SPECTROSCOPY of ELECTRONS and PHONONS in GRAPHENE STRUCTURES

COLUMBIA UNIVERSITY
IN THE CITY OF NEW YORK

Alcatel·Lucent
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NATIONAL ENTERPRISE FOR NANOSCIENCE AND NANOTECHNOLOGY

CONSEJO SUPERIOR DE INVESTIGACIONES CIENTÍFICAS
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Spectroscopy of Electrons and Phonons in Graphene Structures

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(DE-AIO2-04ER46133)

Picture Gallery

Jun Yan

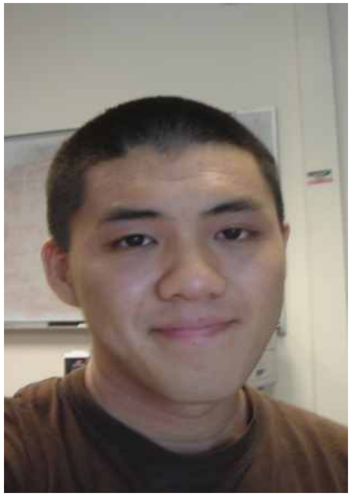
Trevor D. Rhone

Vittorio Pellegrini

Sarah Goler

Picture Gallery

Jorge García



Mason Jiang

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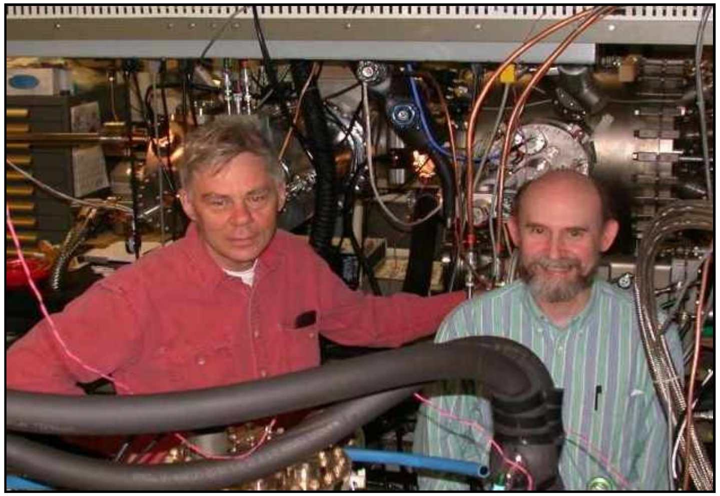


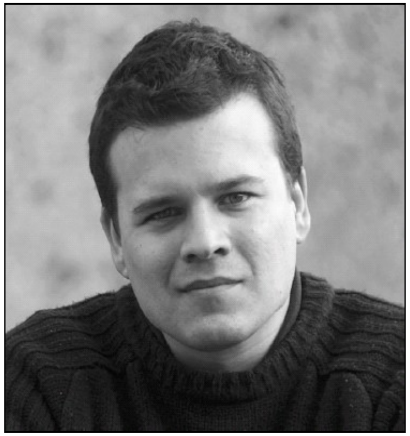
Loren Pfeiffer and Ken West

Picture Gallery

Philip Kim

Yuanbo Zhang

Melinda Han



Pablo Jarrillo-Herrero

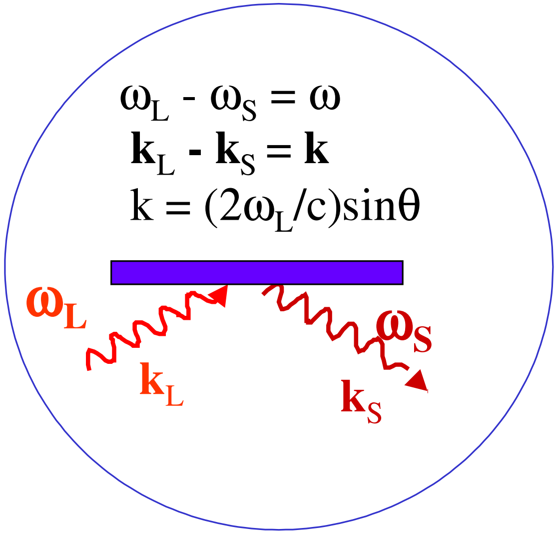
Kirill Bolotin

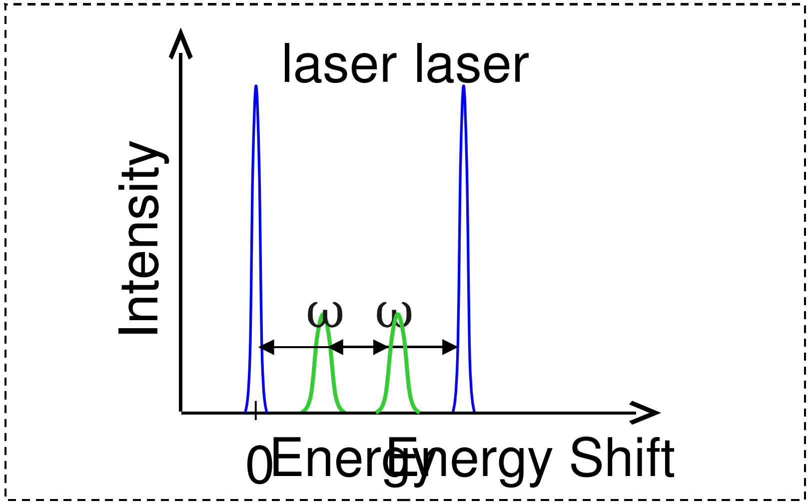
ErikHenriksen

Spectroscopy of Electrons and Phonons in Graphene Structures

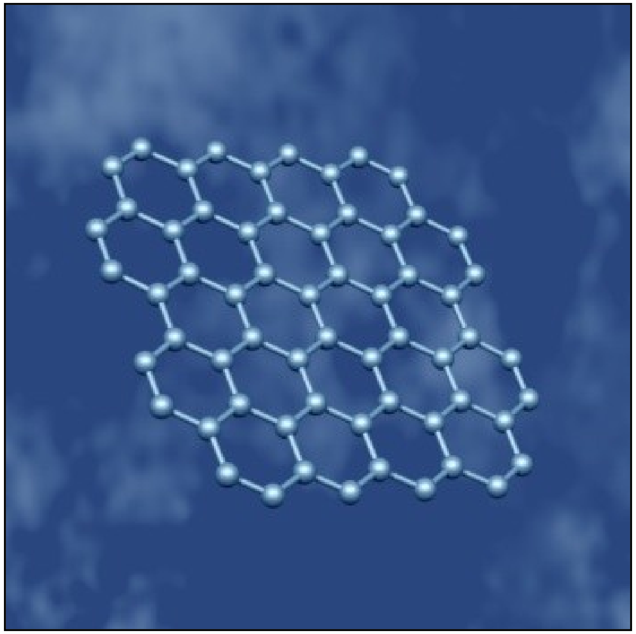
Inelastic Light Scattering

elementary excitation modes are seen directly in the spectra

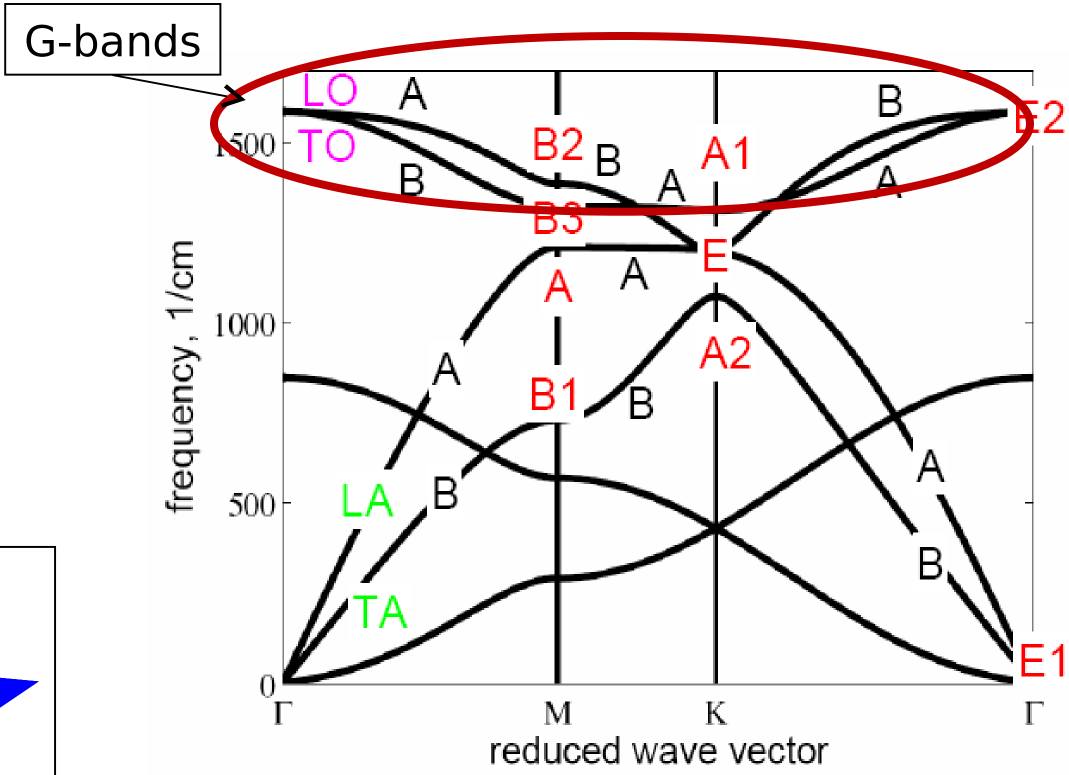


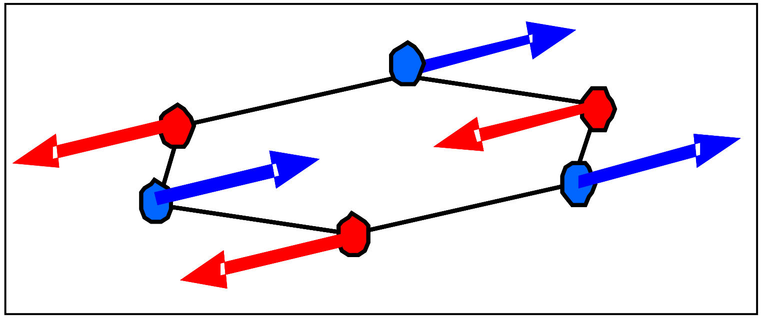


The Bands in Raman Spectra



The Raman bands are due to optical phonon modes

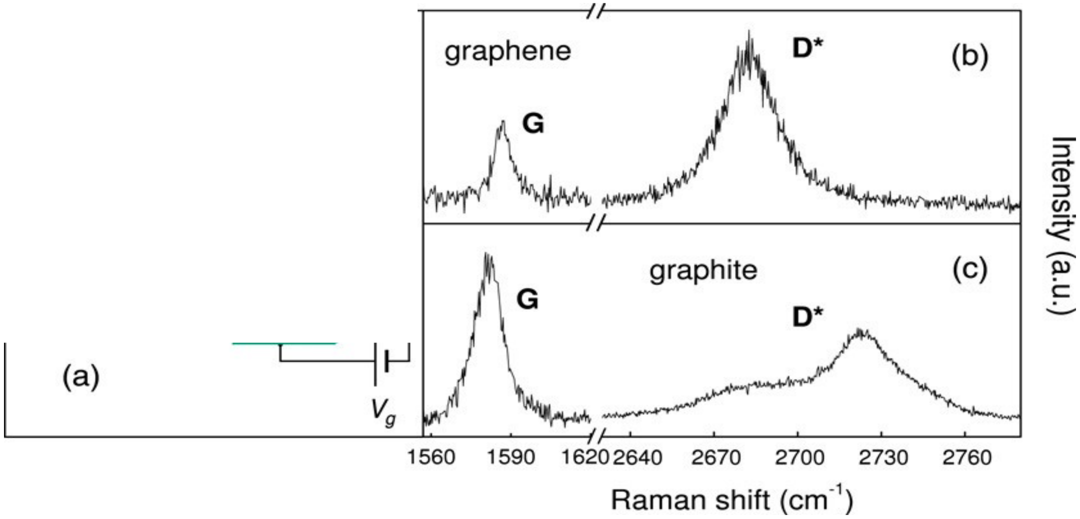




Spectroscopy of Electrons and Phonons in Graphene Structures

Raman scattering by optical phonons

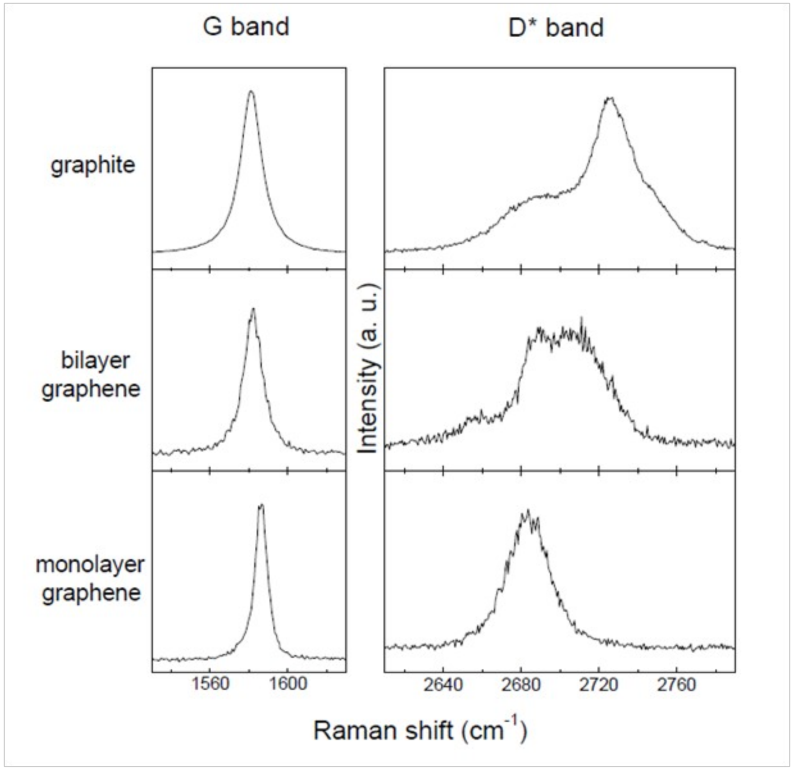
- G-band (q→0 mode)
- D* band (resonant second-order)



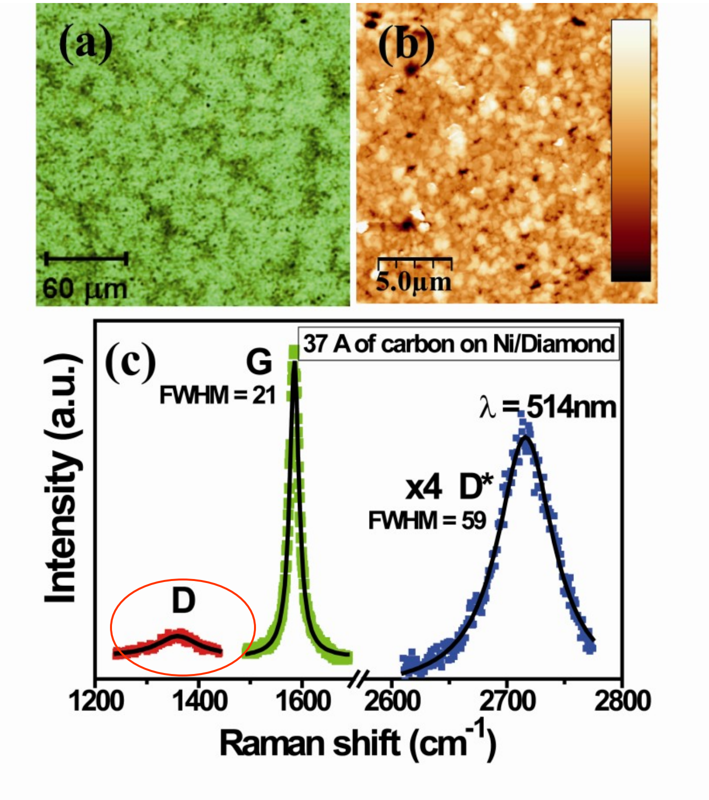
D band at ~1350 cm^{-1} (activated by disorder)

Raman Scattering in Graphene Structures

a wonderful tool



A. Ferrari et al (2006)



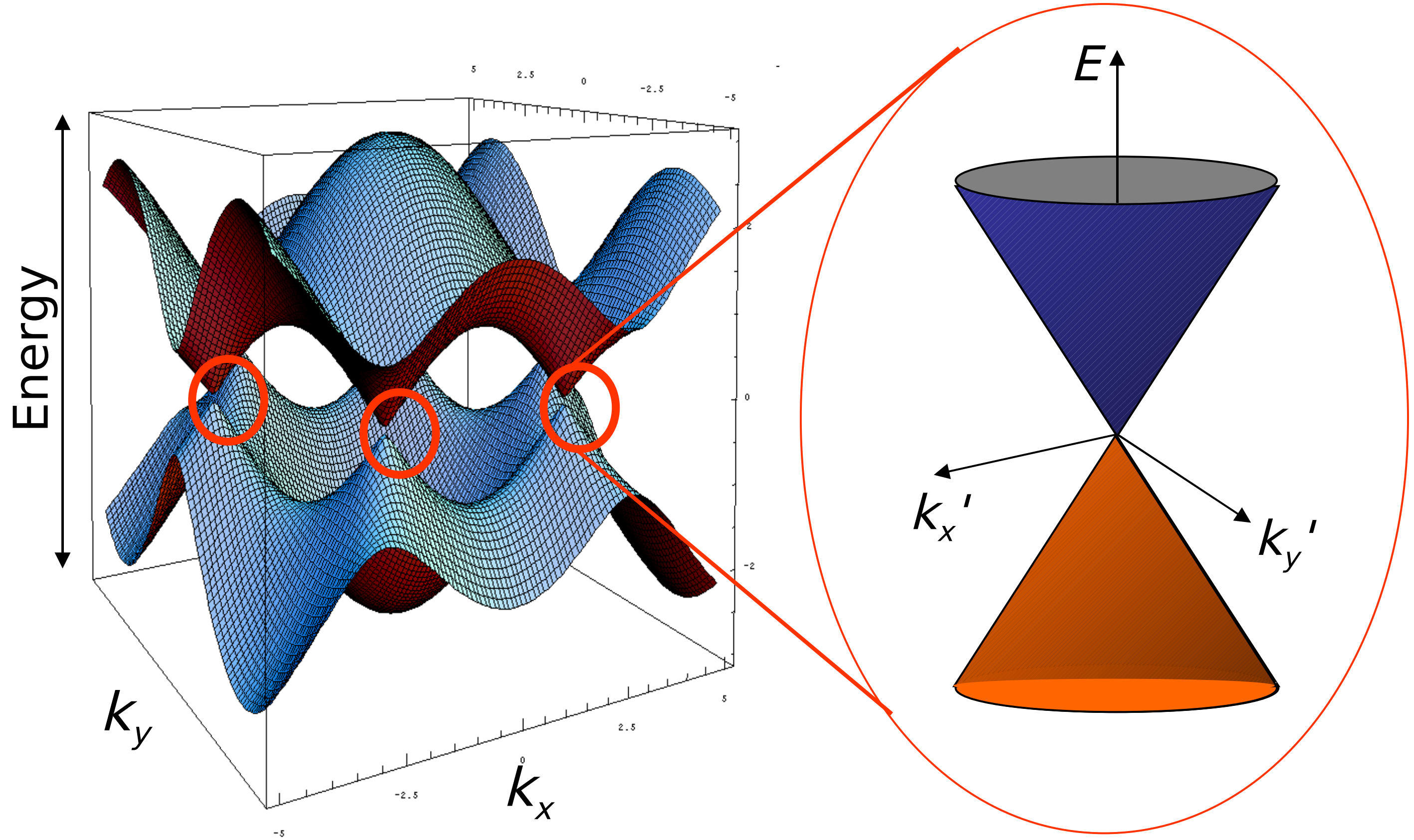
Molecular beam deposition
J. Garcia et al (2009)

