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

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Outline:

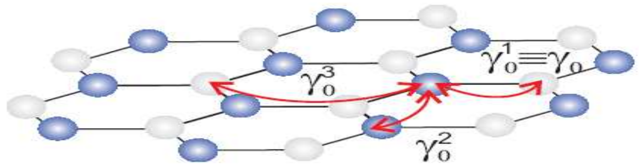
- introduction

- 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

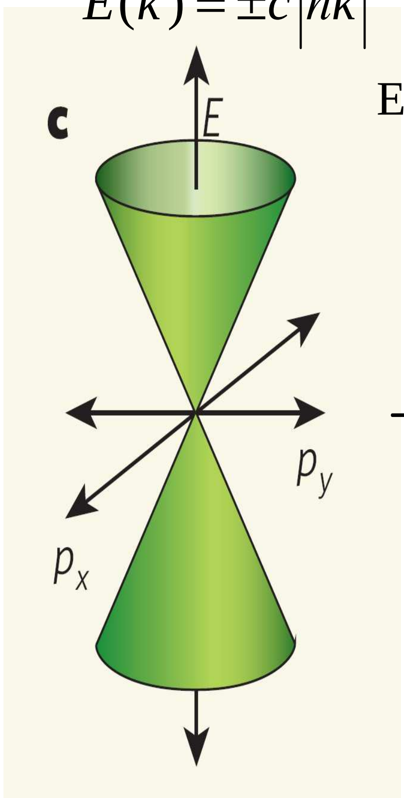
Graphene, Dirac-like energy bands, inter Landau level transitions

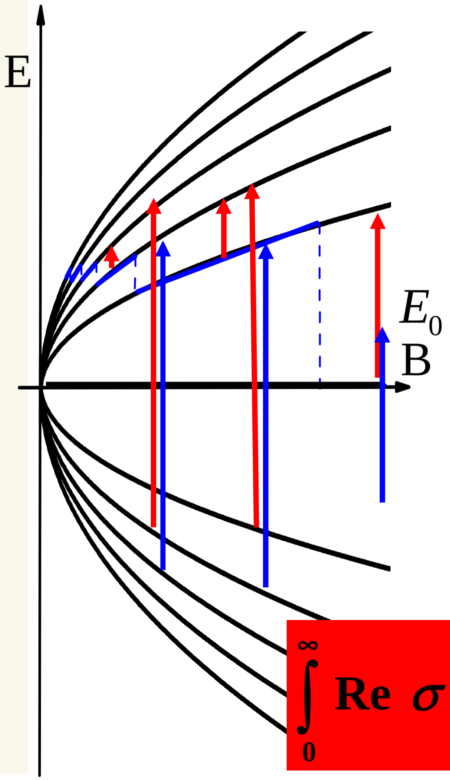


if only $\gamma_0 \quad \tilde{c} = \sqrt{3} a_0 \gamma_0 / 2\hbar$

$E(\vec{k}) = \pm \tilde{c} \left| \hbar \vec{k} \right|$

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





Selection rules : $\quad |n| - |m| = \pm 1 \quad$ (σ^+) (σ^-)

Transition energies :

Cyclotron resonance like :

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

Interband like :

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

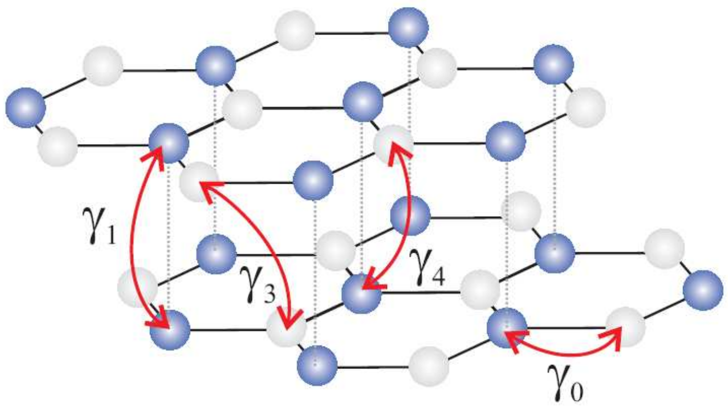
Oscillator strength

$$\int_0^{\infty} \mathbf{Re}\, \sigma_{xx} \, dE \approx e^2 4 G_B \cdot \hbar \cdot \frac{\pi}{\Delta_{tr}} \cdot \frac{1}{4} \cdot \tilde{c}^2 \propto \frac{\sqrt{B}}{\sqrt{n+1} \pm \sqrt{n}}$$

Multimode spectrum
"sqrt(B) rules"

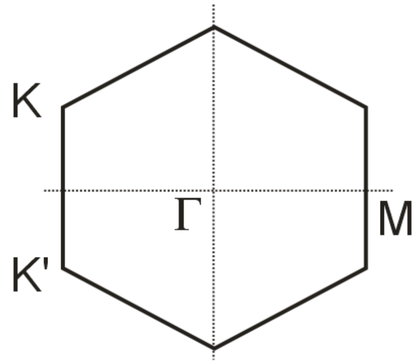
Broadening ~ scattering efficiency

Graphene bilayer, “massive Dirac fermions”

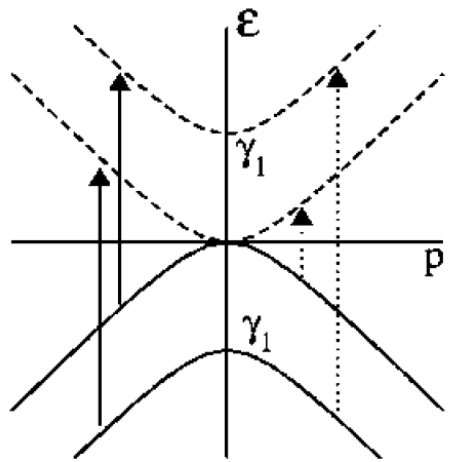


$$\gamma_3, \gamma_4 = 0$$

Brillouin zone:



Parabolic dispersion:



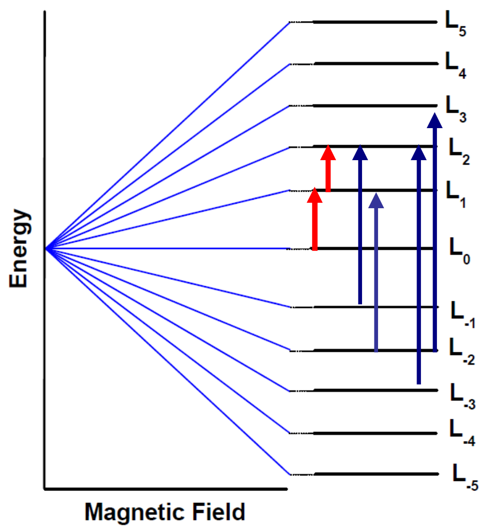
Parabolic dispersion:

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

$$m = \gamma_1/(2\tilde{c}^2) \approx 0.03 m_0$$

Density of states:

$$DOS(E) = \frac{2m}{\pi \hbar^2}$$



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

$$E_n \underset{n \gg 1}{=} \hbar\omega_c (n + 1/2)$$

Doubly degenerated 0th LL

Optically active transitions :

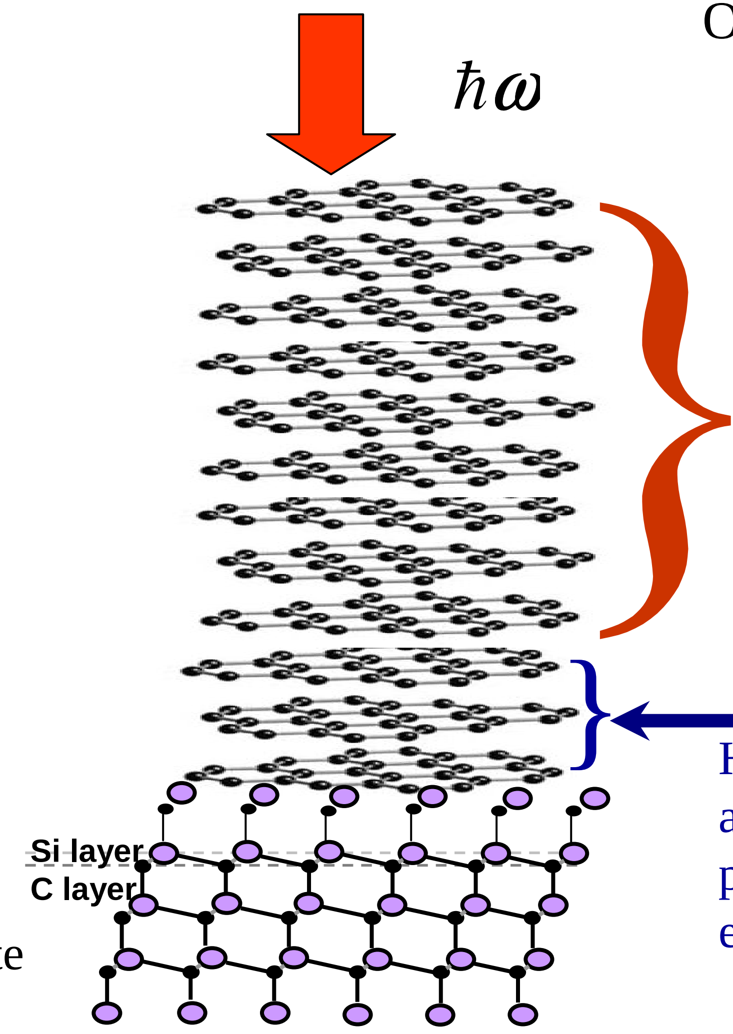
|n |- |m | = +/- 1

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

C-terminated SiC surface

multilayer graphene

Our samples, experiments



SiC substrate

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

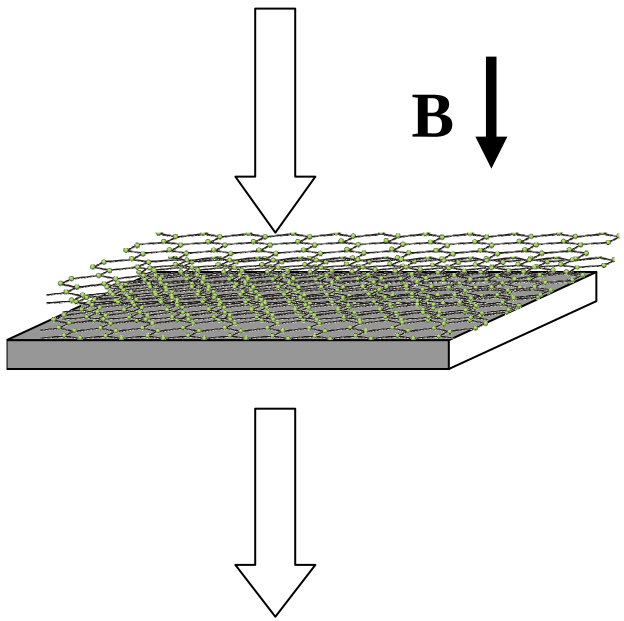
Highly conducting layers at the interface probed in electric transport e.g., C. Berger *et al.*, Science (2006)

$$\frac{T(\hbar\omega, B)}{T(\hbar\omega, B=0)}$$

Magneto-spectroscopy of multilayer graphene (on SiC, C-face)
Dirac-like bands in many layer system !!!

