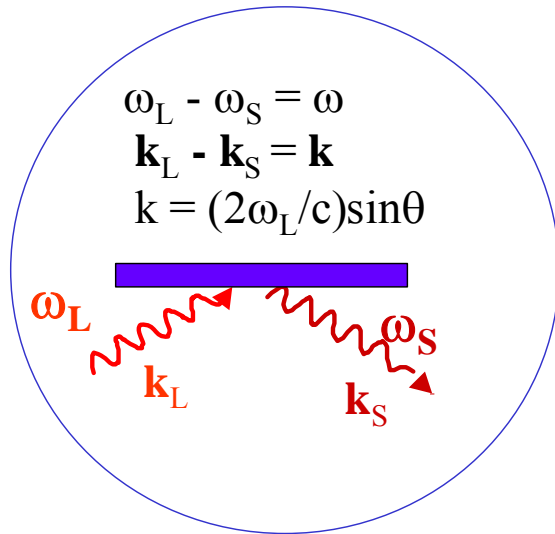


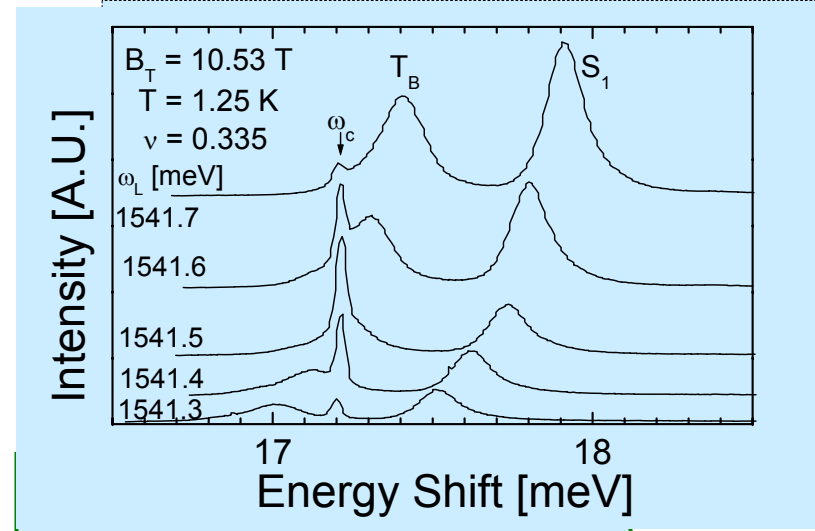
# Spectroscopy of Electrons and Phonons in Graphene Structures

## Inelastic Light Scattering

elementary excitation modes are seen directly in the spectra



2D electron gas in a GaAs quantum well



# Spectroscopy of Electrons and Phonons in Graphene Structures

Why inelastic light scattering studies in graphene?

Raman scattering (optical phonons):  
insights on physics of carbon-based structures

Inelastic light scattering (electrons and holes):  
spin or charge excitations of Dirac fermions