Spectroscopy of Electrons and Phonons in Graphene Structures

inelastic light scattering studies

To probe spin or/and charge excitations of Dirac fermions

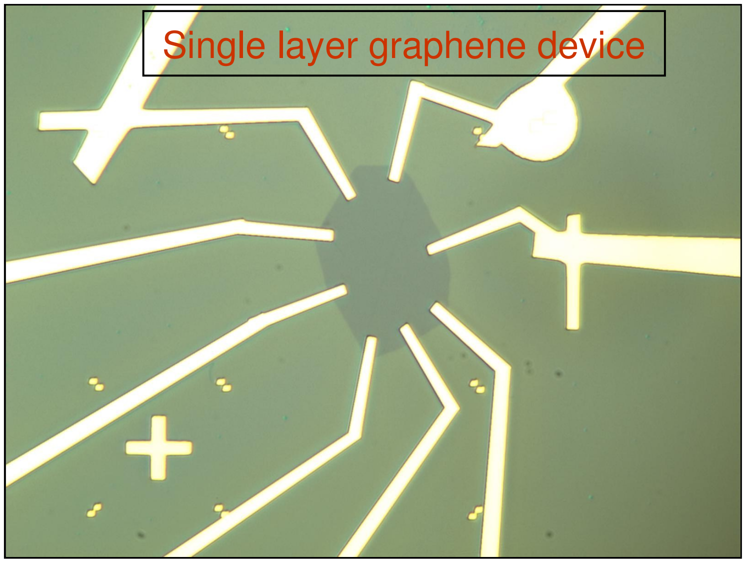
Band structure of graphene

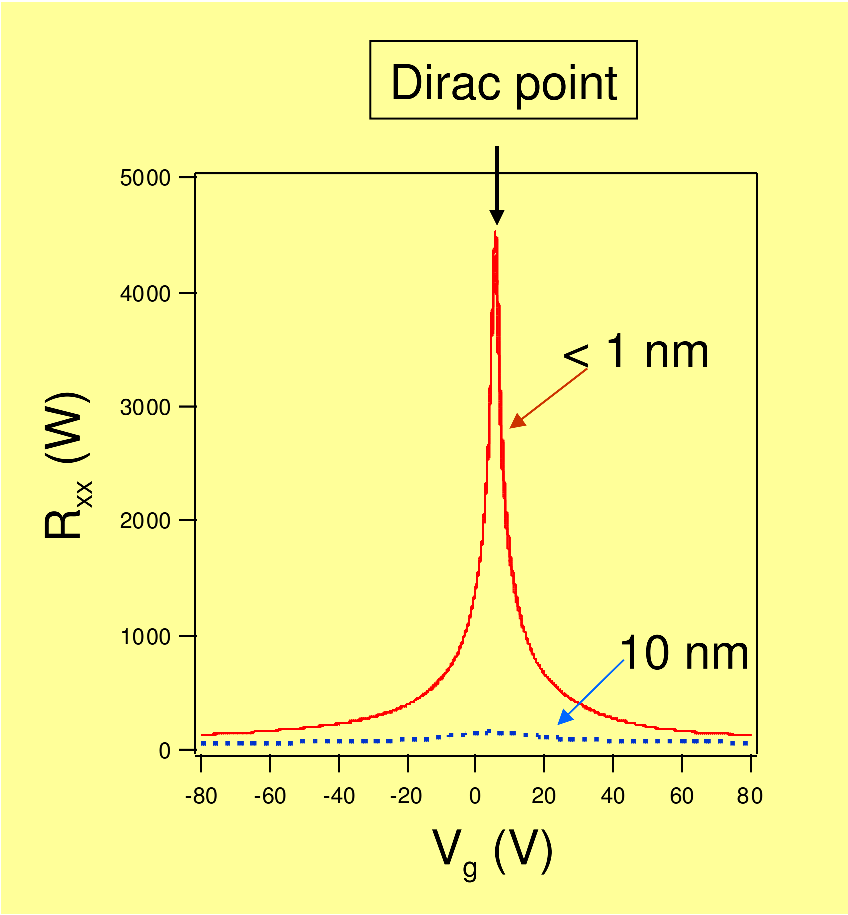


Massless Dirac particles with effective speed of light v_F.

Particle-hole symmetry.

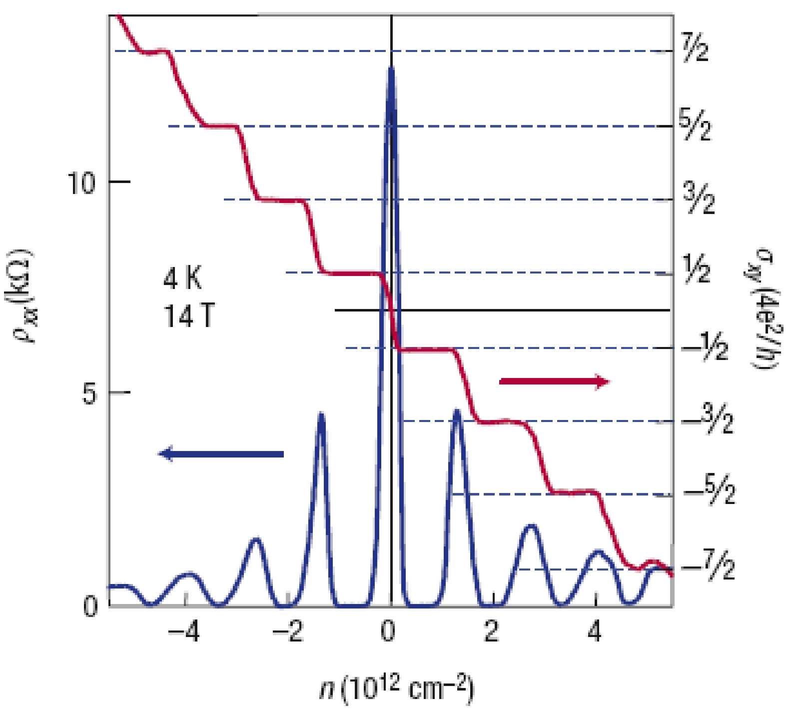
Electric-Field-Effect in Graphene



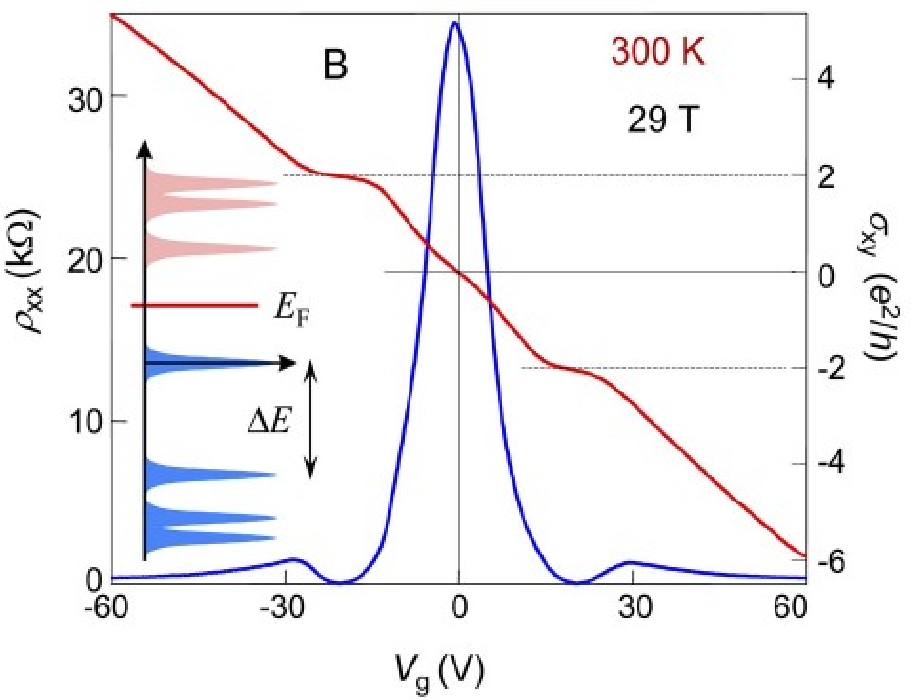


K.S. Novoselov et al Science 306, 666 (2004)
Yuanbo Zhang et al Phys. Rev. Lett 94, 176803 (2005)

Quantum Hall Effect in Graphene

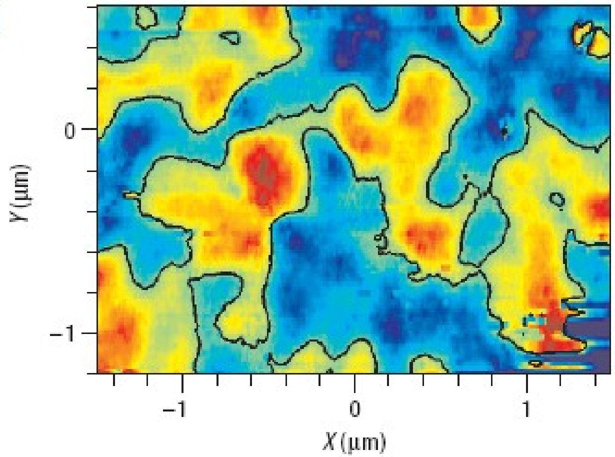


Novoselov et al. Nature (2005)
Zhang et al. Nature (2005)

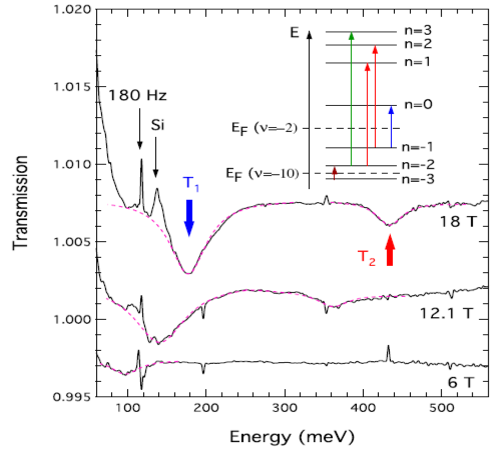


Novoselov et al. Science (2007)

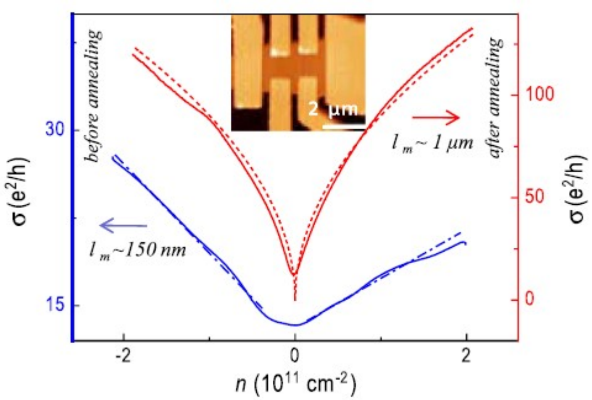
Fermions in Graphene Structures



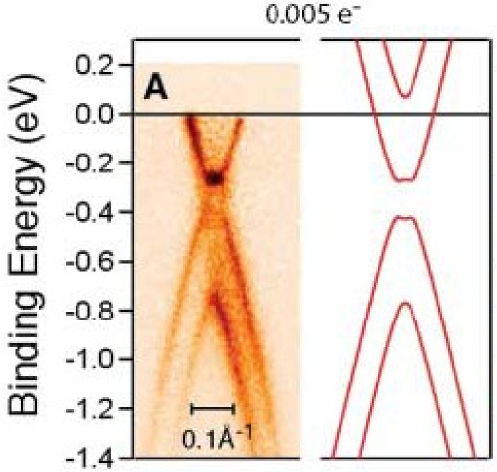
Martin et al. Nature Phys. (2007)



Jiang et al. PRL (2007)



Bolotin et al. PRL (2008)



Ohta et al. Science (2006)

Inelastic Light Scattering Studies in Graphene Structures

To gain insights on Dirac fermions beyond those in:

IR absorption (cyclotron resonance)
Tunneling
Photoemission …

J. Yan et al, Phys. Rev. Lett. 98, 168802 (2007)
J. Yan et al, Phys. Rev. Lett. 101, 136804 (2008)
J. Yan, PhD dissertation, June 2009

Raman scattering studies :

- Electric-field effect
- Electron-phonon coupling:
  - frequency shifts
  - Landau damping
- Magneto-phonon resonances
  - probes of Landau levels
- Broken-symmetry in bilayers

Raman Studies of Graphene Structures

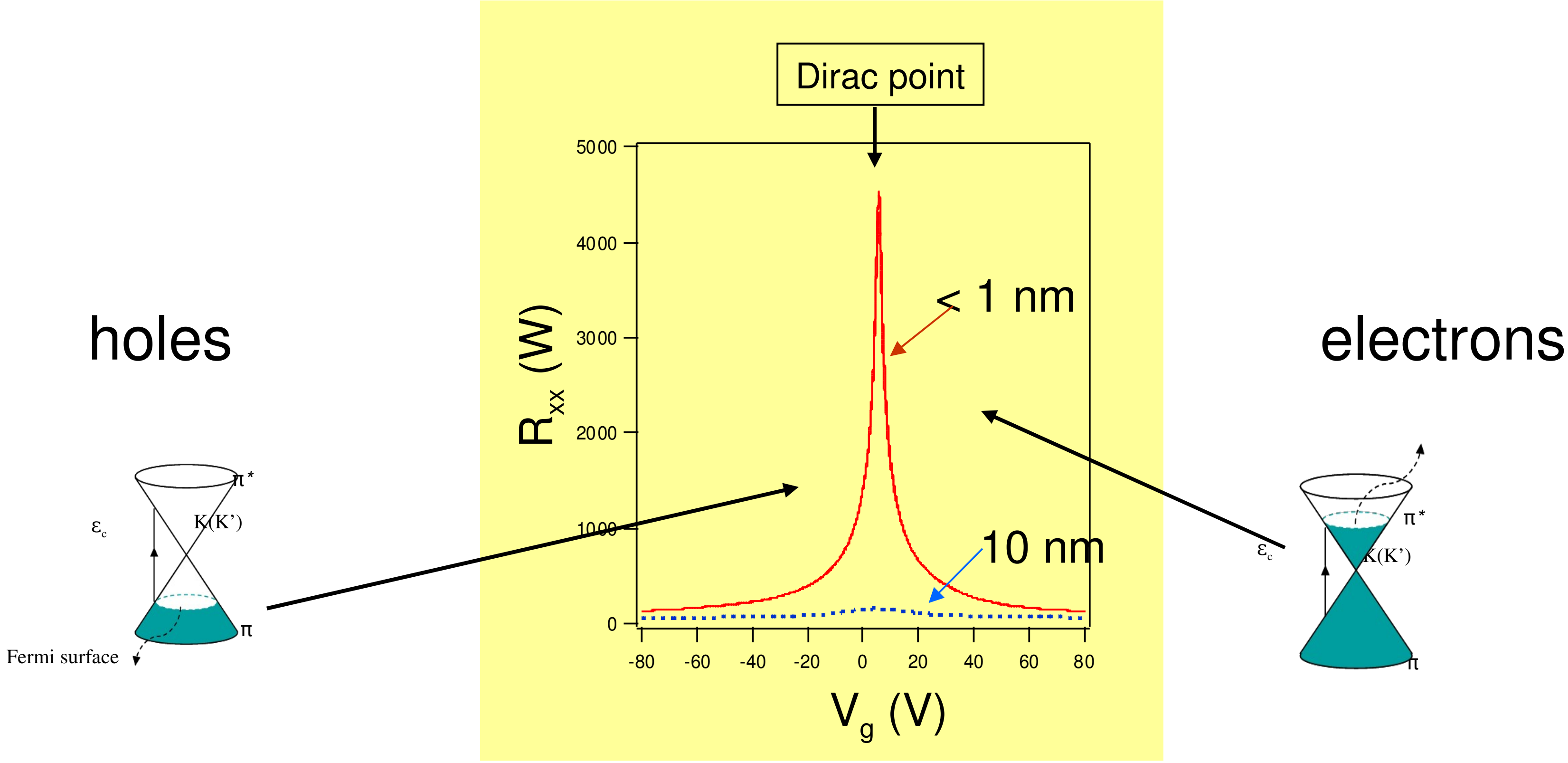
electron-phonon interactions are venues to study carrier properties