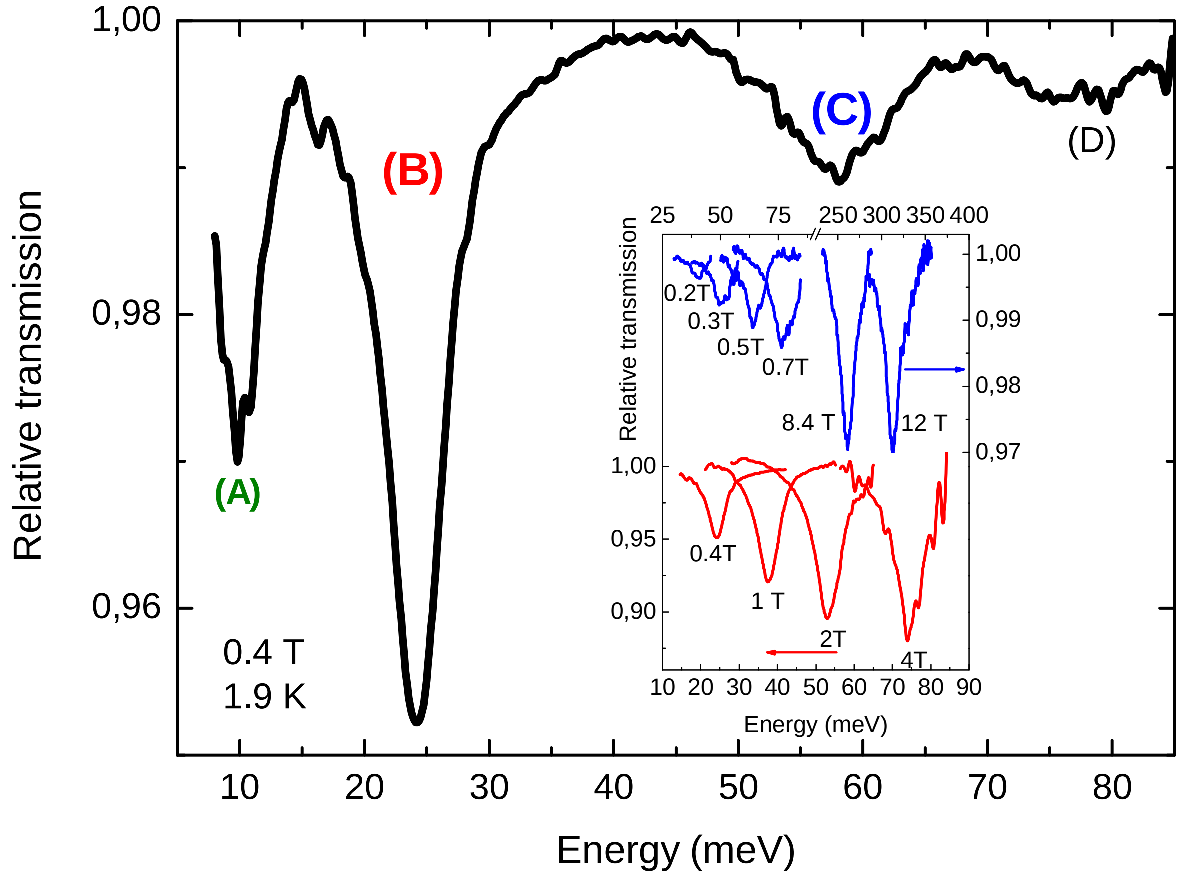
***Landau Level Spectroscopy of Ultrathin Graphite Layers*, M.L. Sadowski, G. Martinez, M. Potemski, C. Berger, and W. A. de Heer, Phys. Rev. Lett. 97, 266405 (2006)**

Magneto-transmission: Fourier transform spectroscopy



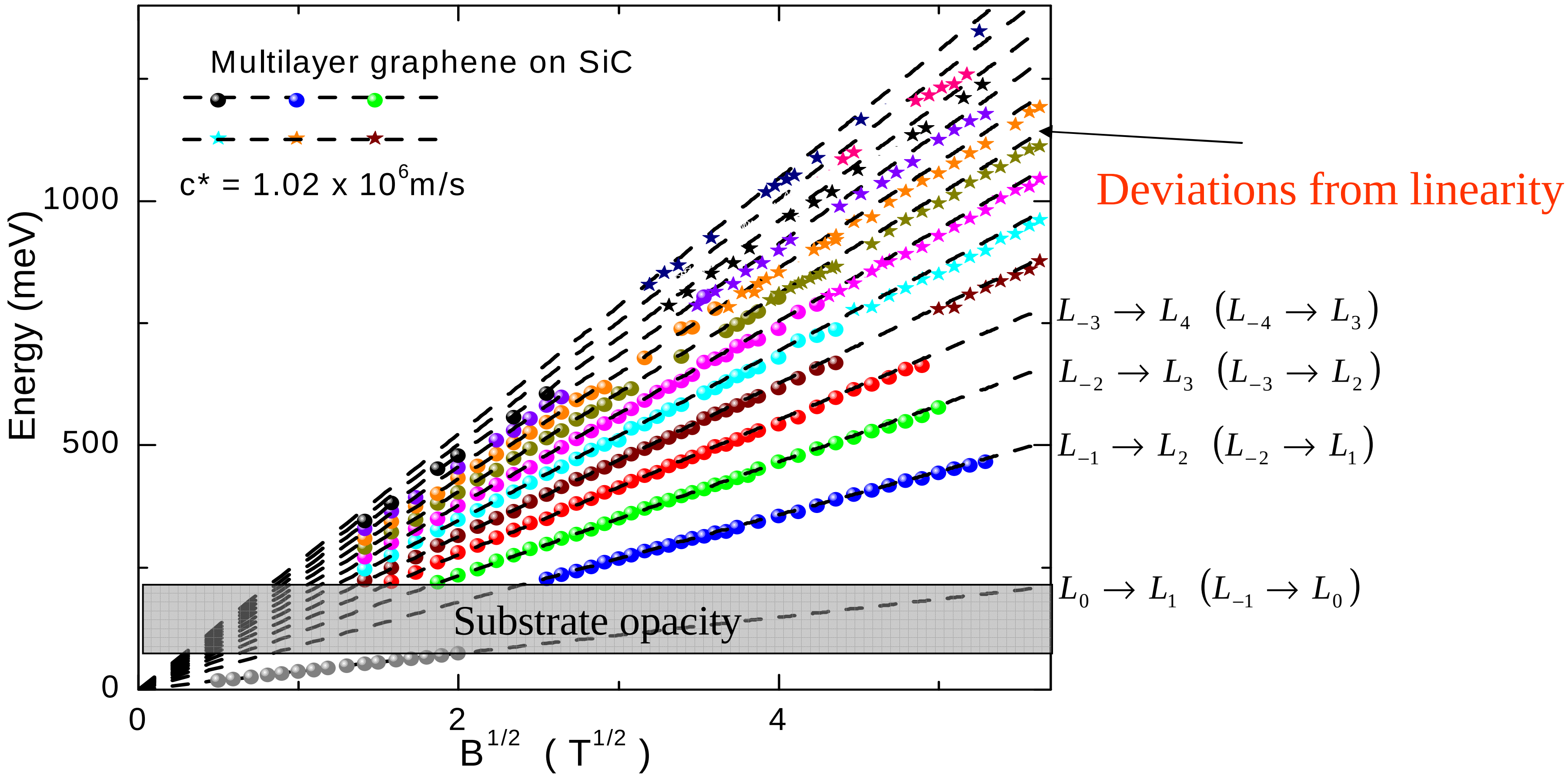
$$\frac{T(\omega, B)}{T(\omega, B=0)}$$

~ 5GL on SiC



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,
M.O. Goerbig, J.-N. Fuchs,C. Berger, and W.A. de Heer, Phys. Rev. Lett. 100, 087401 (2008)

High energy limits of "Dirac like electron dispersions"

+ next nearest neighbor hopping

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

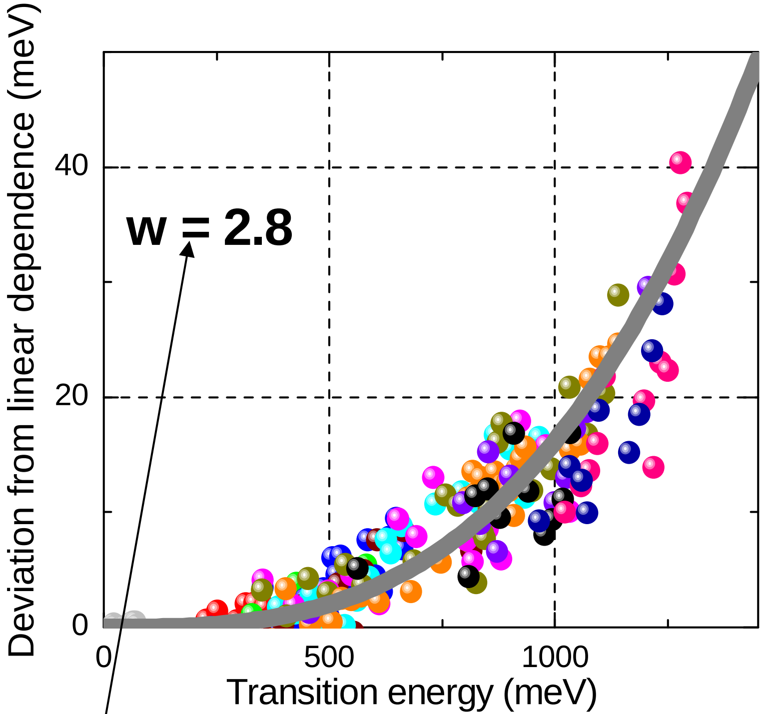
In magnetic fields :

graphene :

tight binding (t)

next nearest neighbor (t`/t ~ 0.1)

$$\mathbf{E}_{\pm,\mathbf{n}} = \pm\mathbf{E}_0\sqrt{\mathbf{n}} \mp \mathbf{E}_0\sqrt{\mathbf{n}}\left\{\frac{3\mathbf{w}^2}{8}\left(\frac{\tilde{\mathbf{a}}}{\mathbf{l}_\mathbf{B}}\right)^2\mathbf{n}\right\} + \mathbf{E}_0\frac{3\mathbf{t}'}{\sqrt{2}\mathbf{t}}\frac{\tilde{\mathbf{a}}}{\mathbf{l}_\mathbf{B}}\mathbf{n}$$

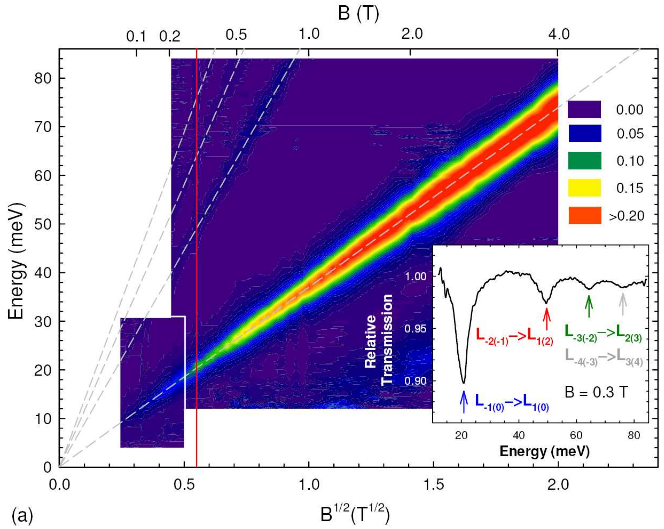


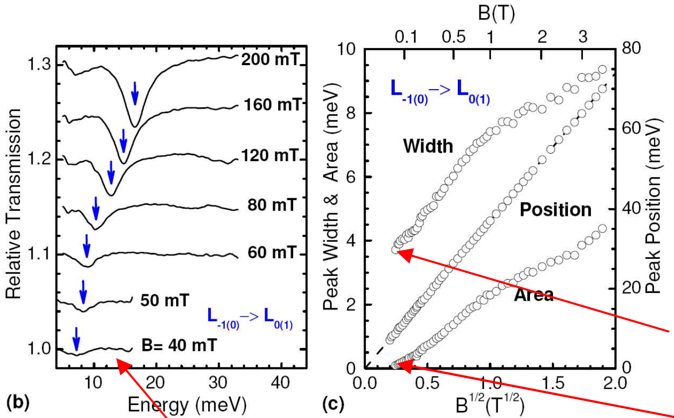
$$\Delta E^{\pm} = \mp\frac{9t'}{2}\left(\frac{\tilde{a}}{l_B}\right)^2 + \frac{3\tilde{a}w^2}{64\hbar^2 v_F}\left(\Delta_n^0\right)^3$$

Electron-hole asymmetry: small and not seen !

