Landau level spectroscopy of graphene
(Raman scattering and far-infrared absorption)
Electron-phonon and electron-electron interactions

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CNRS/UJF/UPS/INSA

SEVENTH FRAMEWORK PROGRAMME

European Research Council
Established by the European Commission

MOMB

GRAPHENE FLAGSHIP

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

Nano Lett. 2014, 14, 4548–4553

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

How Perfect Can Graphene Be?

PRL **103,** 136403 (2009)

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

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

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

C. Faugeras, M. Amado, P. Kossacki, M. Orlita, M. Kühne, A. A. L. Nicolet, Yu. I. Latyshev, and M. Potemski

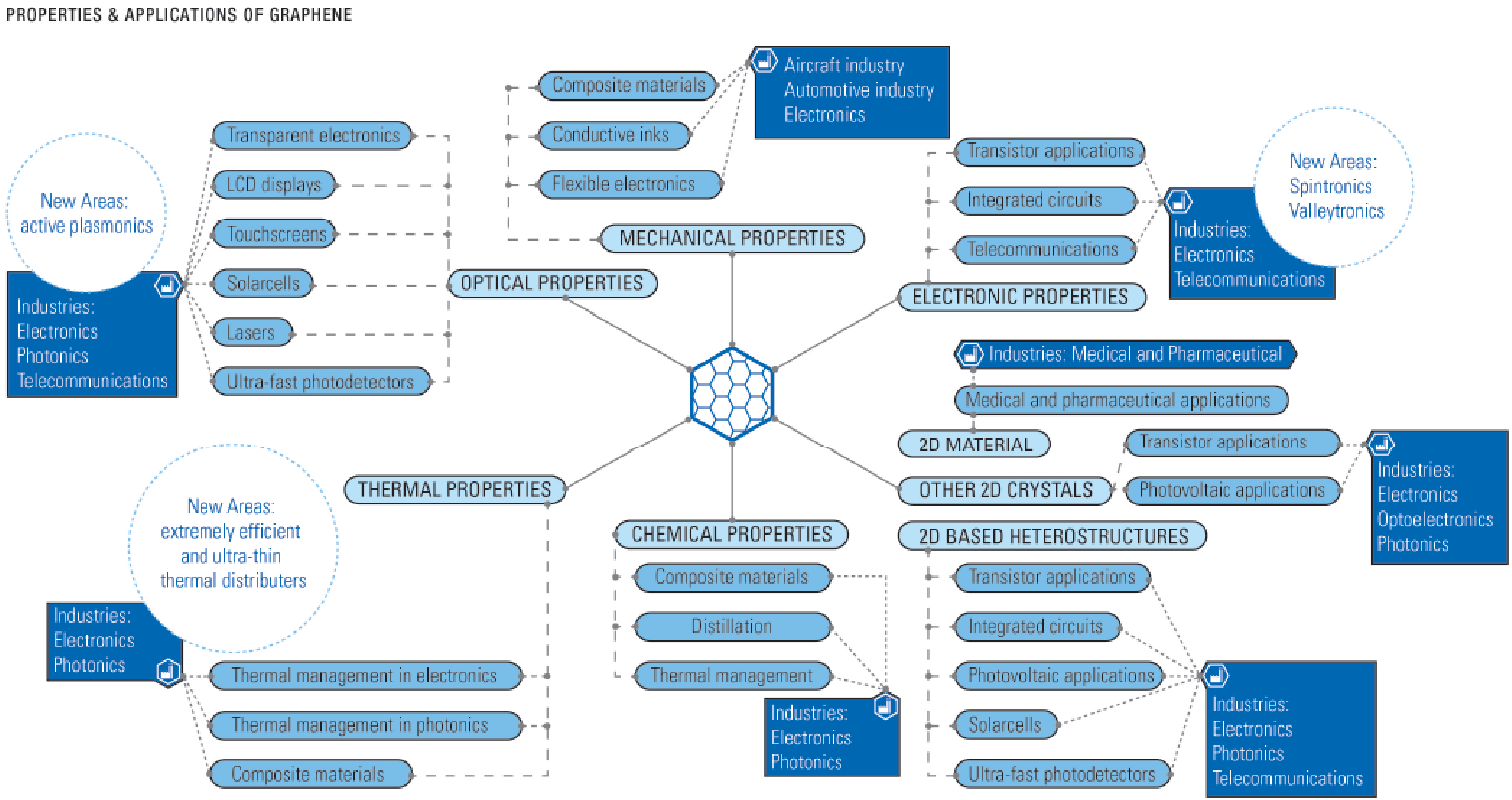
"The ZOO of magneto-phonon resonances in graphene"
D.M. Basko, P. Leszczynski, C. Faugeras… et al., to be published

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. Potemski[1,†]

Why ? Graphene: a truly two-dimensional crystal of sp^2 –bonded carbon



GRAPHENE FLAGSHIP

Graphene-Based Revolutions in ICT And Beyond

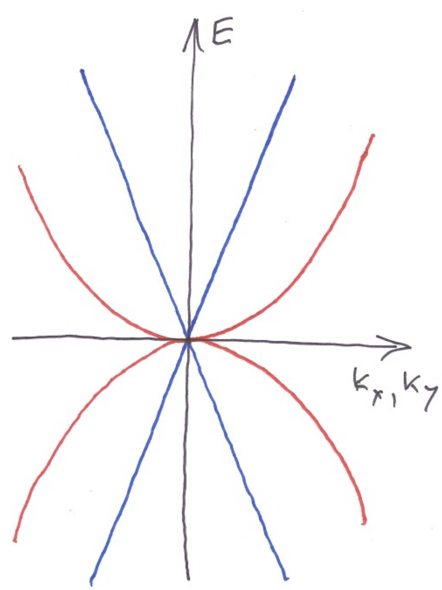
This talk: fundamental properties studied with magnetic fields (spectroscopy)

Dispersion relations and corresponding Landau level ladders

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

~ graphene + (effective) bilayers

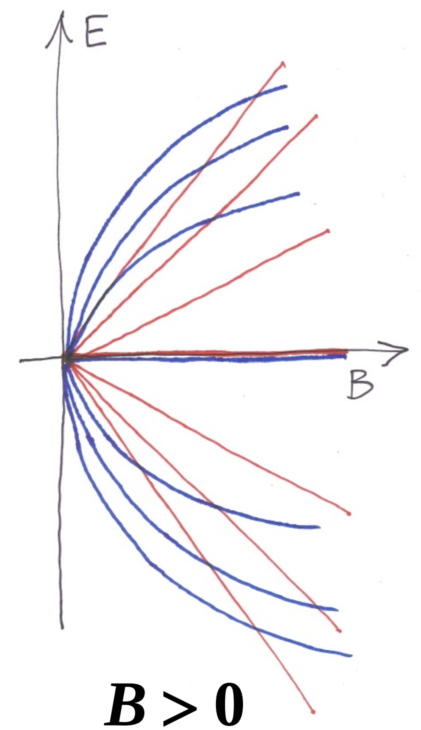
$$E = E(\vec{k})$$



$$\boldsymbol{B = 0}$$

!

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



$$\boldsymbol{B > 0}$$

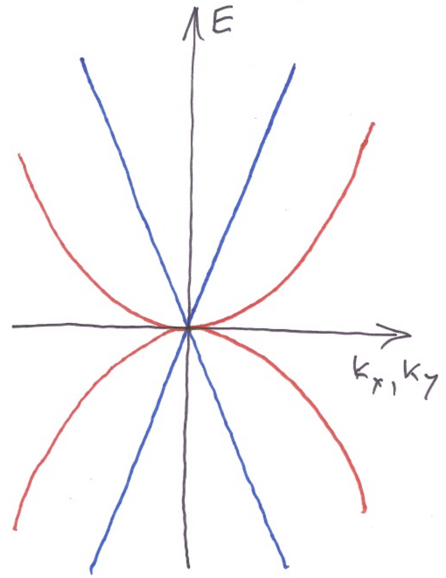
Dispersion relations and corresponding Landau level ladders

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

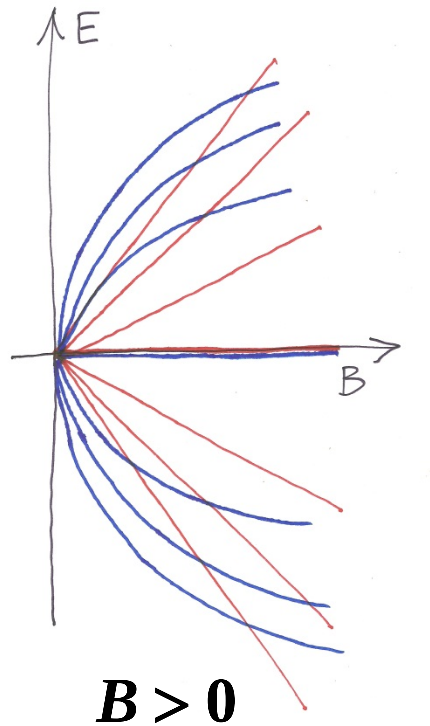
~ graphene + (effective) bilayers

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

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



?!

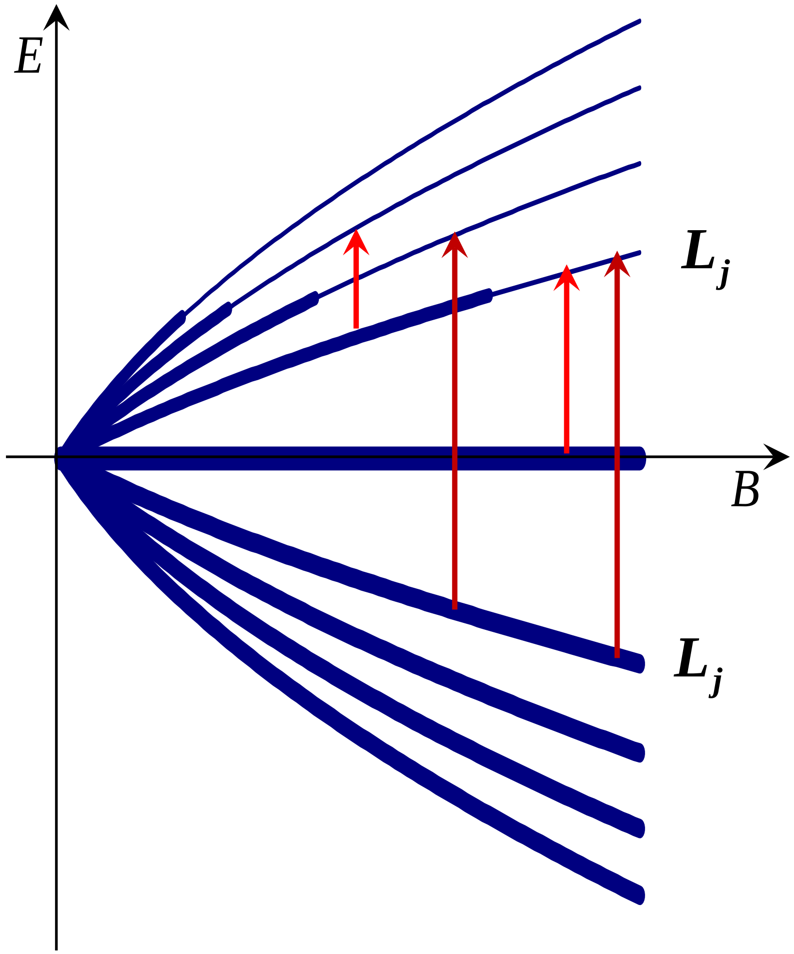


$\boldsymbol{B = 0}$

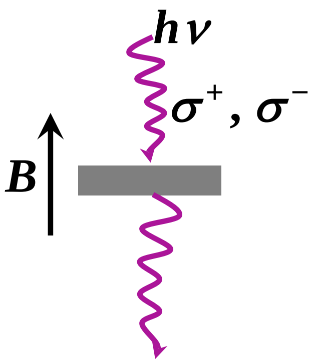
$\boldsymbol{B > 0}$

Landau level spectroscopy

Probing inter Landau level excitations : $L_i \rightarrow L_j$



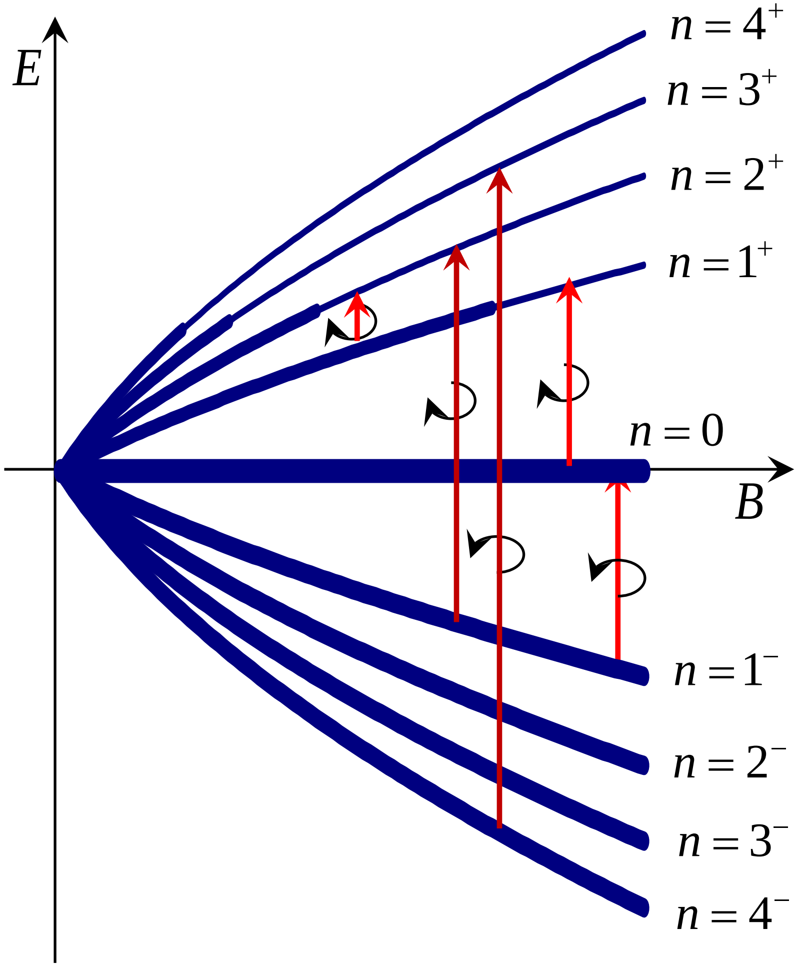
Absorption/transmission



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

resonant when

$h\nu = E_{exc}$



Selection rules

$\Delta n = \pm 1$

$\Delta n = +1 \, : \, \sigma^+$

$\Delta n = -1 \, : \, \sigma^-$

and

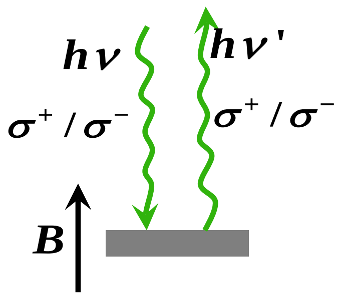
$\Delta n = 2, 4, 5, 7, 8$

if trigonal warping

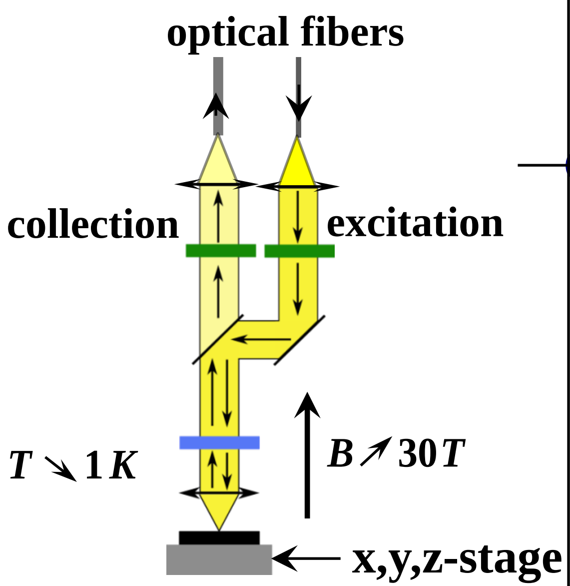
V.P. Gusynin & S.G. Sharapov, PRB, 2006
M. Koshino & T. Ando, PRB, 2008
M. Mucha-Kruczynski *et al.*, J. Phys., 2009