M. S. Dresselhaus, P. C. Eklund, A.C. Ferrari, A. Iorio,
M.A. Pimenta, S. Reich, A. K. Sood, C. Thomsen …

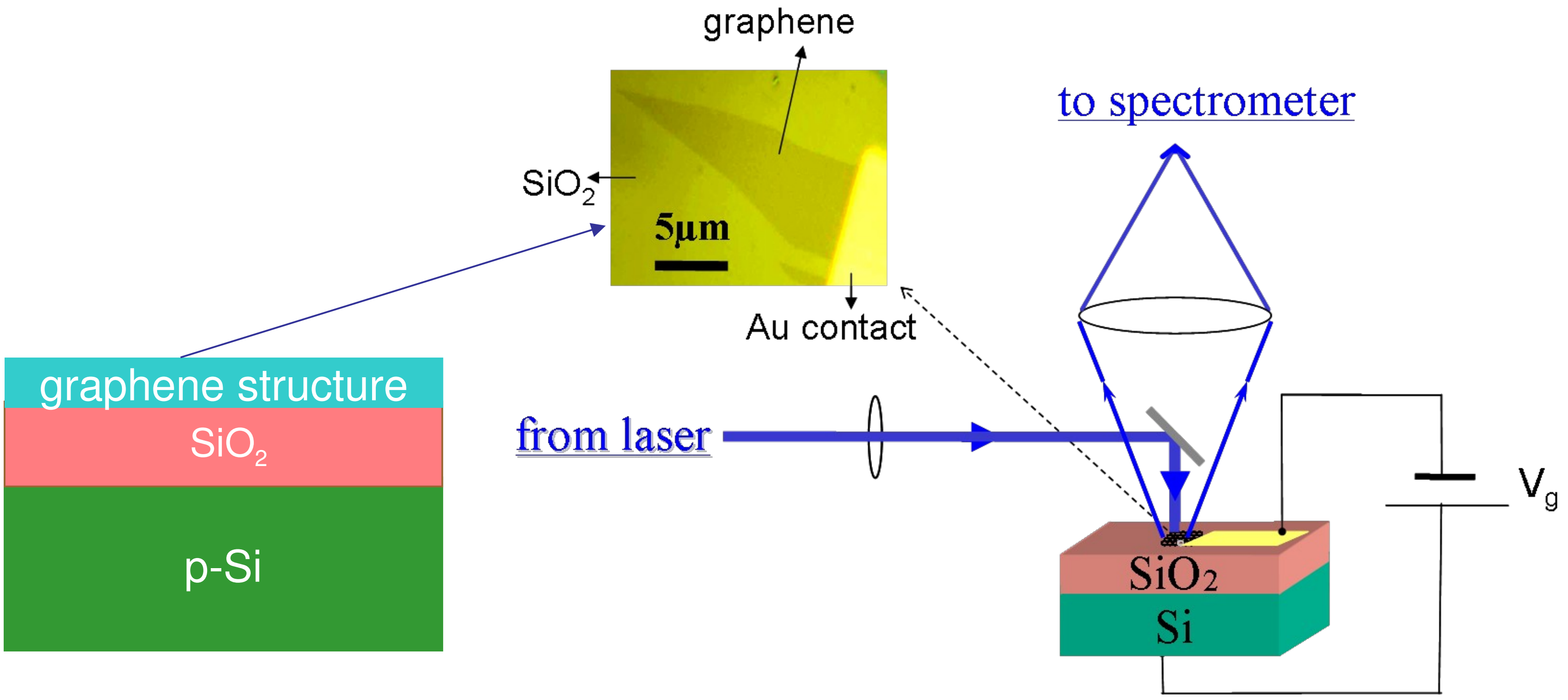
T. Ando, L. Brey, A. Castro Neto, T. Chakraborty,
S. Das Sarma, H. Fertig, F. Guinea, A.H. MacDonald,
F. Maury, …

Electric-Field-Effect in Graphene

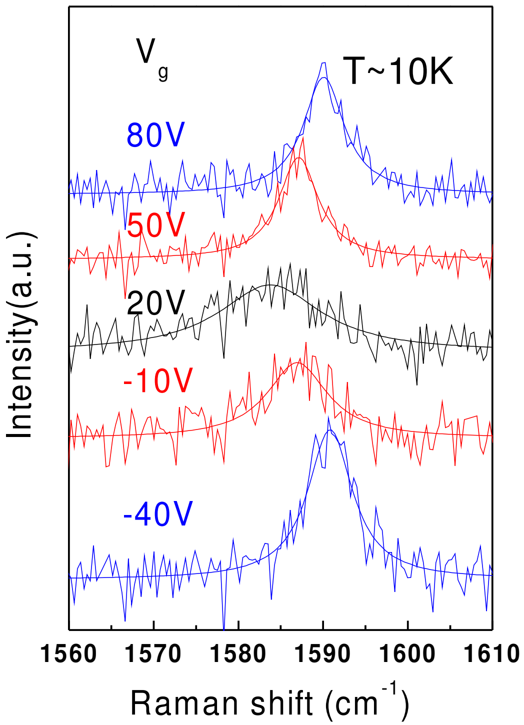
Massless Dirac Fermions



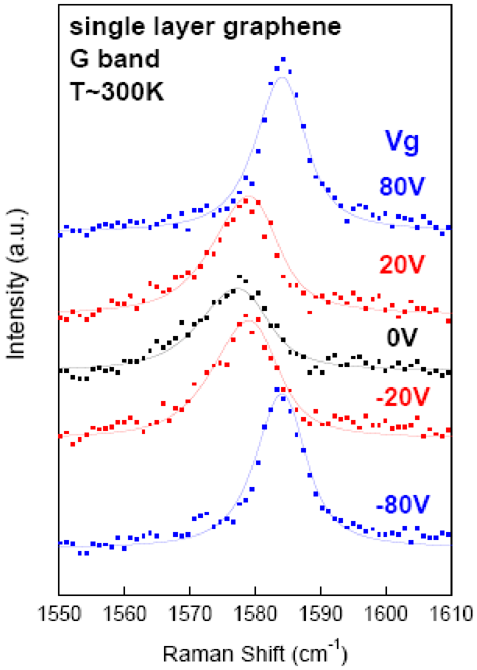
Raman Scattering: electric-field-effect in graphene



Raman Scattering in Gated Single Layer Graphene



Dirac point is at $V_G \to 20V$

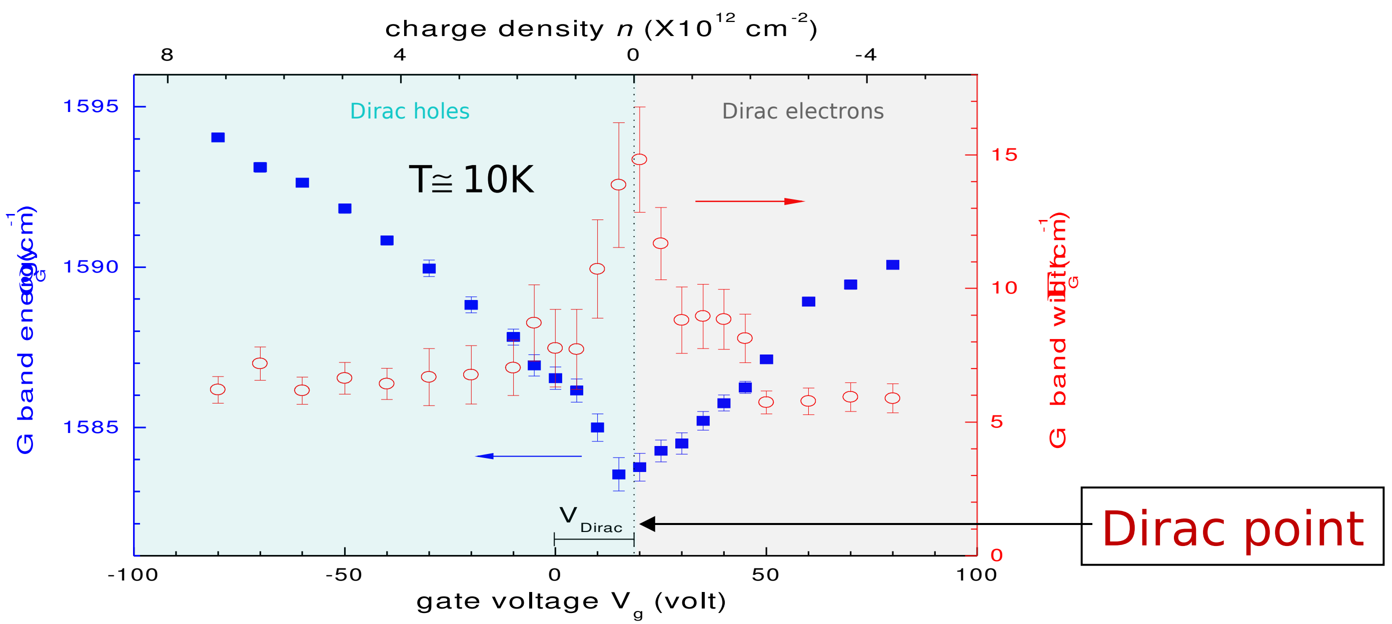


Dirac point is at $V_G \to 0V$

Electric-Field-Effect in Single Layer Graphene: Electron-phonon Coupling

Large dependence of the G band on Dirac fermion density

G band vibration of graphene



Jun Yan et al., Phys. Rev. Lett.2007

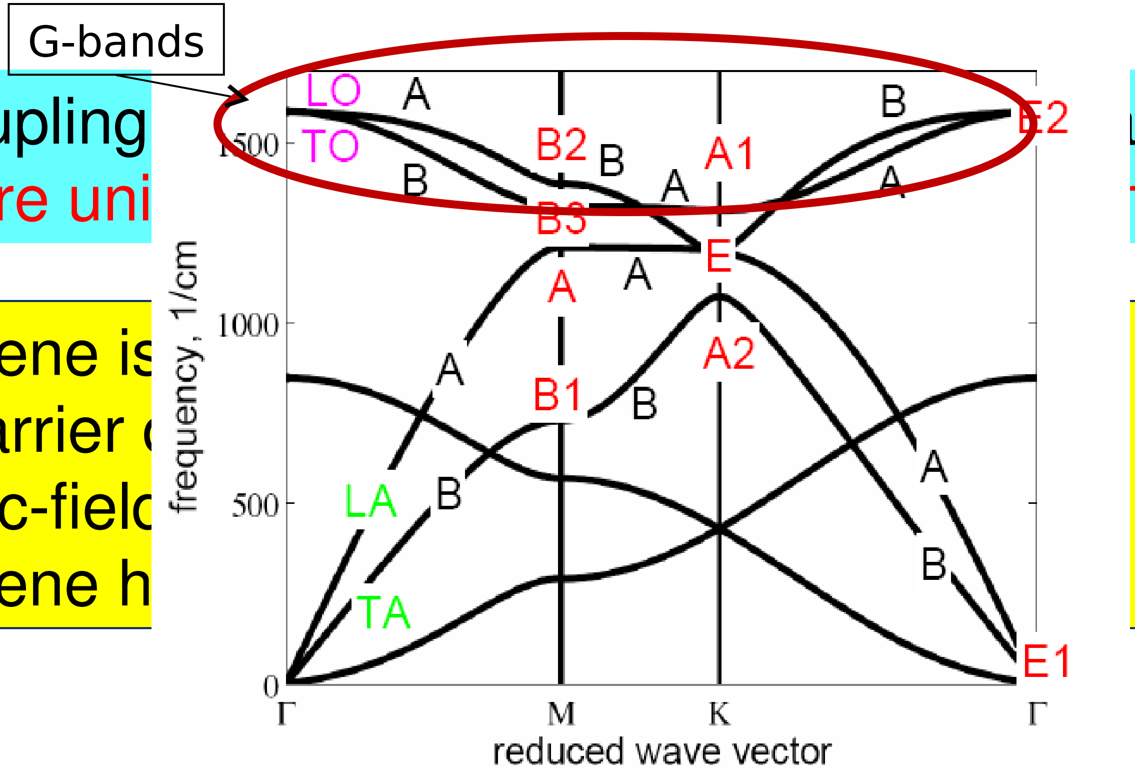
Electron-phonon coupling: the G-band (Raman)

These are non-polar modes because
the ideal graphene lattice has a center of inversion

Electron-phonon coupling is due to the deformation potential interaction

This coupling ... l.
There are uni ... ne:

* Graphene is
* The carrier ... he electric-fielc
* Graphene h

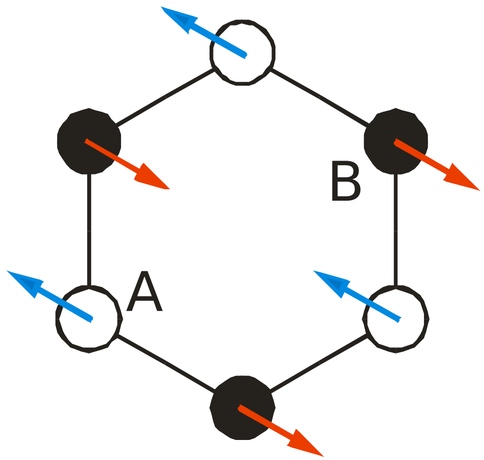


Electron-phonon coupling: the G-band

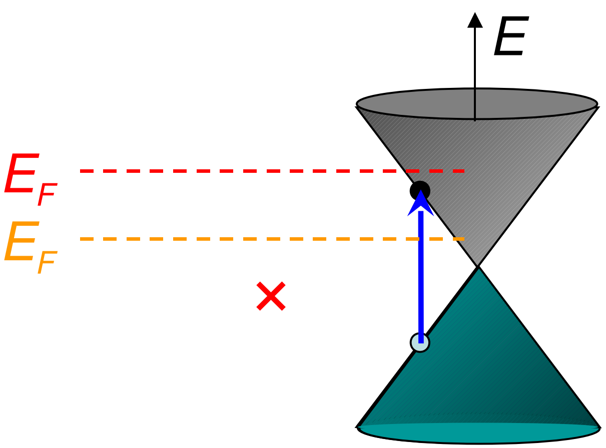
$$\hbar\omega_G = \hbar\omega_{bare} + \Delta(\hbar\omega_G)$$

changes with charge density in graphene

real space view



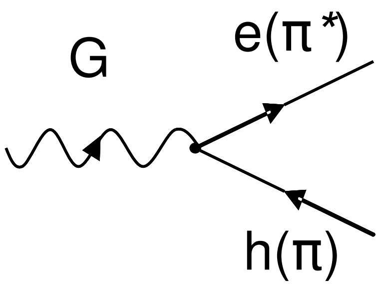
Energy-momentum view

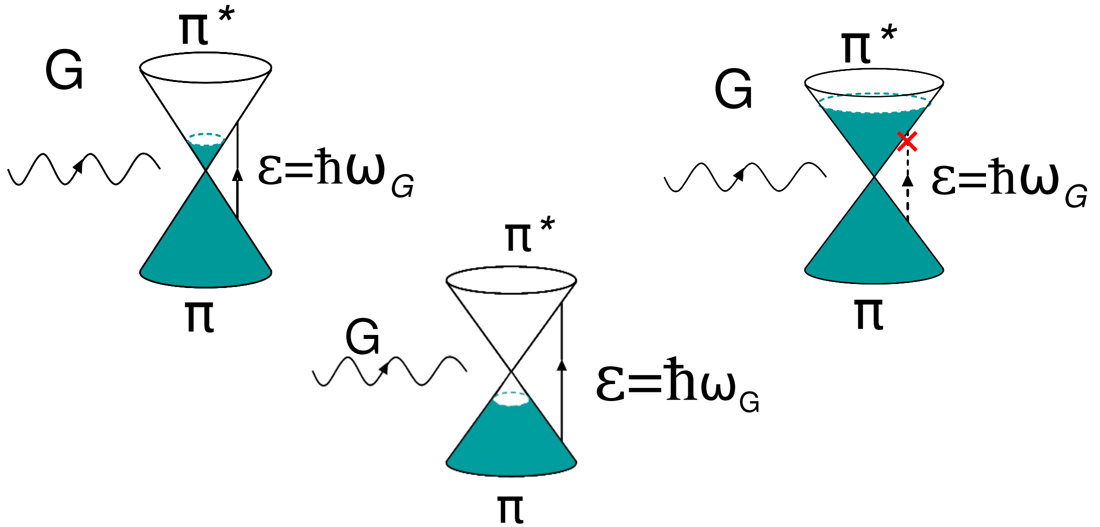


Electron-Phonon Coupling

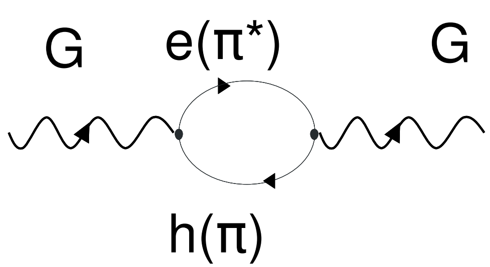
time-dependent perturbation theory

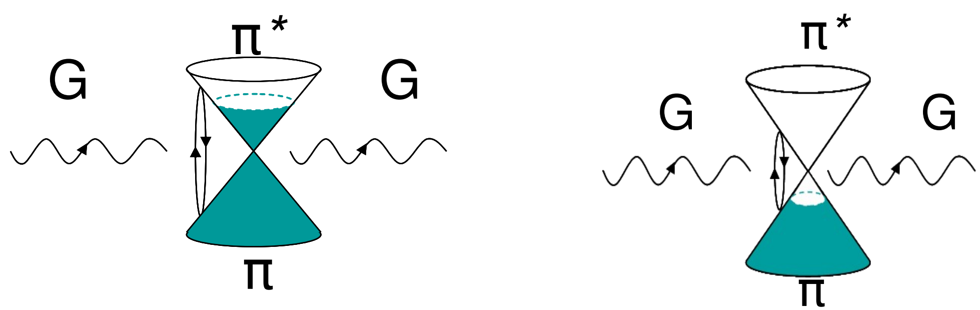
"Landau damping"