# Spectroscopy of Electrons and Phonons in Graphene Structures

## Recent Raman studies of single-layer graphene:

- Electric-field effect
- Electron-phonon coupling

## Future light scattering studies in graphene:

- Dirac fermion excitations
- Graphene hetero-layers

# Spectroscopy of Electrons in Low-Dimensional Structures

## Support from

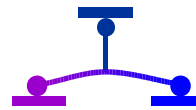
W. M. Keck Foundation

Office of Naval Research

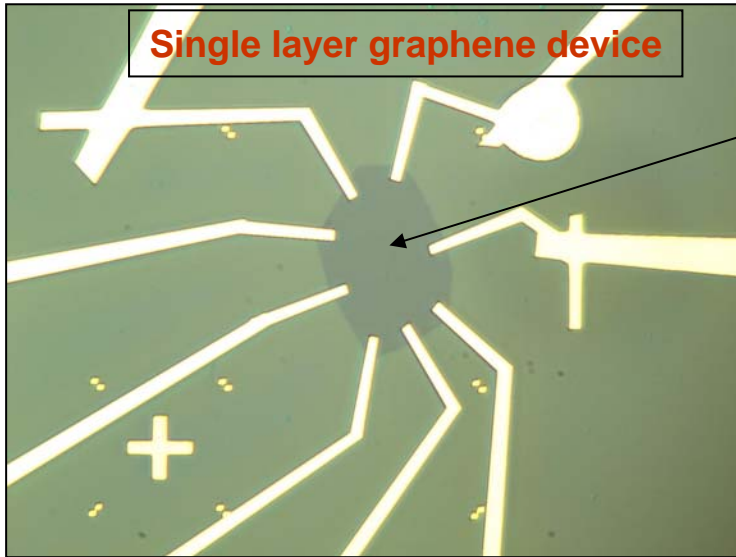
Columbia NSEC (NSF, NYSTAR)

National Science Foundation  
(DMR 03-52738)

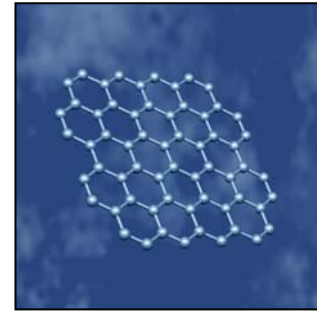
Department of Energy  
(DE-AIO2-04ER46133 )



