

Experiment in qualitative agreement with simulations

\[ v_F = 1.06 \cdot 10^6 \text{ m / s} , \quad \lambda = 3.5 \cdot 10^{-3} \]

\[ E_F < 100 \text{ meV} , \quad n < 2.5 \cdot 10^{12} \text{ cm}^{-2} \]
Magneto-phonon resonance in graphite

Phonon coupling to $\Delta n=\pm 1$ inter Landau band transitions from the vicinity of the K-point + of the H point

P. Kossacki et al., PRB, 2011
Probing the band structure with magneto-phonon resonance

few (?) layer graphene on Si/SiO₂

weakly doped tetra-layer graphene!

Graphene on graphite: magneto-Raman scattering response: electronic excitations

$v_F \sim 1.025 \times 10^6$ m/s
Beyond the standard magneto-$E_{2g}$ phonon resonances
New class of magneto-phonon resonances:
- accelerated relaxation, shortening of the final/initial states

Interactions

\[ E_{-1,2} = E_{-1,0} + E_{\text{ph}} \]
\[ E_{k=0} = E_{K,K} + E_{\Gamma} \]
\[ E_{k=0} = E_{K,K'} + E_{K} \]

both $K$- and $\Gamma$-phonons involved

two-particle excitations, triple resonances
intra and inter-valley scattering

- learning more on carrier dynamics

D.M. Basko, P. Leszczynski, C. Faugeras, et al
Electron – electron interactions?
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Landau level spectroscopy of electron-electron interactions in graphene

C. Faugeras,1 S. Berciaud,2 P. Leszczynski,1 Y. Henni,1 K. Nogajewski,1 M. Orlita,1 T. Taniguchi,3 K. Watanabe,3 C. Forsythe,4 P. Kim,4 R. Jalil,5 A.K. Geim,5 D.M. Basko,6,* and M. Potemski1,†

Electronic inter Landau level excitations
Magneto Raman scattering

suspended graphene $\varepsilon = 1$  $E_C/E_{\text{kin}} = \alpha_\varepsilon = (c/v)(\alpha/\varepsilon) \sim 2/\varepsilon$

graphene encapsulated in hBN $\varepsilon = 5$

graphene on graphite $\varepsilon = 10$?
Electron – electron interactions?

\[ \hbar \omega_n = 2v\sqrt{2e\hbar}\sqrt{B}\sqrt{n} = 2\sqrt{2n} \frac{\hbar v}{l_B} \]

\[ \nu_n^{exp} = \omega_{-n,n}^{exp} \frac{l_B}{\sqrt{8n}} \]
Electron – electron interactions

$B = 0$

First order perturbation theory with respect to $\alpha_\varepsilon = (c/v)(\alpha/\varepsilon)$

\[
\frac{v}{v_0} = 1 - \frac{\alpha_\varepsilon}{4} \ln \frac{|E|}{W}
\]

\[
v = v_0 - \frac{\alpha c}{4\varepsilon} \ln \frac{|E|}{W}
\]
Electron – electron interactions!

B = 0

First order perturbation theory with respect to \( \alpha_\varepsilon = \left( \frac{c}{v} \right) \left( \alpha / \varepsilon \right) \)

\[
\frac{v}{v_0} = 1 - \frac{\alpha_\varepsilon}{4} \ln \left| \frac{E}{W} \right|
\]

\[
v = v_0 - \frac{\alpha c}{4\varepsilon} \ln \left| \frac{E}{W} \right|
\]

Beyond FOPT

1/(N=4) expansion:


RPA:


\[
\frac{\alpha_\varepsilon}{4} \rightarrow \frac{2}{\pi^2} \left[ 1 - \frac{1}{\alpha_\varepsilon} + \frac{2}{\pi \alpha_\varepsilon} \frac{\arccos \left( \pi \alpha_\varepsilon / 2 \right)}{\sqrt{1 - \left( \pi \alpha_\varepsilon / 2 \right)^2}} \right]
\]

\[
\varepsilon \rightarrow \varepsilon_{1/N} = \varepsilon + 1.28 \alpha c / v_0 = \varepsilon + 3 \approx \varepsilon_*
\]
Denis Basko

First order perturbation theory with respect to 
\[ \alpha_\varepsilon = \left(\frac{c}{v}\right)\left(\frac{\alpha}{\varepsilon}\right) \]

\[ v_n \equiv \frac{\omega_{n,n} l_B}{\sqrt{8n}} = v_0 + \frac{\alpha c}{4\varepsilon} (\mathcal{L} - \ln \frac{l_{B_0}}{l_B}) + \frac{\alpha c}{4\varepsilon} C_n \]

\[ C_1 = -0.4, \quad C_2 = -0.2 \]

\[ v_2 > v_1 \]

Phenomenology to match the data (numbers)

\[ v_n = v_0 + \frac{\alpha c}{4\varepsilon_*} (\mathcal{L} - \ln \frac{l_{B_0}}{l_B}) + \frac{\alpha c}{4\varepsilon_{\delta v}} C_i \]

\[ \varepsilon_* \approx 3.9, 7, 12 \]

\[ \varepsilon_{\delta v} \approx 1.3, 3.7, 12 \]

\[ \varepsilon_0 = 0.88 \times 10^6 \text{ m/s} \]

\[ W = \left(\frac{\hbar v_0}{l_{B_0}}\right) \mathcal{L} = 3.1 \text{ eV} \]
Conclusions

Magneto-optics is a useful tool to study the "unconventional" and conventional graphene structures.

- band structure
- scattering efficiency
- electron-phonon interaction
- electron-electron interactions