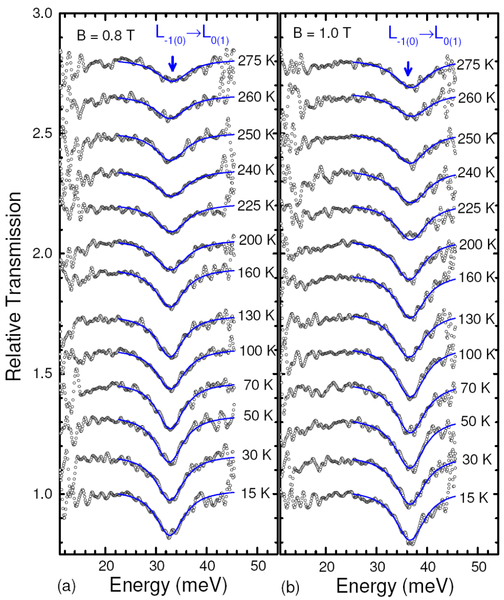
High-Energy Limit of Massless Dirac Fermions in Multilayer Graphene,
P. Plochocka, C. Faugeras, M. Orlita, M.L. Sadowski, G. Martinez, M. Potemski, M.O. Goerbig, J.-N. Fuchs,C. Berger, and W.A. de Heer, Phys. Rev. Lett. 100, 087401 (2008)

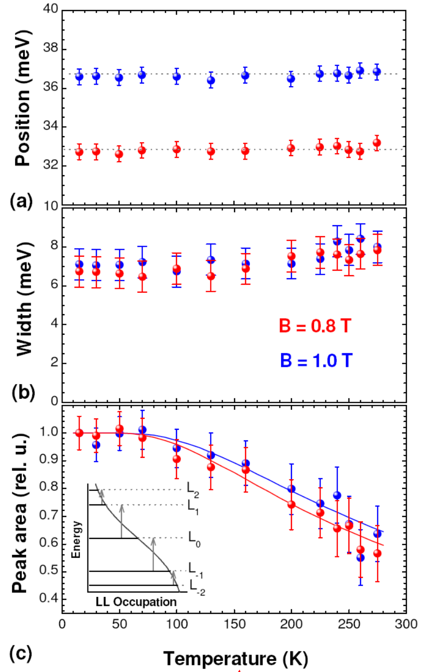
Multilayer epitaxial graphene : quasi neutral layers on C-face SiC quality of electronic states ?





Carrier scattering from cyclotron resonance measurements





$$\sigma \approx 10 e^2 / h$$

$$\Delta\omega \cdot \tau = 1 \rightarrow \tau = 200\ fs$$

no temperature activated scattering

$$\mu B > 1 \rightarrow \mu > .25 \cdot 10^6 \frac{cm^2}{Vs}$$

$$n_e \cong 0.5 \cdot 10^{10} cm^{-2}, \quad E_F \cong 6meV$$

Record RT-mobility !

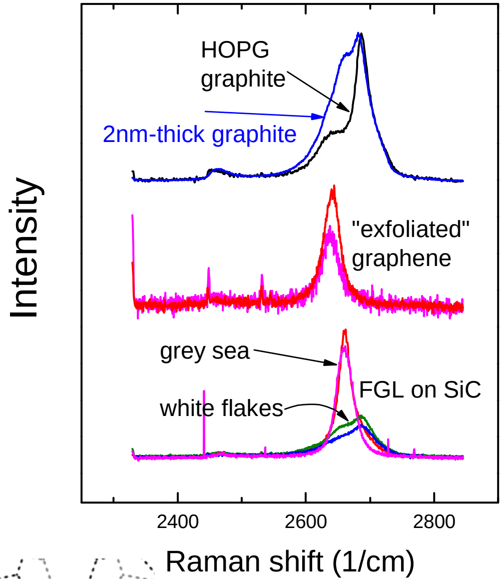
Ultrahigh mobility in multilayered epitaxial graphene: a step towards graphene based electronics

M. Orlita, C. Faugeras, P. Plochocka, P. Neugebauer, G. Martinez, D.K. Maude, A.-L. Barra, M. Sprinkle C. Berger, W.A. de Heer, M. Potemski, Phys. Rev. Lett. 100, 136403 (2008).

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

C-terminated SiC surface

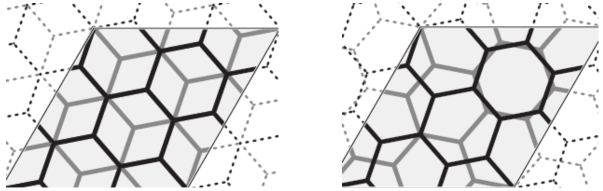
Multilayered graphene



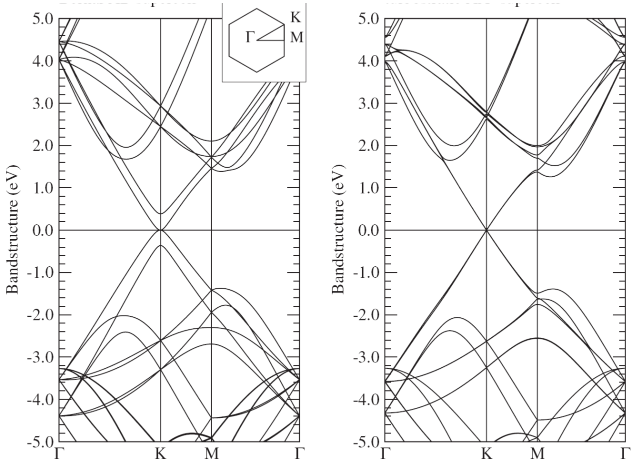
C. Faugeras et al., APL, 2007

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

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



Bernal AB turbostratic AA' bilayer



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

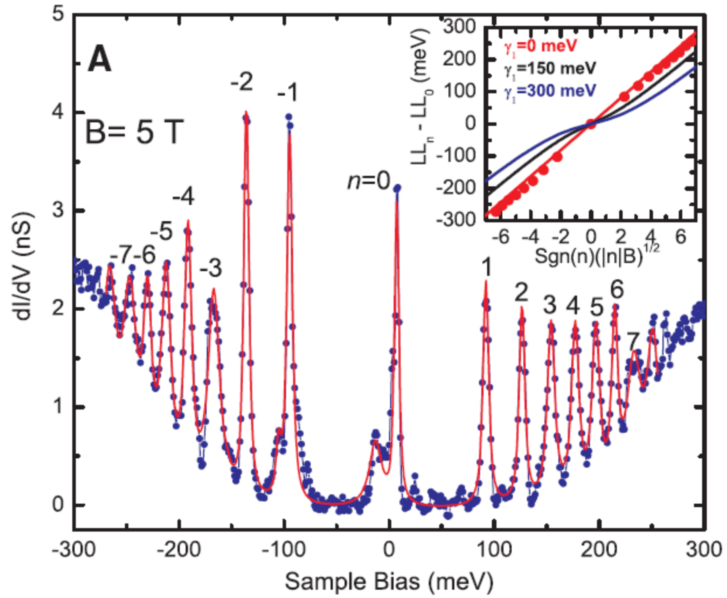
Latest confirmations of Dirac-like electronic bands

15 MAY 2009 VOL 324 SCIENCE www.sciencemag.org

Observing the Quantization of Zero Mass Carriers in Graphene

David L. Miller,[1]* Kevin D. Kubista,[1]* Gregory M. Rutter,[2] Ming Ruan,[1] Walt A. de Heer,[1]
Phillip N. First,[1]† Joseph A. Stroscio[2]†

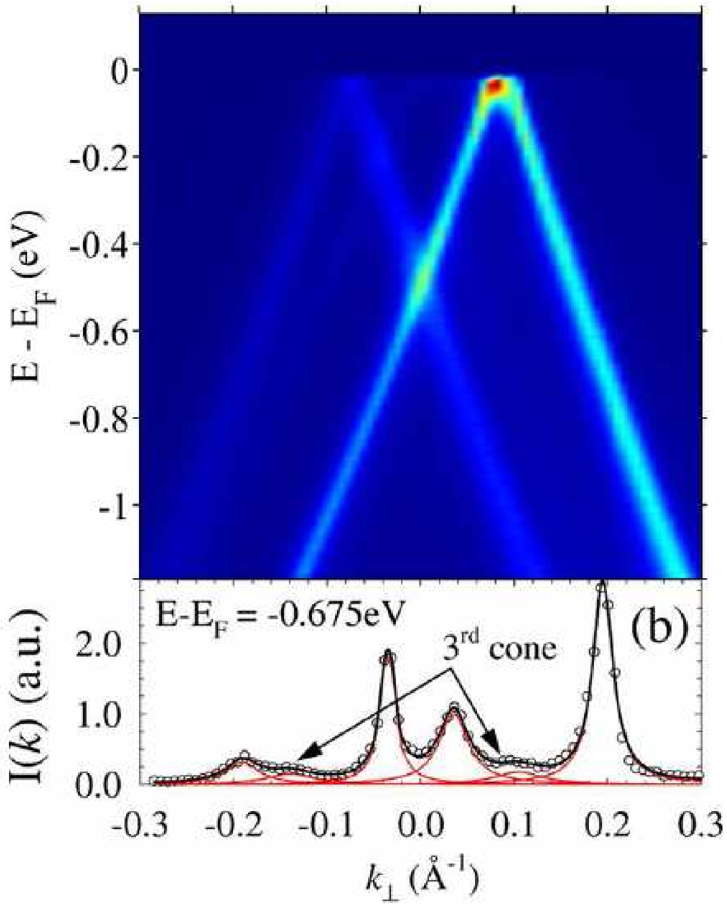
Application of a magnetic field to conductors causes the charge carriers to circulate in cyclotron orbits with quantized energies called Landau levels (LLs). These are equally spaced in normal metals and two-dimensional electron gases. In graphene, however, the charge carrier velocity is independent 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

M. Sprinkle,[1] D. Siegel,[2,3] Y. Hu,[1] J. Hicks,[1] P. Soukiassian,[4] A. Tejeda,[5,6] A. Taleb-Ibrahimi,[7] P. Le Fèvre,[6] F. Bertran,[6] C. Berger,[1,8] W.A. de Heer,[1] A. Lanzara,[2,3] and E.H. Conrad[1]



Electron-phonon interaction

Journal of the Physical Society of Japan
Vol. 75, No. 12, December, 2006, 124701

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

nature materials | VOL 6 | MARCH 2007 | www.nature.com/naturematerials

SIMONE PISANA[1], MICHELE LAZZERI[2], CINZIA CASIRAGHI[1], KOSTYA S. NOVOSELOV[3], A. K. GEIM[3], ANDREA C. FERRARI[1]* AND FRANCESCO MAURI[2]*

PRL **98,** 166802 (2007) PHYSICAL REVIEW LETTERS week ending 20 APRIL 2007

Electric Field Effect Tuning of Electron-Phonon Coupling in Graphene

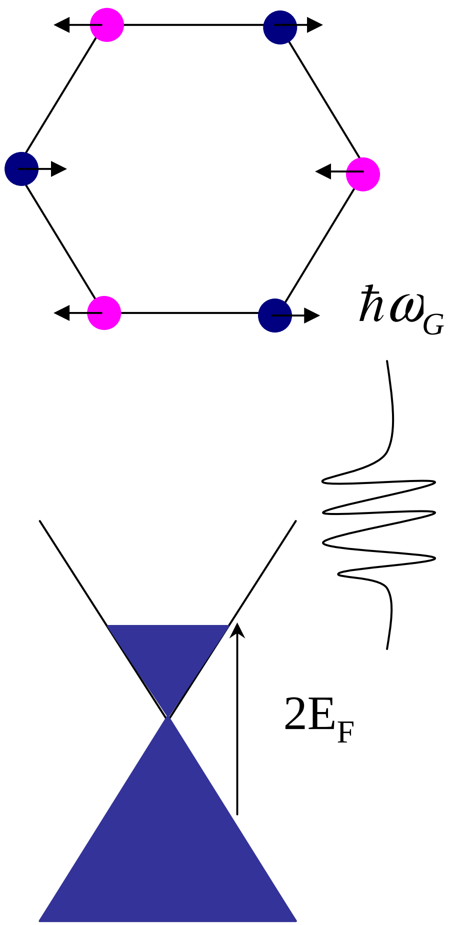
Jun Yan,[1] Yuanbo Zhang,[1] Philip Kim,[1] and Aron Pinczuk[1,2]