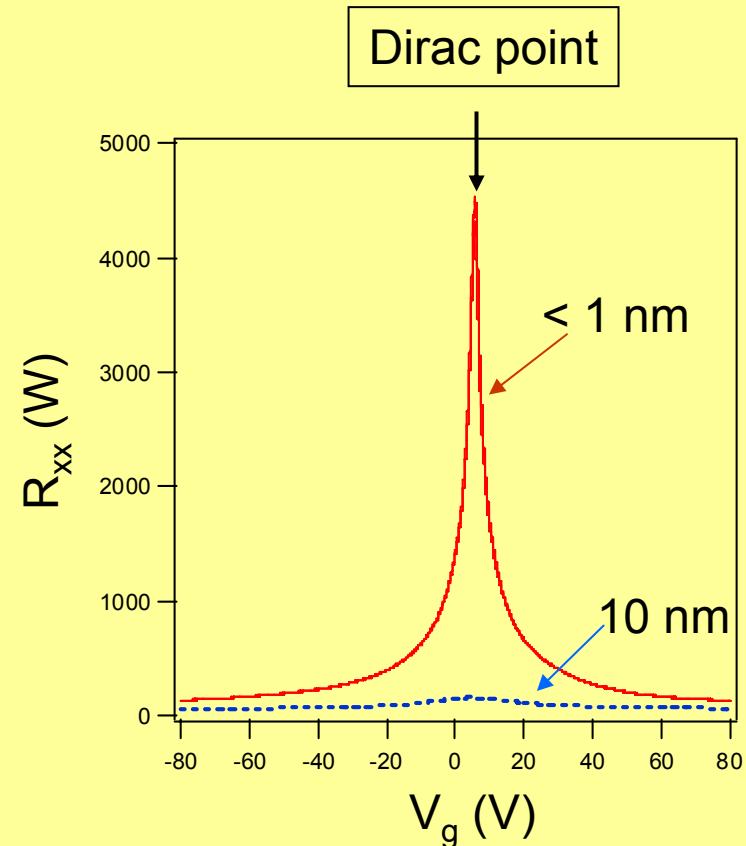
# Electric-Field-Effect in Graphene



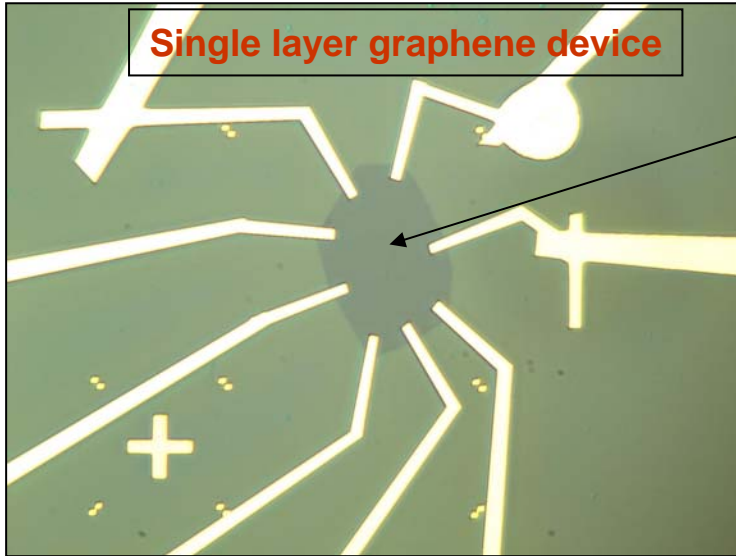
Single atomic layer of  
carbons  
in a honeycomb lattice!



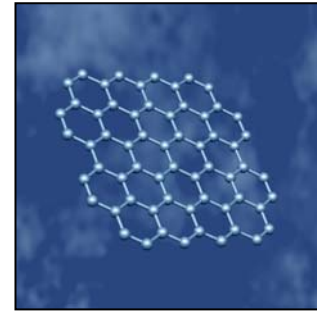
K.S. Novoselov et al Science **306**, 666 (2004)  
Yuanbo Zhang et al Phys. Rev. Lett **94**, 176803 (2005)  
K.S. Novoselov et al Nature **438**, 197 (2005)  
Yuanbo Zhang et al Nature **438**, 201 (2005)



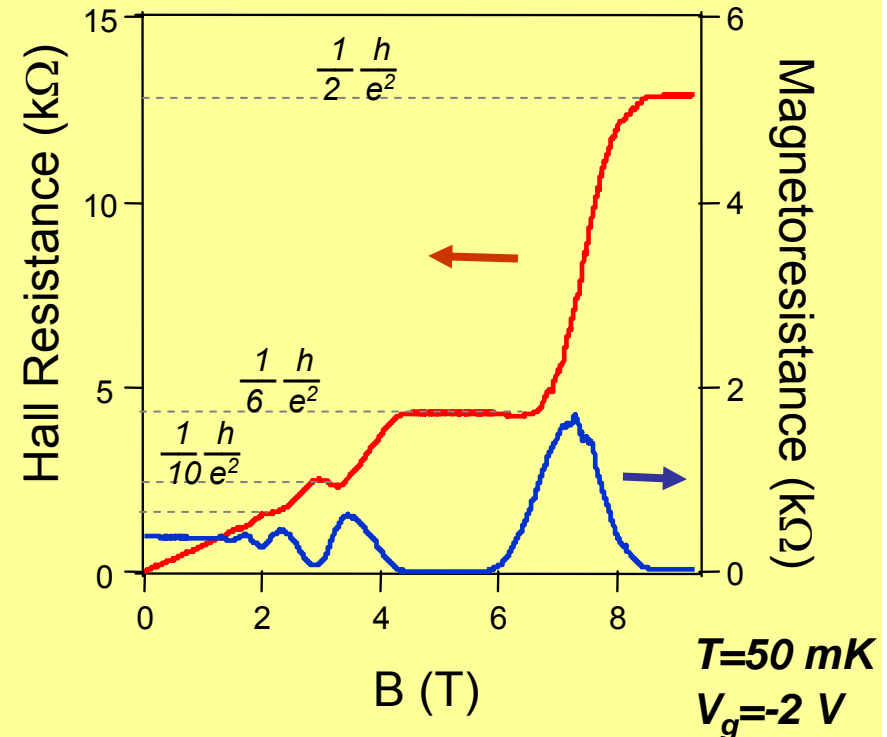
# Electric-Field-Effect in Graphene



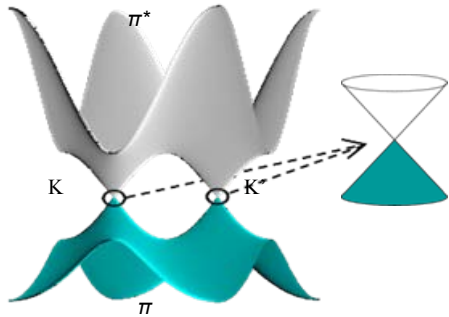
Single atomic layer of  
carbons  
in a honeycomb lattice!