M.L. Sadowski *et al.*, SSC, 2007

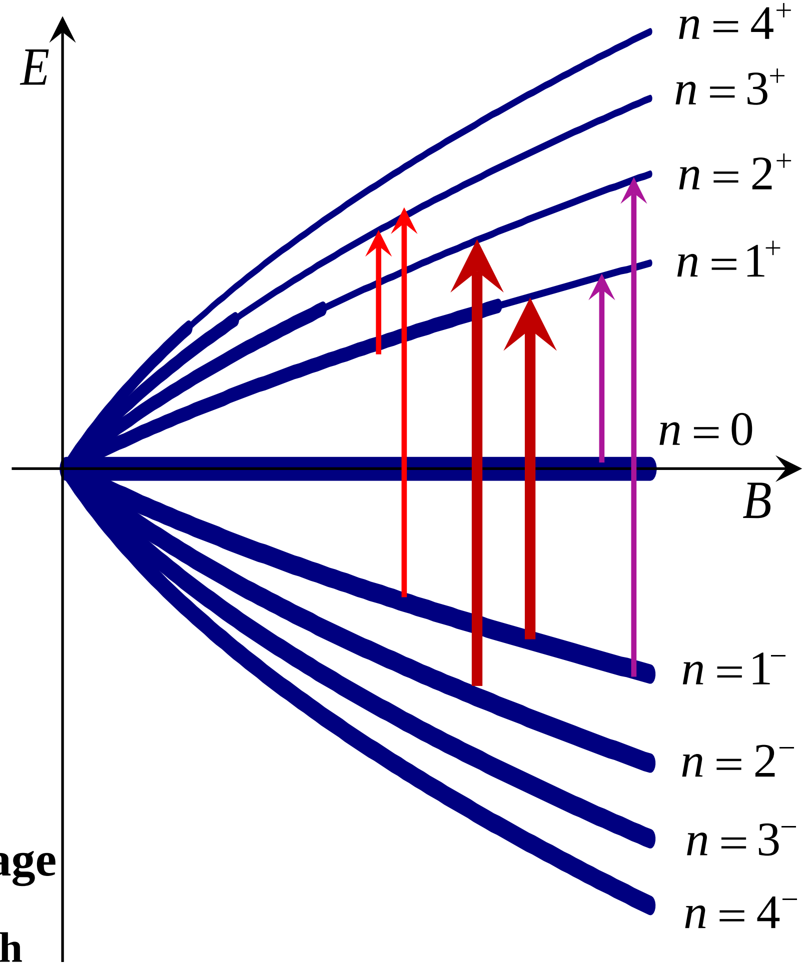
Raman scattering



$$E_{exc} = h\nu' - h\nu$$



miniaturized optical bench



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

Faugeras, Kossacki, Breslavetz, …

O. Kashuba & V.I. Falko PRB, 2009; M. Mucha-Kruczynski *et al.*, PRB, 2010

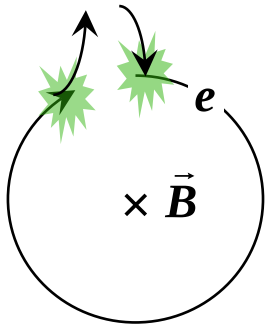
What can be learned from magneto-optics ?

Band structure √

Scattering: efficiency (mechanism) ?

Scattering ?

Classical condition for observation of cyclotron resonance (Landau quantization)



$$\tau_{scattering} > T_{cyclotron}$$

$$\tau_S > 1/\omega_C$$

$\mu > 1/B_{\min}$ rough estimate of carrier mobility

More general :

Spectral broadening $\Gamma \leftarrow 1/\tau_{scat}$

Scattering mechanisms $\leftarrow \Gamma = \Gamma(B, E)$

What can be learned ?

Band structure √

Scattering: efficiency (and mechanism) √

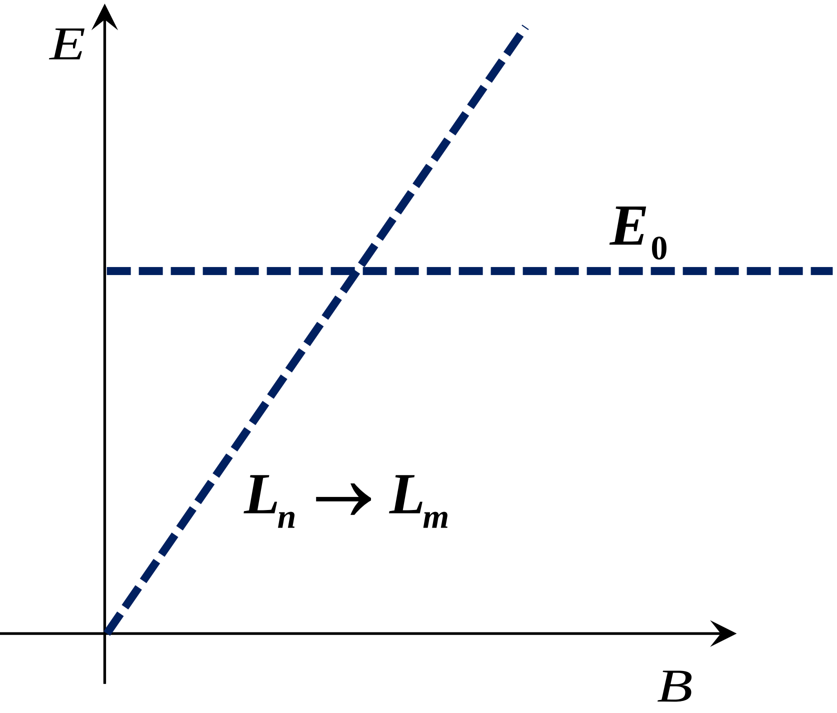
Interactions (?) :

electron-phonon

electron-electron

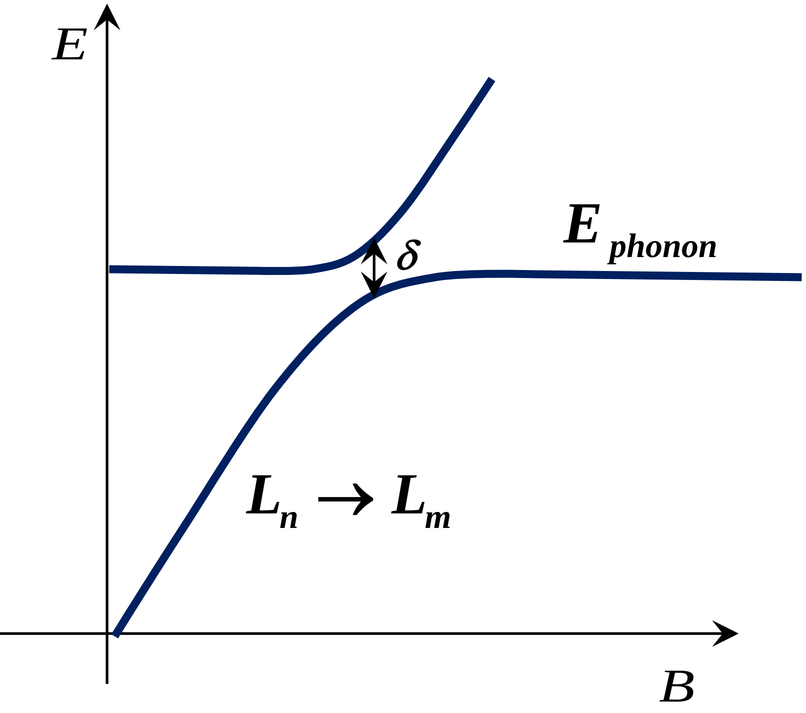
Interactions ?

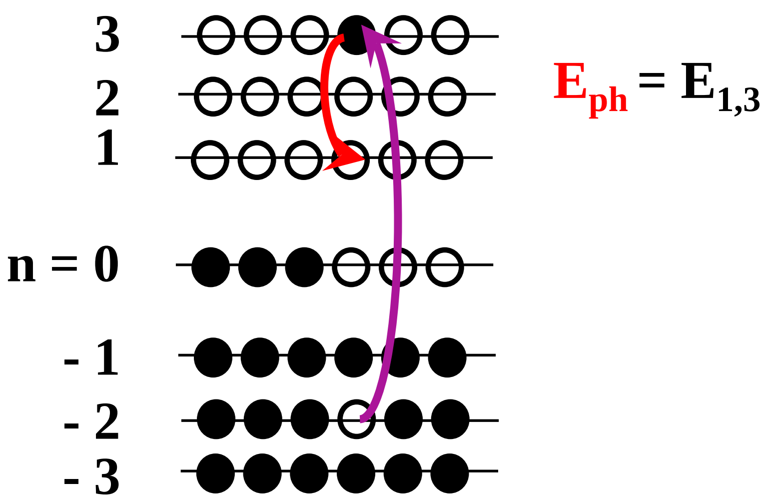
tuning the excitations in resonance



Interactions ?

resonant electron-phonon coupling ?





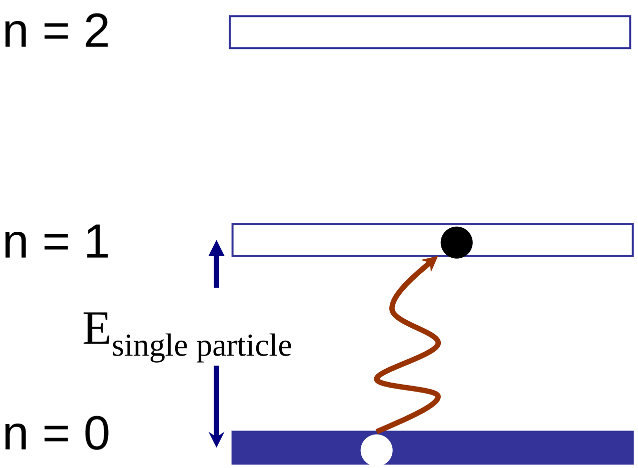
+ more than this !!

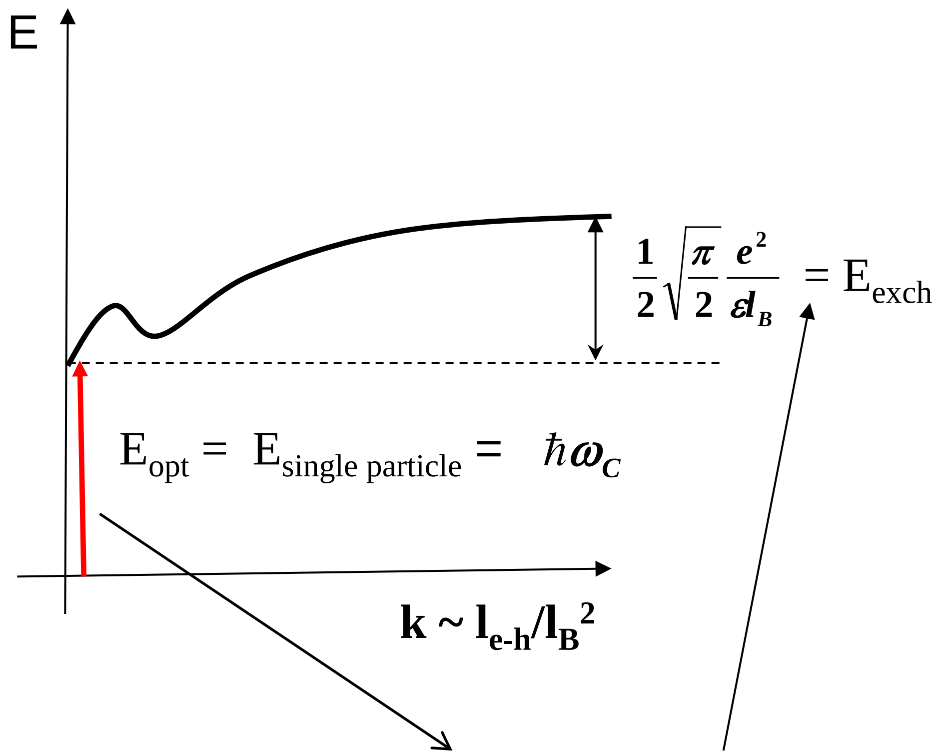
strength of interaction ← δ

other magneto-phonon" resonance

Electron-electron interactions and inter Landau level transitions

Parabolic dispersions
equidistant LLs





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

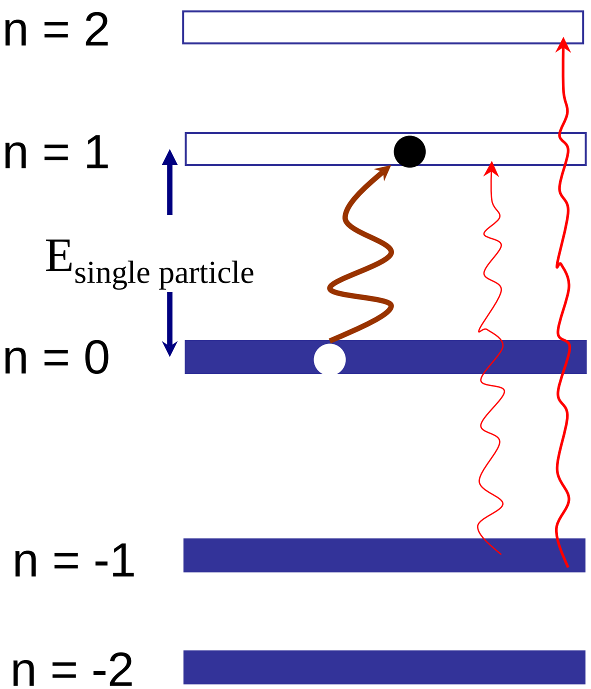
$$k_{opt} \sim 1/\lambda \ll k_{coll} \sim 1/l_B$$

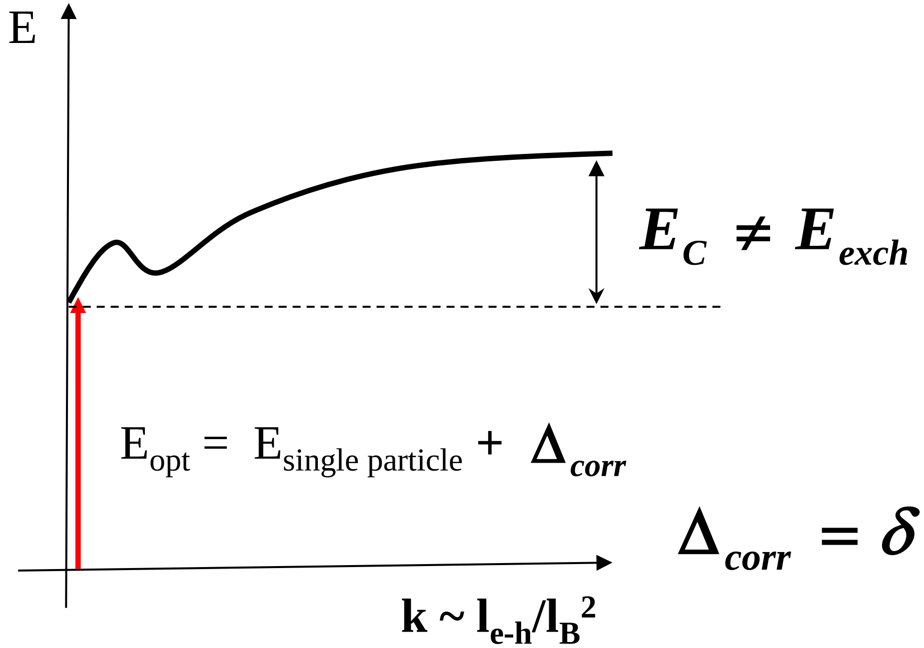
Optics is useless to study the many-body effects !?

e.g., C. Kalin, B.I. Halperin, PRB , 1984, Bychkov, Eliashberg, Iordanskii, (JETP Letters, 1981)

Electron-electron interactions and inter Landau level transitions

Linear dispersions
non-equidistant spacing





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

A. Iyengar, *et al.*, PRB, 2008

Yu.A. Bychkov, G. Martinez, PRB, 2008

R. Roldan et al., PRB, 2010

J. Sari, C. Toke, PRB, 2013

YU.E. Lozovik, A.A. Sokolik, Nanoscale Research Lett., 2012

Expectations:

Rather large deviations from effective single electron model ?

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

Graphene: Electron-electron interactions at B=0

nature physics

LETTERS

PUBLISHED ONLINE: 24 JULY 2011 | DOI: 10.1038/NPHYS2049

Dirac cones reshaped by interaction effects in suspended graphene

D. C. Elias[1], R. V. Gorbachev[1], A. S. Mayorov[1], S. V. Morozov[2], A. A. Zhukov[3], P. Blake[3], L. A. Ponomarenko[1], I. V. Grigorieva[1], K. S. Novoselov[1], F. Guinea[4]* and A. K. Geim[1,3]

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 applicable[1,2]. 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 proposed[3-5]. 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 nanotubes[6]. 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.