Electron-phonon interaction

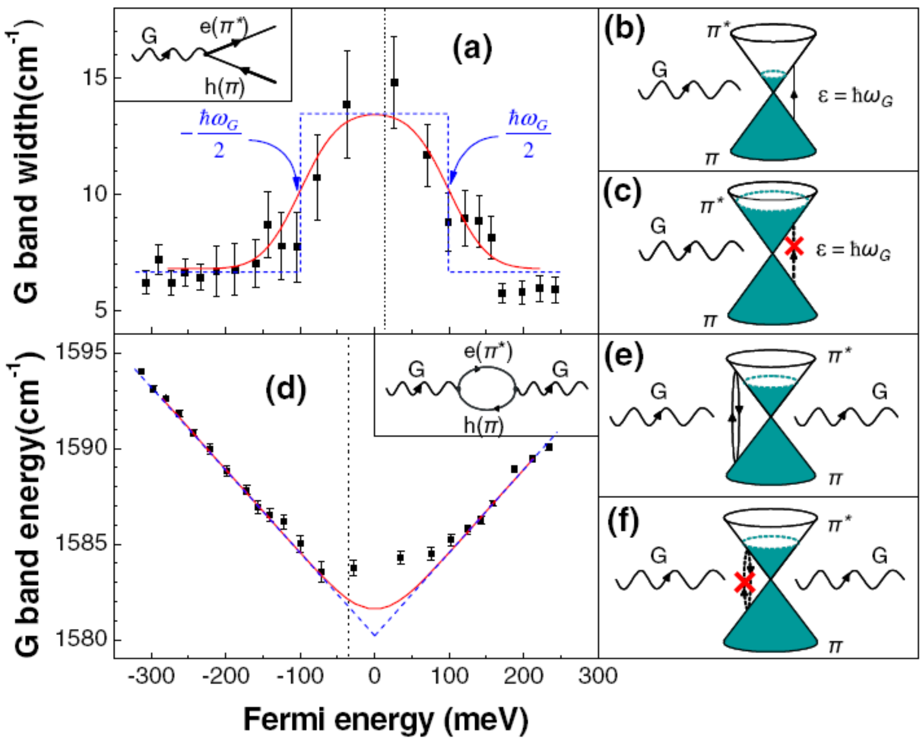
PRL **98,** 166802 (2007) PHYSICAL REVIEW LETTERS week ending 20 APRIL 2007

Electric Field Effect Tuning of Electron-Phonon Coupling in Graphene

Jun Yan,[1] Yuanbo Zhang,[1] Philip Kim,[1] and Aron Pinczuk[1,2]



$$\hbar\omega_G \ vs. \ 2E_F$$

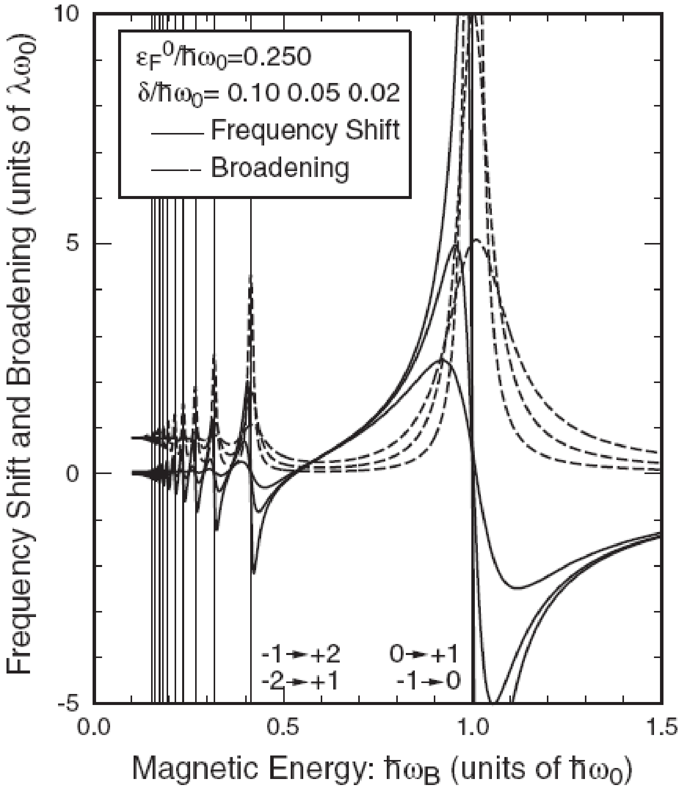


Tuning electron-phonon coupling with magnetic field in graphene

Journal of the Physical Society of Japan
Vol. 76, No. 2, February, 2007, 024712

Magnetic Oscillation of Optical Phonon in Graphene

Tsuneya ANDO



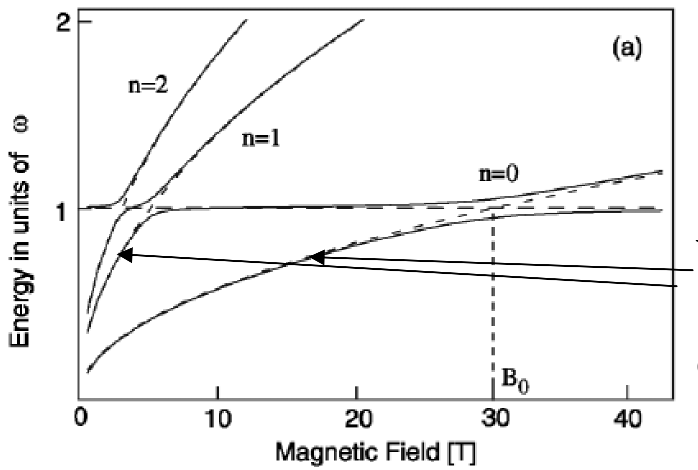
G-phonon couples to
inter Landau level transitions
which follow the same selection rules as
optically active transitions

Tuning electron-phonon coupling with magnetic field in graphene

PRL **99**, 087402 (2007) PHYSICAL REVIEW LETTERS

Filling-Factor-Dependent Magnetophonon Resonance in Graphene

M. O. Goerbig,[1] J.-N. Fuchs,[1] K. Kechedzhi,[2] and Vladimir I. Fal'ko[2]



valley asymmetric
inter-Landau level transitions
optically inactive

$$\hbar\omega^{\pm}(n) = \frac{1}{2}(\Omega_n + \hbar\omega_\Gamma) \mp \sqrt{\frac{1}{4}(\Omega_n - \hbar\omega_\Gamma)^2 + g^2(n)}$$

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

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

g = 0.2 eV

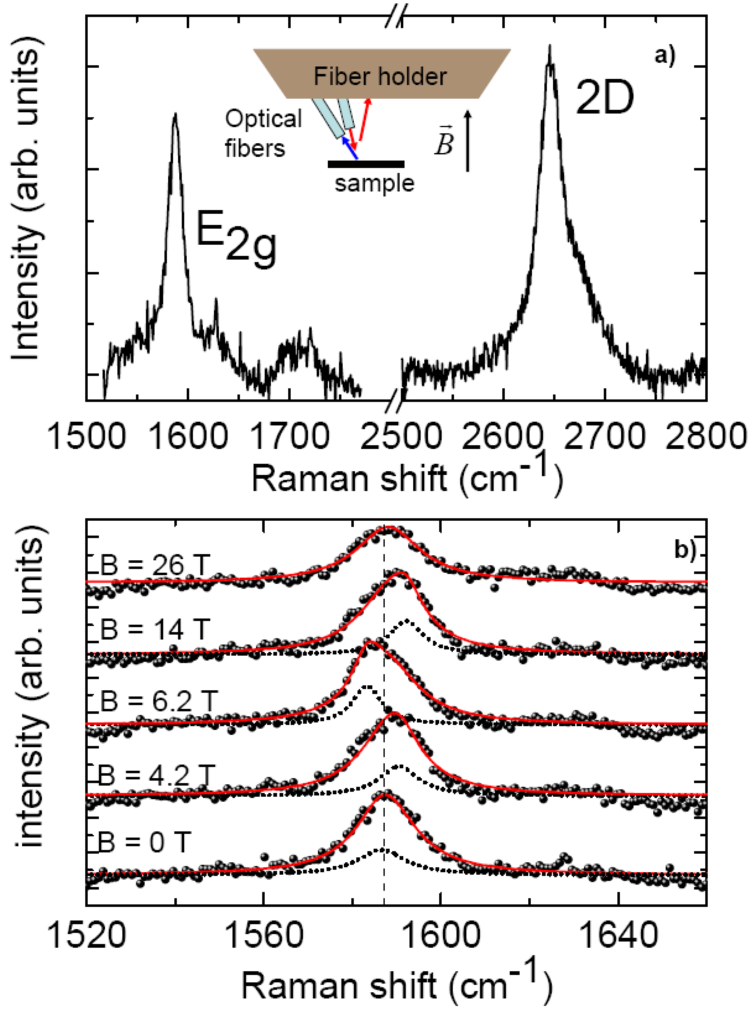
$$A_n = \begin{cases} 2 & if \quad n = 0 \\ 1 & if \quad n \neq 0 \end{cases}$$

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)



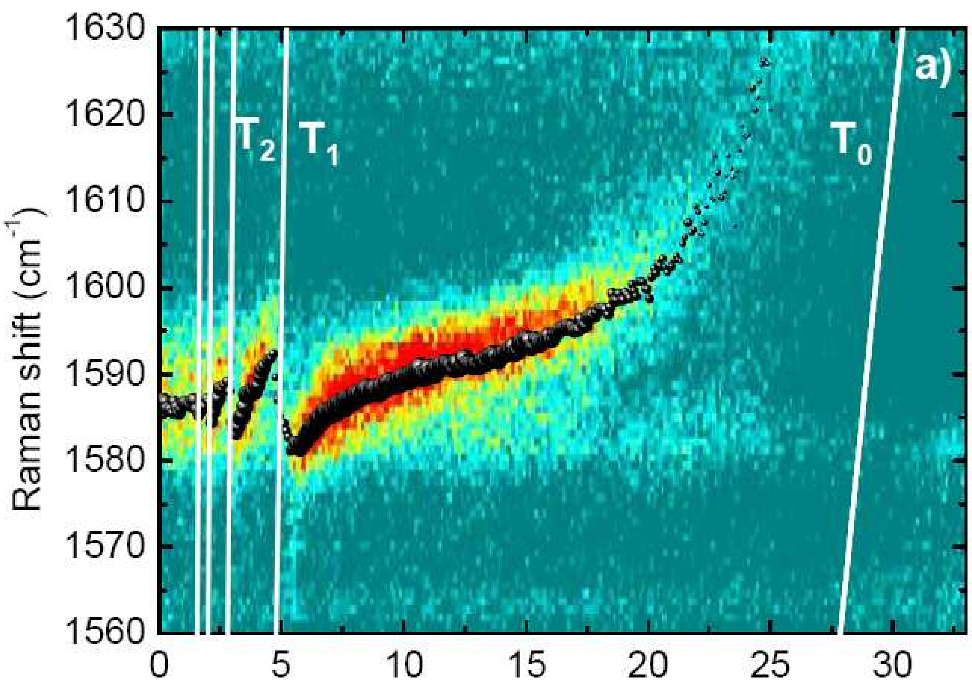
Oscillatory (~ 40%) +
Field independent component

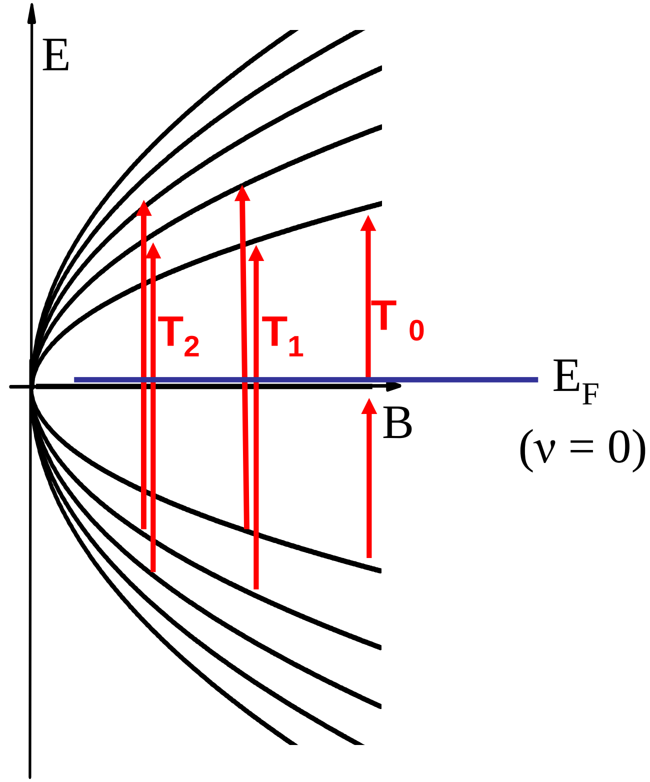
Tuning electron-phonon coupling with magnetic field