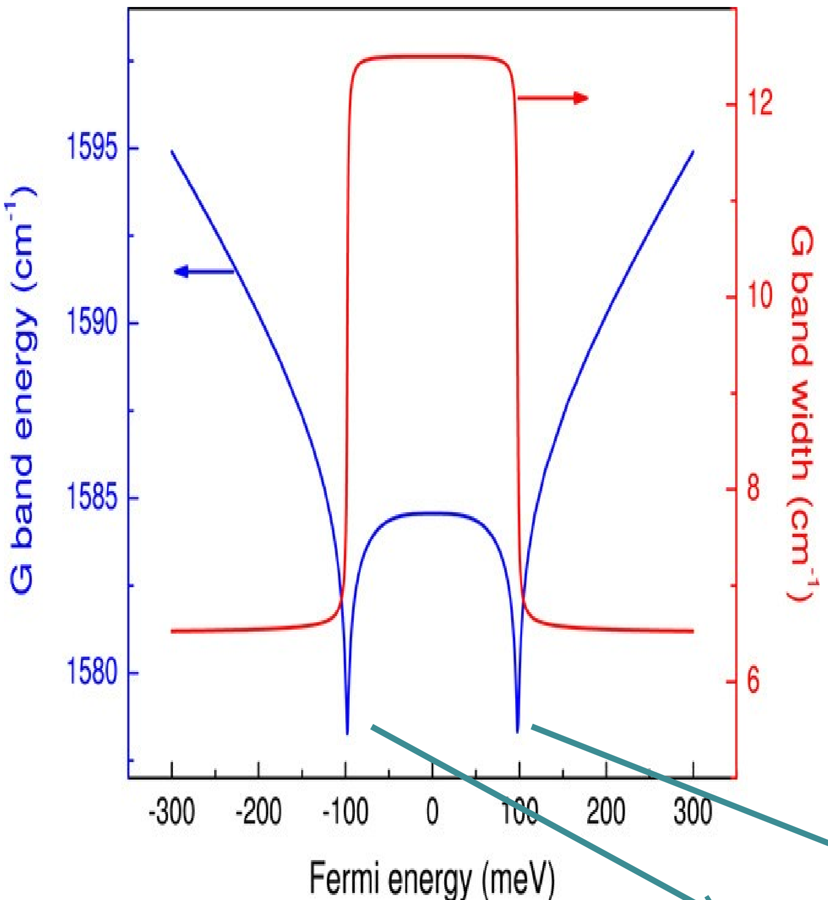


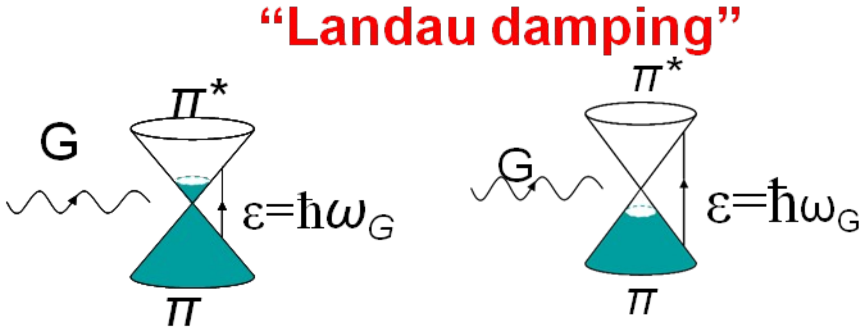
Phonon energy shift

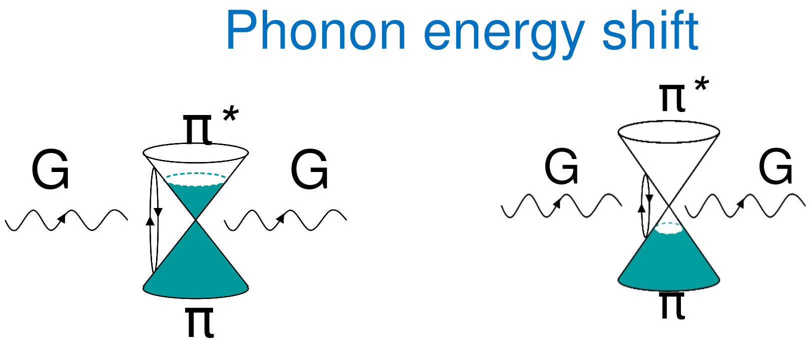




Electron-Phonon Coupling



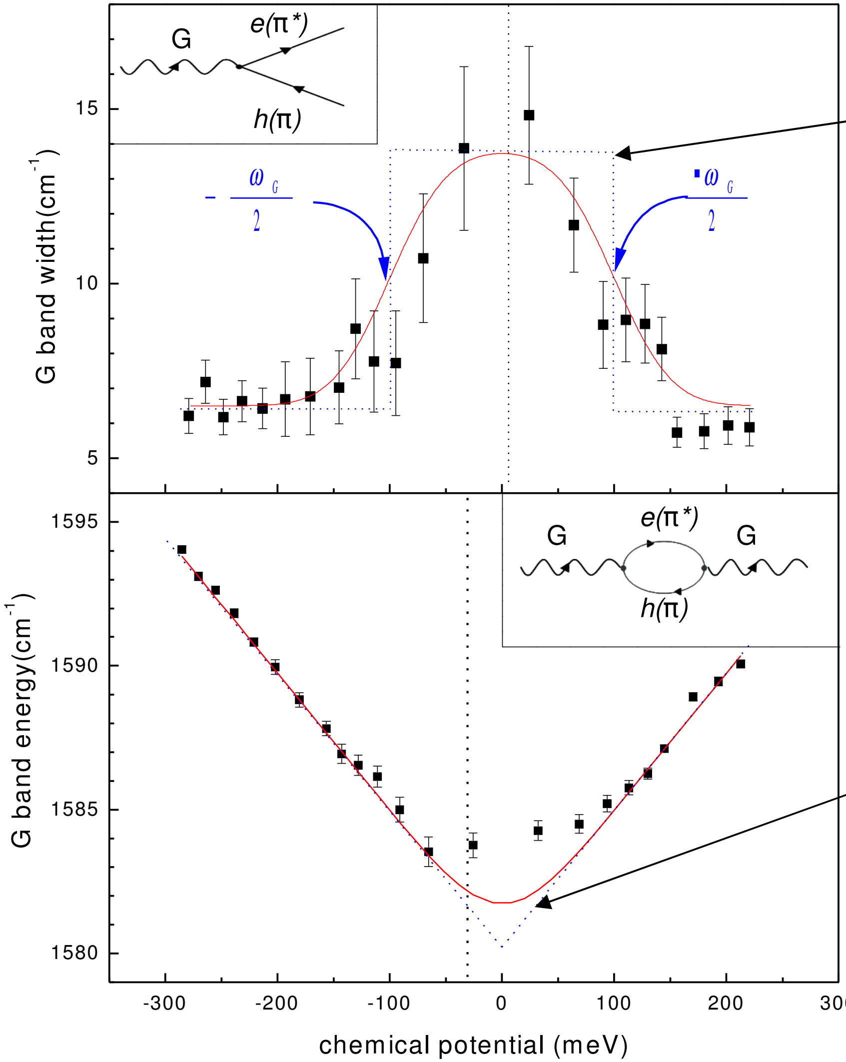




$$\Delta(\hbar\omega_G)=\lambda\left\{|E_F|+\ln\left|\frac{2|E_F|-\hbar\omega_G}{2|E_F|+\hbar\omega_G}\right|\right\}$$

<u>logarithmic divergence</u> due to <u>resonant</u> electron-phonon coupling ('phonon anomaly')

Electron-Phonon Coupling



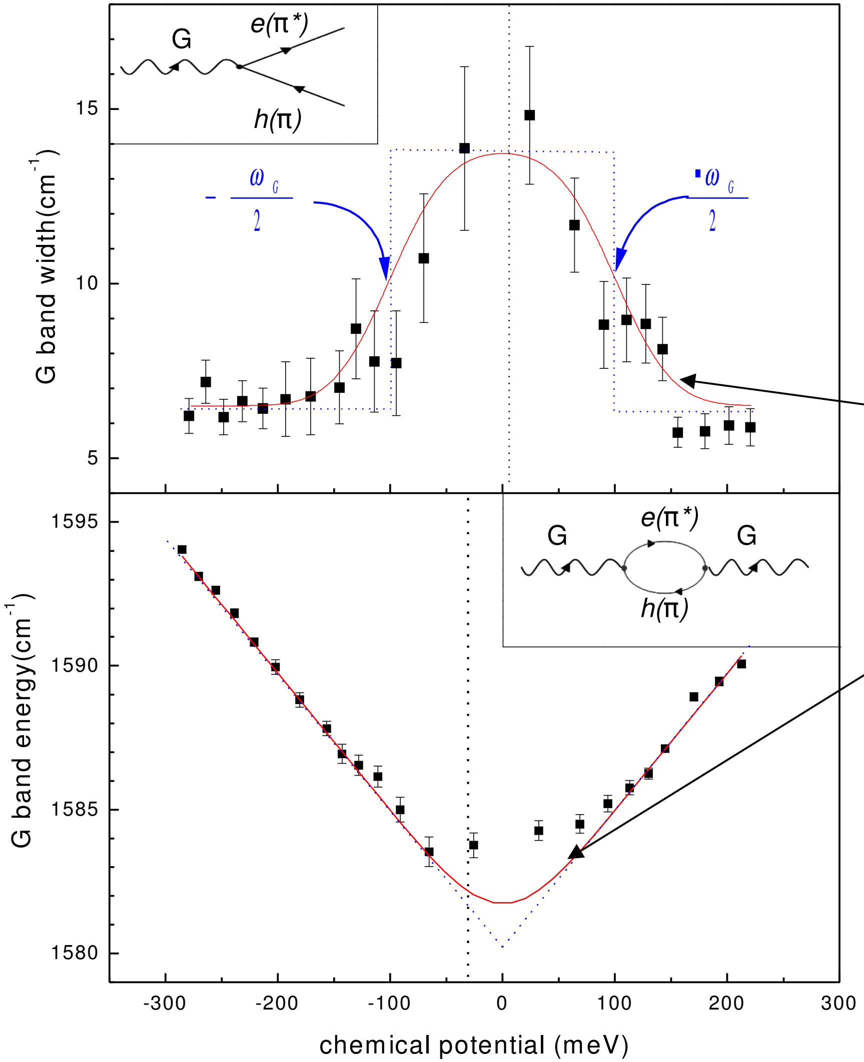
$$\Delta\Gamma_G = \frac{A_{uc}\overline{D^2}}{2Mv_F^2}$$

$$\hbar\omega_G = \hbar\omega_G^0 + \frac{A_{uc}\overline{D^2}}{\pi\hbar\omega_G^0 M v_F^2}|E_F|$$

('phonon anomaly' term is not included)

J. Yan et al, Phys. Rev. Lett. 98, 168802 (2007)

Electron-Phonon Coupling



$$\Delta\Gamma_G = \frac{A_{uc}\overline{D^2}}{2Mv_F^2}$$

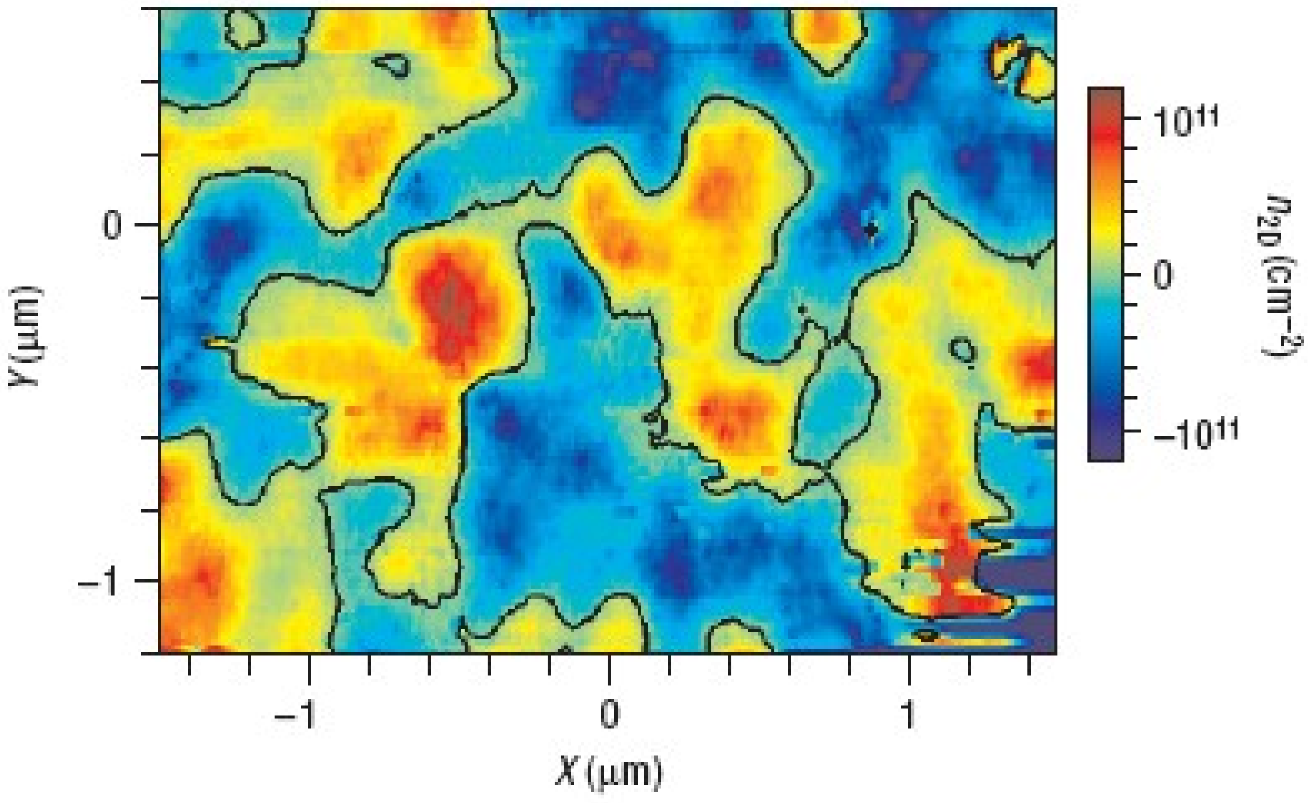
Non-uniformity

$$g(\varphi) = \frac{1}{\sqrt{\pi}}\frac{\sqrt{2}}{\Delta\varphi}e^{-\left(\frac{\varphi}{\Delta\varphi/\sqrt{2}}\right)^2}$$

$$\hbar\omega_G = \hbar\omega_G^0 + \frac{A_{uc}\overline{D^2}}{\pi\hbar\omega_G^0 M v_F{}^2}|E_F|$$

Non-uniformity removes the 'phonon anomaly'

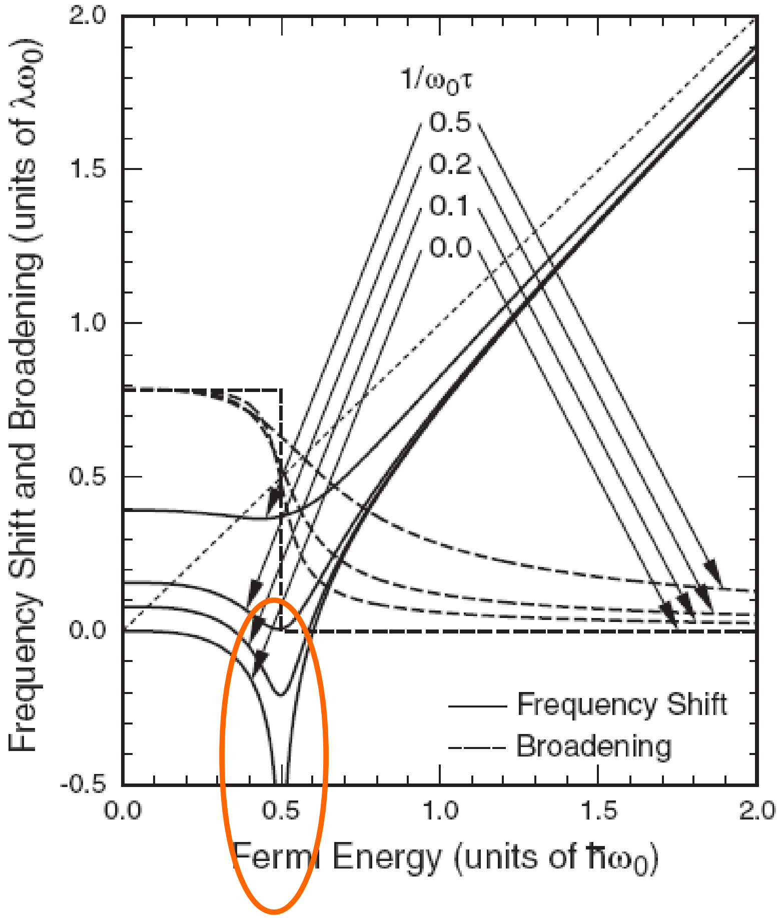
Electron and hole puddles in graphene



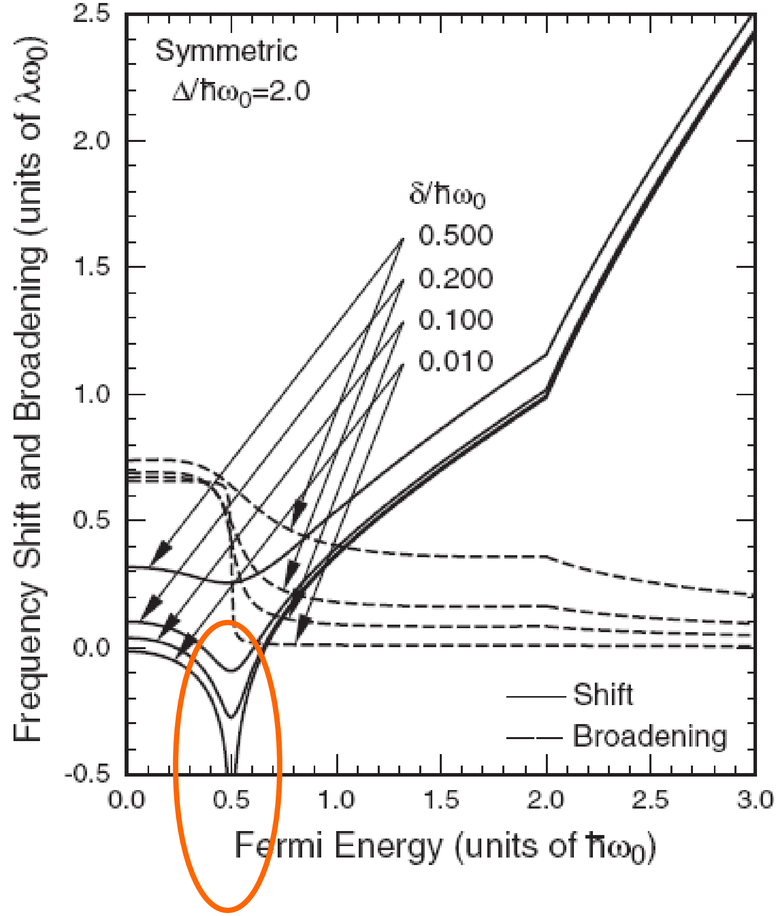
Martin et al Nature Phys. 4, 144 (2007)

