Magneto-spectroscopy of multilayer epitaxial graphene, of graphite and of graphene

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Outline:
- introduction
- magneto-spectroscopy
  - inter Landau level transitions
  - magneto-Raman scattering of optical phonons
- discussion
  - band structure parameters and scattering efficiency
  - electron-phonon coupling
  - electron-electron interactions
- conclusions and acknowledgments
Graphene, Dirac-like energy bands, inter Landau level transitions

\[ E(\vec{k}) = \pm \tilde{c} |\hbar k| \]

if only \( \gamma_0 \) \( \tilde{c} = \sqrt{3} a_0 \gamma_0 / 2 \hbar \)

\[ \sigma_{\pm}(\omega, B) = \frac{4 G_B e^2}{\omega} \sum_{m,n} \frac{(f_n - f_m)}{E_m - E_n - (\hbar \omega + i\gamma)} \langle n | \hat{v}_\pm | m \rangle \langle m | \hat{v}_\pm^* | n \rangle \]

Selection rules:

\[ |n| - |m| = \pm 1 \]

(\( \sigma^+ \))

(\( \sigma^- \))

Transition energies:

Cyclotron resonance like:

\[ \tilde{c} \sqrt{2e\hbar B} \left( \sqrt{|n+1|} - \sqrt{|n|} \right) \]

Interband like:

\[ \tilde{c} \sqrt{2e\hbar B} \left( \sqrt{|n+1|} + \sqrt{|n|} \right) \]

Oscillator strength

\[ \int_0^\infty \text{Re} \sigma_{xx} dE \approx e^2 4 G_B \cdot \hbar \cdot \frac{\pi}{\Delta r} \cdot \frac{1}{4} \cdot \tilde{c}^2 \propto \frac{\sqrt{B}}{\sqrt{n + 1} \pm \sqrt{n}} \]

Multimode spectrum

“sqrt(B) rules”

Broadening ~ scattering efficiency
Graphene bilayer, “massive Dirac fermions”

\[ \gamma_3, \gamma_4 = 0 \]

**Parabolic dispersion:**

\[ E(p) = \pm \frac{p^2}{2m} \]

\[ m = \frac{\gamma_1}{(2c^2)} \approx 0.03m_0 \]

**Density of states:**

\[ \text{DOS}(E) = \frac{2m}{\pi\hbar^2} \]

**Brillouin zone:**

**Parabolic dispersion:**

\[ E_n = \hbar\omega_c \sqrt{n(n+1)} \]

\[ E_n \approx \frac{\hbar\omega_c (n + 1/2)}{n \gg 1} \]

Doubly degenerated 0th LL

Optically active transitions:

\[ |n| - |m| = +/- 1 \]
Multilayer epitaxial graphene: quasi neutral layers on C-face SiC

C-terminated SiC surface

multilayer graphene

Our samples, experiments

(Essentially) neutral, majority graphene layers probed in transmission (these experiments)

Highly conducting layers at the interface probed in electric transport e.g., C. Berger et al., Science (2006)
Magneto-spectroscopy of multilayer graphene (on SiC, C-face)
Dirac-like bands in many layer system !!!


Magneto-transmission: Fourier transform spectroscopy

\[ \frac{T(\omega, B)}{T(\omega, B = 0)} \]
Multilayer epitaxial graphene: quasi neutral layers on C-face SiC

Accurateness of the “Dirac cone”? Far- and near-infrared magneto-optics

High-Energy Limit of Massless Dirac Fermions in Multilayer Graphene,
P. Plochocka, C. Faugeras, M. Orlita, M. L. Sadowski, G. Martinez, M. Potemski,
High energy limits of “Dirac like electron dispersions”

+ next nearest neighbor hopping

\[ E_s(k) = \pm \hbar v_F \sqrt{k^2 + \frac{a^2}{16}k^4 + \frac{a}{2}s \left( k_x^3 - 3k_xk_y^2 \right)}, \]

In magnetic fields:
- graphene:
  - tight binding (t)
  - next nearest neighbor (t’/t \sim 0.1)

\[ E_{\pm,n} = \pm E_0 \sqrt{n} \mp E_0 \sqrt{n} \left\{ \frac{3w^2}{8} \frac{\tilde{a}}{l_B} n \right\} + E_0 \frac{3t'}{\sqrt{2t}l_B} \tilde{a} n \]

\[ \Delta E^\pm = \mp \frac{9t'}{2} \left( \frac{\tilde{a}}{l_B} \right)^2 + \frac{3\tilde{a}w^2}{64\hbar^2v_F} \left( \Delta_n^0 \right)^3 \]

Electron-hole asymmetry: small and not seen!