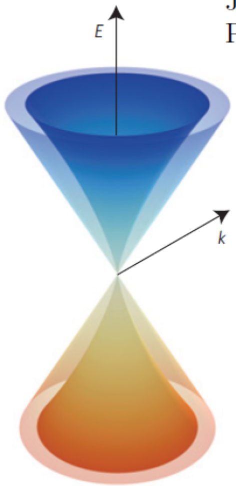


Figure 1 | Sketch of graphene's electronic spectrum with and without taking into account e–e interactions. The outer cone is the single-particle

J. González, F. Guinea, and M. A. H. Vozmediano, Mod. Phys. Lett. B **7**, 1593 (1993).

$$v = v_0 - \frac{\alpha c}{4\varepsilon} \ln \frac{|E|}{W}$$

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 !

NANO LETTERS

Letter

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

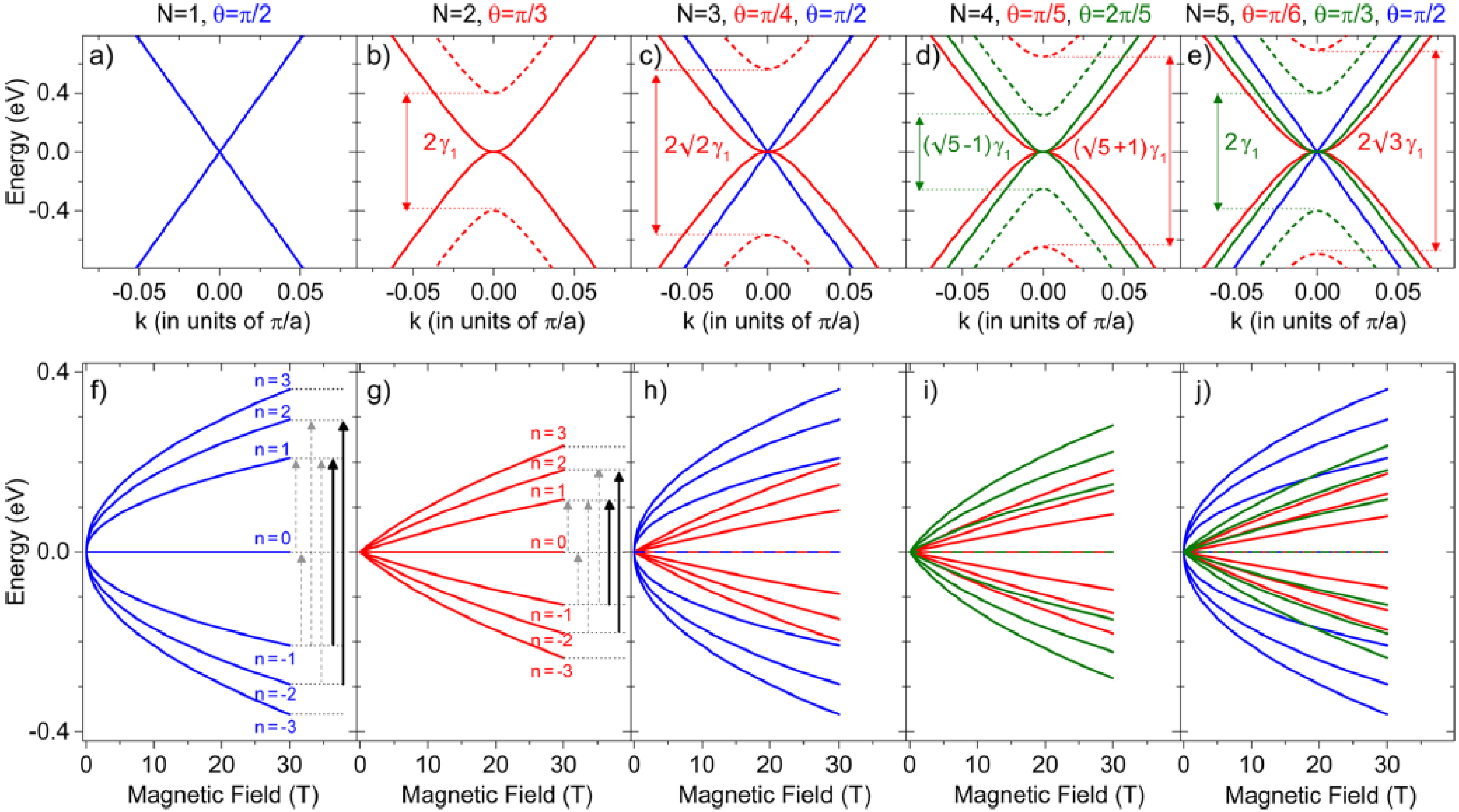
Stéphane Berciaud,*,† Marek Potemski,‡ and Clément Faugeras*,‡

†Institut de Physique et Chimie des Matériaux de Strasbourg and NIE, UMR 7504, Université de Strasbourg and CNRS, 23 rue du Lœss, BP43, 67034 Strasbourg Cedex 2, France

‡Laboratoire National des Champs Magnétiques Intenses, CNRS/UJF/UPS/INSA, Grenoble F-38042, France

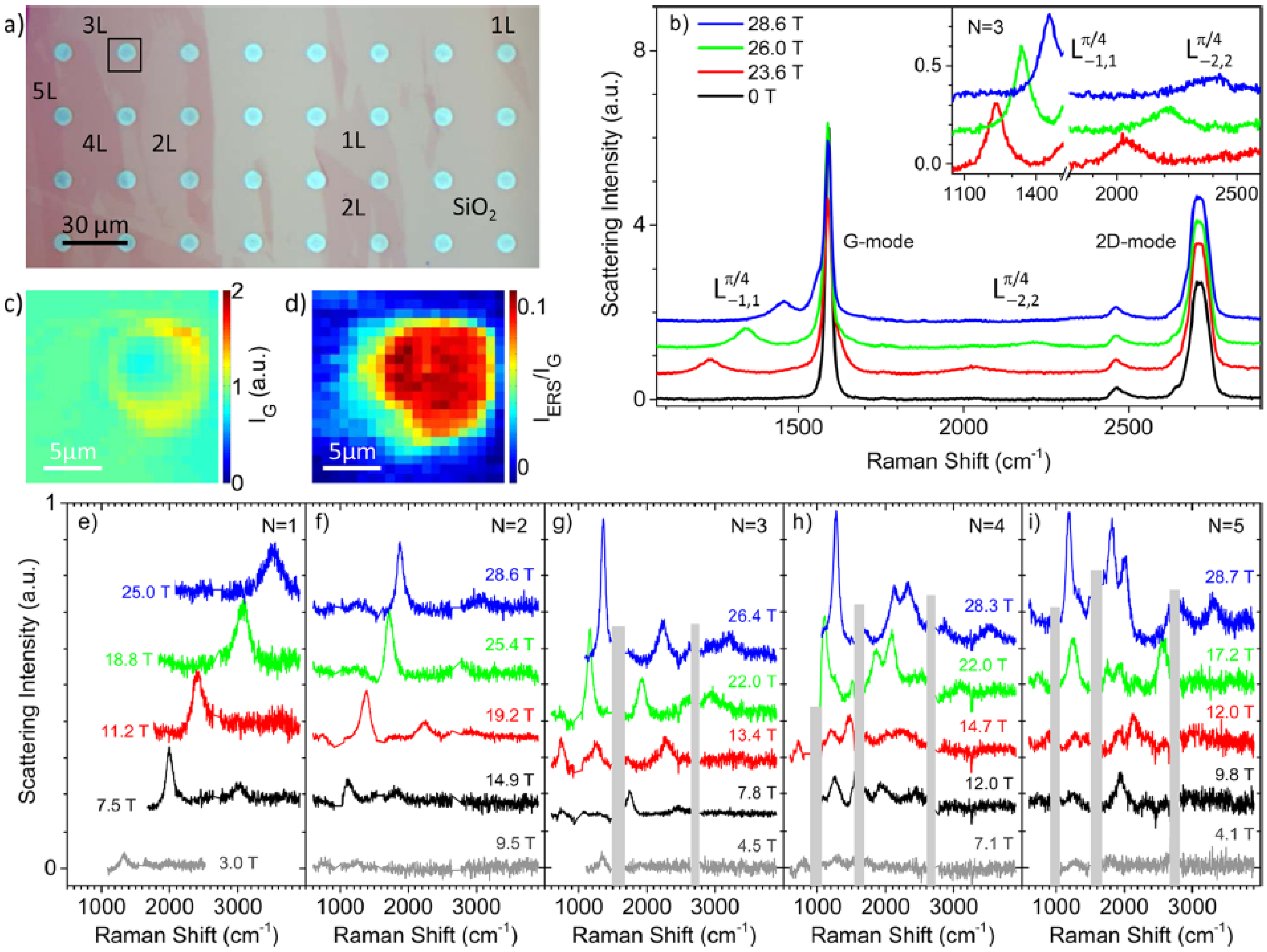
What can be learned from magneto-optics ?

Band structure !



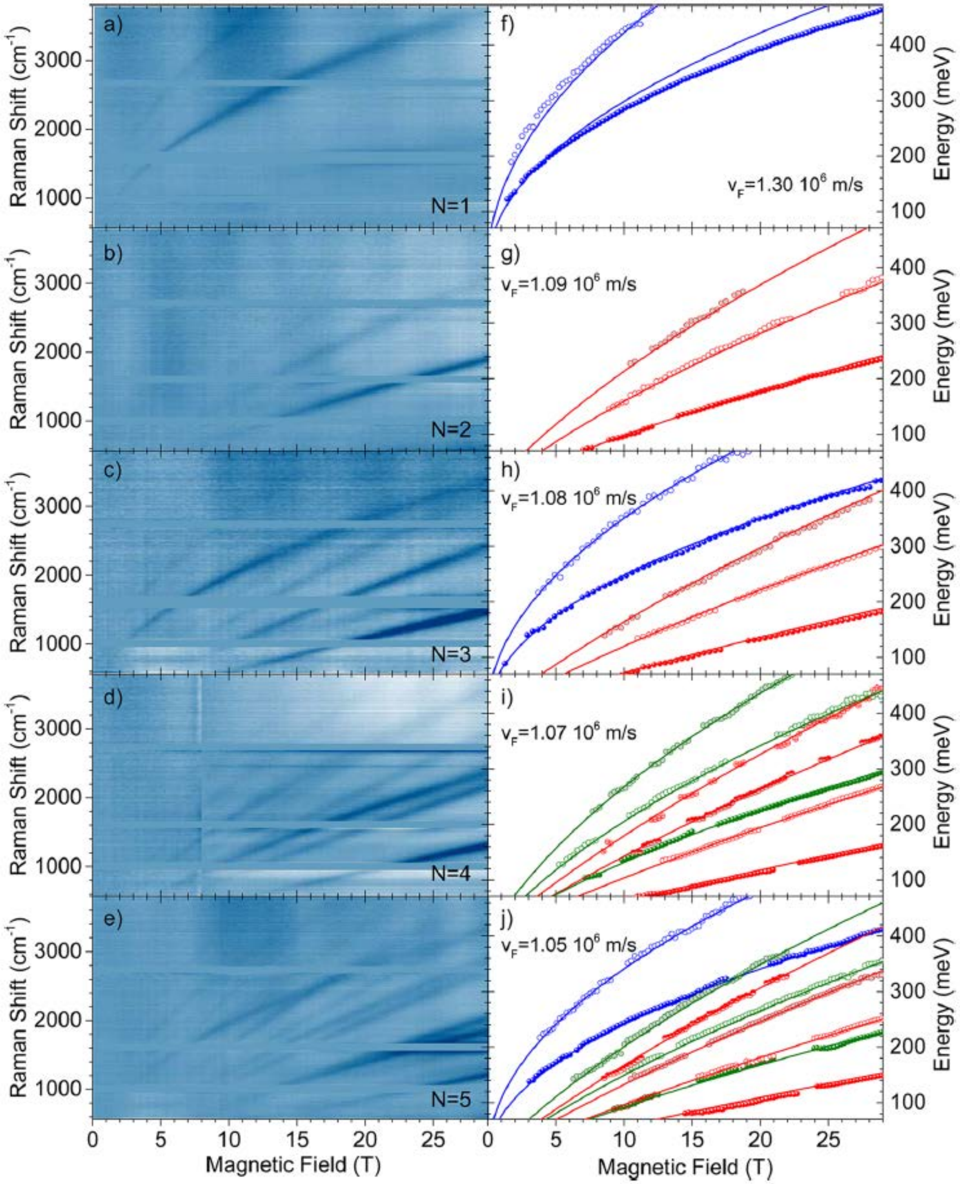
What can be learned from magneto-optics ?

Band structure !



What can be learned from magneto-optics ?

Band structure !



What can be learned from magneto-optics ?
Scattering: efficiency (mechanism) !

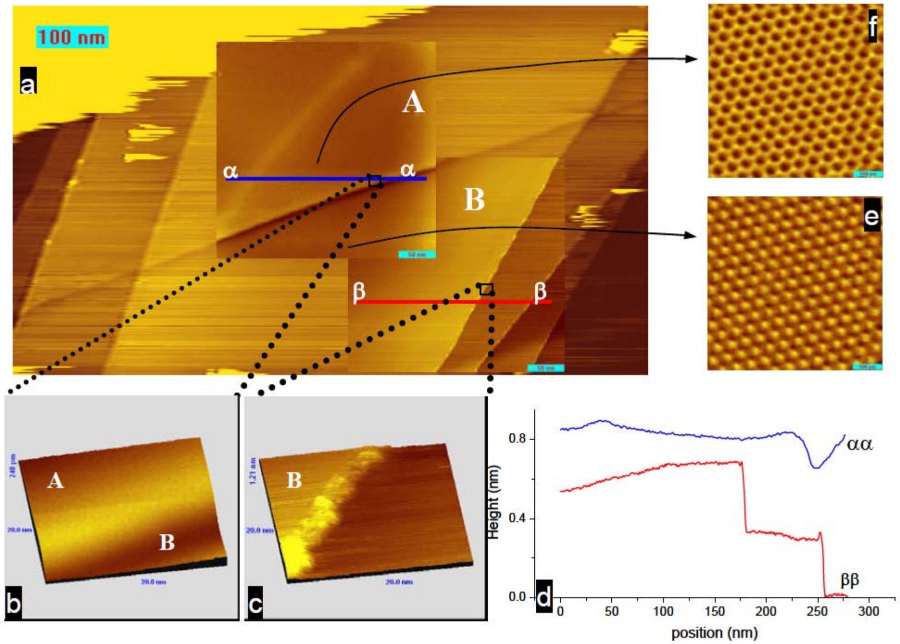
PRL **103,** 136403 (2009) PHYSICAL REVIEW LETTERS week ending 25 SEPTEMBER 2009

How Perfect Can Graphene Be?

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

Graphene on graphite: best ever seen graphene !!