predictions of phonon anomaly in graphene

- single-layer
- bi-layer

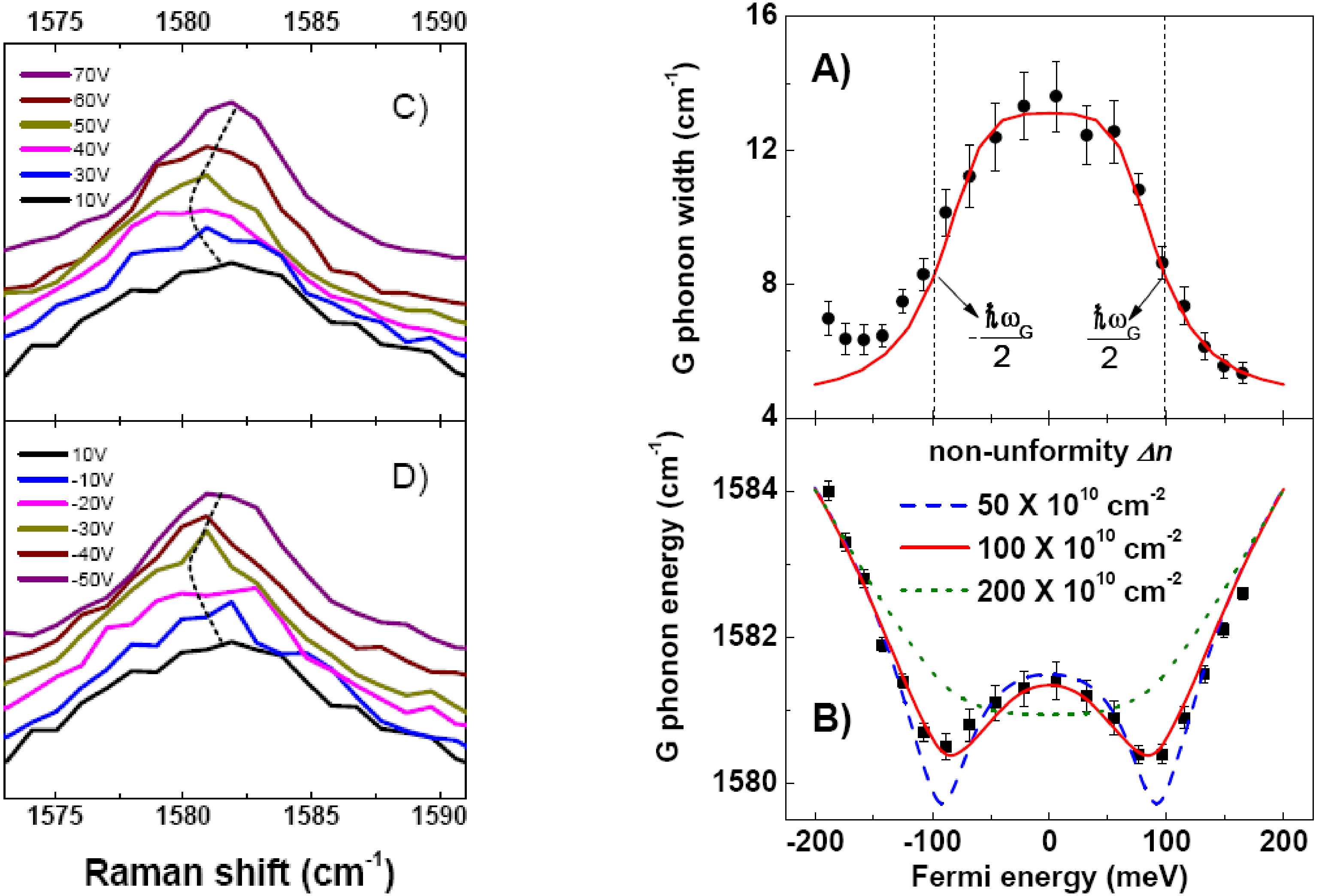


T. Ando JPSJ 75, 124701 (2006)



T. Ando JPSJ 76, 104711 (2007)

‘phonon anomaly’ in bi-layer graphene



Jun Yan et al, Phys. Rev. Lett. (2008)
Jun Yan, PhD dissertation (2009)

Raman scattering in gated graphene

- Electron-phonon coupling in G-band

  * frequency shift linear in E_F:
    $\sim n^{1/2}$ (Dirac fermions)

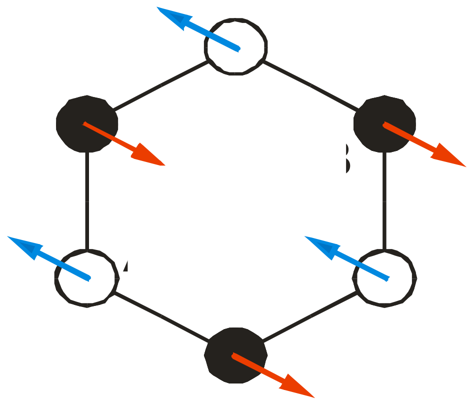
  * Landau damping

  * Particle-hole symmetry

  * Inhomogeneous broadening

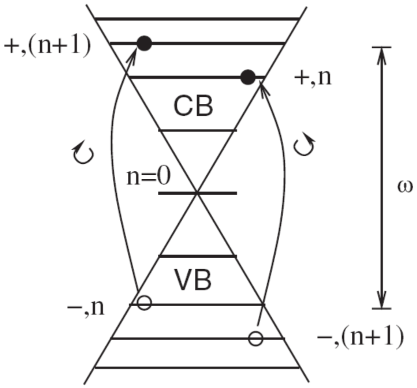
  * ‘phonon anomaly’

magneto-phonon-resonance

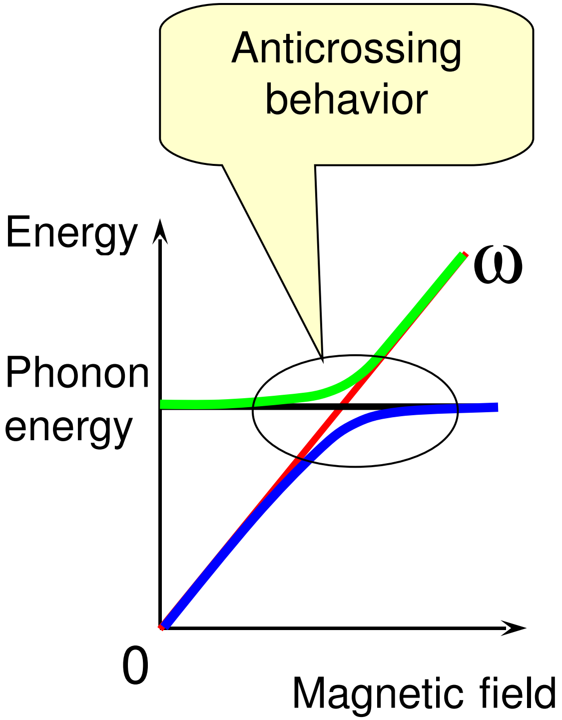


G-band phonon

add a magnetic field

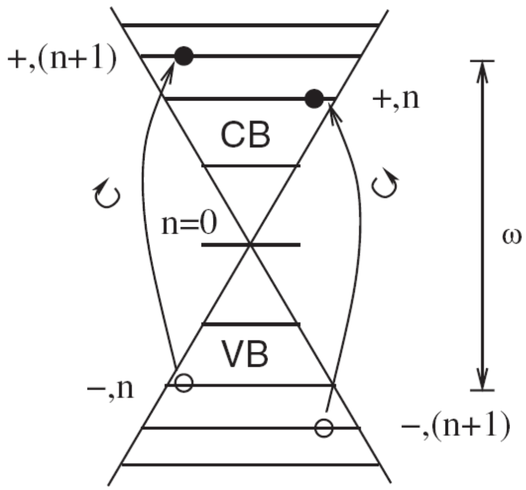


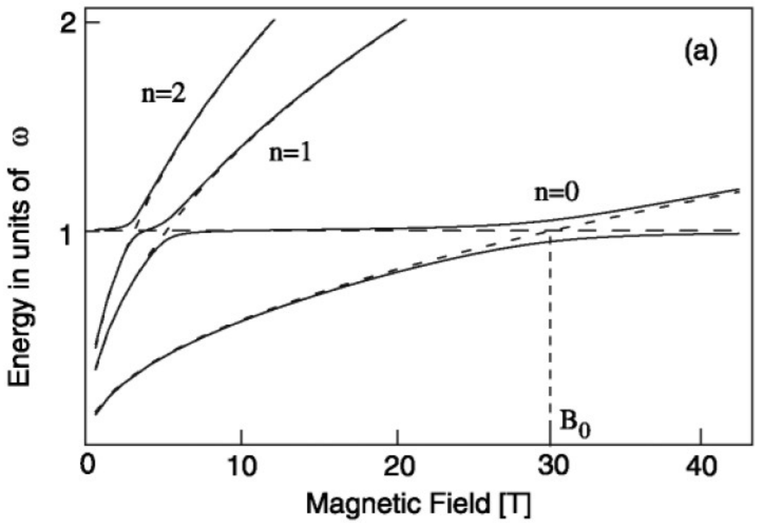
Inter-Landau level transition



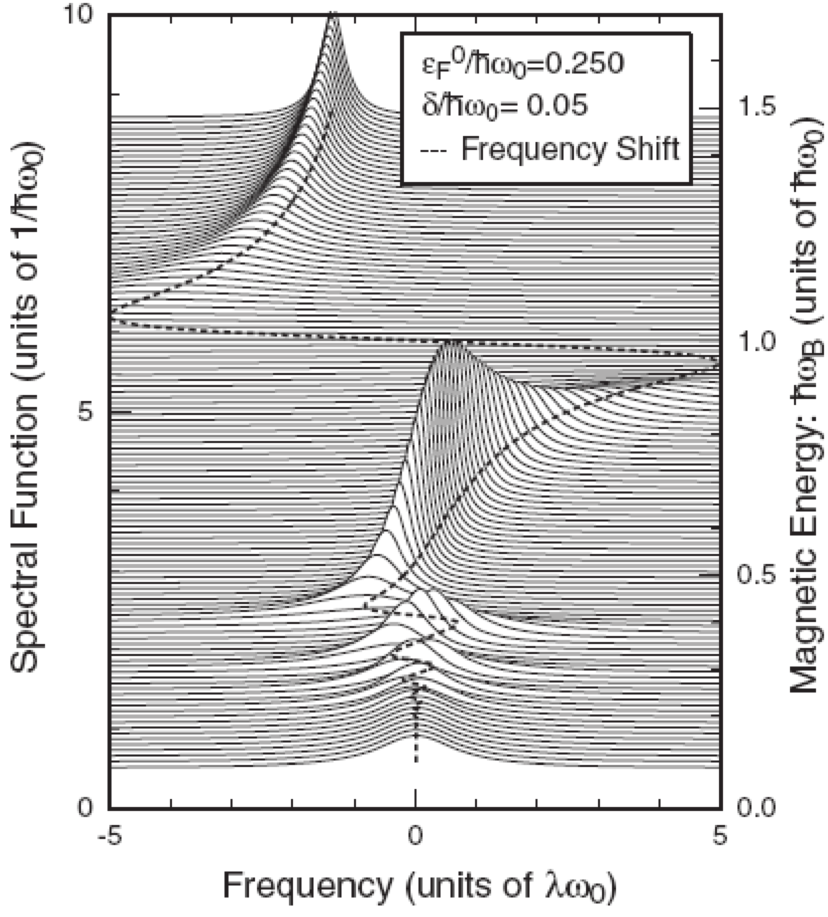
- Magneto-phonon-resonance (MPR) from electron-phonon coupling
- MPR probes Landau level physics

theoretical predictions of MPR in graphene



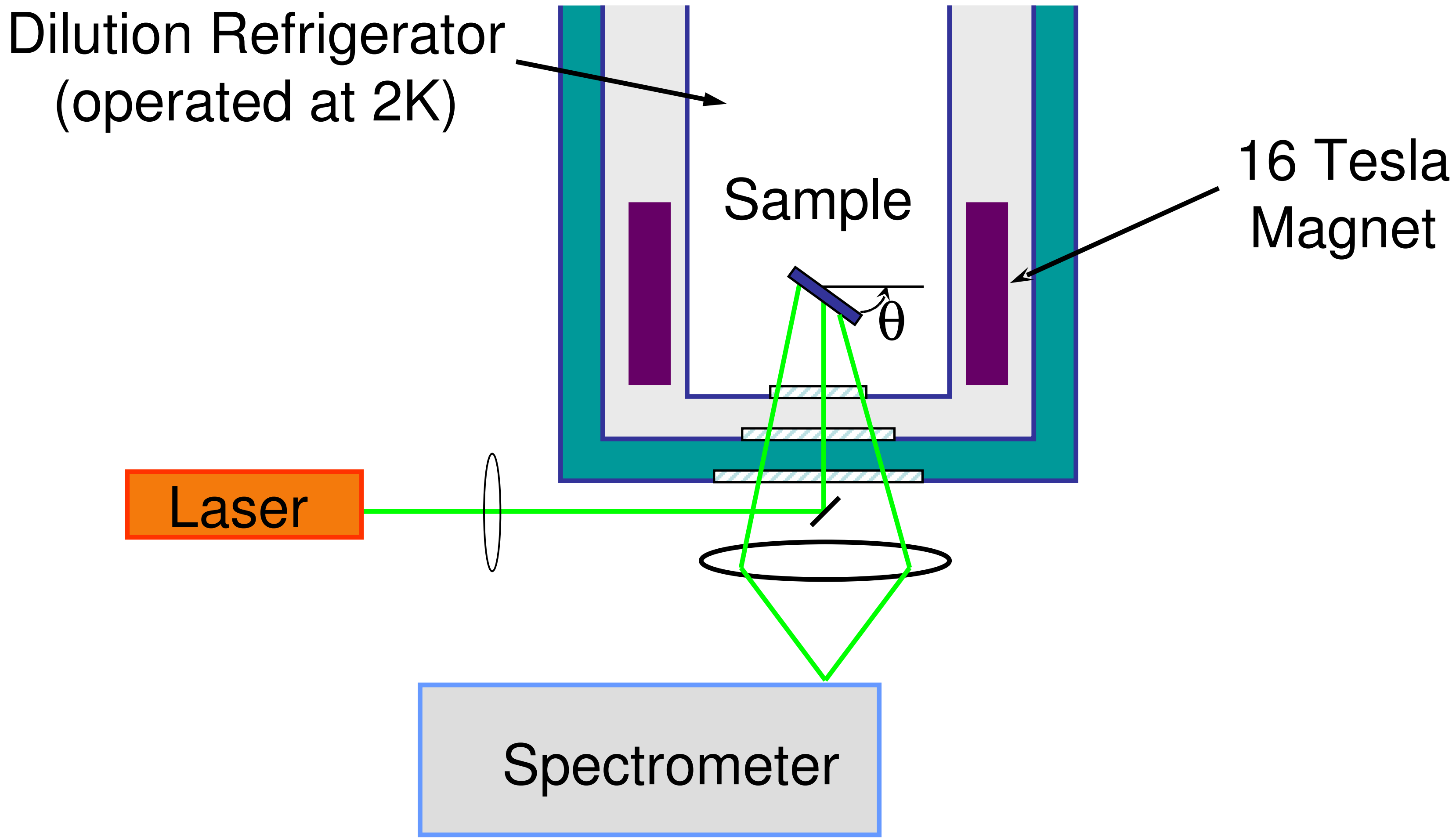


Goerbig et al PRL 99, 087402 (2007)