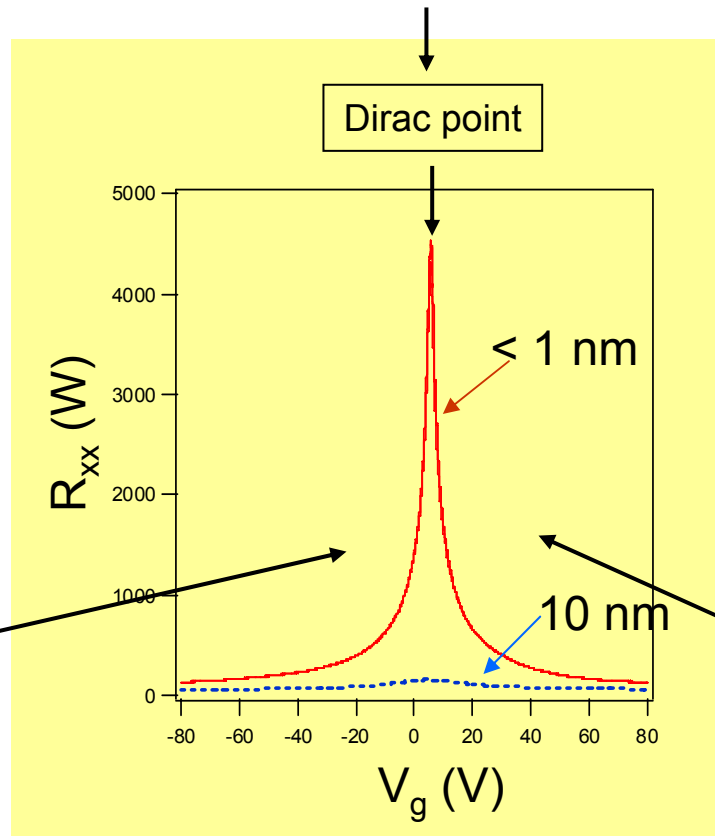
- K.S. Novoselov et al Science **306**, 666 (2004)  
Yuanbo Zhang et al Phys. Rev. Lett **94**, 176803 (2005)  
K.S. Novoselov et al Nature **438**, 197 (2005)  
Yuanbo Zhang et al Nature **438**, 201 (2005)