G. Li *et al.*, PPRL, 2008



Cyclotron resonance absorption : high temperature but well resolved LLs

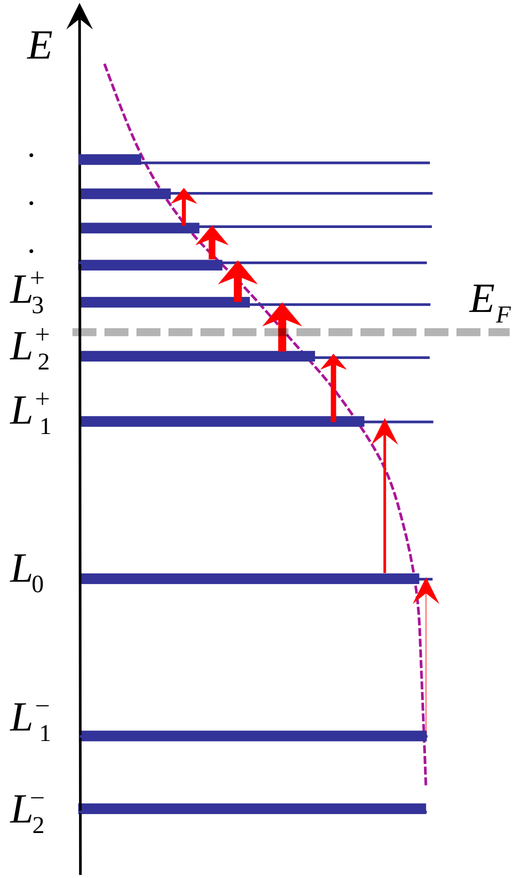
LL spacing > kT

LL broadening < LL spacing

non-equidistant spacing

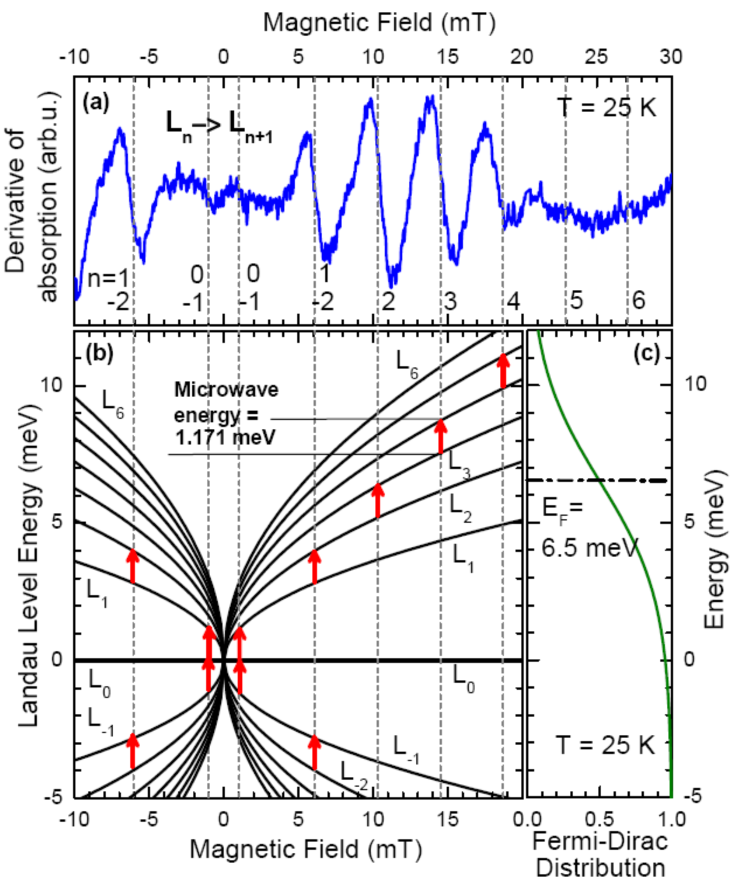
↓

multimode
cyclotron resonance absorption

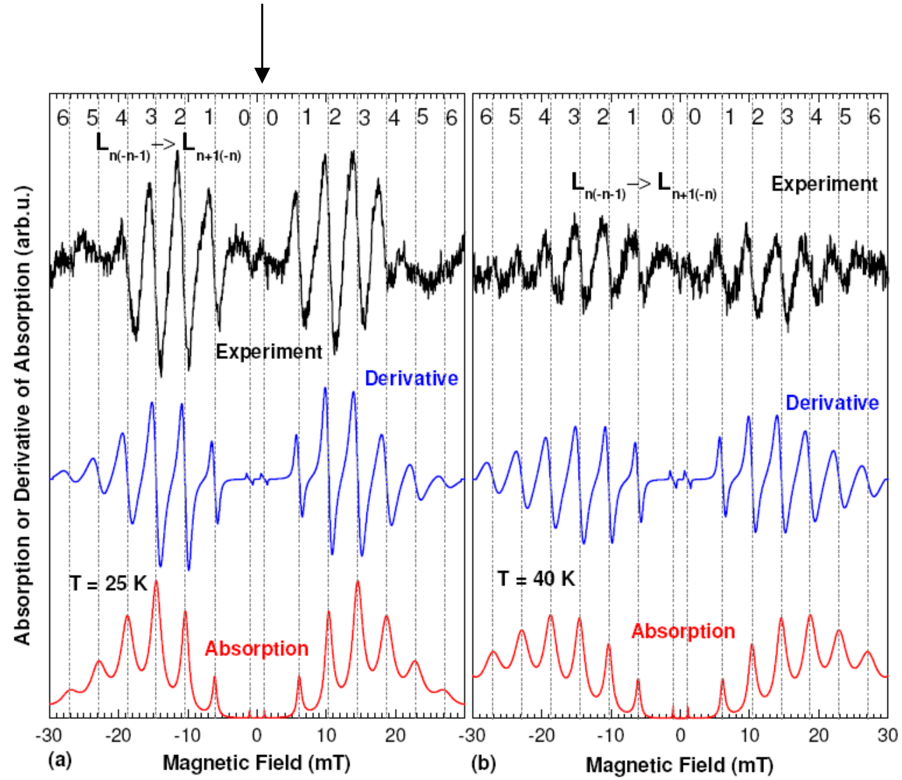


Graphene on graphite

(Very) low field cyclotron resonance absorption



1 mT



perfect Dirac states :

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

LL broadening : $\Gamma \approx 35\ \mu eV\ \ (0.4\,K)$

P. Neugebauer *et. al.*, PRL, 2009

How perfect can graphene be

$$\left(E_F \cong 6.5\ meV,\quad n \cong 3\cdot 10^9\ cm^{-2},\quad m^* = E_F / v_F^2 \cong 1.3\cdot 10^{-3}\, m_e \right)$$

Landau level quantization down to $B_0 = 1$ mT $\longrightarrow$ $\mu > \frac{1}{1mT} = 10^7 \frac{cm^2}{V\cdot s}$

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

Also at 50 K !

$$\gamma = E_1 \longrightarrow B = 1\,\mu T$$

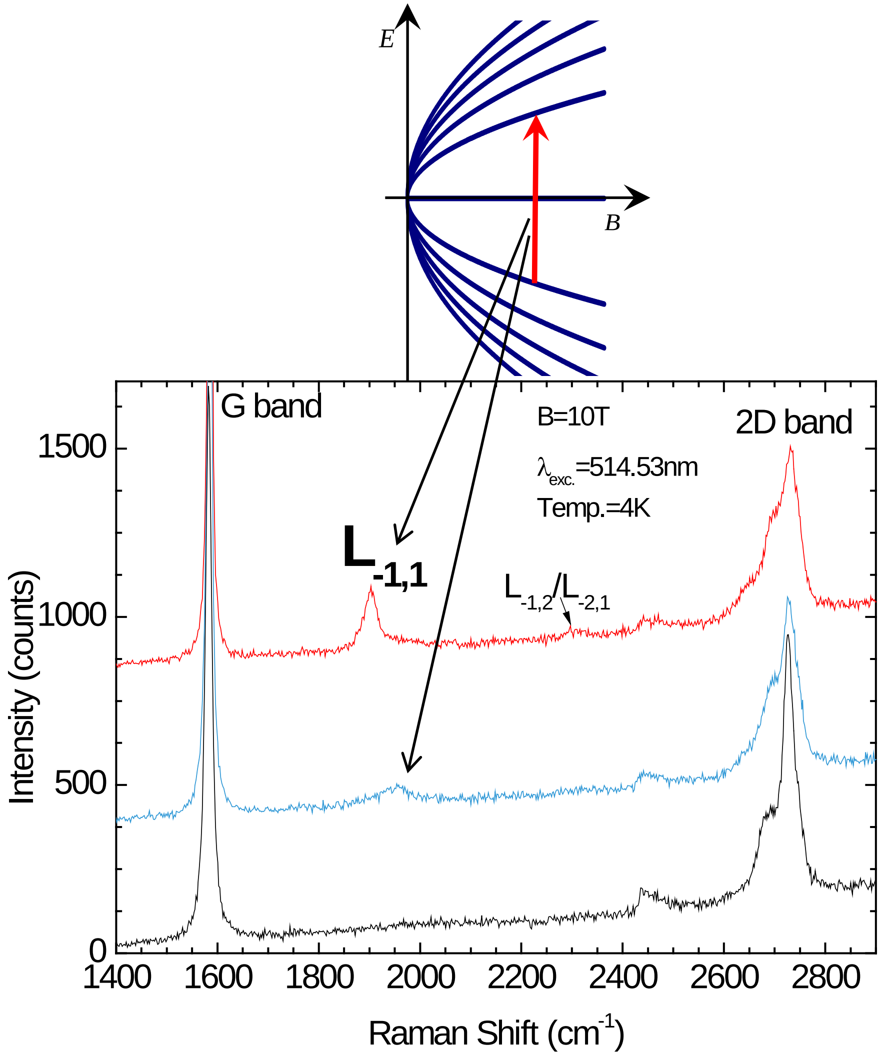
$$B_{Earth} \approx 50\,\mu T \longrightarrow E_1 \approx 0.25\, meV = 3\,K \ > \ \gamma = 0.4\,K$$

Pronounced Landau quantization in the magnetic field of the Earth

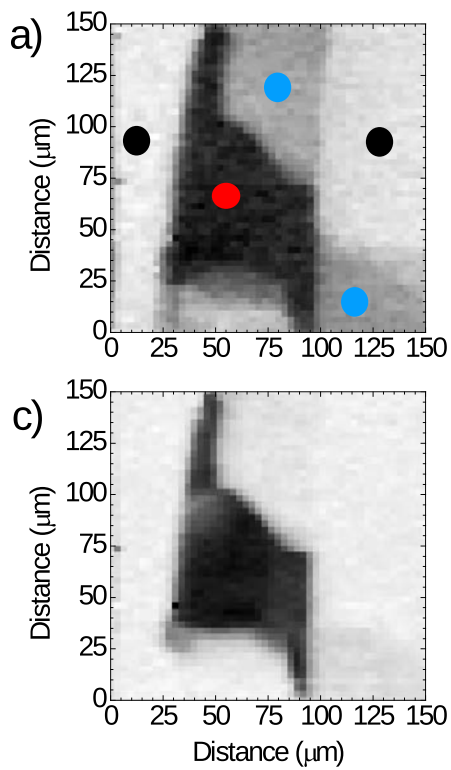
P. Neugebauer *et. al.*, PRL, 2009

Graphene on graphite: magneto Raman scattering response
phonons + search for a characteristic electronic response

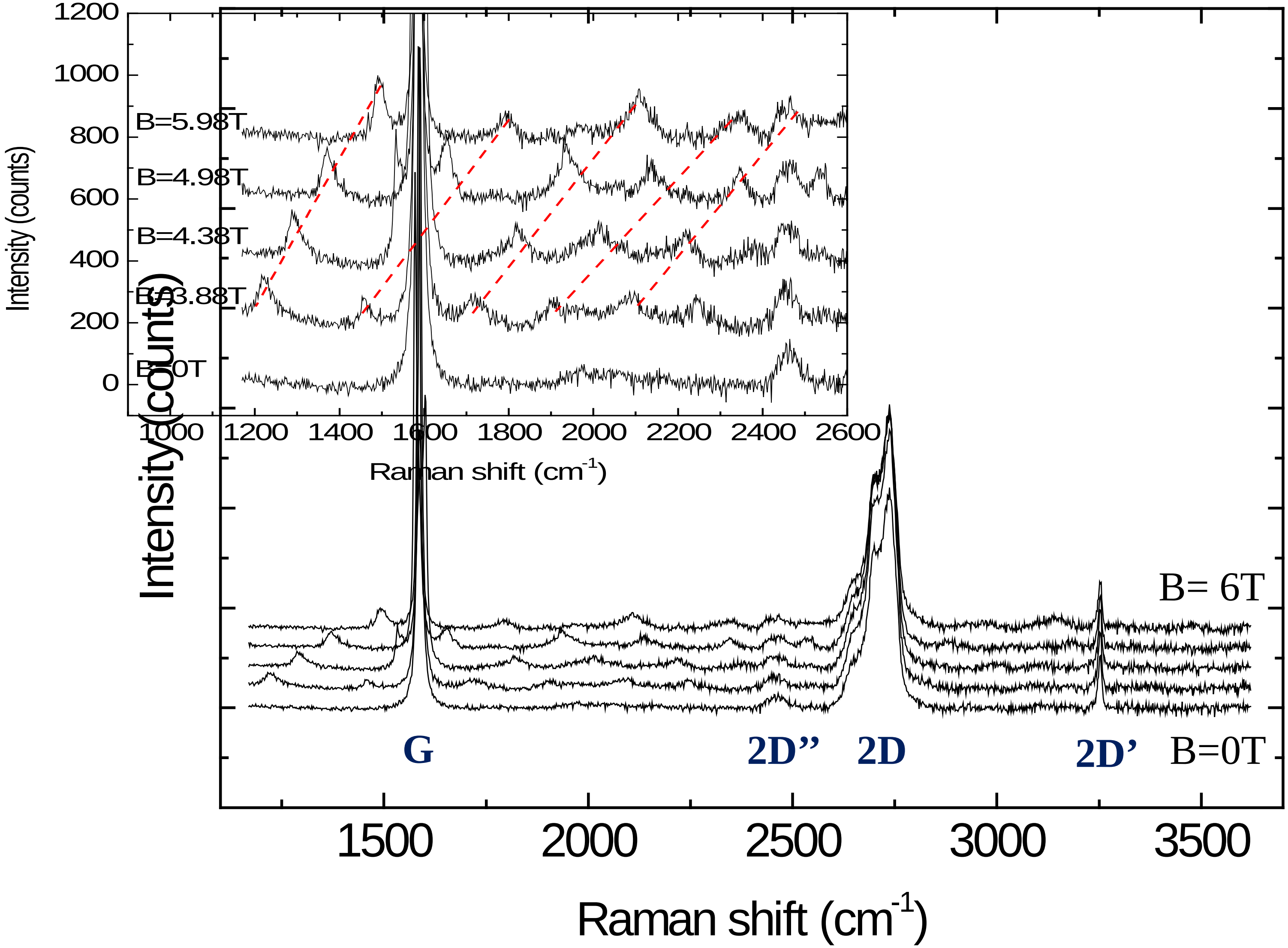
e.g., $L_{-1} \rightarrow L_1$ inter Landau level excitation