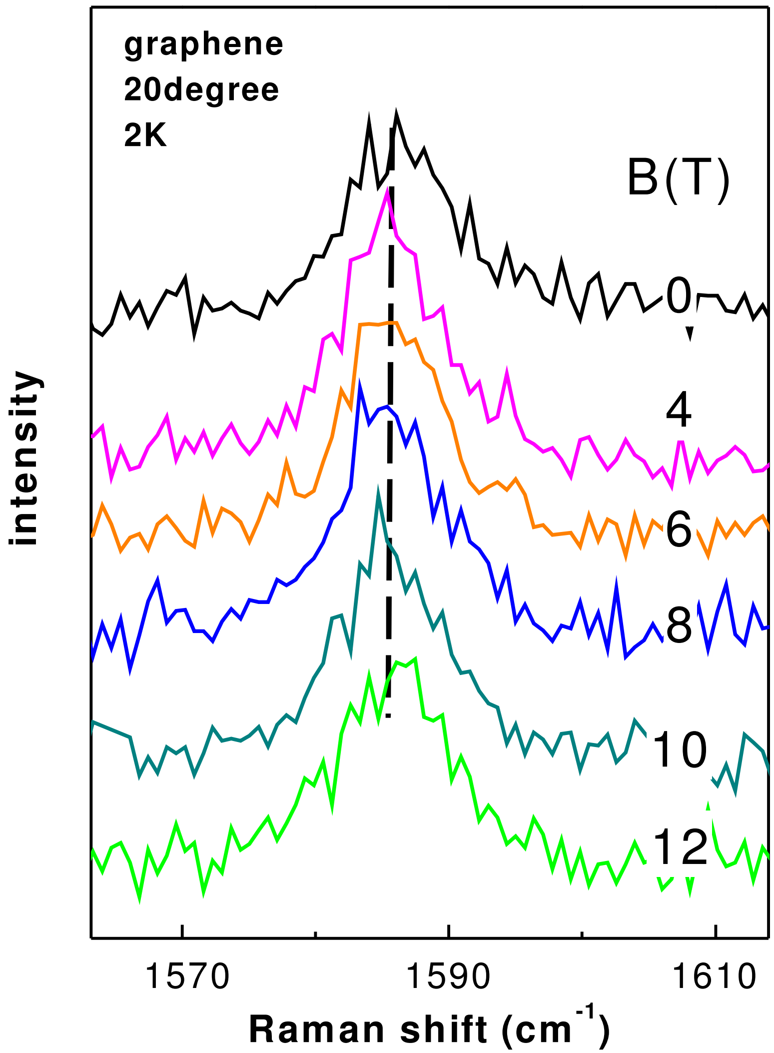


T. Ando JPSJ 76, 024712 (2007)

Experimental setup



preliminary results of MPR in graphene



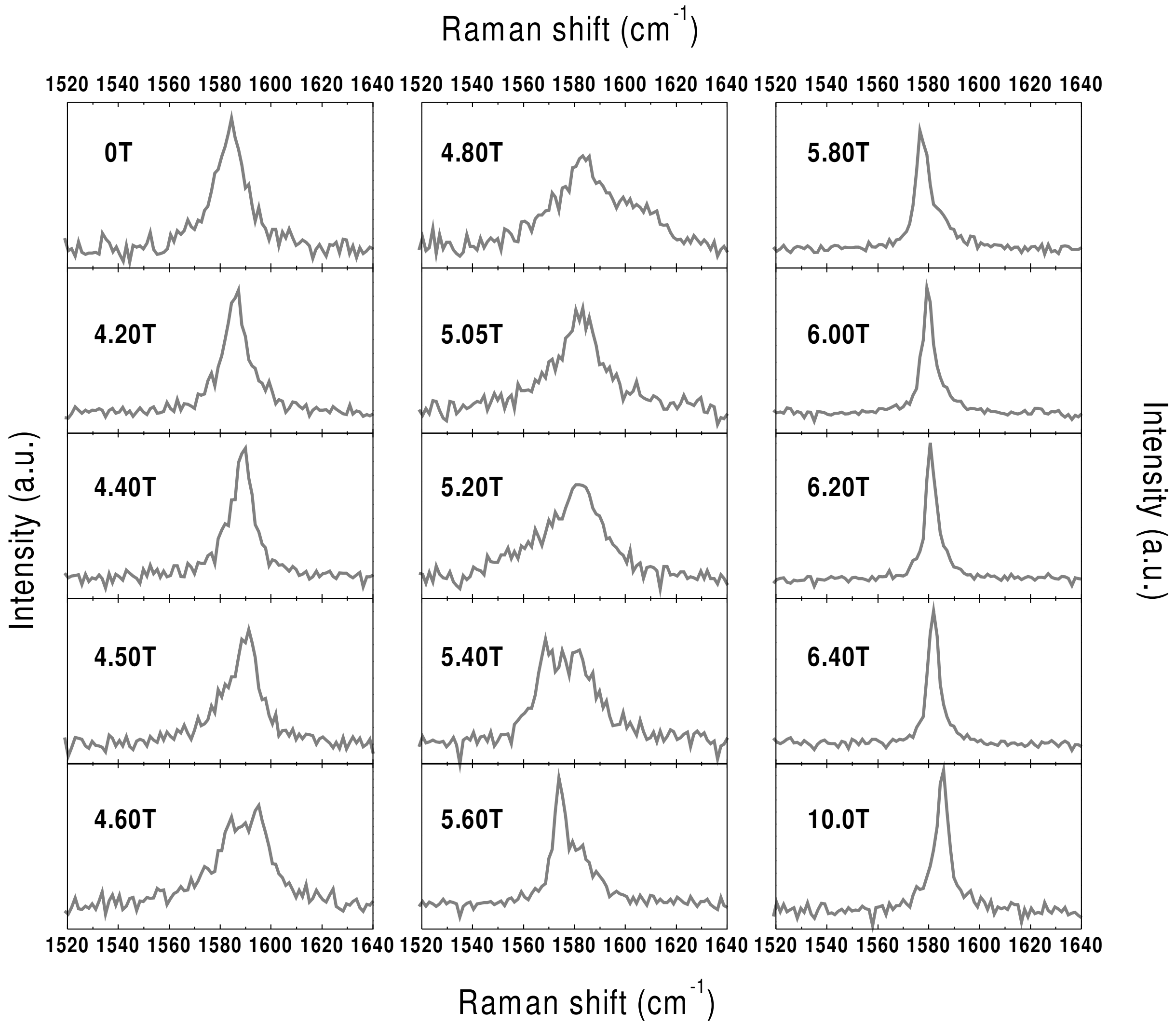
- Quite small MPR effects

J. Yan, PhD dissertation, June 2009

MPR in 'Kish' graphite

Kish graphite has marked position dependence

<u>region I</u> has large MPR

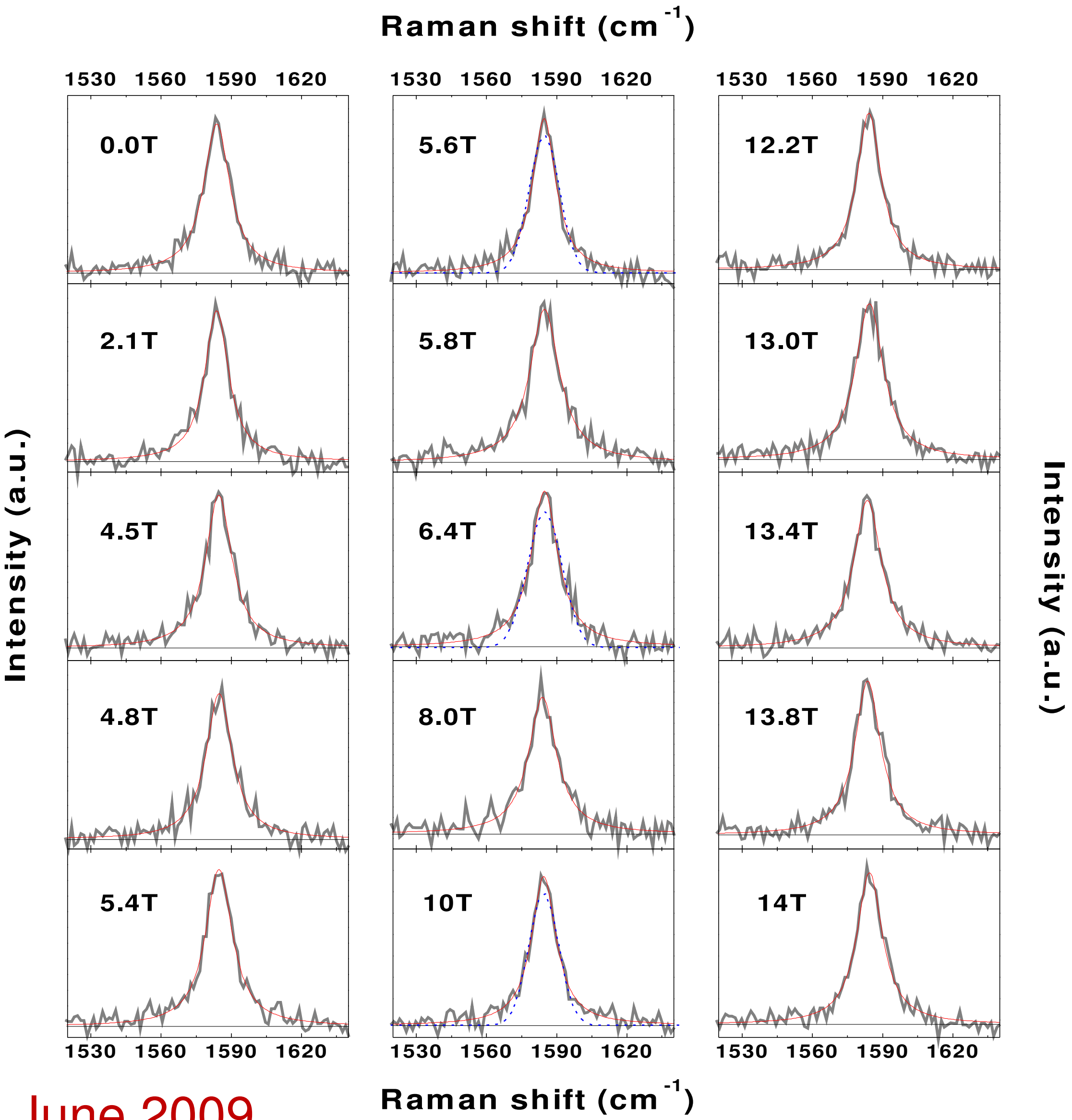


J. Yan, PhD dissertation, June 2009

MPR in ‘Kish’ graphite

Kish graphite has marked position dependence

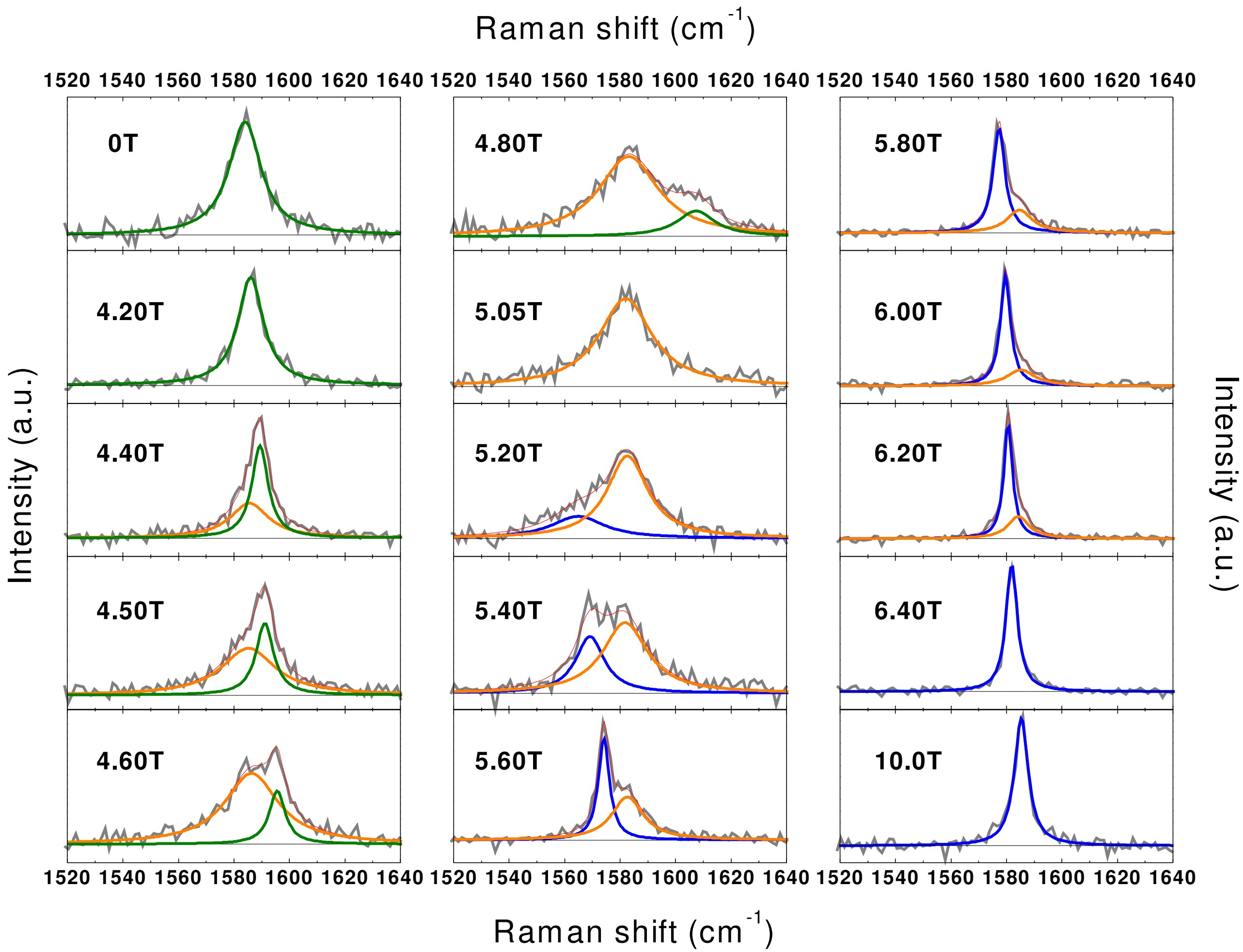
<u>region II</u> has small MPR



J. Yan, PhD dissertation, June 2009

MPR in graphite: region I

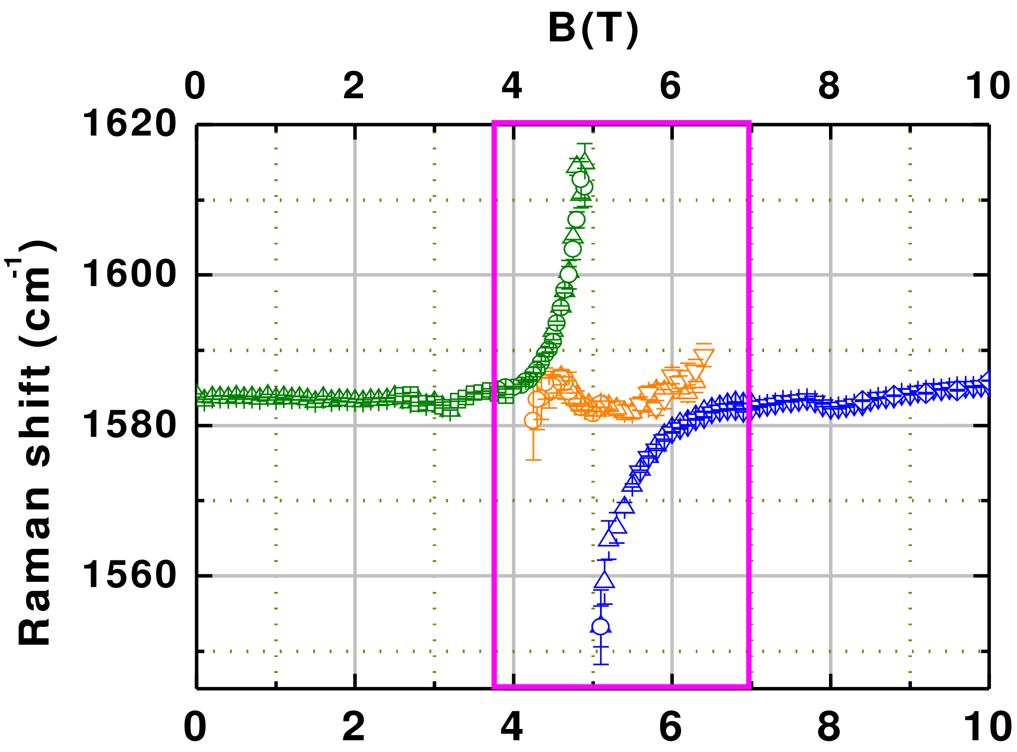
line-shape analysis of the MPR



A ‘marked MPR’ overlaps with a ‘minor’ one

J. Yan, PhD dissertation, June 2009

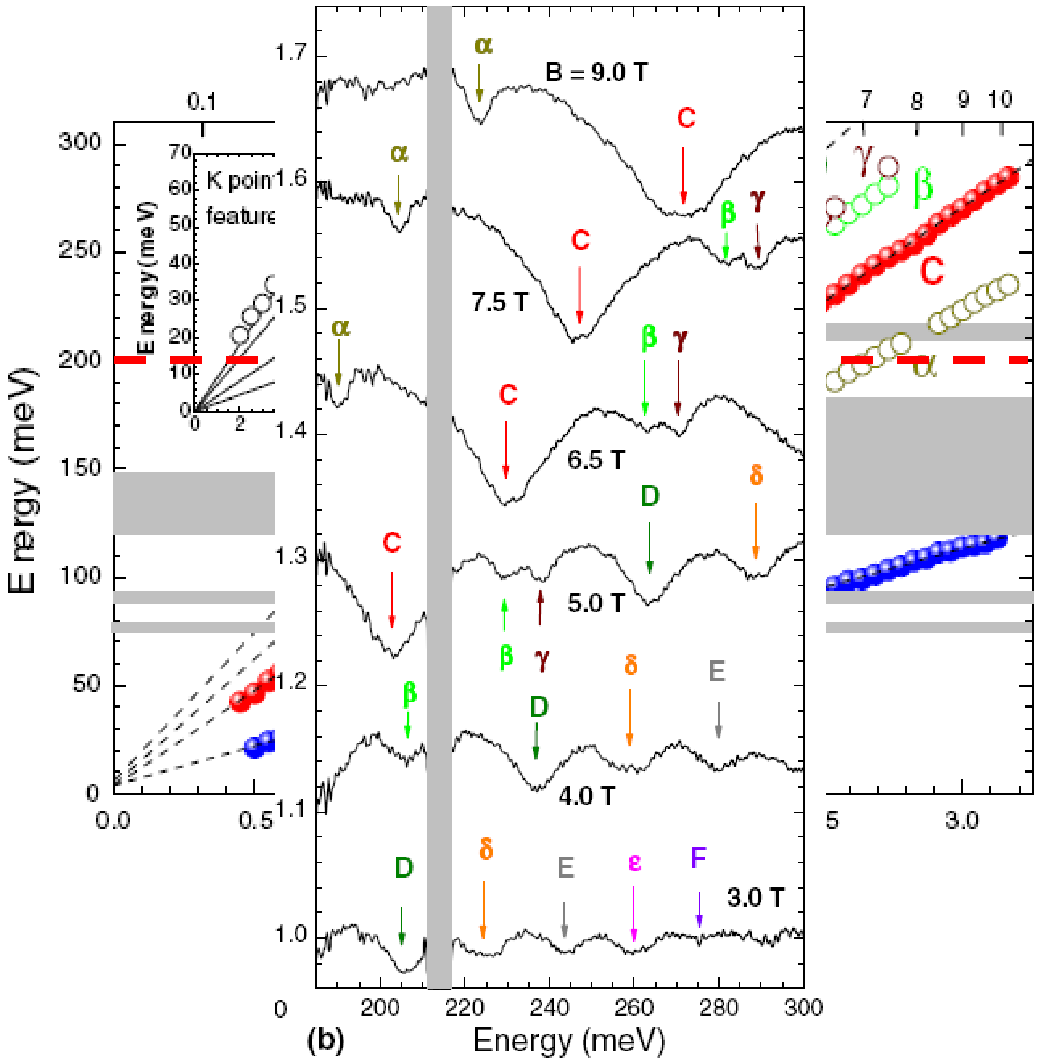
MPR in graphite: region I



The ‘marked’ MPR occurs at a resonance field B=5T

J. Yan, PhD dissertation, June 2009

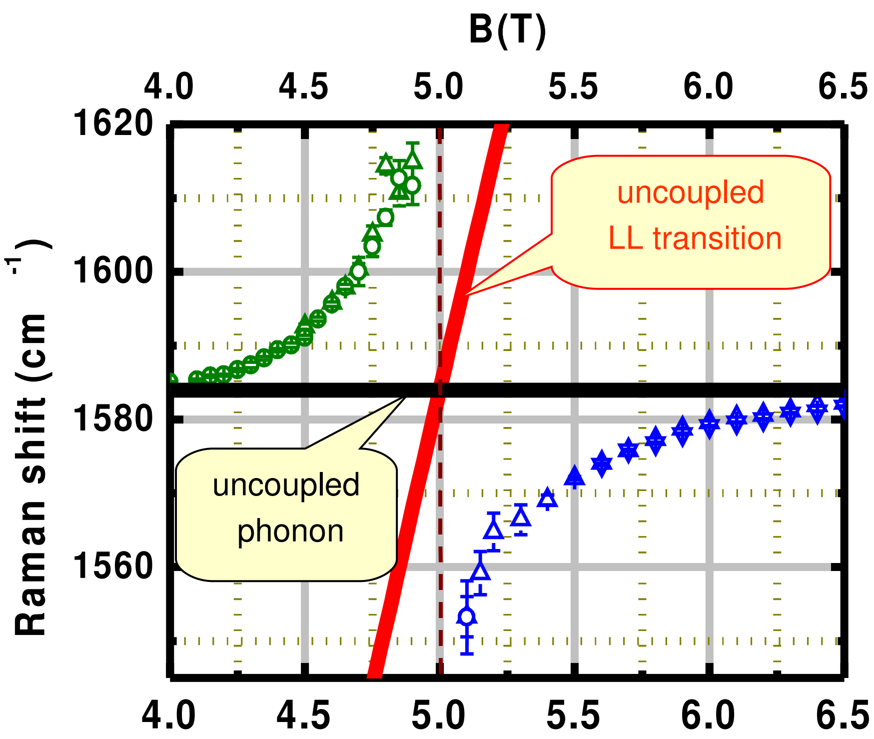
Dirac fermions in graphite



Cyclotron resonance: HOPG graphite

Orlita et al. PRL 100, 136403 (2008)

MPR in graphite: region I



The MPR in graphite is due to transitions of Dirac Fermions!