Carrying scattering from cyclotron resonance measurements

\[ \sigma \approx 10 e^2 / h \]
\[ \Delta \omega \cdot \tau = 1 \rightarrow \tau = 200 \text{fs} \]

\[ \mu B > 1 \rightarrow \mu > 0.25 \cdot 10^6 \text{ cm}^2 / \text{V} \text{s} \]
\[ n_e \approx 0.5 \cdot 10^{10} \text{ cm}^{-2}, \quad E_F \approx 6 \text{meV} \]

Record RT-mobility!

**Ultrahigh mobility in multilayered epitaxial graphene: a step towards graphene based electronics**

Multilayer epitaxial graphene : quasi neutral layers on C-face SiC

**C-terminated SiC surface**

**Multilayered graphene**

C. Faugeras et al., APL, 2007

Raman scattering:
- fingerprint of “simple, (Dirac like ?) band”
- + graphite inclusions

blue shift as compared to graphene (smaller $c^*$ ?)

Rotational stacking !!! (not Bernal)
(misoriented, turbostratic, incommensurate) of graphene layers
with electronic bands of a single sheet !
(but slightly smaller $c^*$ ?)

J. Haas et al., PRL (2007)
J.M.B. Lopes dos Santos et al., PRL (2008)
S. Latil et al., PRB (2008)
Z. Ni et al., PRB (2008)
Multilayer epitaxial graphene: quasi neutral layers on C-phase SiC

Latest confirmations of Dirac-like electronic bands

15 MAY 2009 VOL 324 SCIENCE www.sciencemag.org

Observing the Quantization of Zero Mass Carriers in Graphene

David L. Miller, Kevin D. Kubista, Gregory M. Rutter, Ming Ruan, Walt A. de Heer,
Phillip N. First, Joseph A. Stroscio

Application of a magnetic field to conductors causes the charge carriers to circulate in cyclotron orbits with quantized energies called Landau levels (LLs). These are equally spaced in normal metals and two-dimensional electron gases. In graphene, however, the charge carrier velocity is independent of their energy (like massless photons). Consequently, the LL energies are not equally spaced and include a characteristic zero-energy state (the $n = 0$ LL). With the use of scanning tunneling spectroscopy of graphene grown on silicon carbide, we directly observed the discrete, non-equally-spaced energy-level spectrum of LLs including the hallmark zero-energy state of graphene. We also detected characteristic magneto-oscillations in the tunneling conductance and mapped the electrostatic potential of graphene by measuring spatial variations in the energy of the $n = 0$ LL.

Cond/mat, 30 July 2009

First direct observation of a nearly ideal graphene band structure

Electron-phonon interaction

Anomaly of Optical Phonon in Monolayer Graphene

Tsuneya ANDO

PHYSICAL REVIEW B 75, 045404 (2007)

Electron-phonon coupling and Raman spectroscopy in graphene

A. H. Castro Neto
Department of Physics, Boston University, 590 Commonwealth Avenue, Boston, Massachusetts 02215, USA

Francisco Guinea
Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco E28049 Madrid, Spain

Breakdown of the adiabatic Born–Oppenheimer approximation in graphene

SIMONE PISANA¹, MICHELE LAZZERI², CINZIA CASIRAGHI¹, KOSTYA S. NOVOSELOV³, A. K. GEIM³, ANDREA C. FERRARI¹* AND FRANCESCO MAURI²*

PRL 98, 166802 (2007)

Electric Field Effect Tuning of Electron-Phonon Coupling in Graphene

Jun Yan,¹ Yuanbo Zhang,¹ Philip Kim,¹ and Aron Pinczuk¹,²
Electric Field Effect Tuning of Electron-Phonon Coupling in Graphene