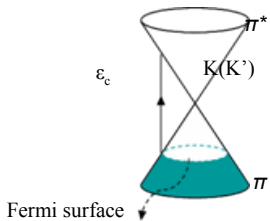
# Electric-Field-Effect in Graphene



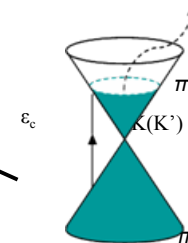
Massless Dirac Fermions



holes



particles



# Spectroscopy of Electrons and Phonons in Graphene Structures

## Raman Scattering

- Electric-field effect in graphene
- Electron-phonon coupling

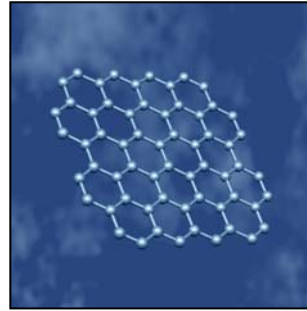
## Inelastic Light Scattering

- Dirac fermion excitations
- Graphene hetero-layers

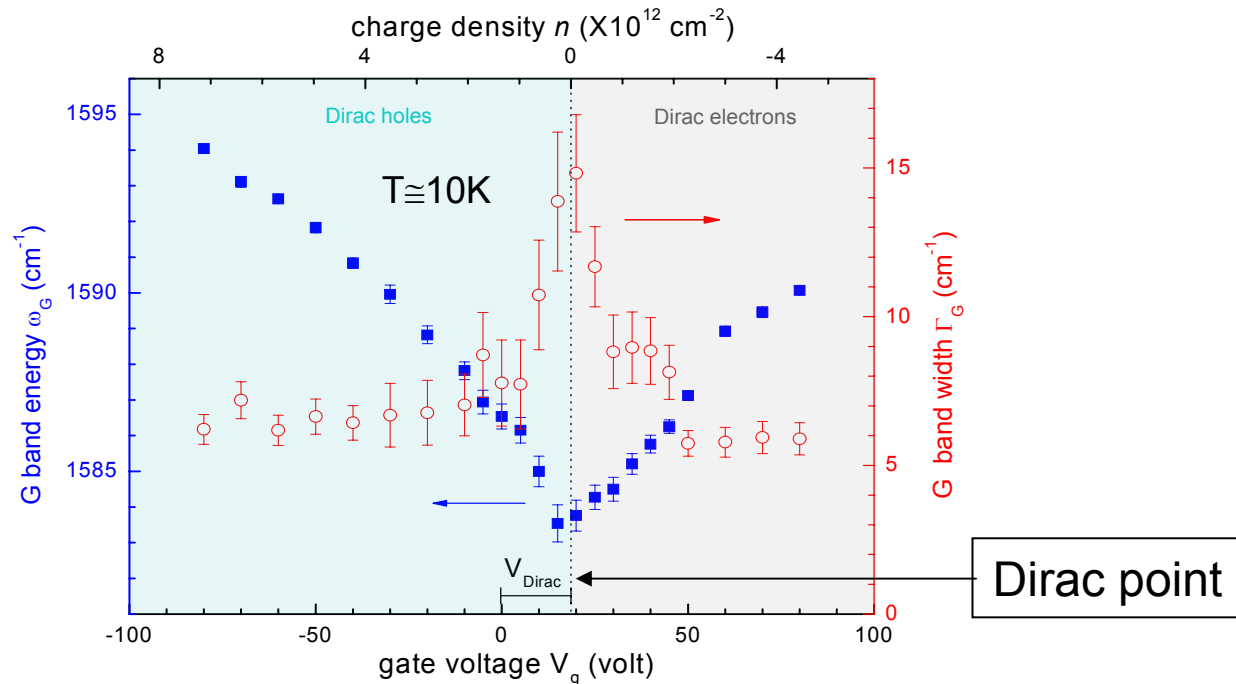
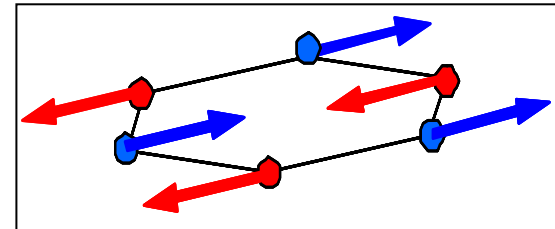


# Electric-Field-Effect in Single Layer Graphene: Electron-phonon Coupling

Single atomic layer of carbons in a honeycomb lattice!