J. Yan, PhD dissertation, June 2009

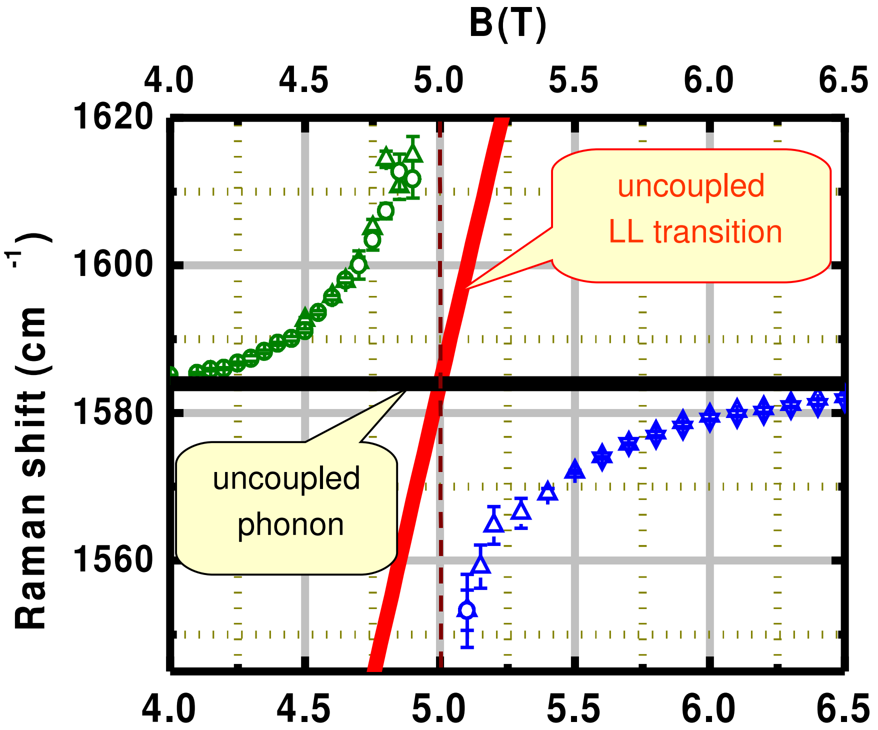
MPR in graphite: region I

coupled mode interpretation

'bare'
LL transition

$$\begin{bmatrix} E_{LL}+i\delta & g \\ g & \hbar\omega_G+i\eta \end{bmatrix}$$

'bare'
phonon



MPR in graphite: region I

coupled mode interpretation

'bare' LL transition

$$\begin{bmatrix} E_{LL}+i\delta & g \\ g & \hbar\omega_G+i\eta \end{bmatrix}$$

'bare' phonon

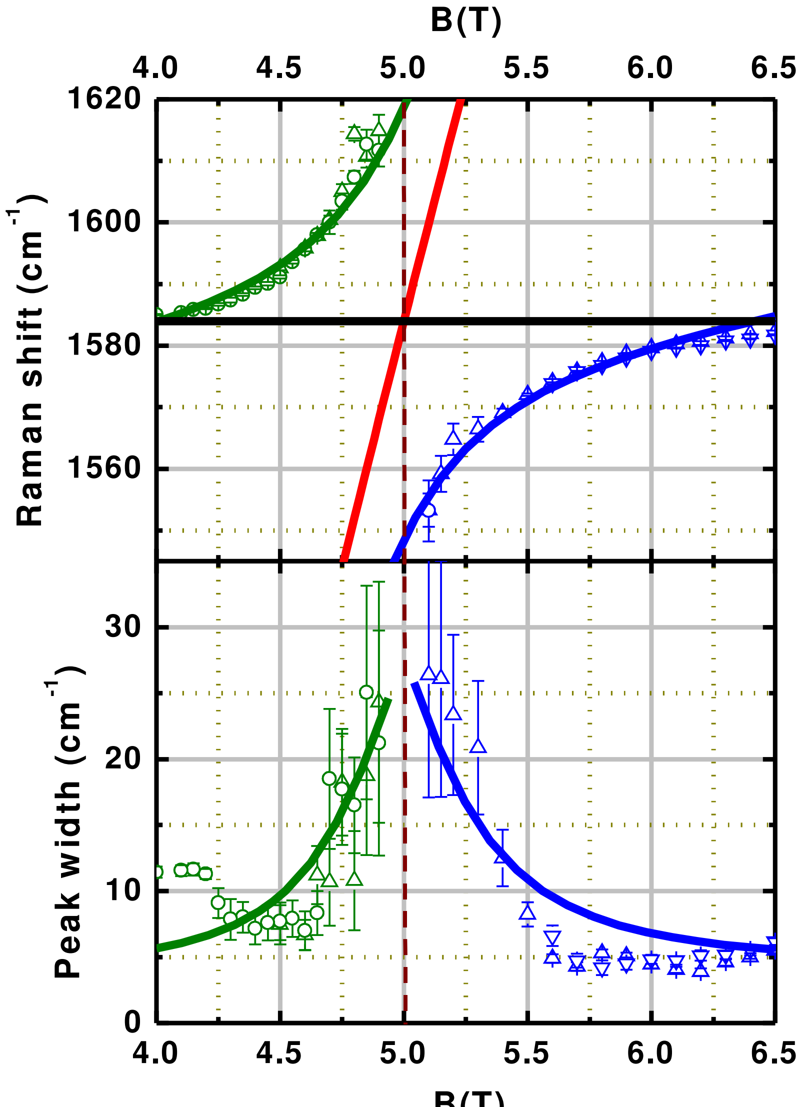
Fitting parameters

$$\delta = 26\,cm^{-1}$$

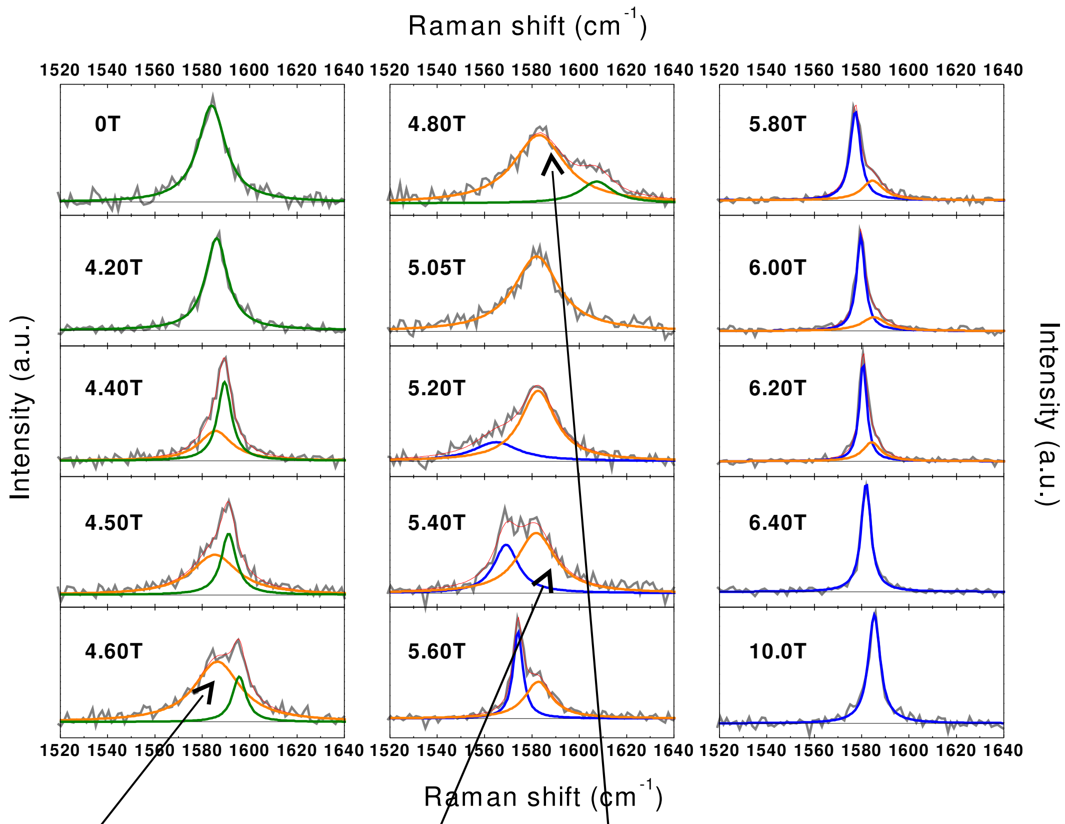
$$\eta = 2\,cm^{-1}$$

$$g = 37\,cm^{-1}$$

$$\frac{\partial t}{\partial l} = 6.7\,eV/\mathring{A}$$



MPR in graphite: region III?



There is an additional mode in the spectra!

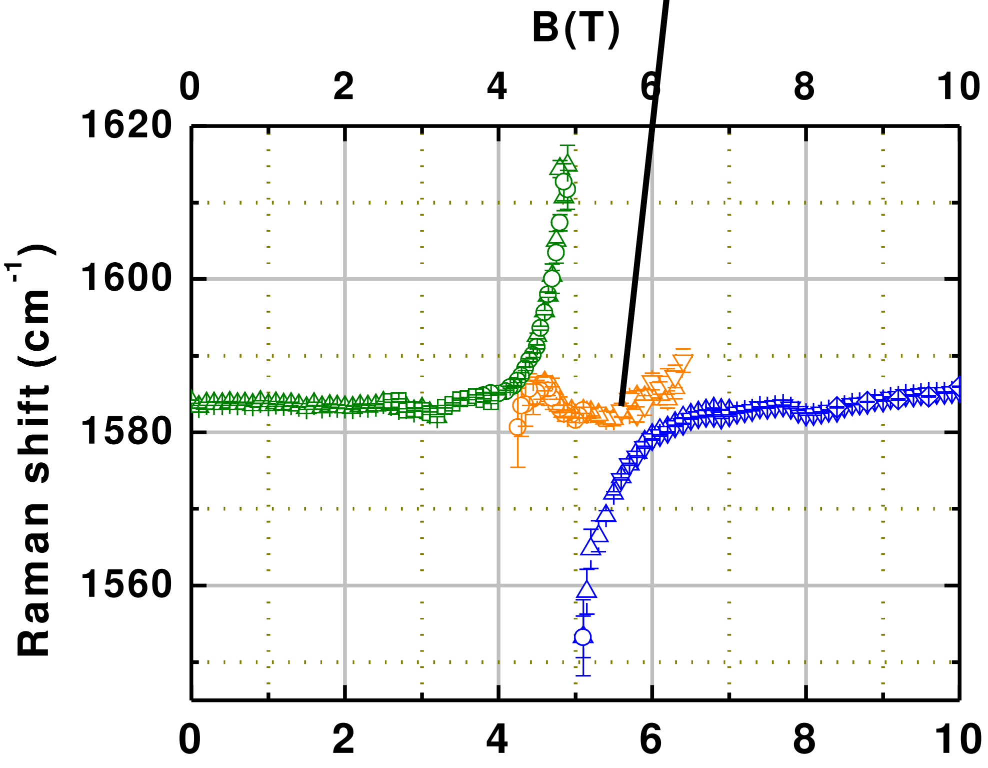
MPR in graphite: region III?

'bare'
LL transition

$$\begin{bmatrix} E_{LL}+i\delta & g \\ g & \hbar\omega_G+i\eta \end{bmatrix}$$

'bare'
phonon

mode from region III

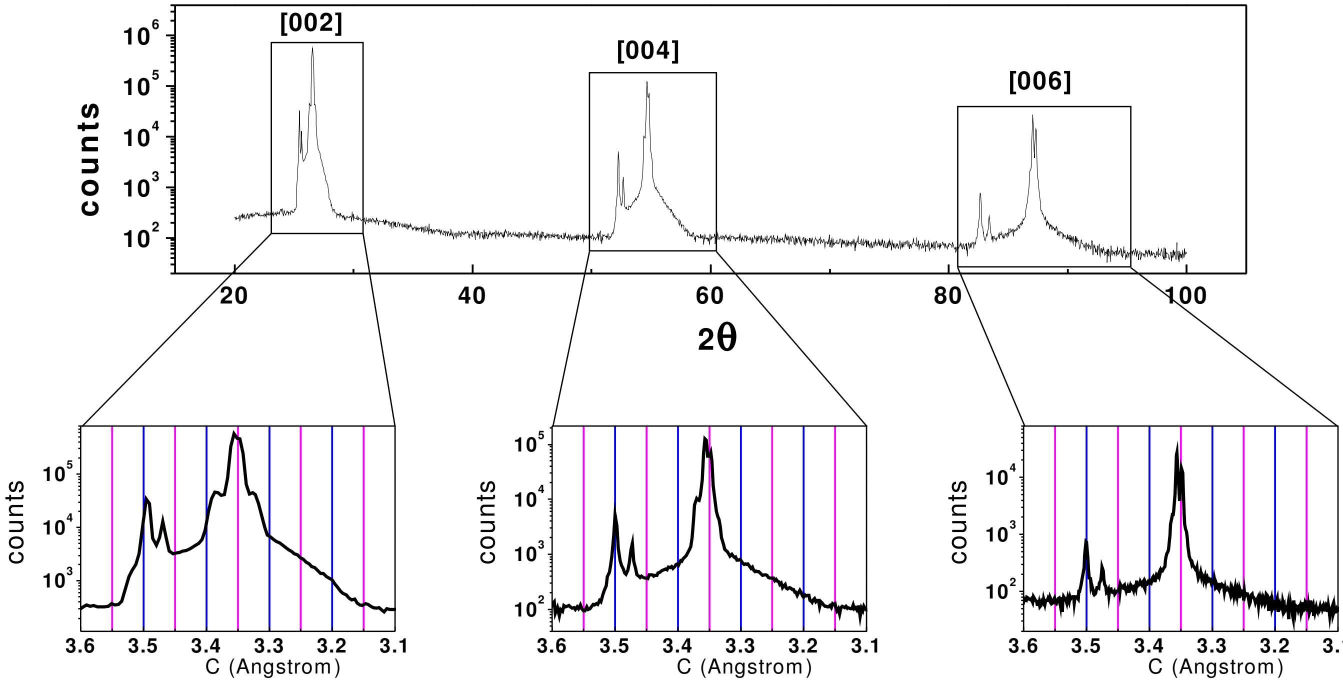


Region III needs to have g<<δ

Why several regions in Kish graphite ?

XRD of Kish graphite

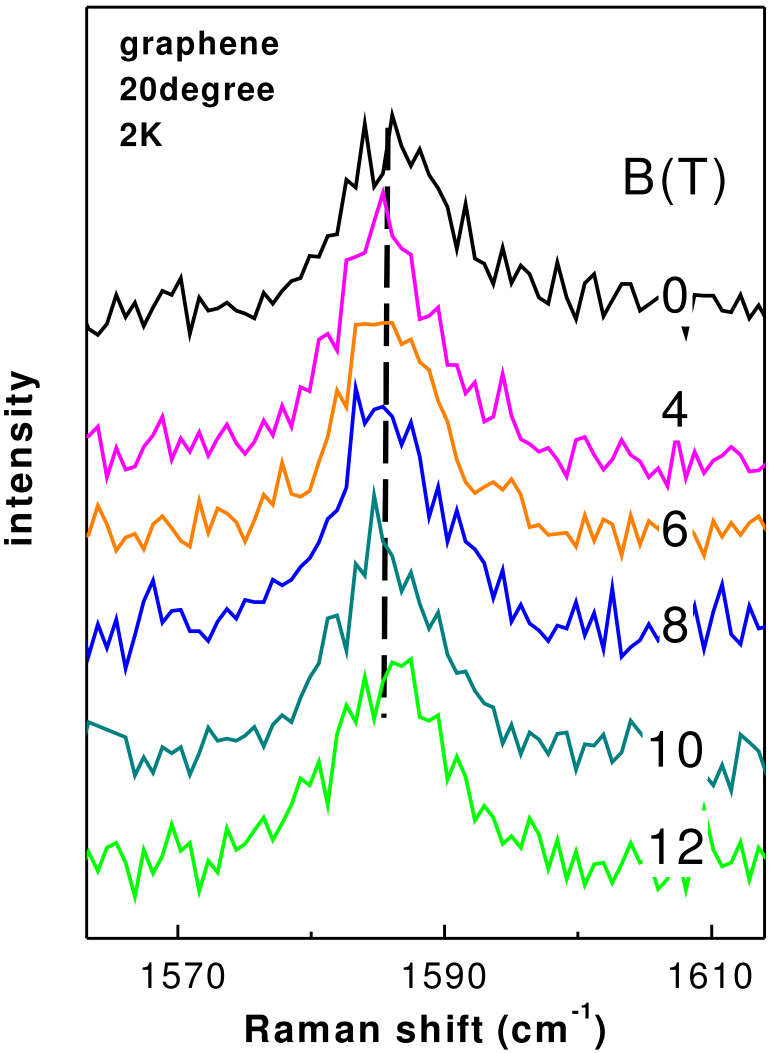
(with C. Noyan and S. Polvino)



Stacking with different inter-graphene plane distances:

Is this the reason for several regions in MPR response?

preliminary results of MPR in graphene



- Quite small MPR effects
- Impact of broadening of Landau levels due to large disorder?