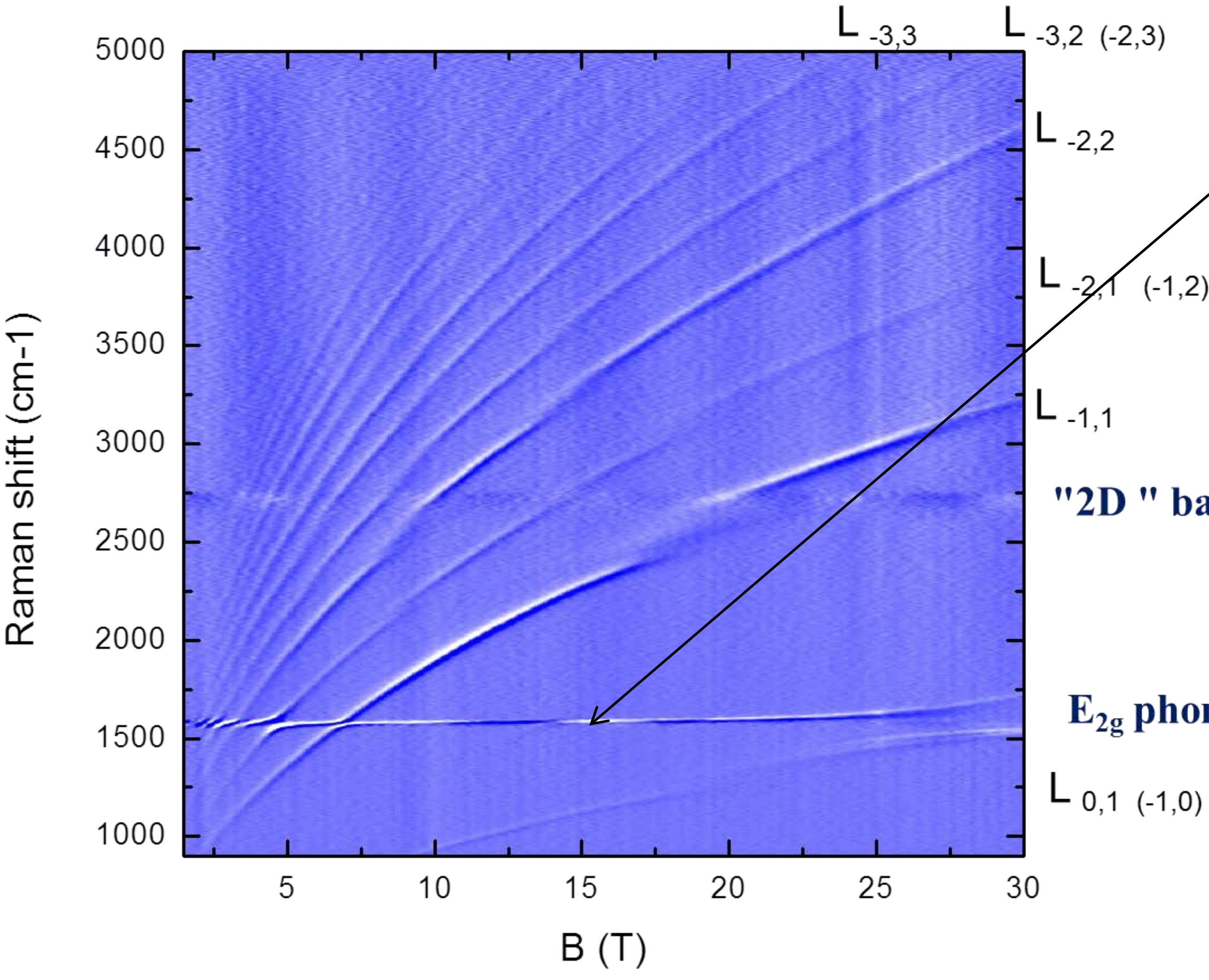
$L_{-1} \rightarrow L_1$



Graphene on graphite: magneto-Raman scattering response: an overview



Graphene on graphite: magneto-Raman scattering response: an overview

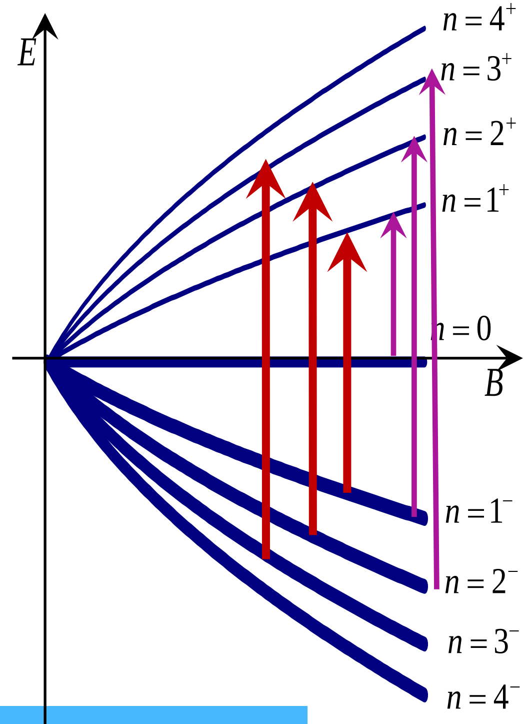


E_{2g} phonon

"2D " band

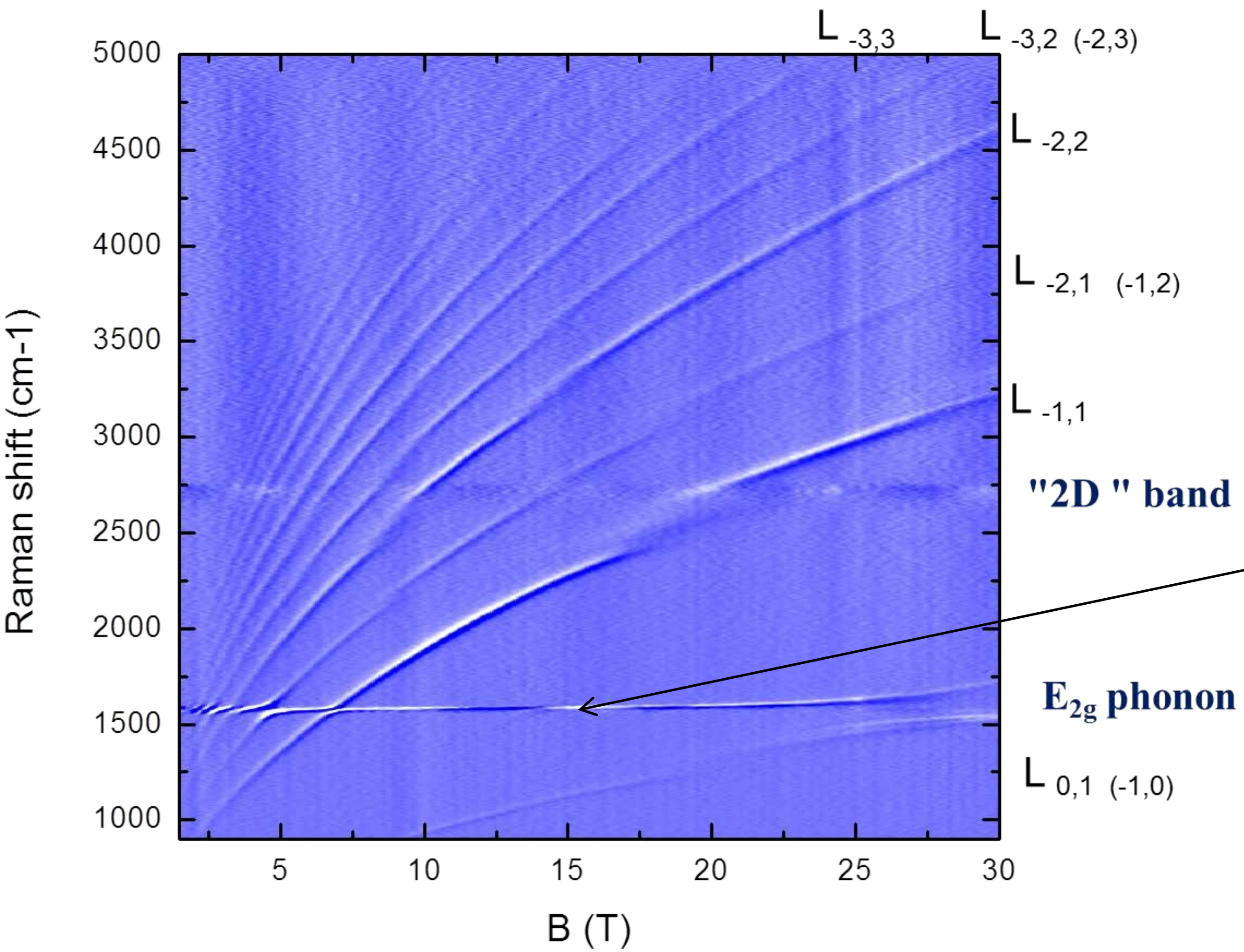
E_{2g} phonon

+ electonic 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

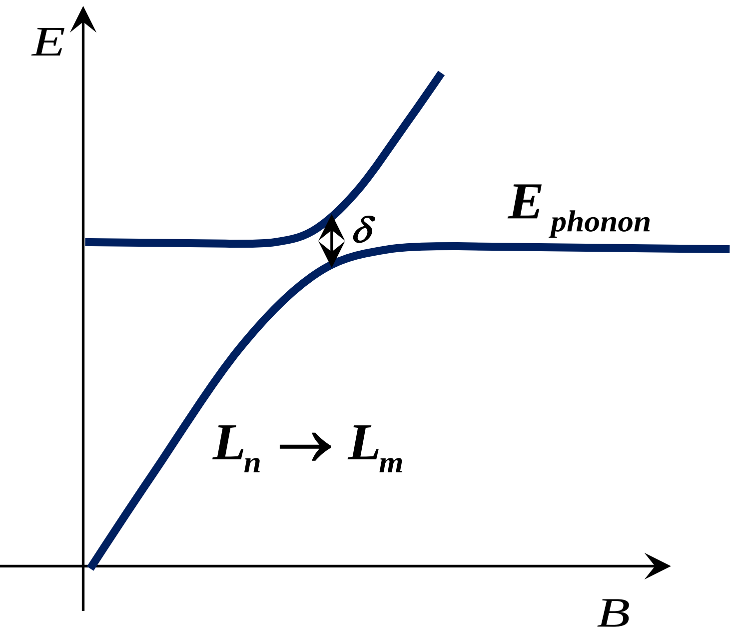


focus on E_{2g} phonon

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

Interactions ?

resonant electron-phonon coupling !



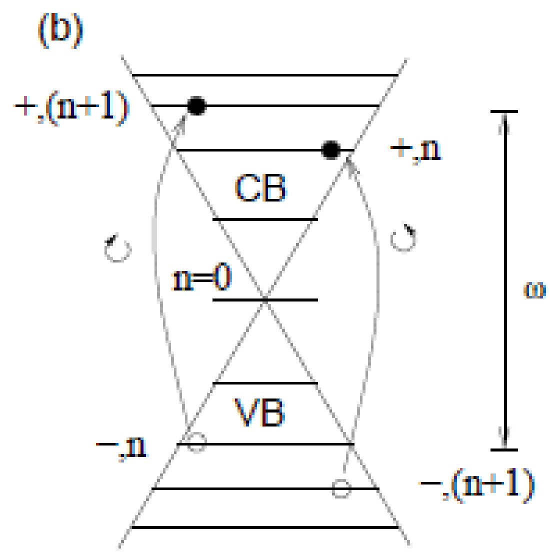
strength of interaction $\leftarrow \delta$

In magnetic fields

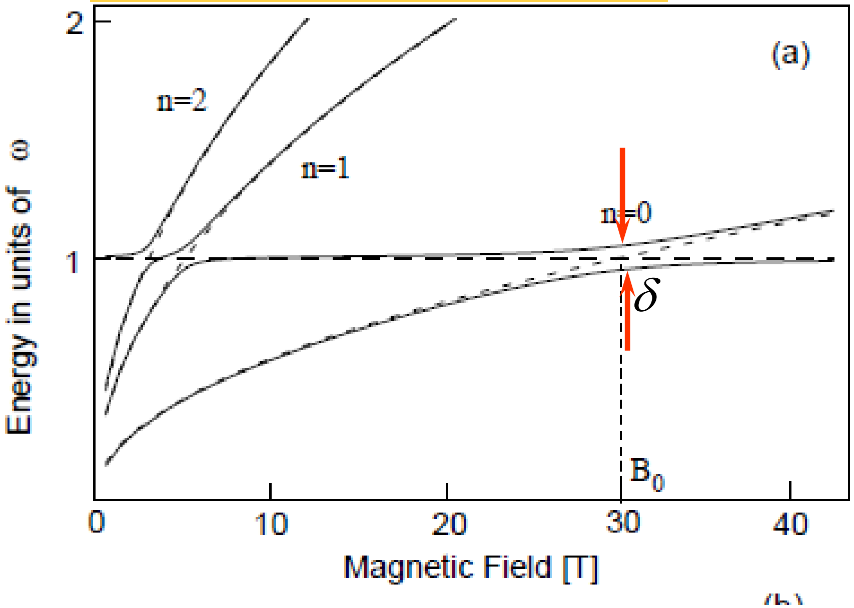
Resonant coupling of E_{2g} phonon ("optical") with Δn=±1 inter Landau level excitations

Theoretical predictions :

T. Ando, JPSJ, 2007
M.O. Goerbig, *et al.*, PRL , 2007

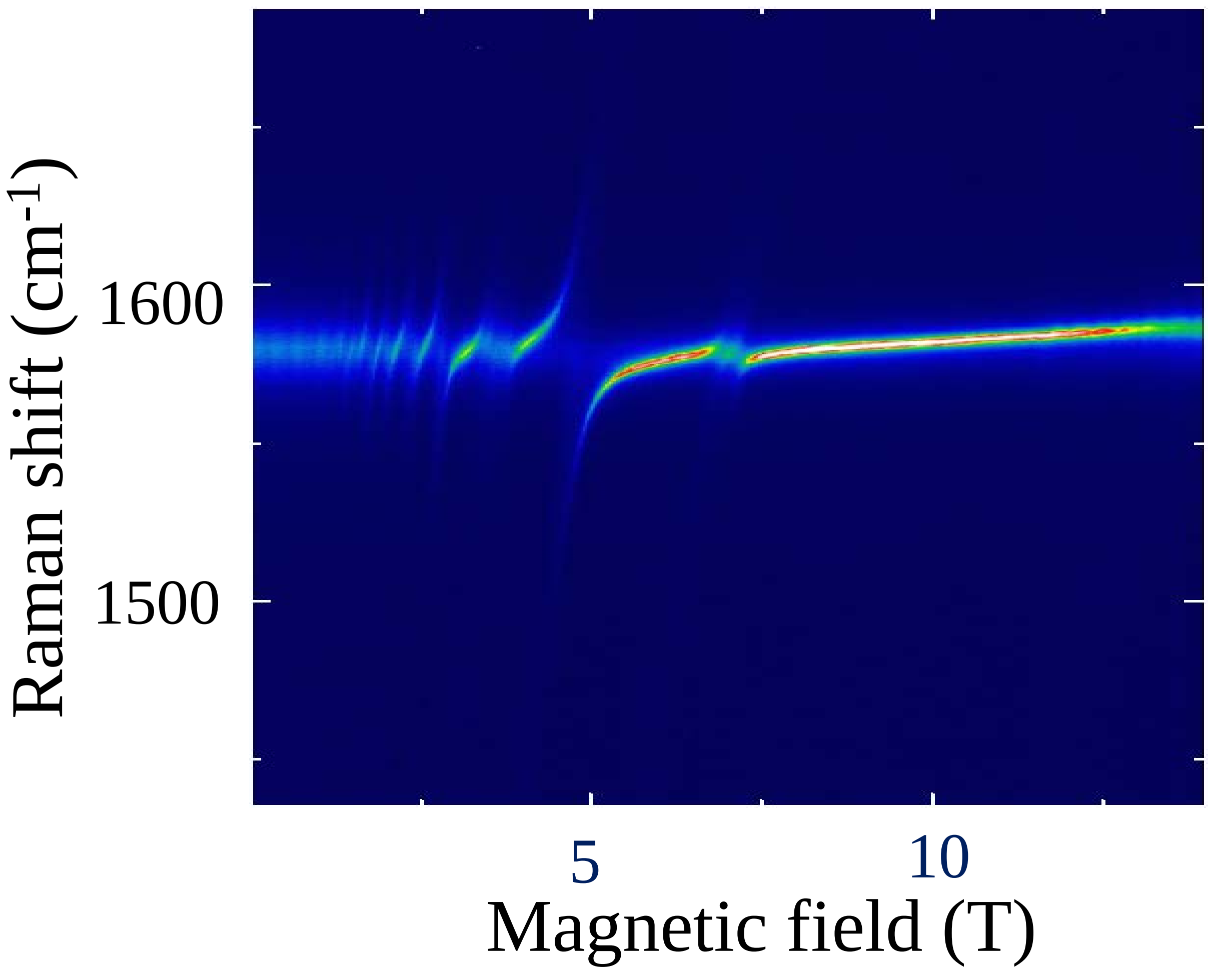


σ^- / σ^+ σ^+ / σ^-



$$\delta \sim \sqrt{2\lambda} \cdot E_1(B_{res}) \cdot \sqrt{(1-f_f)f_i} \sim \sqrt{\lambda \cdot B_{res}} \cdot \sqrt{(1-f_f)f_i}$$

Magneto-phonon resonance: graphene on graphite

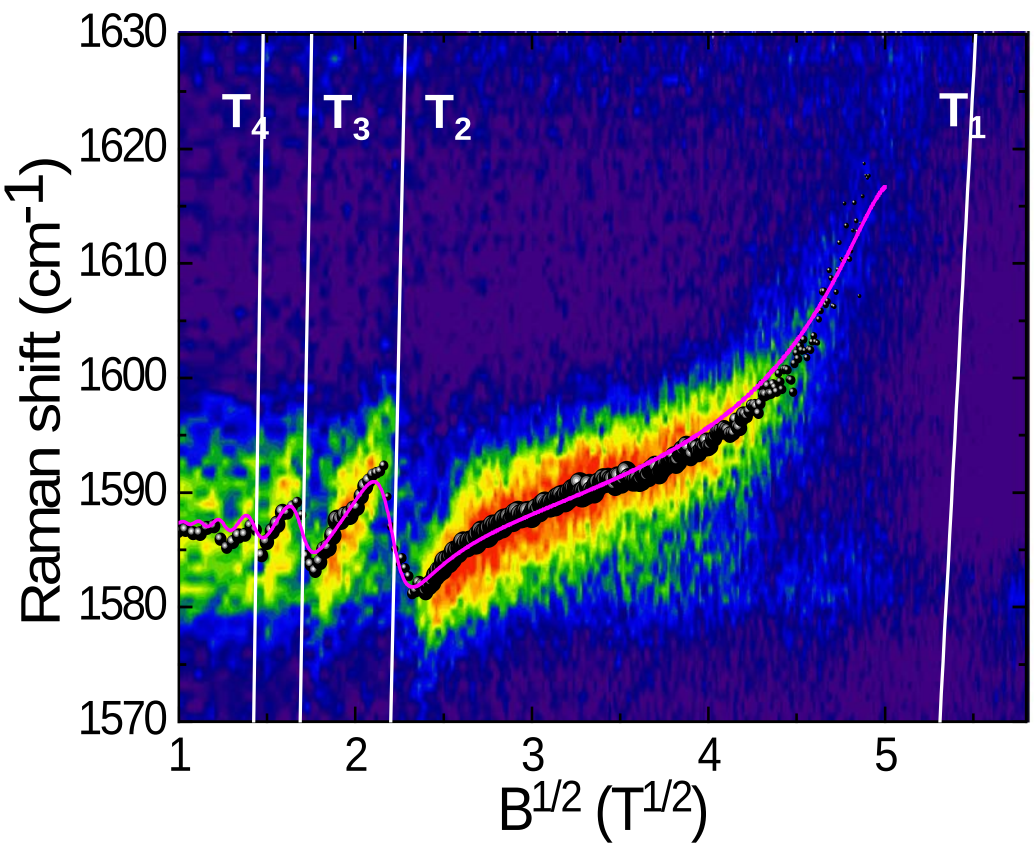


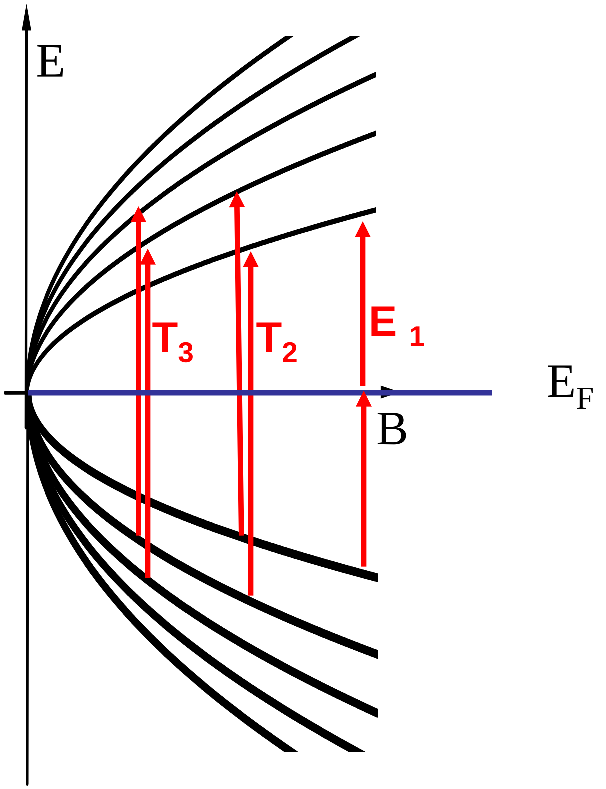
Graphene on graphite: an electronic system of unprecedented quality !