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

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

Oscillatory component



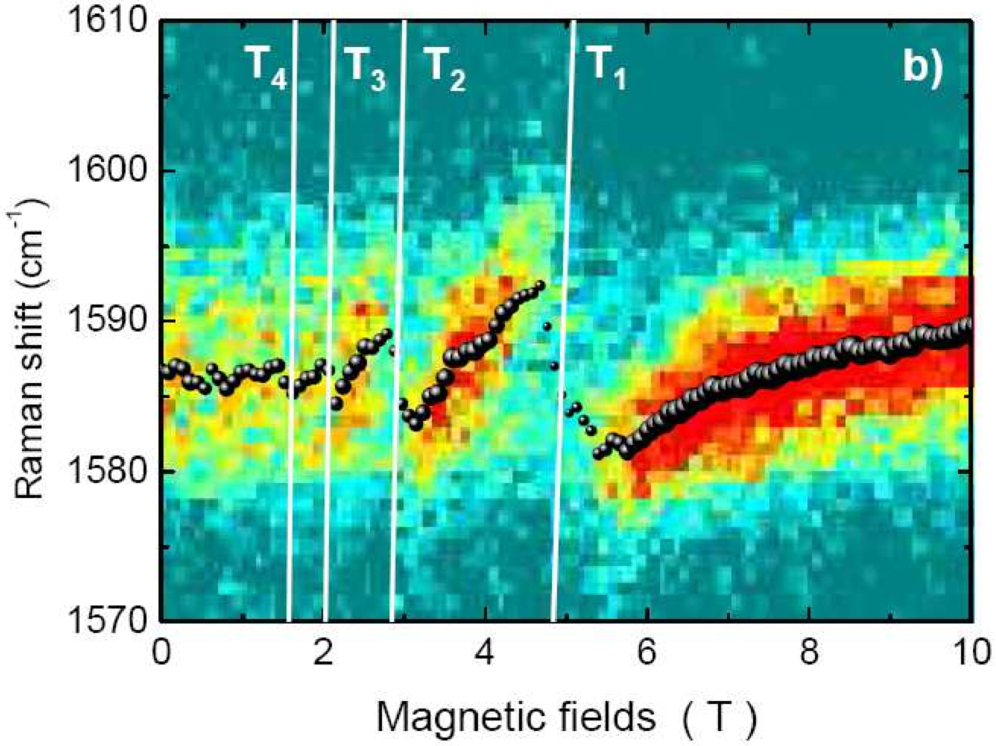


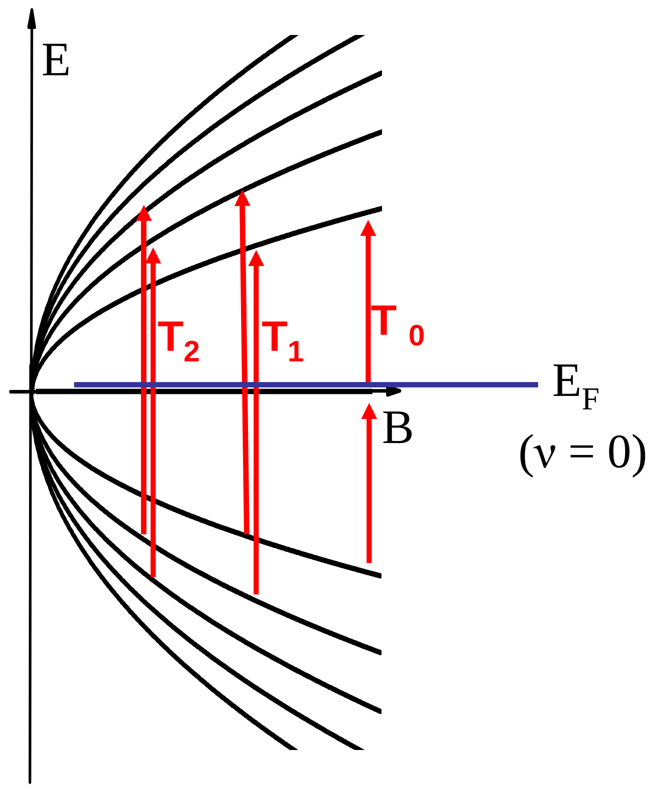
Tuning electron-phonon coupling with magnetic field

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

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

Oscillatory component





Tuning electron-phonon coupling with magnetic field

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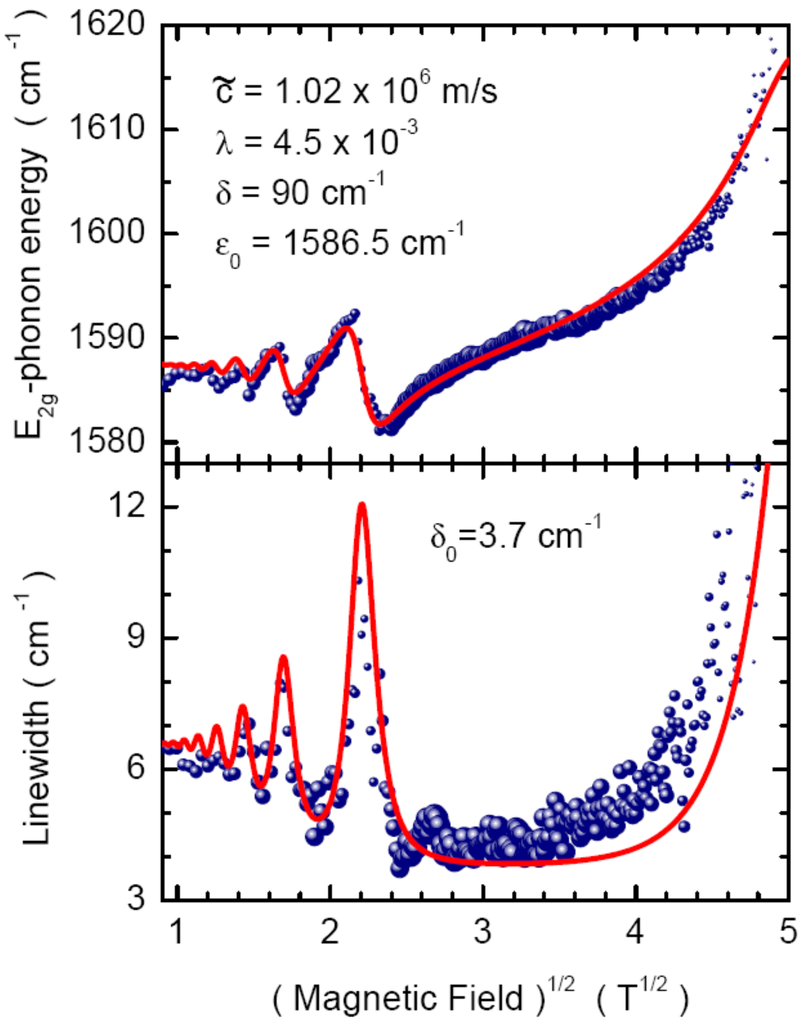
C. Faugeras *et al.*, cond/mat, 2009



approach of Ando [12] to our neutral Dirac-like system and derive the phonon energy ϵ and broadening parameter Γ by extracting $\tilde{\epsilon} = \epsilon - i\Gamma$ from the equation which defines the poles of the phonon Green's function:

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

where ϵ_0 stands for the phonon energy of the neutral system at B=0 T and δ 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 δ_0 accounts for other, than electron-phonon coupling, broadening mechanisms.

Cleaner systems ?

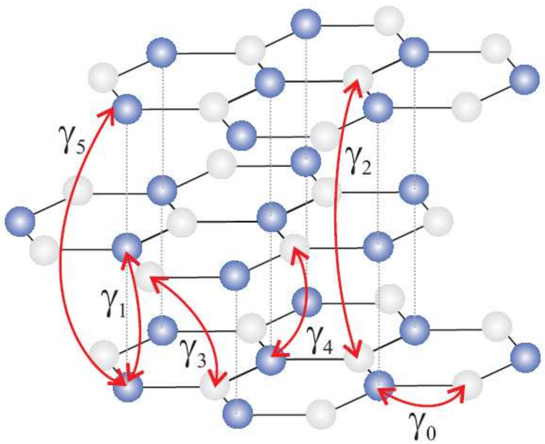
Digression on

(Bulk) graphite ?
higher mobilities, controversial reports

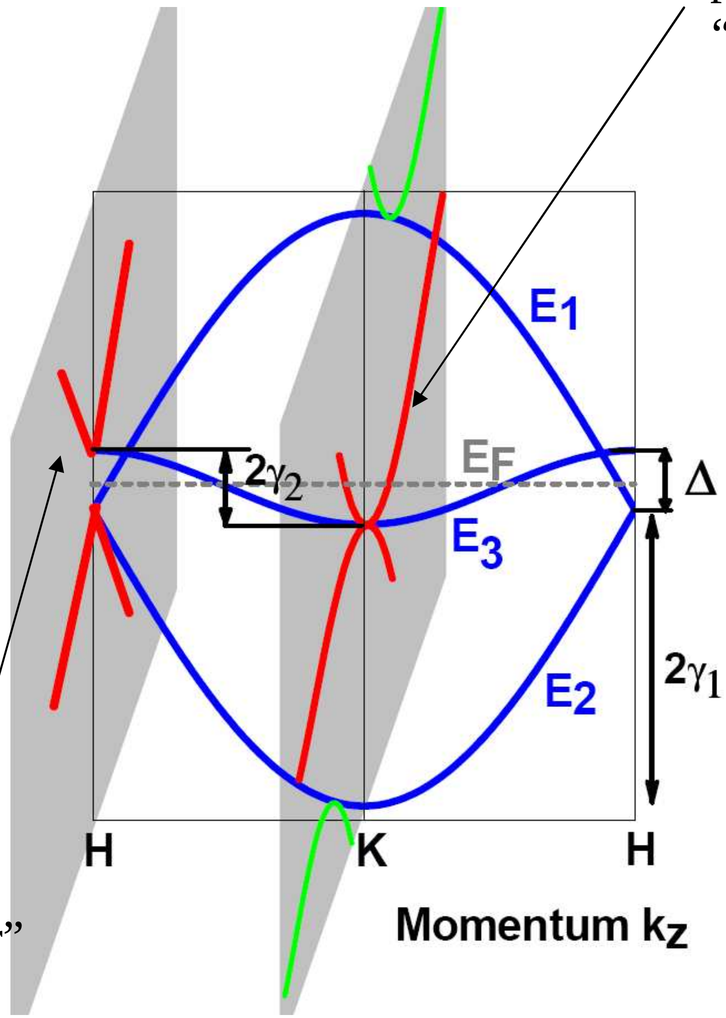
more complex, 3D material

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

J. C. Slonczewski and P. R. Weiss, Phys. Rev. 109, 272 (1958).
J.W. McClure (1964)



parabolic "bilayer"