J. Yan, PhD dissertation, June 2009

mode coupling in high magnetic fields

- Graphene

  * weak MPR (probably due to disorder)

- Graphite

  * Well-defined MPR with narrow Landau level transitions

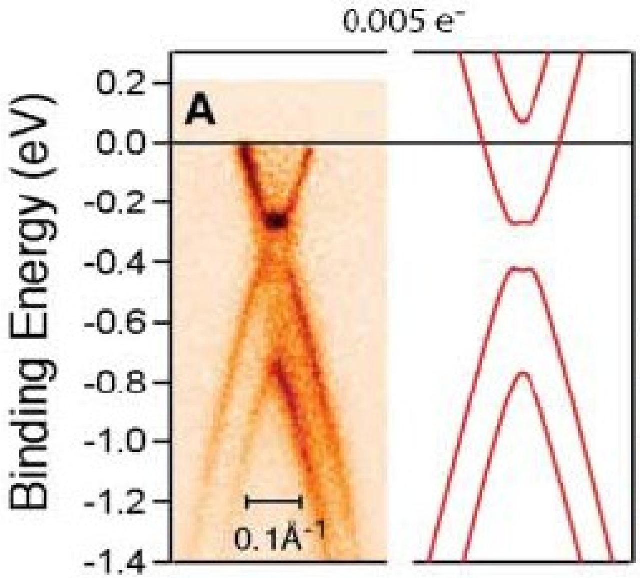
  * Several regions with different MPR response (inhomogeneous Kish graphite)

Broken-symmetry in graphene bi-layers

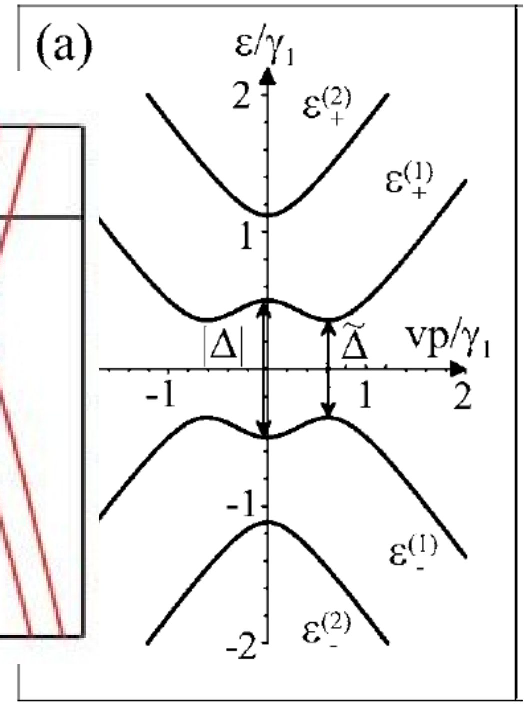
when the two layers are not equivalent

A gap opens when
between
on-site ene

$\Delta = \varepsilon_2 - \varepsilon$



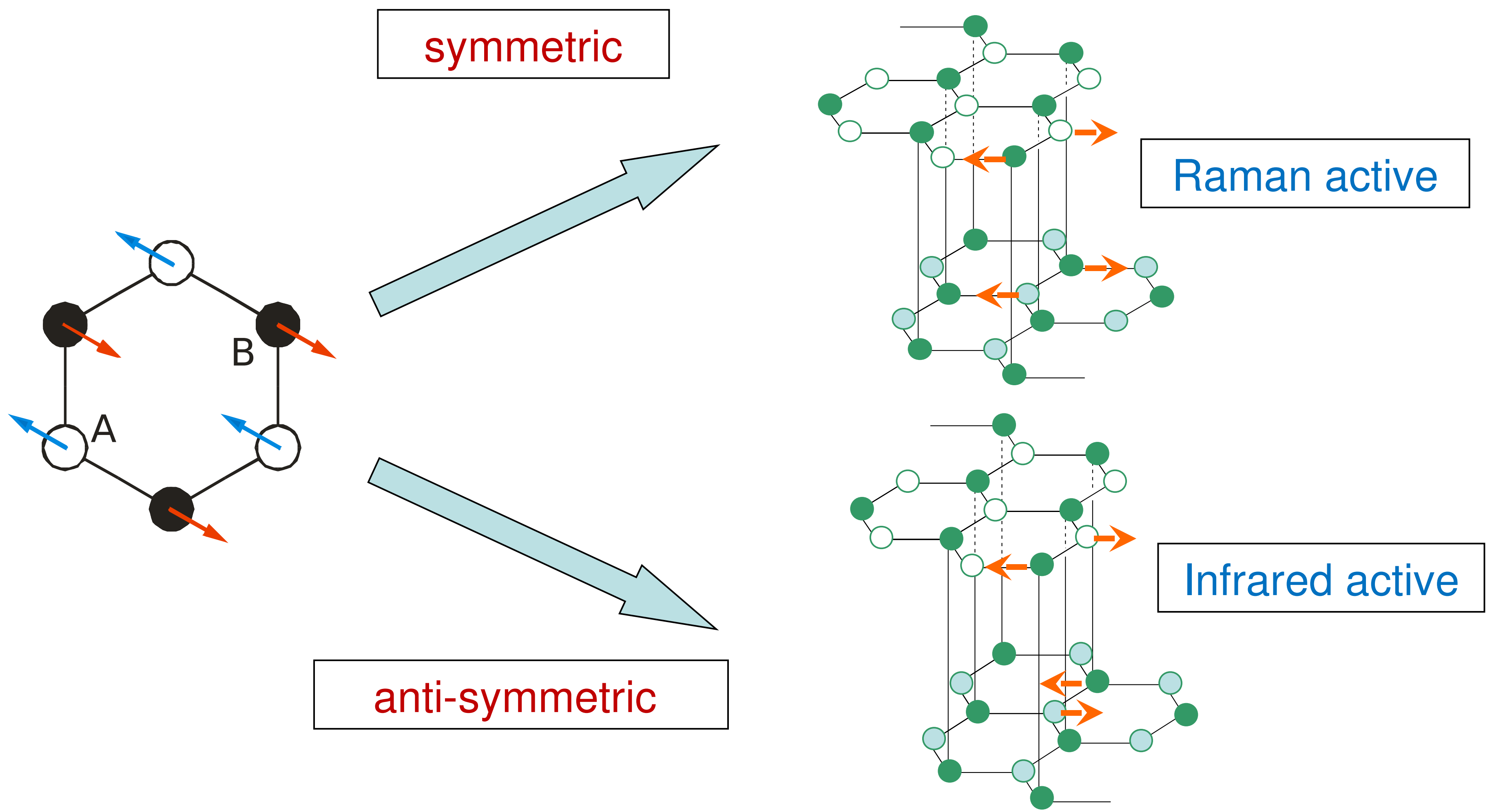
Ohta et al. Science (2006)



E. McCann, PRB 74,161403 (2006)

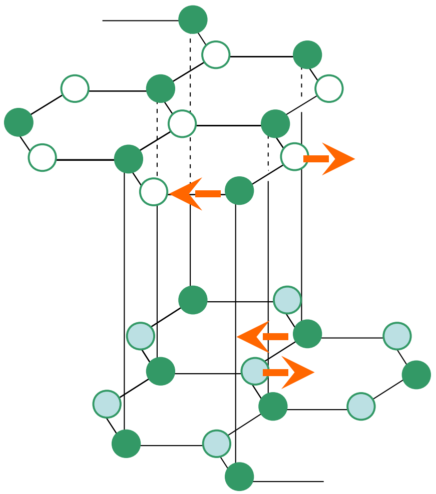
Symmetric graphene bi-layers

G-band optical phonons

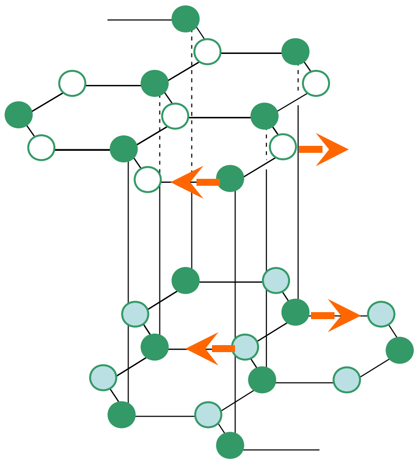


Broken-symmetry in graphene bi-layers

when the two layers are not equivalent



coupled



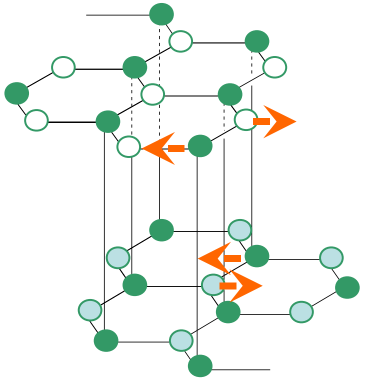
anti-symmetric
(odd-parity)

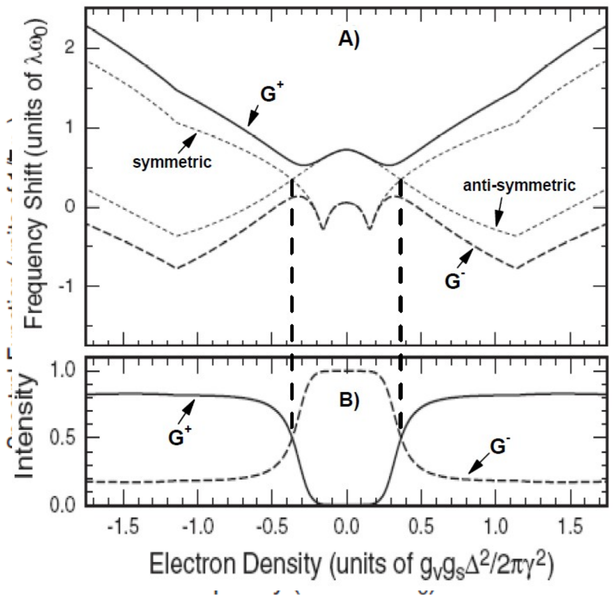
symmetric
(even-parity)

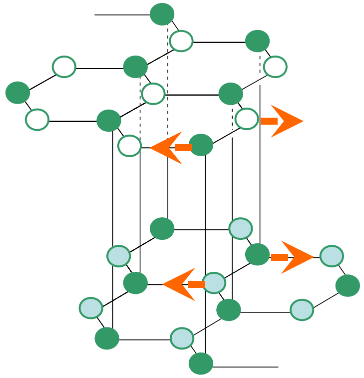
'entanglement' of even- and odd-parity phonons

Broken-symmetry in graphene bi-layers

when the two layers are not equivalent







anti-symmetric
(odd-parity)

coupled

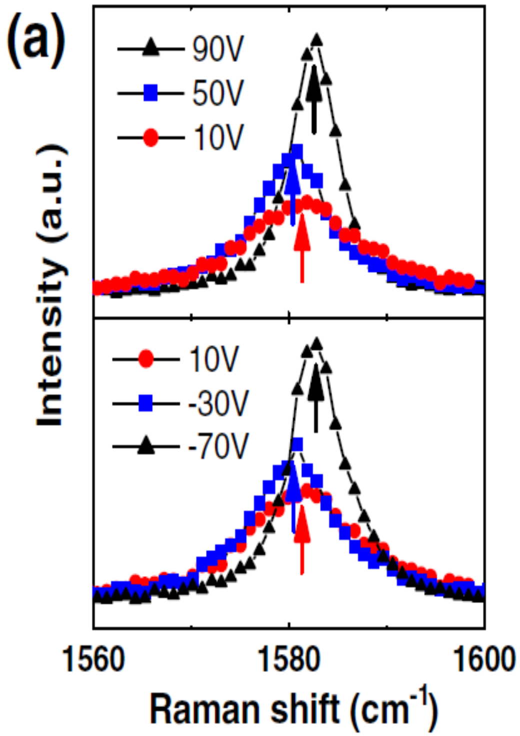
symmetric
(even-parity)

T. Ando and M. Koshino, JSPS 78, 034709 (2009)
P. Gava et al, preprint, July 2009.

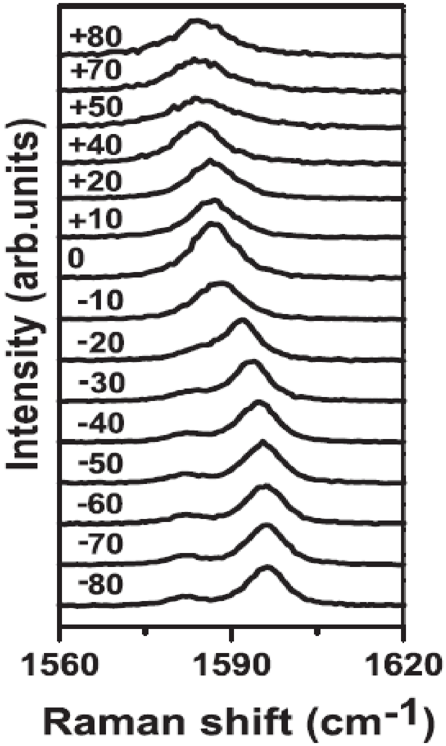
coupled G^+ and G^- modes should appear in Raman spectra

gated graphene bi-layers

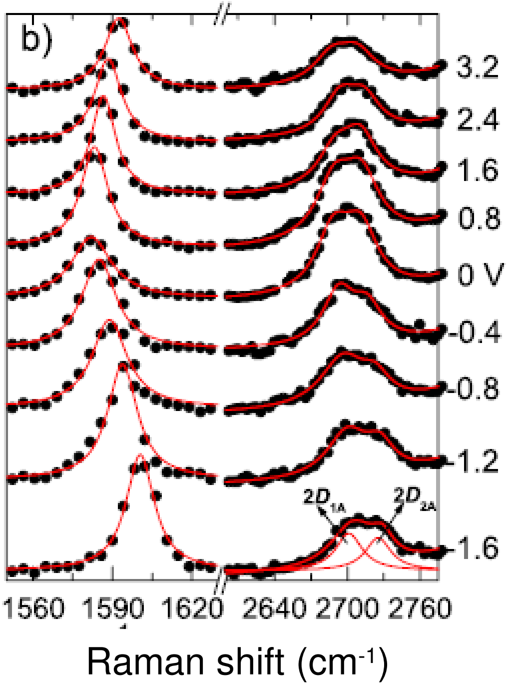
recent Raman results



Jun Yan et al
PRL (2008)



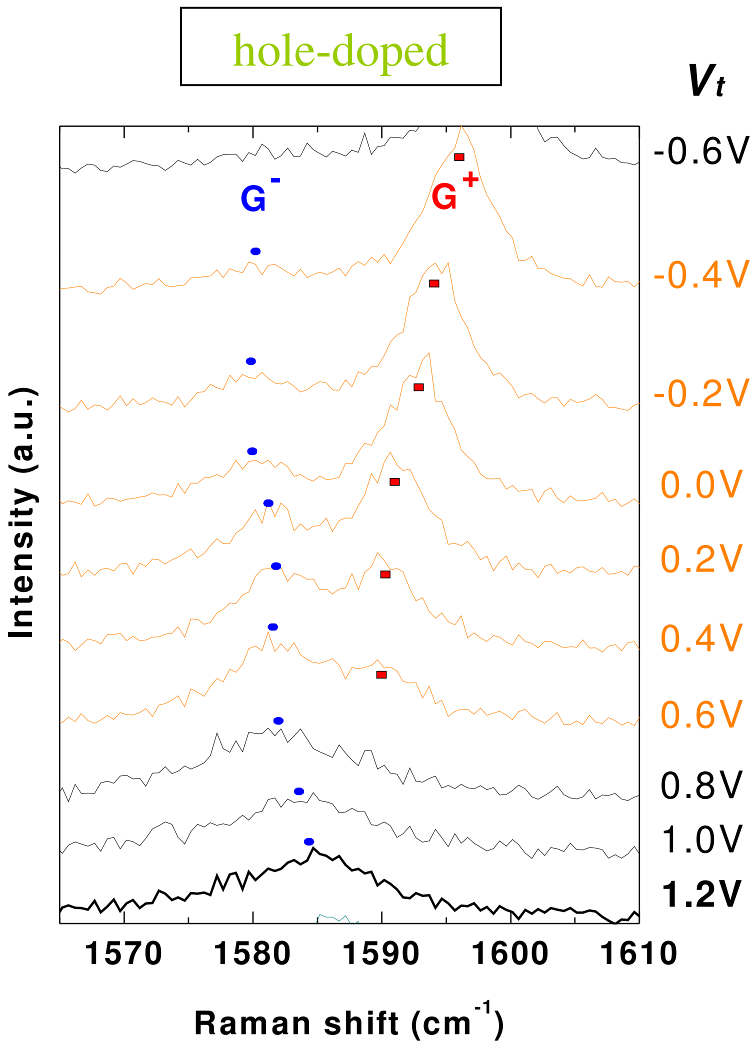
L.M. Malard et al
PRL (2008)

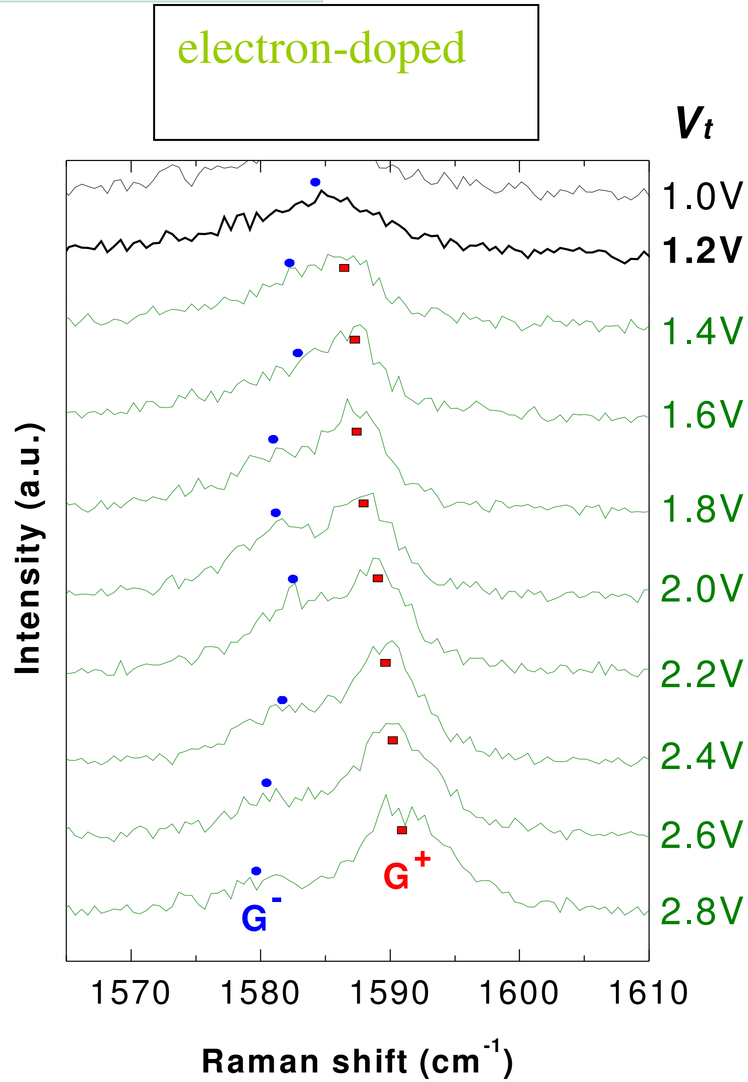


A. Das et al
PRB (2009)

gated graphene bi-layers

more recent Raman results





Jun Yan, PhD dissertation, June 2009

Broken-symmetry in graphene bi-layers

- Coupled G+/G- modes observed

- Questions

  * quantitative links of G^+/G^- splitting with gap opening

  * splitting is not observed in some Raman experiments. Why?