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

J. Yan *et al.*, PRL , 2010

Experiment: magneto-phonon resonance in epitaxial graphene





Neutral graphene

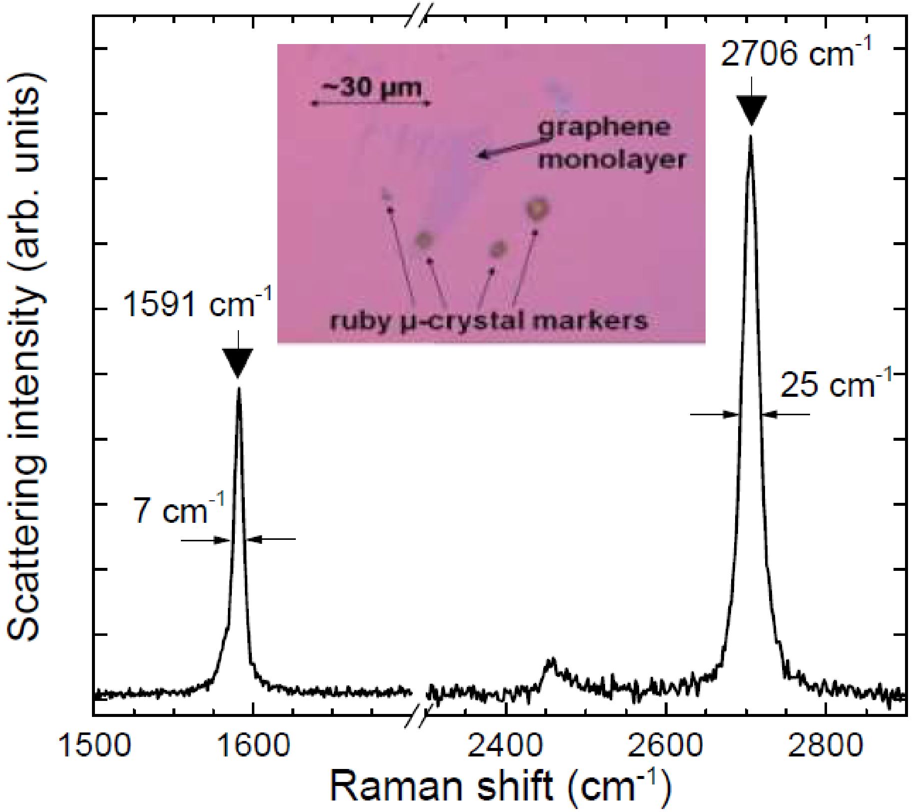
$$\lambda = 4.5 \cdot 10^{-3}$$

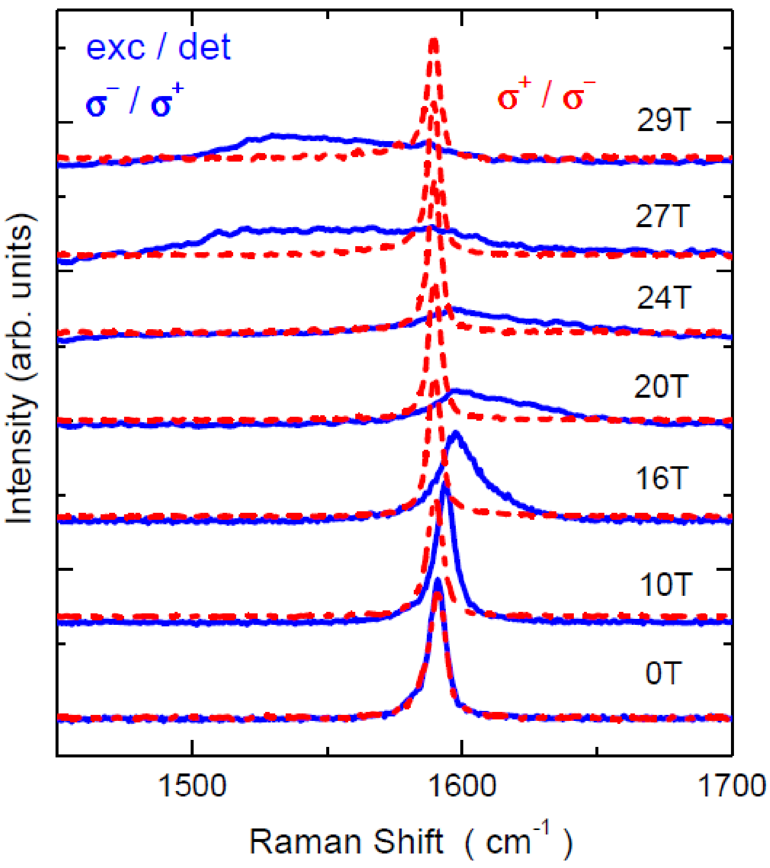
C. Faugeras, et al., PRL, 2009

Magneto-phonon resonance in doped graphene

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

Graphene flake on Si/SiO$_2$



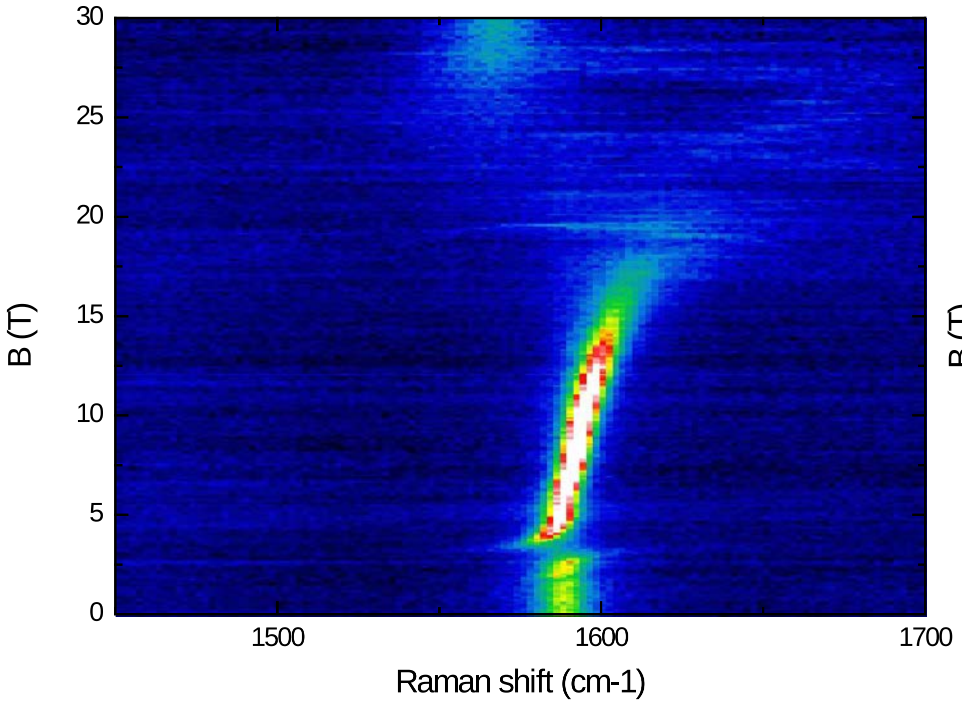


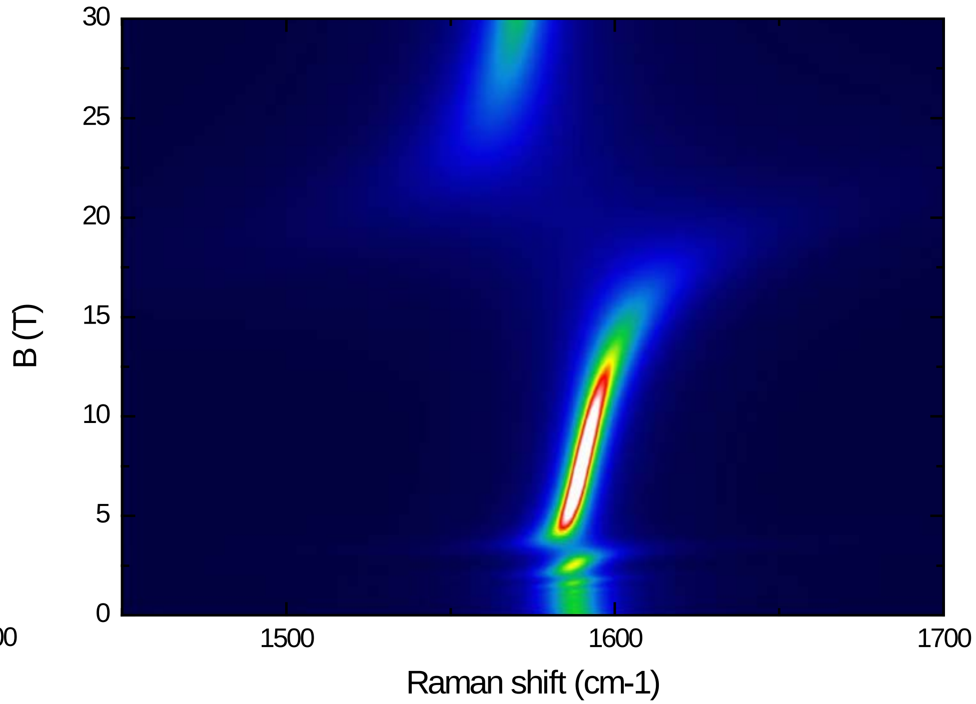
P. Kossacki *et al.*, Phys. Rev. B, 2012

Magneto-phonon resonances: graphene on h-BN

~ neutral and better electronic quality

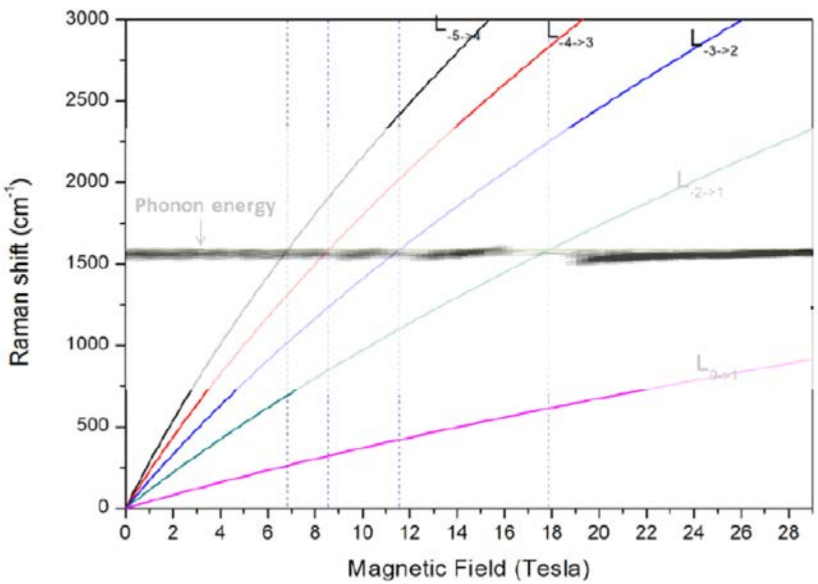
Experiment **in qualitative agreement with** **simulations**

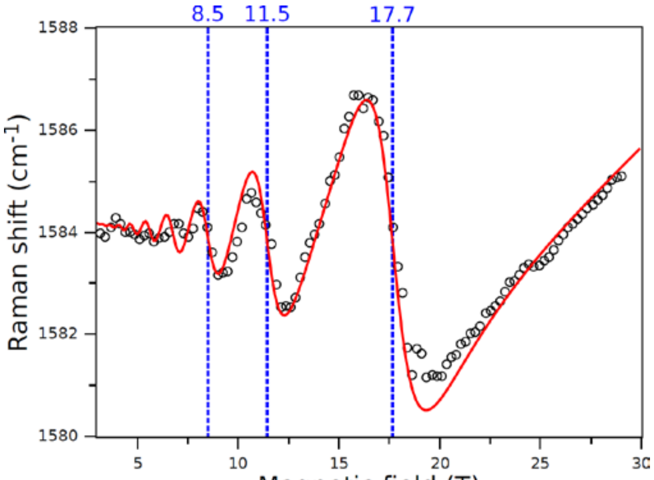


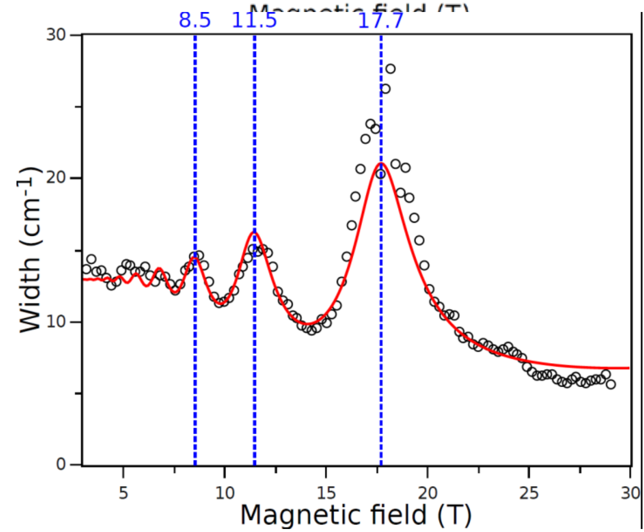


P. Leszczynski, A. Nicolet, C. Faugeras *et al.*, to be published

Magneto-phonon resonances: bilayer graphene on h-BN







Experiment in <u>qualitative</u> agreement

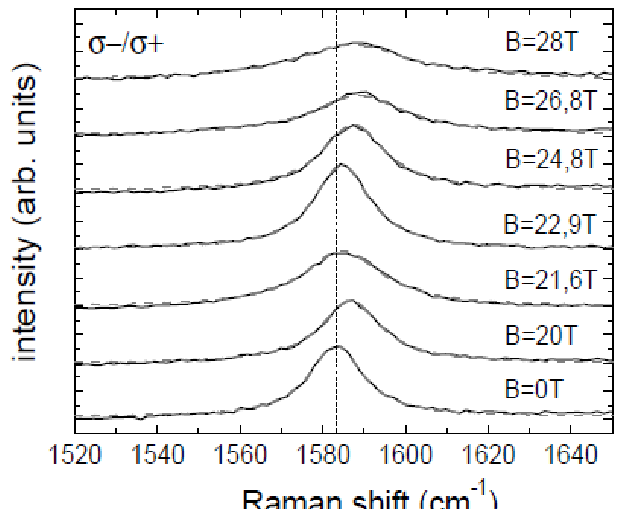
with simulations

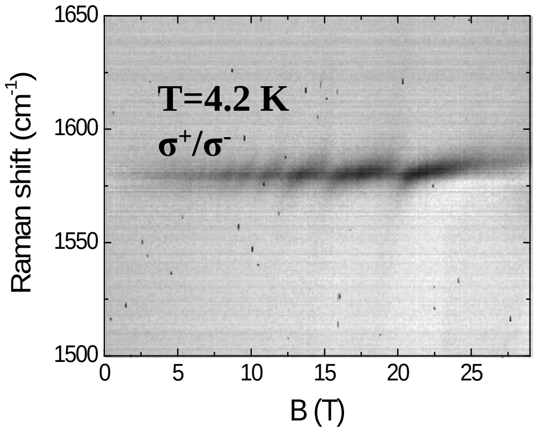
$$v_F = 1.06 \cdot 10^6\, m/s\ , \quad \lambda = 3.5 \cdot 10^{-3}$$