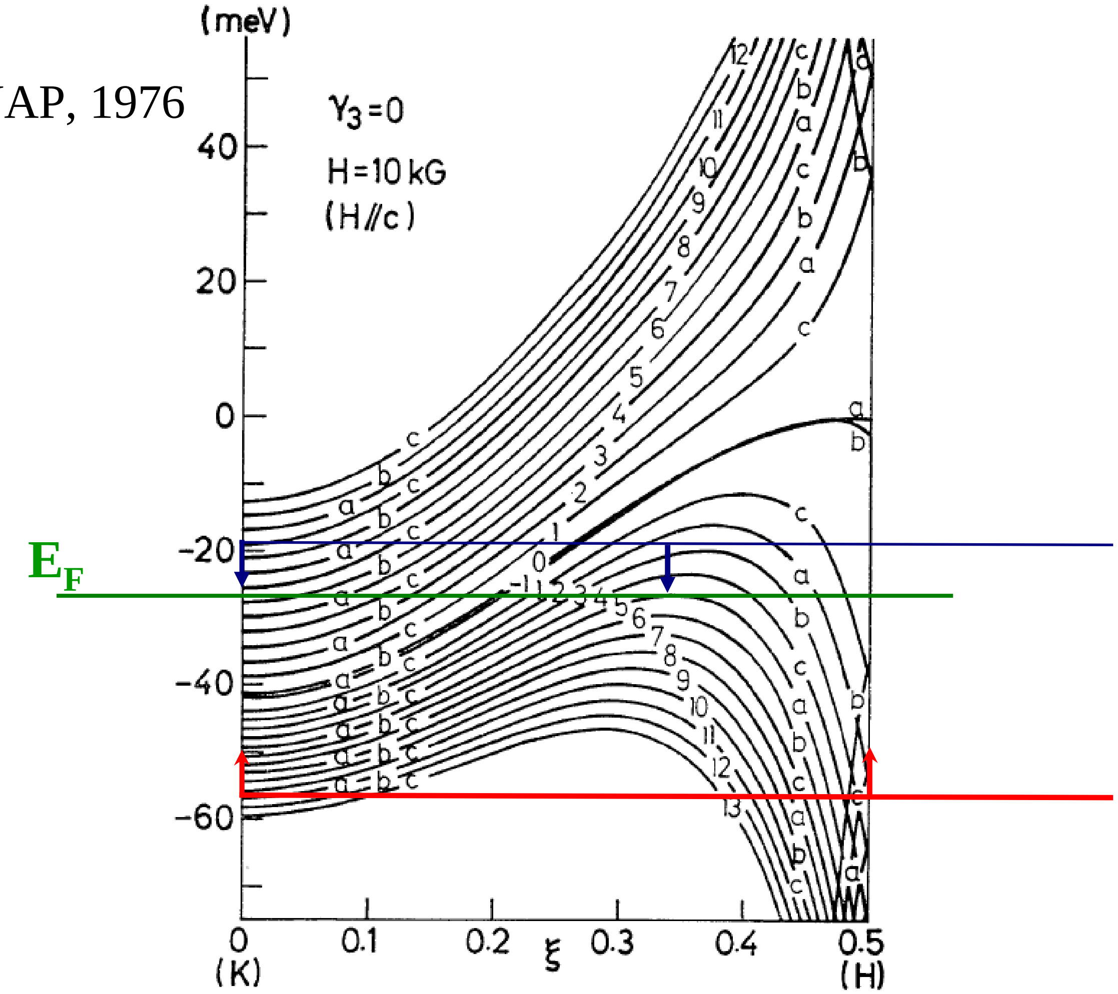


linear "monolayer"

What about graphite: more complex 3D material

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

Nakao, JAP, 1976



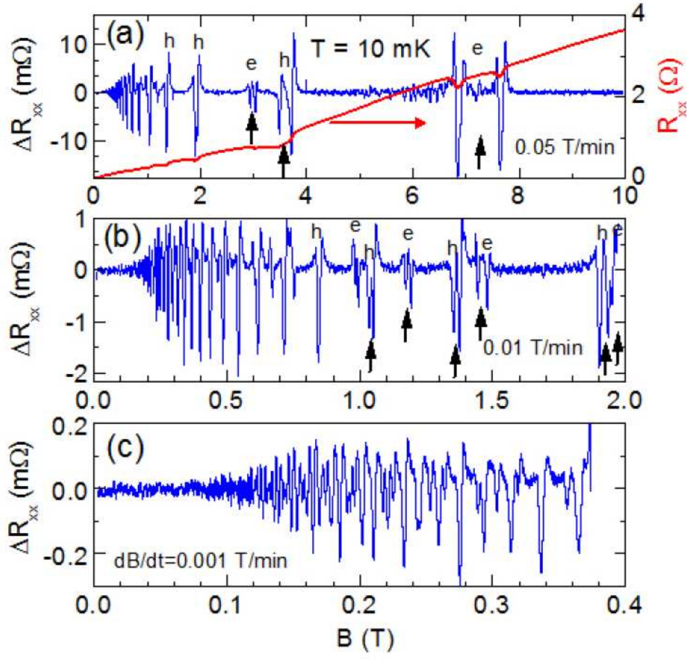
Transport (SdH oscillations): spectroscopy :

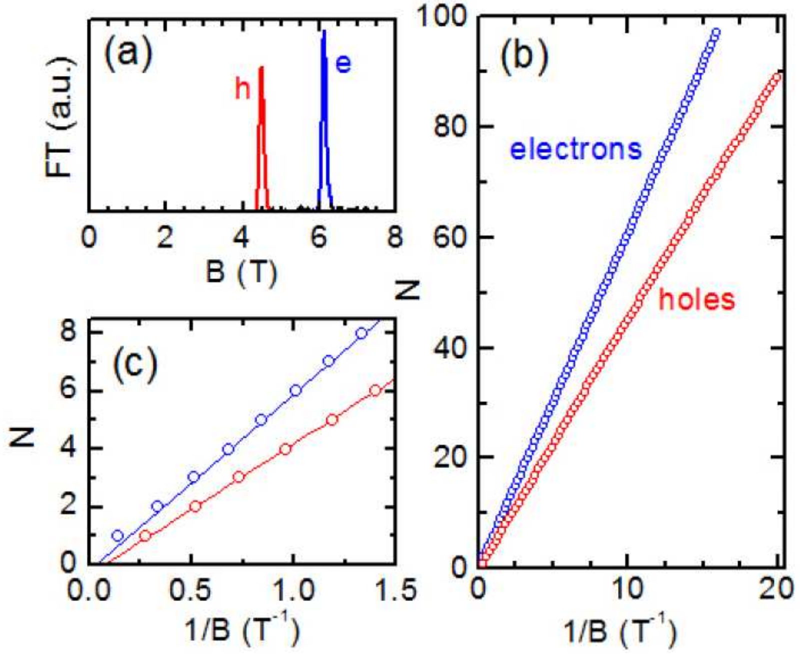
maxima in density of states

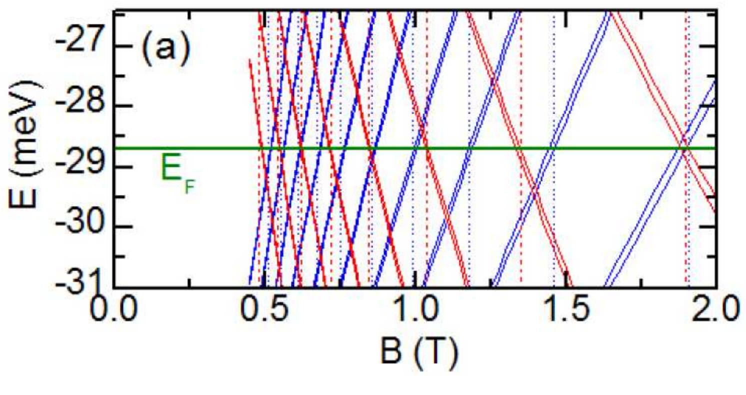
Landau level spectroscopy :

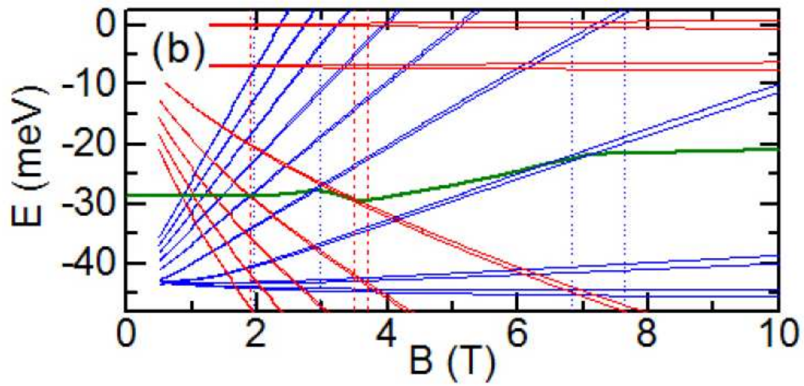
maxima in <u>joint</u> density of states

Graphite: SdH pattern, consistent with SWM model





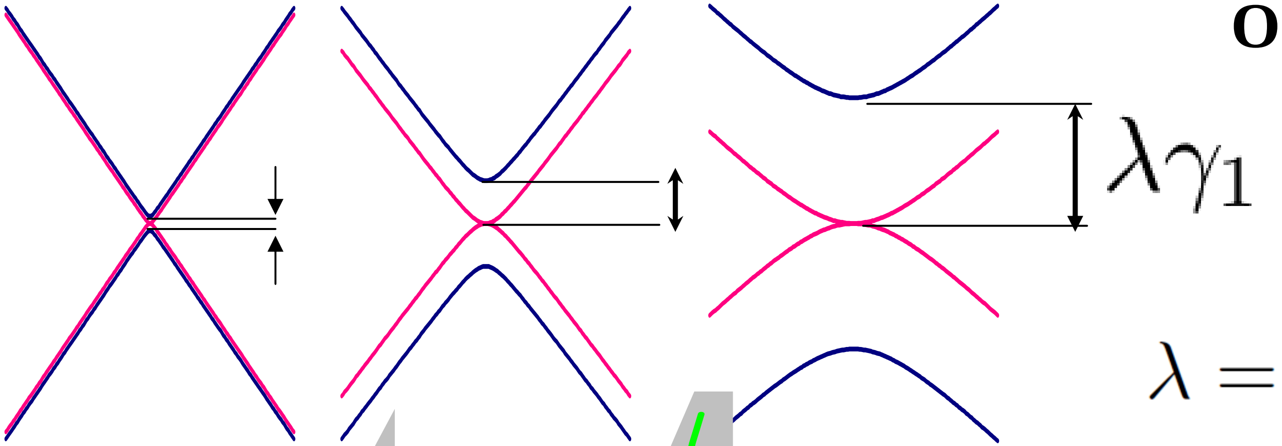




J.M. Schneider, M. Orlita, M. Potemski, D.K. Maude, PRL 102, 166403 (2009)

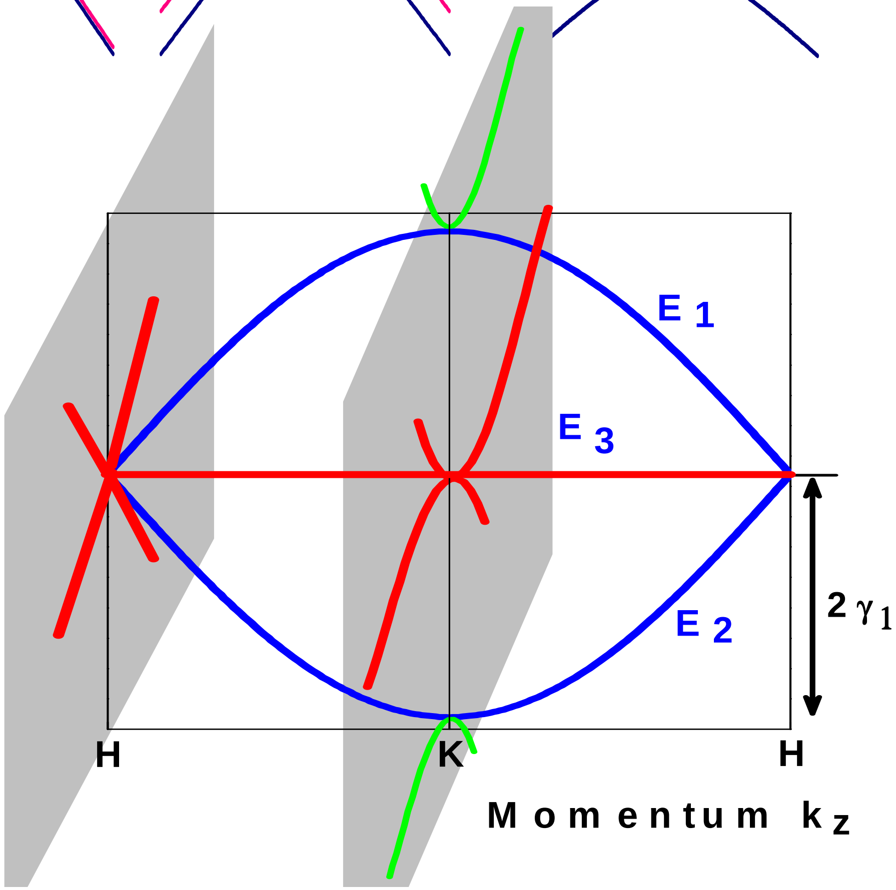
Graphite: simplified model: "effective graphene bilayer"