Jun Yan,¹ Yuanbo Zhang,¹ Philip Kim,¹ and Aron Pinczuk¹,²

\[ \hbar \omega_G \text{ vs. } 2E_F \]
G-phonon couples to inter Landau level transitions which follow the same selection rules as optically active transitions.
Tuning electron-phonon coupling with magnetic field in graphene

Filling-Factor-Dependent Magnetophonon Resonance in Graphene

M. O. Goerbig, J.-N. Fuchs, K. Kechedzhi, and Vladimir I. Fal’ko

\[ \hbar \omega^\pm(n) = \frac{1}{2}(\Omega_n + \hbar \omega_T) \pm \sqrt{\frac{1}{4}(\Omega_n - \hbar \omega_T)^2 + g^2(n)} \]

\[ g(n) = g \sqrt{\frac{1}{2} A_n \gamma \sqrt{\nu_{n+1} - \nu_n}} \]

\[ \gamma = \frac{3 \sqrt{3} a^2}{2 \pi \ell_B^2} \]

\[ g = 0.2 \text{ eV} \]

\[ A_n = \begin{cases} 2 & \text{if } n = 0 \\ 1 & \text{if } n \neq 0 \end{cases} \]
Tuning electron-phonon coupling with magnetic field
Magneto-Raman scattering experiments
Multilayer epitaxial graphene: quasi neutral layers on C-face SiC

C. Faugeras et al., cond/mat, 2009

Experiment on multilayer epitaxial graphene (C-face of SiC)

Oscillatory (~ 40%) + Field independent component
Tuning electron-phonon coupling with magnetic field
Multilayer epitaxial graphene: quasi neutral layers on C-face SiC

C. Faugeras et al., cond/mat, 2009

Oscillatory component
Oscillatory component

Tuning electron-phonon coupling with magnetic field
Multilayer epitaxial graphene : quasi neutral layers on C-face SiC
C. Faugeras et al., cond/mat, 2009
approach of Ando [12] to our neutral Dirac-like system and derive the phonon energy $\epsilon$ and broadening parameter $\Gamma$ by extracting $\tilde{\epsilon} = \epsilon - i\Gamma$ from the equation which defines the poles of the phonon Green’s function:

$$\tilde{\epsilon}^2 - \epsilon_0^2 = 2\epsilon_0\lambda E_1^2 \sum_{k=0}^{\infty} \left\{ \frac{T_k}{(\tilde{\epsilon} + i\delta)^2 - T_k^2} + \frac{1}{T_k} \right\}$$

where $\epsilon_0$ stands for the phonon energy of the neutral system at $B=0$ T and $\delta$ accounts for the broadening characteristic for electronic excitations. The measured linewidth has been assumed as a convolution sum $\sqrt{\delta_0^2 + \Gamma^2}$, where $\delta_0$ accounts for other, than electron-phonon coupling, broadening mechanisms.
Cleaner systems?

Digression on

(Bulk) graphite?
higher mobilities, controversial reports

more complex, 3D material
(Bulk) graphite?: higher mobilities, controversial reports
more complex, 3D material

J.W. McClure (1964)
What about graphite: more complex 3D material

Dispersion of Landau bands across the layers (z-direction)

Nakao, JAP, 1976

Transport (SdH oscillations):
spectroscopy :
maxima in density of states

Landau level spectroscopy :
maxima in joint density of states
Graphite: SdH pattern, consistent with SWM model

J.M. Schneider, M. Orlita, M. Potemski, D.K. Maude, PRL 102, 166403 (2009)
Graphite: simplified model: “effective graphene bilayer”

Only $\gamma_0$ and $\gamma_1$

$$\lambda = 2 \cos(\pi k_z)$$

$$m = \lambda \gamma_1 / (2\tilde{c}^2).$$

$\lambda \gamma_1$
Graphite from viewpoint of Landau level spectroscopy: An effective graphene bilayer and monolayer

\[
\gamma_0 = (3.20 \pm 0.06) \text{ eV} \quad \gamma_1 = (375 \pm 10) \text{ meV} \\
m = \frac{\gamma_1}{\tilde{c}^2} \approx 0.063m_0
\]