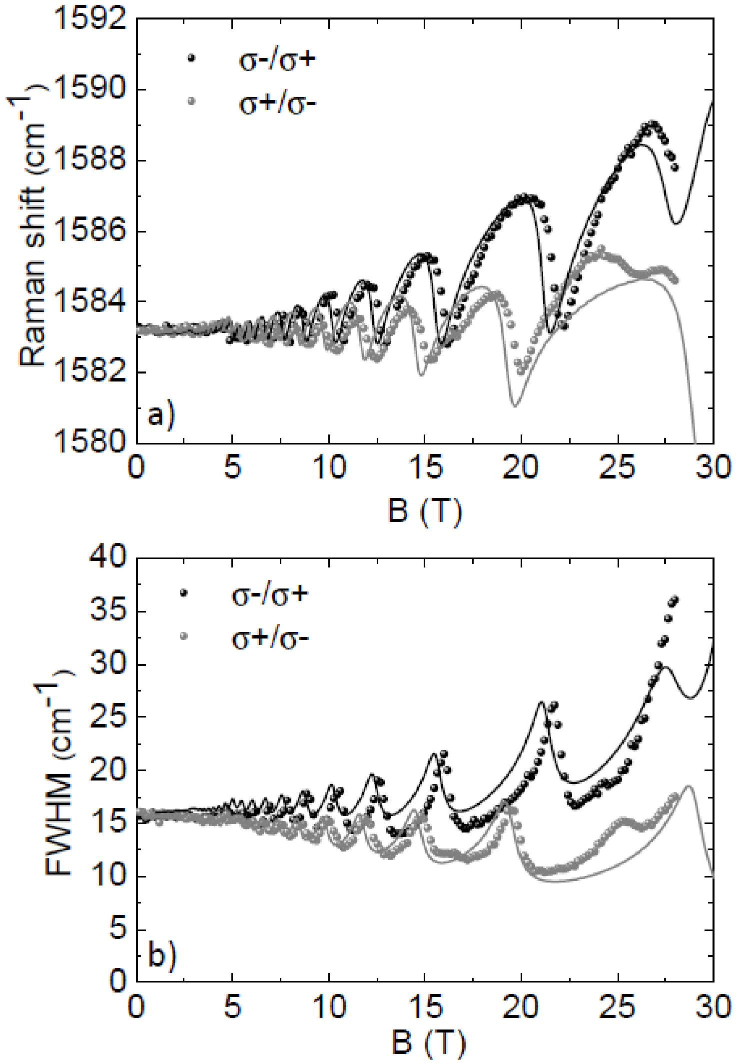
$$E_F < 100\, meV\ , \quad n < 2.5 \cdot 10^{12}\, cm^{-2}$$

A. Nicolet, Y. Henni, C. Faugeras *et al.*, to be published

Magneto-phonon resonance in graphite





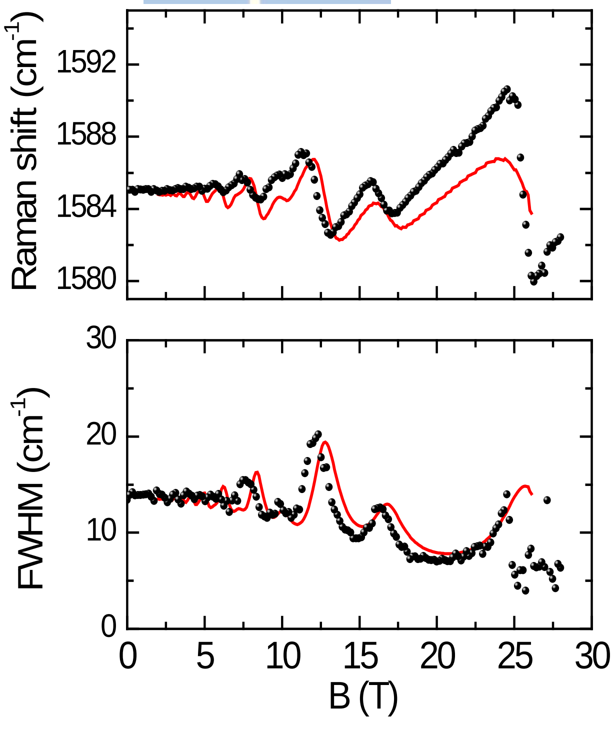


Phonon coupling to Δn=±1 inter Landau <u>band</u> transitions from the vicinity of the K-point + of the H point

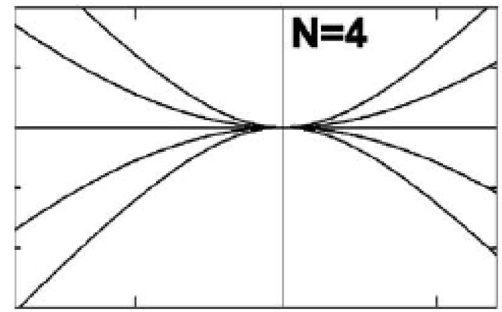
P. Kossacki *et al.*, PRB, 2011

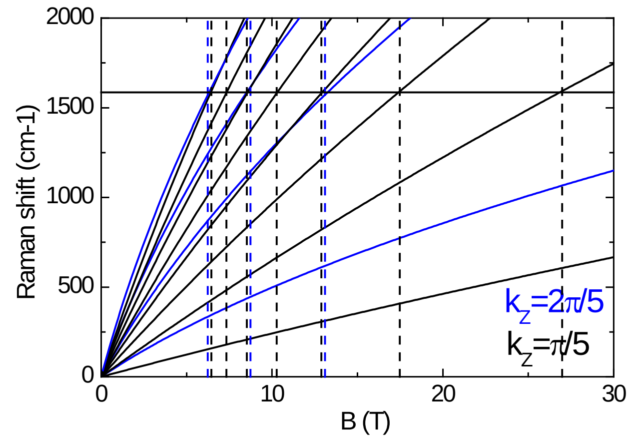
Probing the band structure with magneto-phonon resonance

few (?) layer graphene on Si/SiO_2



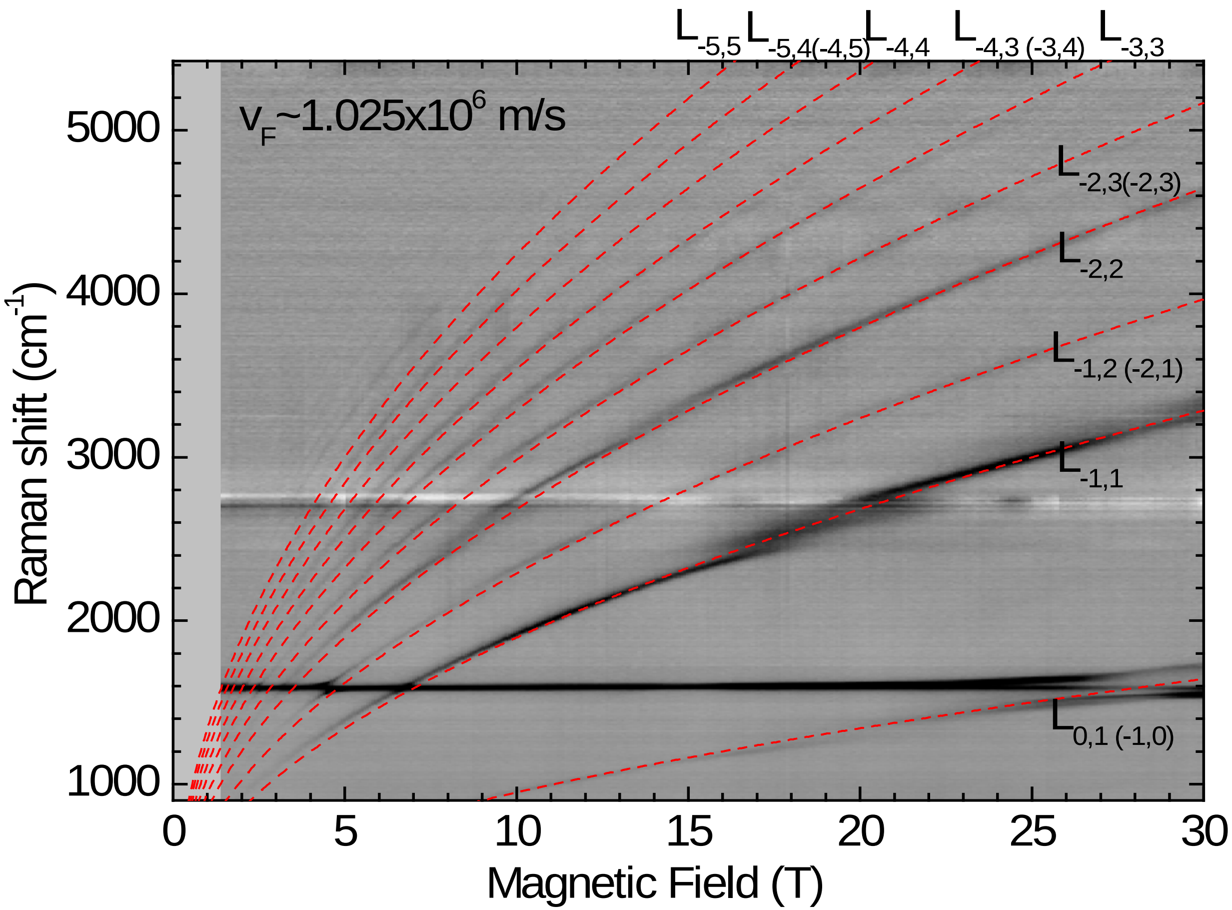
weakly doped
tetra-layer graphene !

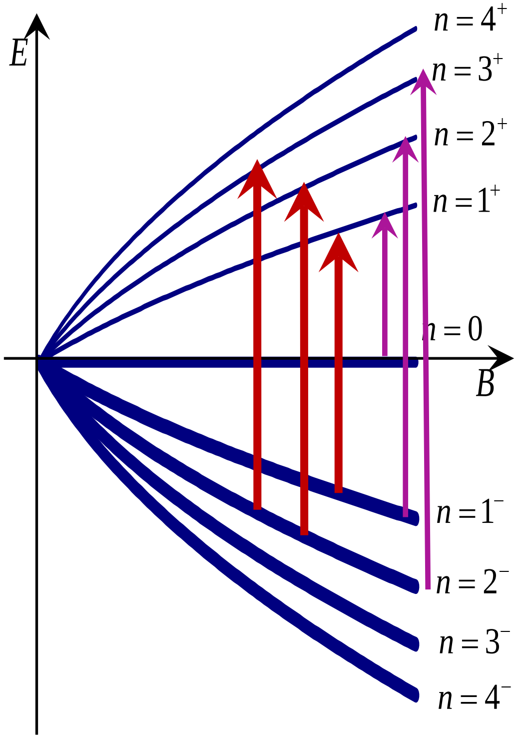




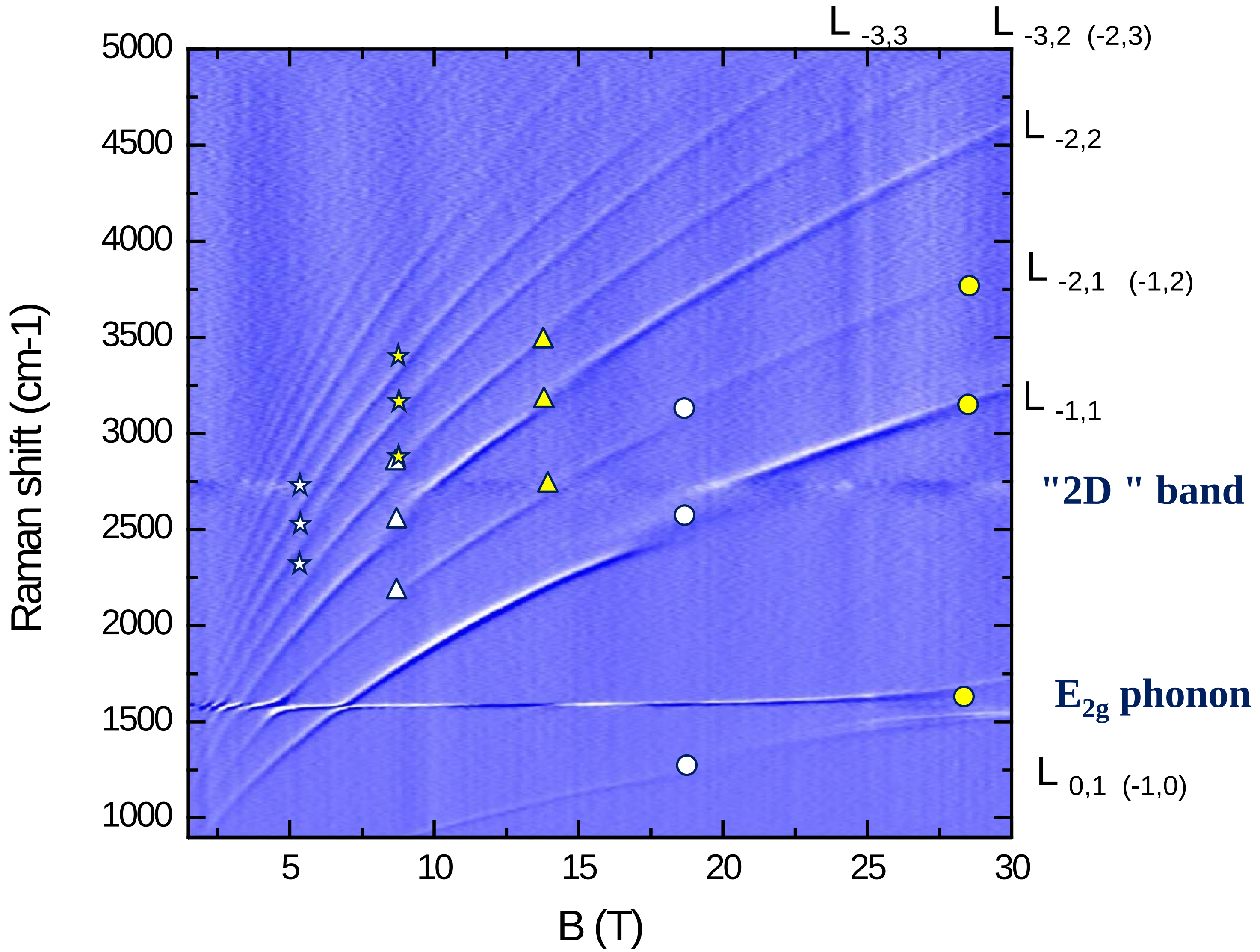
C. Faugeras *et al.*, New J. Phys., 2012

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



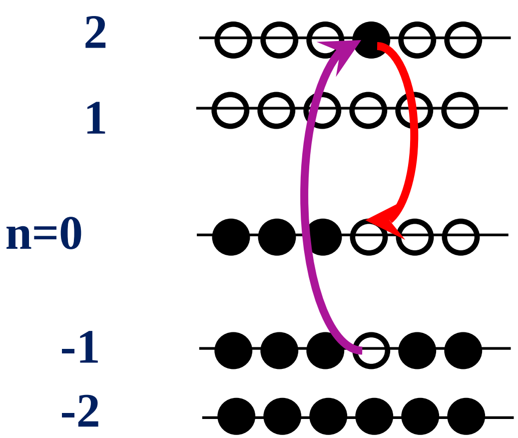


Beyond the standard magneto-E_{2g} phonon resonances



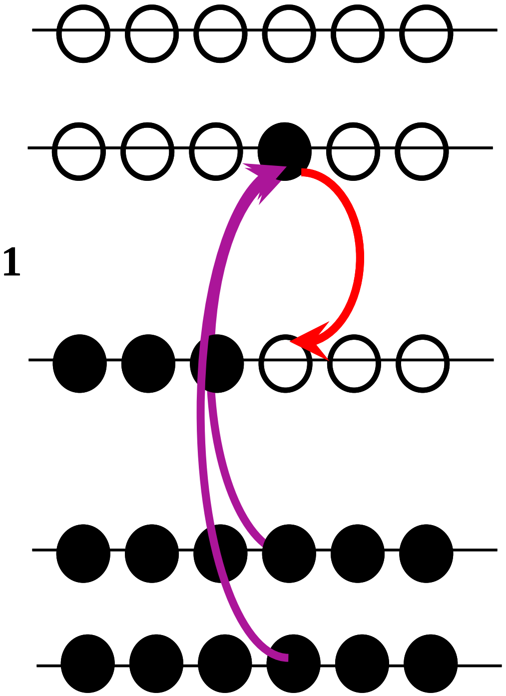
New class of magneto-phonon resonances:

- accelerated relaxation, shortening of the final/initial states



$$E_{ph} = E_{0,2}$$

$$E_{ph} = E_{0,1}$$



Interactions

$$E_{-1,\,2} = E_{-1,\,0} + E_{ph}$$

$$E_{k=0} = E_{K,\,K} + E_{\Gamma}$$

$$E_{k=0} = E_{K,\,K'} + E_{K}$$

$$E_{-1,1} = E_{-1,\,0} + E_{ph} = 2 \times E_{ph}$$

both K- and Γ-phonons invloved
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 ?