Only γ_0 and γ_1

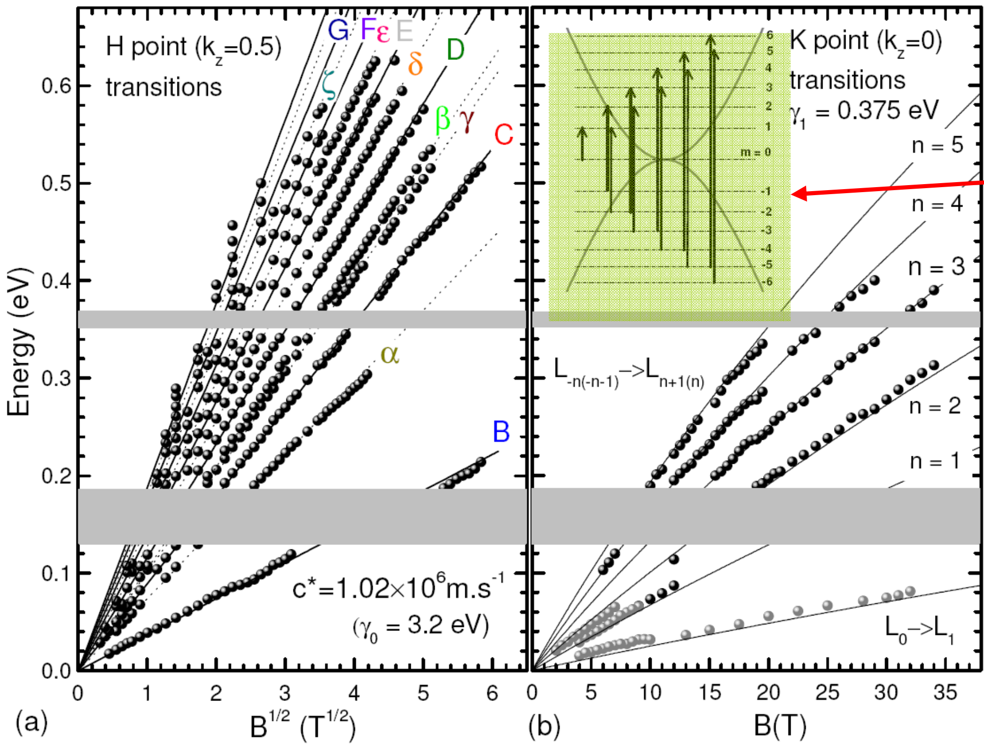


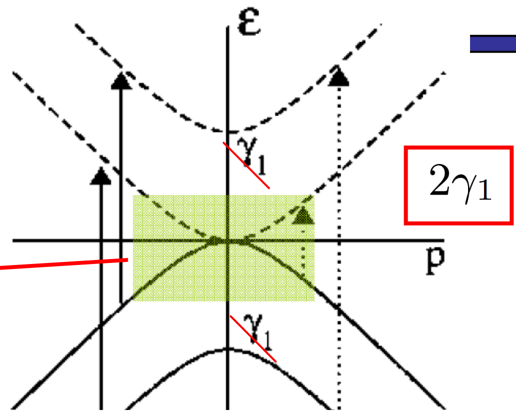
$$\lambda = 2\cos(\pi k_z)$$

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

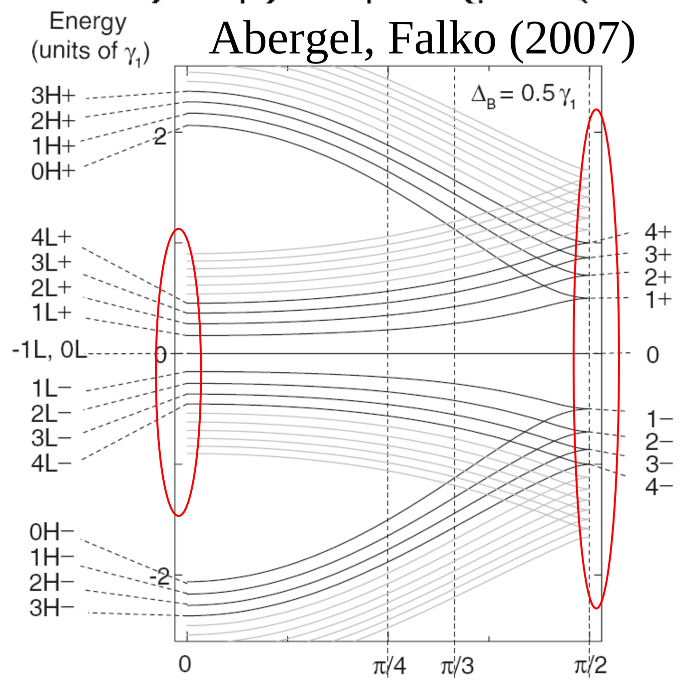


Graphite from viewpoint of Landau level spectroscopy: An effective graphene bilayer and monolayer





Abergel, Falko (2007)



M. Koshino and T. Ando, Phys. Rev. B (2008)

K point **H point**

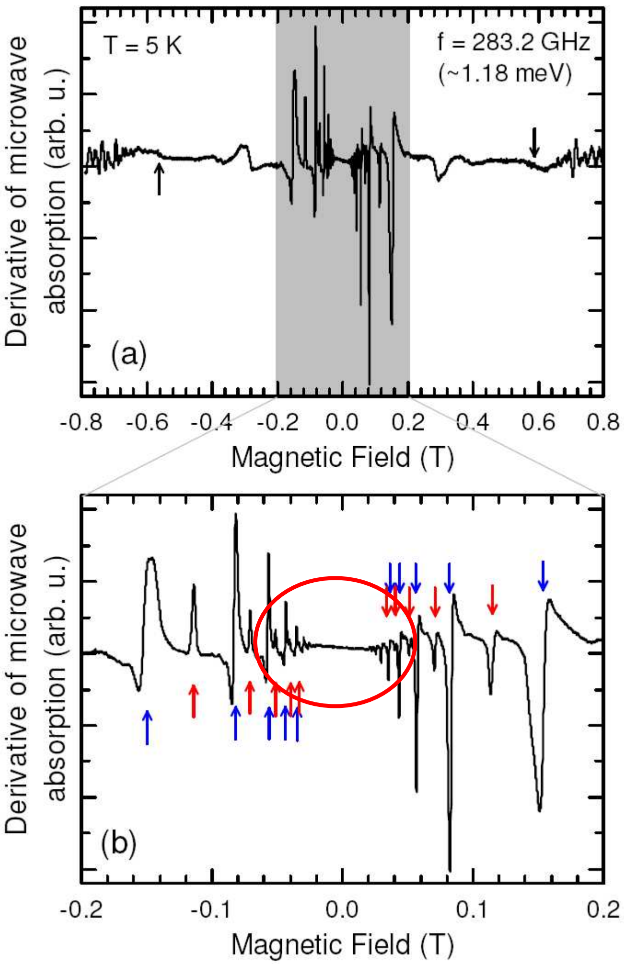
$$\gamma_0 = (3.20 \pm 0.06)\ \text{eV} \qquad \gamma_1 = (375 \pm 10)\ \text{meV}$$

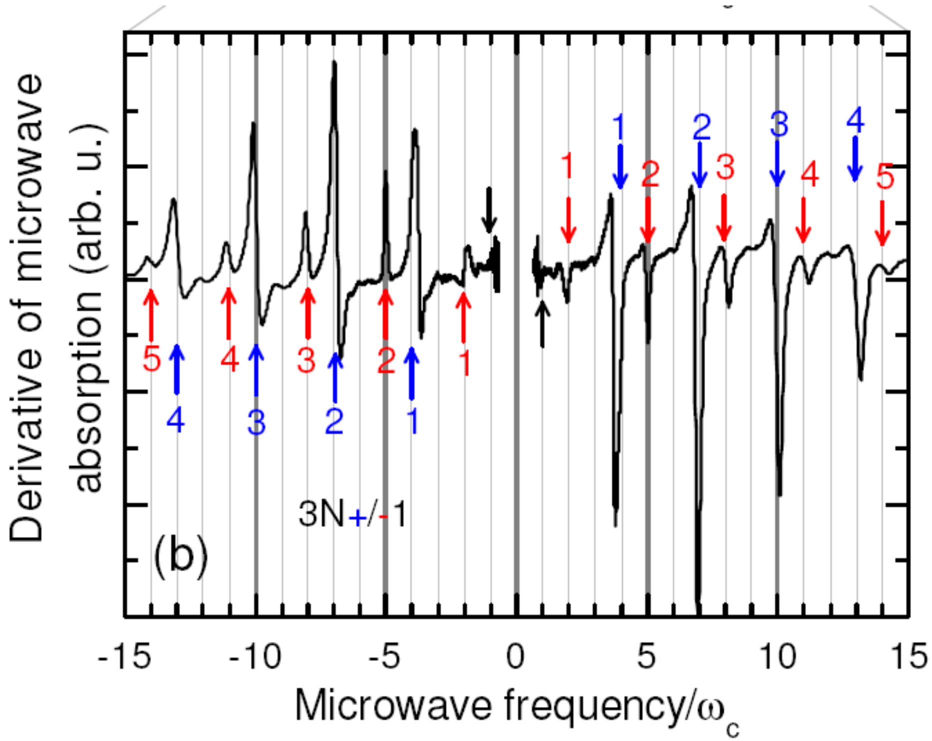
$$m = \gamma_1/\tilde{c}^2 \approx 0.063 m_0$$

For a bilayer : $m = \gamma_1/(2\tilde{c}^2) \approx 0.03 m_0$

M. Orlita, C. Faugeras, J. M. Schneider, G. Martinez, D. K. Maude, and M. Potemski , PRL 102, 166401 (2009)

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





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

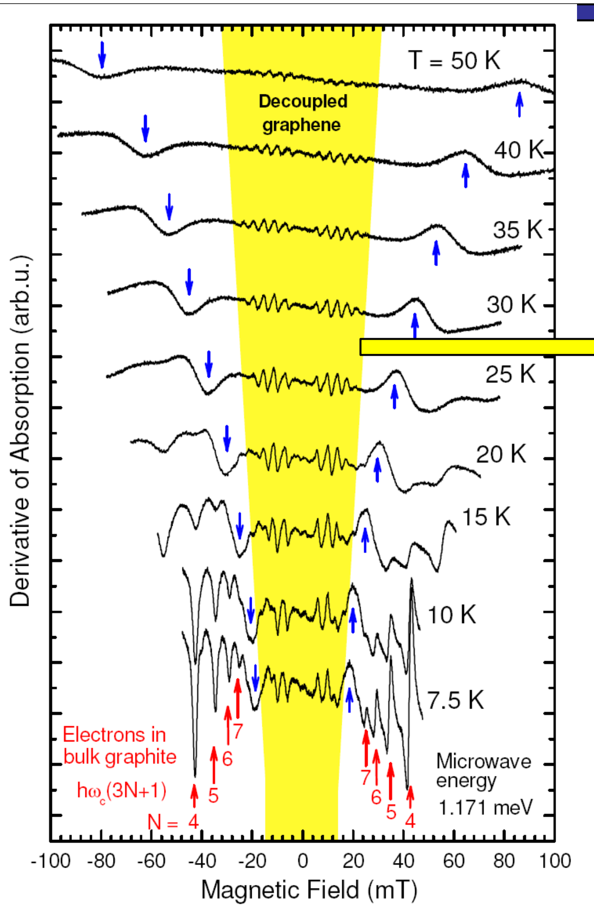
J.K. Galt et al., (1956) P. Nozieres (1958)