For a bilayer: \[m = \frac{\gamma_1}{(2\tilde{c}^2)} \approx 0.03m_0\]

Natural graphite probed with more subtle tools
$E_{\text{exc}} \sim 1\text{meV}$, very low fields (EPR-type spectroscopy)

All but 3N harmonics of K-point electron cyclotron resonance (due to trigonal warping)

J.K. Galt et al., (1956) P. Nozieres (1958)

Expected also for graphene bilayer: Abergel, Falko (2007)
The cleanest, ever seen Dirac-like system

On graphite surface
Natural graphite probed with more subtle tools $E_{\text{exc}} \sim 1\text{meV}$, very low fields (EPR-type spectroscopy)

Harmonics of K-electron CR


Grahene flakes on graphite !!!!

P. Neugebauer, M. Orlita, C. Faugeras, A-L. Barra, and M. Potemski, Cond/Mat (2009)
Grahene flakes on graphite !!!

\[ \tilde{c} = 1.00 \times 10^6 \text{ m.s}^{-1} \]

Gap < 1 meV

\[ \gamma = 35 \mu \text{eV (0.4 K)} \]
Grahene flakes on graphite !!!

Landau level quantisation down to $B = 1 \text{ mT}$

$\sigma \approx 400 e^2 / h \ ???$

$\gamma = 35 \mu\text{eV} \rightarrow B = (\gamma/E_1)^2 \approx 1 \mu\text{T}$

$\Delta \approx 0.3 \text{ meV}$

$\gamma = 35 \mu\text{eV} \rightarrow \tau \approx 20 \text{ ps} \rightarrow \mu = e\tau\tilde{c}^2/E_F \approx 3 \times 10^7 \text{ cm}^2/(\text{V.s})$

Also at 50 K

$n_0 = 10^{11} \text{ cm}^{-2} \rightarrow \mu \approx 5 \times 10^6 \text{ cm}^2/(\text{V.s})$, 

$\mu B > 1 \rightarrow \mu > 10^7 \text{ cm}^2/(\text{V.s})$

$E_F = 6.5 \text{ meV}$

$n_0 \approx 3 \times 10^9 \text{ cm}^{-2}$

$m = E_F/\tilde{c}^2 \approx 1.3 \times 10^{-3} m_0$
H-point of graphite?
Extremely unlikely: pseudogap ~ 5meV, $E_f$ ~ 20 meV, T-dependence

Several samples investigated
Seen for different frequencies
Fast thermal cooling helps (enhanced signal)
Collective excitations?

Simple, one particle approach is applicable? Magneto-plasmons (e.g., size confined modes) (apparent under similar conditions in a 2DEG in GaAs)

Very likely not seen here, but ...

**E_{exc} \sim B/T**

Collective but magnetic???
Why single particle approach works so well?

It does not work

e-e interactions leads to modification of dispersion relations at B=0
c ~ 0.86x10^6 m/s (expected) → c*~1.02x10^6 m/s (experiment)
magnetic field acts on “modified dispersions”
(approach characteristic for “dirty” systems or for electrons with linear dispersions?)

“New” ‘Kohn theorem’ cancellation rules? (specific of “Dirac electrons”)
restoring the one particle character of inter Landau level transitions?

$$\frac{E_{\text{kin}}}{E_{\text{int}}} \sim \frac{E_F}{\frac{1}{r_n}} \sim \frac{\sqrt{n}}{\sqrt{n}} = \text{const} \quad \left(\sim \sqrt{n} \quad \text{for a "conventional" 2DEG}\right)$$

$$\frac{E_{01}}{e^2/\kappa l_B} \sim \frac{\sqrt{B}}{\sqrt{B}} = \text{const} \quad \left(\frac{\hbar \omega_c}{e^2/\kappa l_B} \sim \sqrt{B} \quad \text{for a "conventional" 2DEG}\right)$$

Abstract: EP2DS-2009-Kobe

Approximate validity of Kohn’s theorem in cyclotron resonance in graphene

Kenichi Asano* and Tsuneya Ando**
Conclusions

Magneto-spectroscopy helps to understand the electronic properties of two-dimensional carbon allotropes.

Not much of electron-electron interactions in graphene as far But clear effects of electron-phonon interaction

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