Electron – electron interactions ?

PRL **114**, 126804, (2015)

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. Potemski[1,†]

Electronic inter Landau level excitations
Magneto Raman scattering

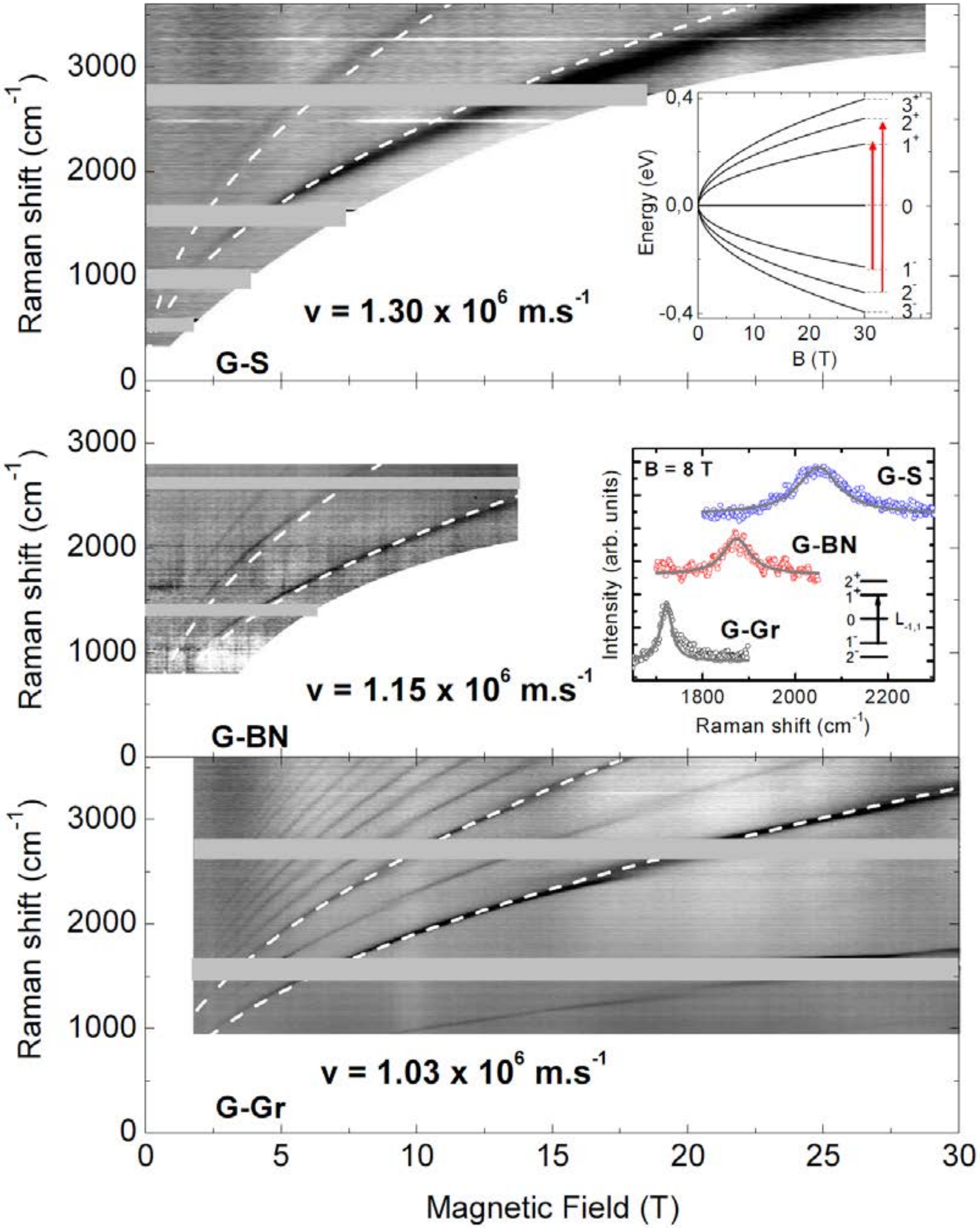
suspended graphene	**ε = 1**
graphene encapsulated in hBN	**ε = 5**
graphene on graphite	**ε = 10 ?**

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

Electron – electron interactions ?

$$\hbar\omega_n = 2v\sqrt{2e\hbar}\sqrt{B\,n}$$
$$= 2\sqrt{2n}\,\hbar v / l_B$$

$$v_n^{exp} = \omega_{-n,n}^{exp} l_B / \sqrt{8n}$$



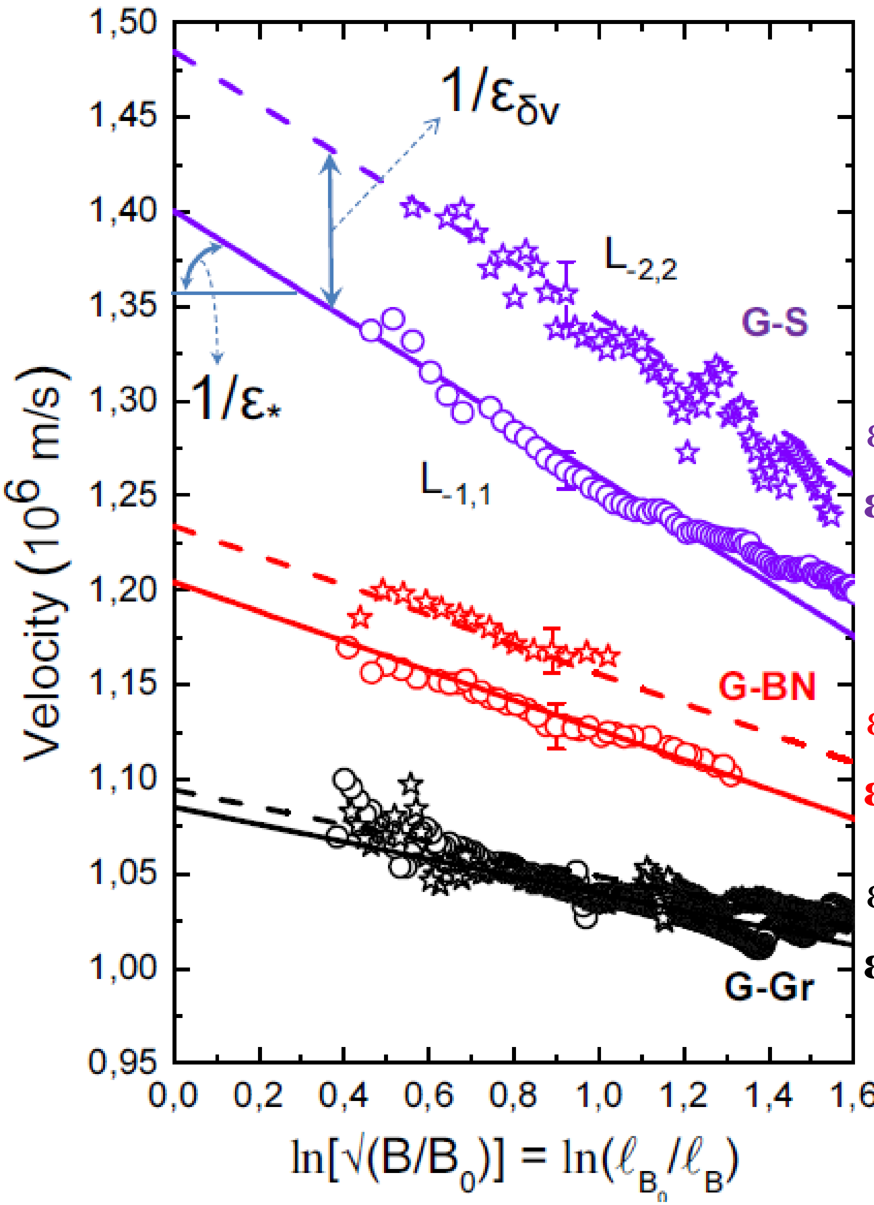
Electron – electron interactions !

B = 0

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

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

$$v = v_0 - \frac{\alpha c}{4\varepsilon} \ln \frac{|E|}{W}$$



ε = 1

ε* = 3.9

ε = 5

ε* = 7

ε = 10 ?

ε* = 12

Electron – electron interactions !

B = 0

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

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

$$v = v_0 - \frac{\alpha c}{4\varepsilon} \ln \frac{|E|}{W}$$

Beyond FOPT

1/(N=4) expansion :

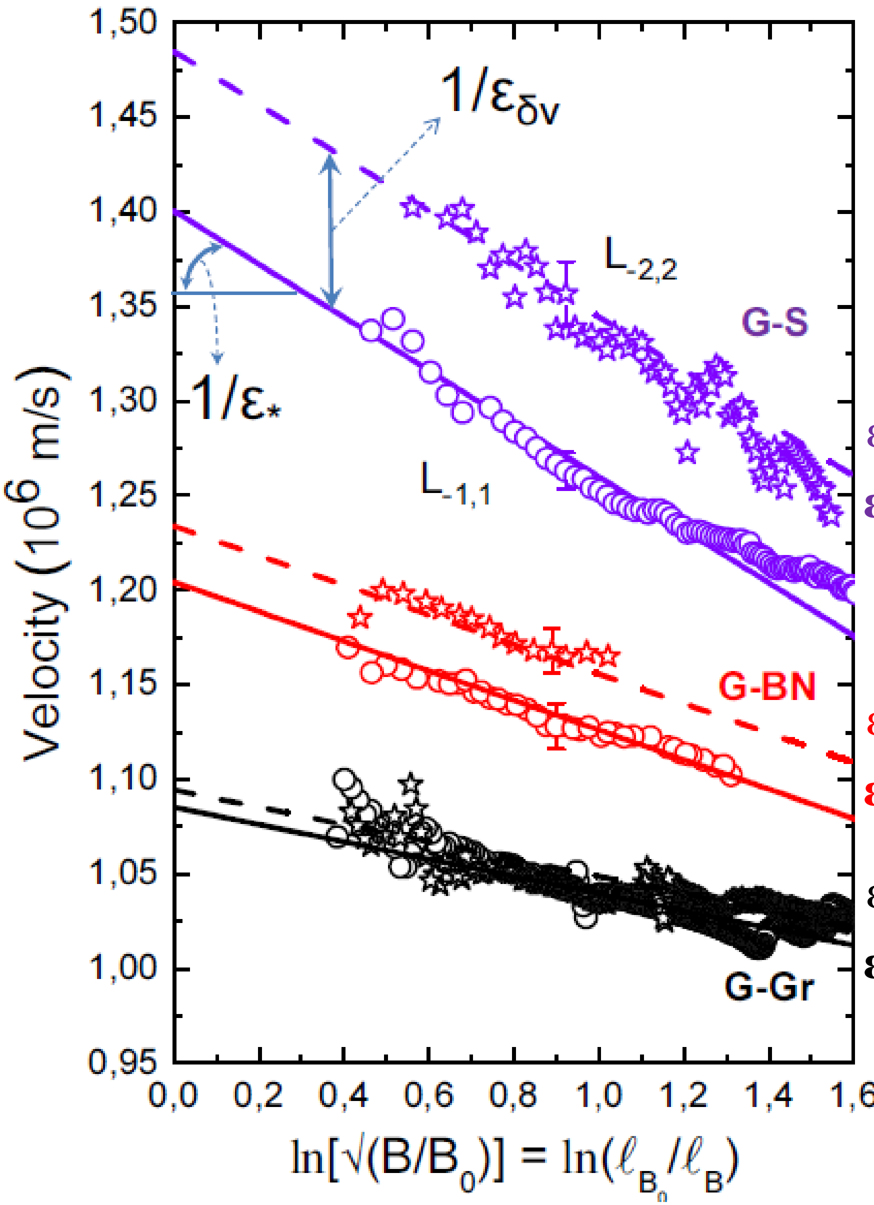
J. González, F. Guinea, and M. A. H. Vozmediano, Phys. Rev. B **59**, R2474 (1999).

RPA :

J. Hofmann, E. Barnes, and S. Das Sarma, Phys. Rev. Lett. **113**, 105502 (2014).

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

$\varepsilon \rightarrow \varepsilon_{1/N} = \varepsilon + 1.28\alpha c/v_0$ **= ε + 3** **≈** $\boldsymbol{\varepsilon_*}$



ε = 1
ε$_*$ = 3.9
ε = 5
ε$_*$ = 7
ε = 10 ?
ε$_*$ = 12

B > 0

K. Shizuya, Phys. Rev. B **81**, 075407 (2010).

Denis Basko

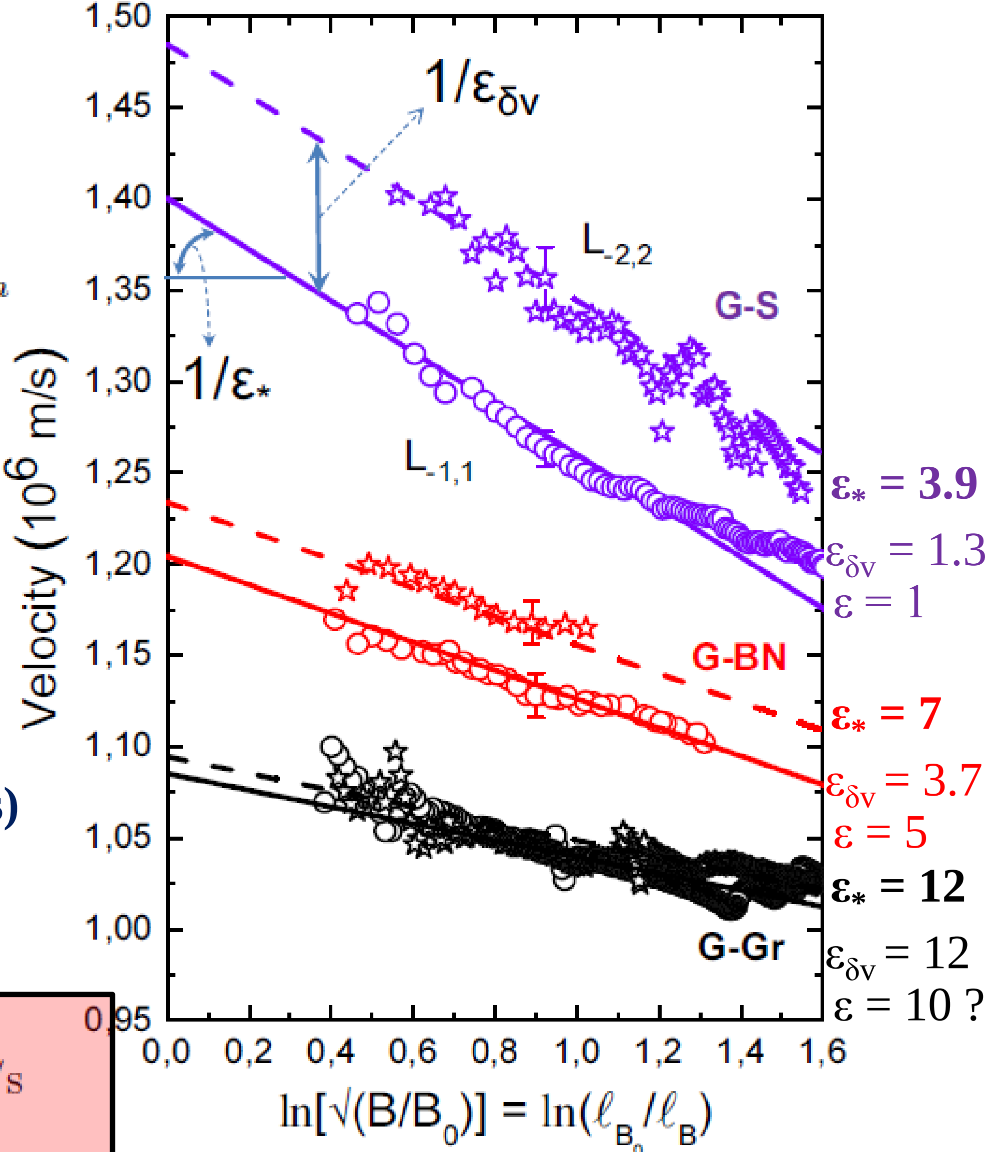
First order perturbation theory with respect to $\alpha_\varepsilon = (c/v)(\alpha/\varepsilon)$

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

C_1 = - 0.4, C_2 = - 0.2

↓

$$v_2 > v_1$$



Phenomenology to match the data (numbers)

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

$\varepsilon_* = 3.9, 7, 12$ **≈** $\boldsymbol{\varepsilon_{1/N}}$ $\quad v_0 = 0.88 \times 10^6$ m/s

$\varepsilon_{\delta v} = 1.3, 3.7, 12$ **≈** $\boldsymbol{\varepsilon}$ $\quad W = (\hbar v_0 / l_{B_0})^{\mathcal{L}} = 3.1$ 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

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