Expected also for graphene bilayer: Abergel, Falko (2007)

The cleanest, ever seen Dirac-like system

On graphite surface

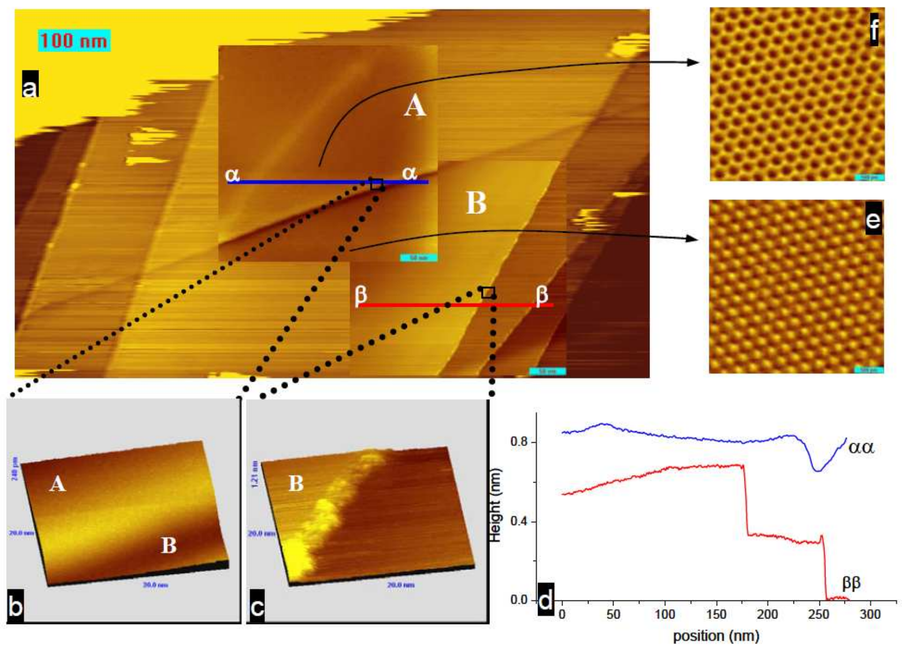
Natural graphite probed with more subtle tools
E_{exc}~ 1meV, very low fields (EPR-type spectroscopy)



Harmonics of K-electron CR

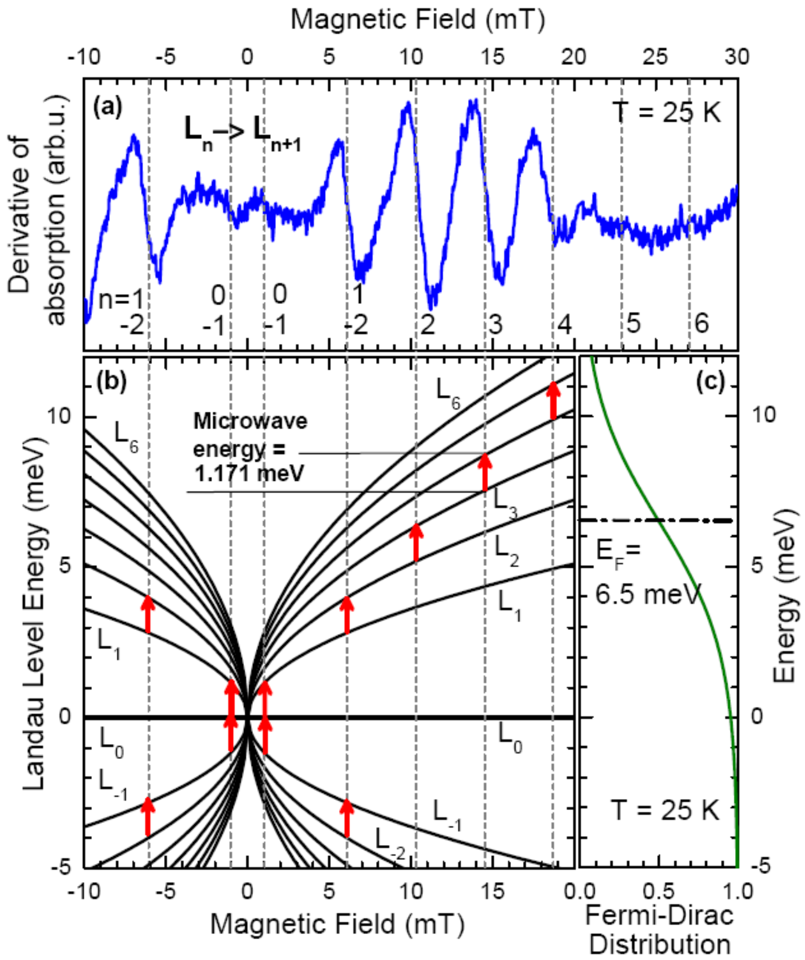
Grahene flakes on graphite !!!

G. Li et al., Phys. Rev. Lett. 102, 170864 (2009)

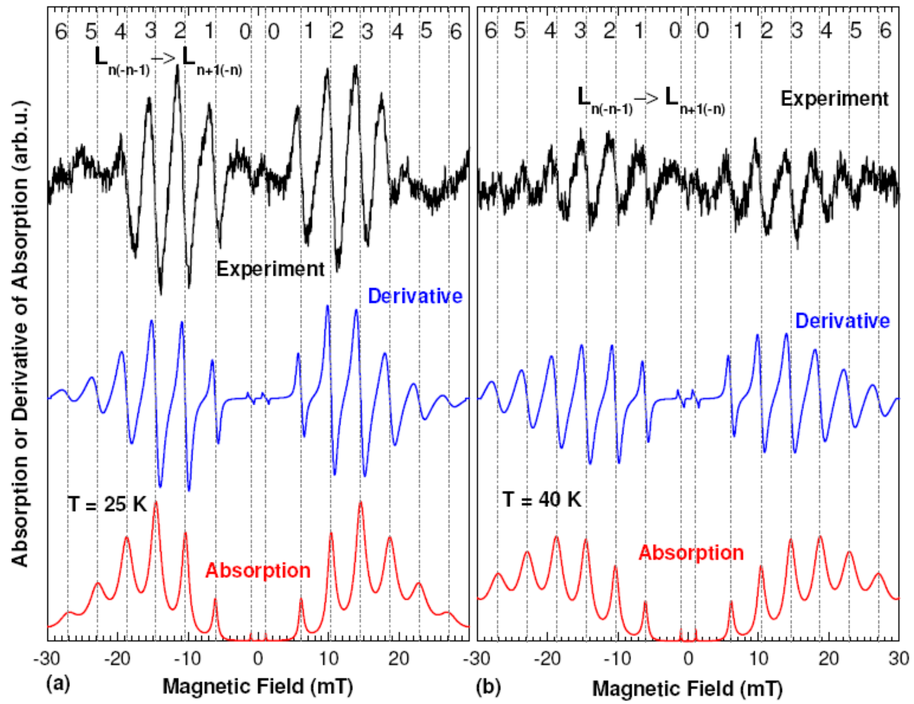


P. Neugebauer, M. Orlita, C. Faugeras, A-L. Barra, and M. Potemski, Cond/Mat (2009)

Grahene flakes on graphite !!!



1 mT



$$\sigma_{xx}(\omega, B) \propto (B/\omega) \sum_{m,n} M_{m,n} \frac{f_n - f_m}{E_m - E_n - (\hbar\omega + i\gamma)},$$

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

Gap < 1 meV

$$\gamma = 35 \ \mu\text{eV} \quad (0.4 \text{ K})$$

Grahene flakes on graphite !!!

Landau level quantisation down to B = 1 mT → $\mu B > 1$ → $\mu > 10^7 \ \mathrm{cm^2/(V.s)}$

$E_F = 6.5 \ \mathrm{meV}$ $n_0 \approx 3 \times 10^9 \ \mathrm{cm^{-2}}$

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

$m = E_F/\tilde{c}^2 \approx 1.3 \times 10^{-3} m_0$

$\gamma = 35 \ \mu\mathrm{eV}$ → $B = (\gamma/E_1)^2 \approx 1 \ \mu\mathrm{T}$ → B_{Earth} $50 \ \mu\mathrm{T}$

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

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

Also at 50 K

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

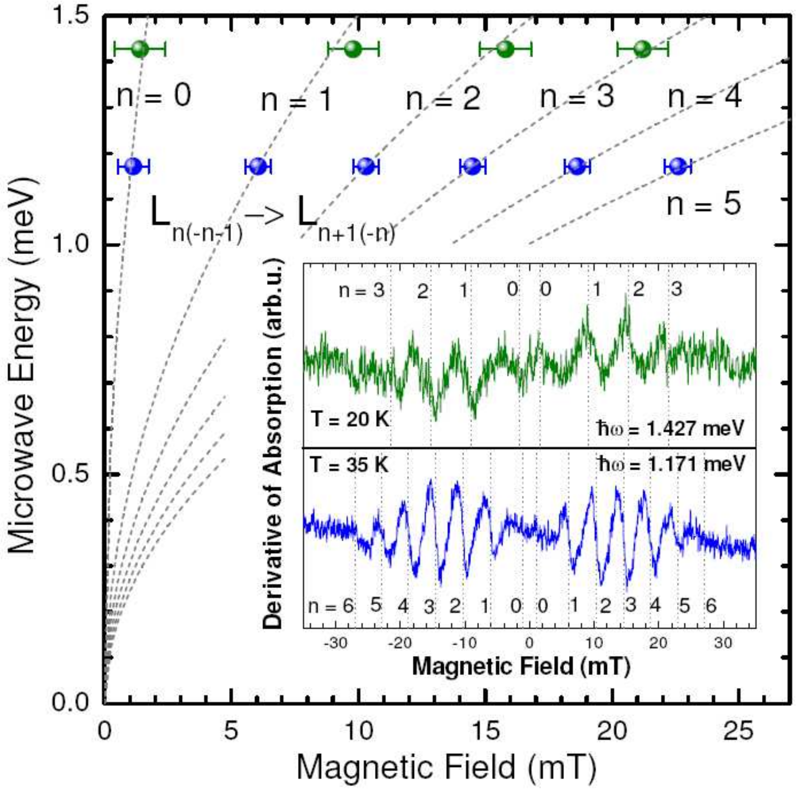
Grahene flakes on graphite ?

H-point of graphite ?
Extremely unlikely: pseudogap ~ 5meV, E_f ~ 20 meV, T-dependence

Several samples investigated

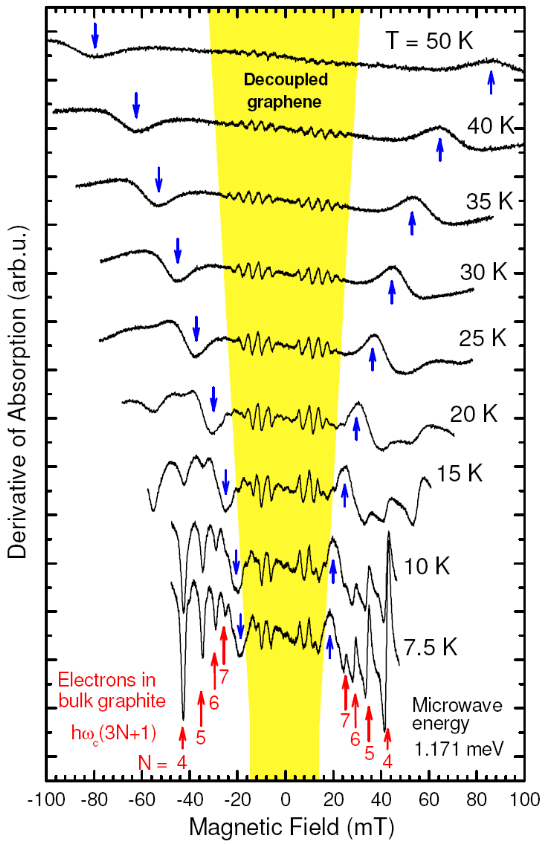
Seen for different frequencies

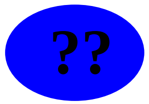
Fast thermal cooling helps (enhanced signal)



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 ...





is likely of collective origin

E_{exc} ~ 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 ?

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

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

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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