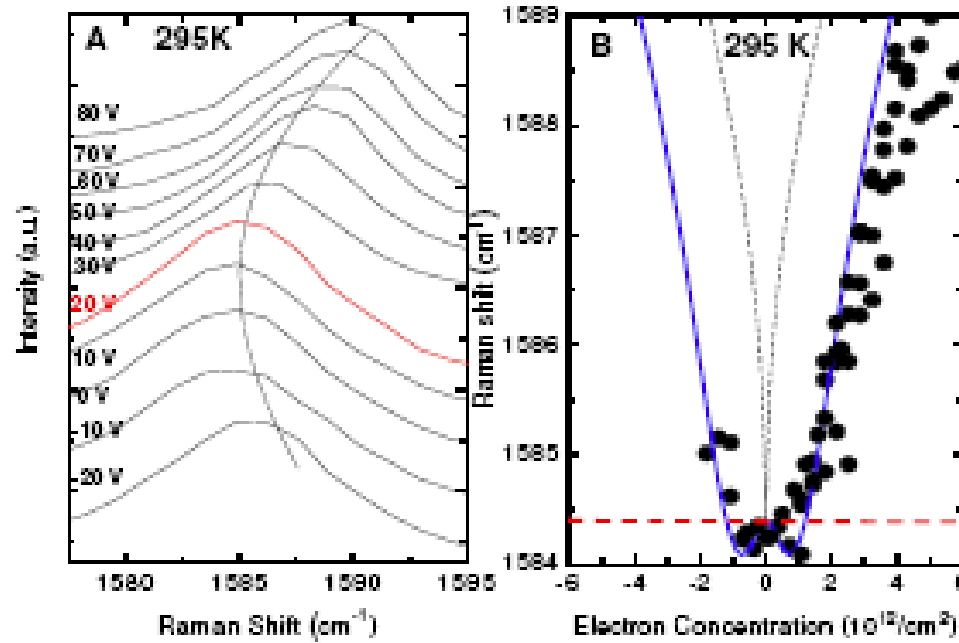
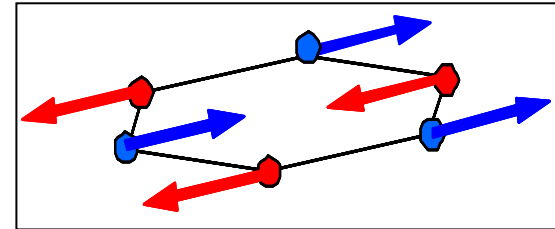
G band vibration of graphene



Jun Yan et al submitted (October 2006)

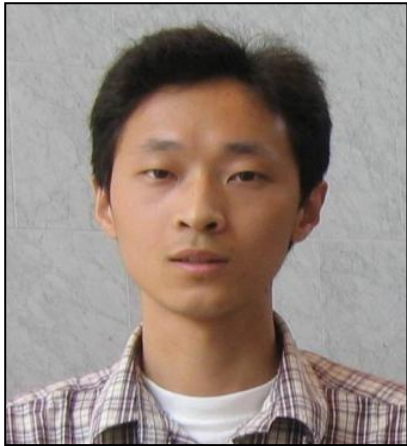
# Electric-Field-Effect in Single Layer Graphene: Electron-phonon Coupling

G band vibration of graphene



S. Pisana et al, submitted

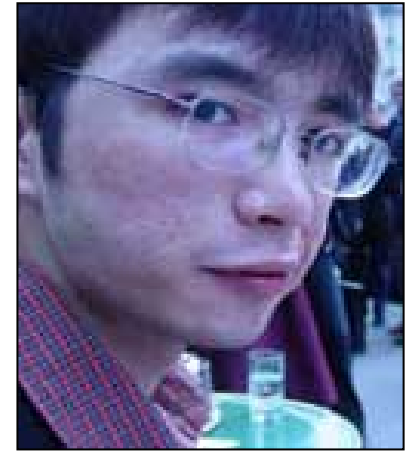
# Picture Gallery



Jun Yan



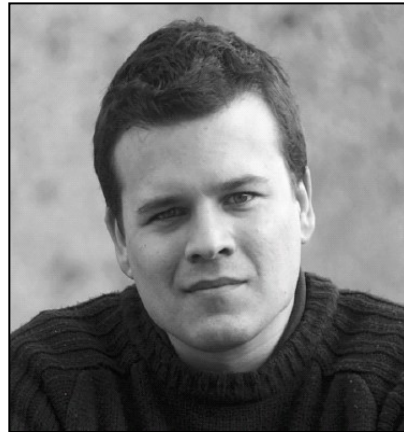
Philip Kim



Yuanbo Zhang



Melinda Han



Pablo Jarrillo-Herrero

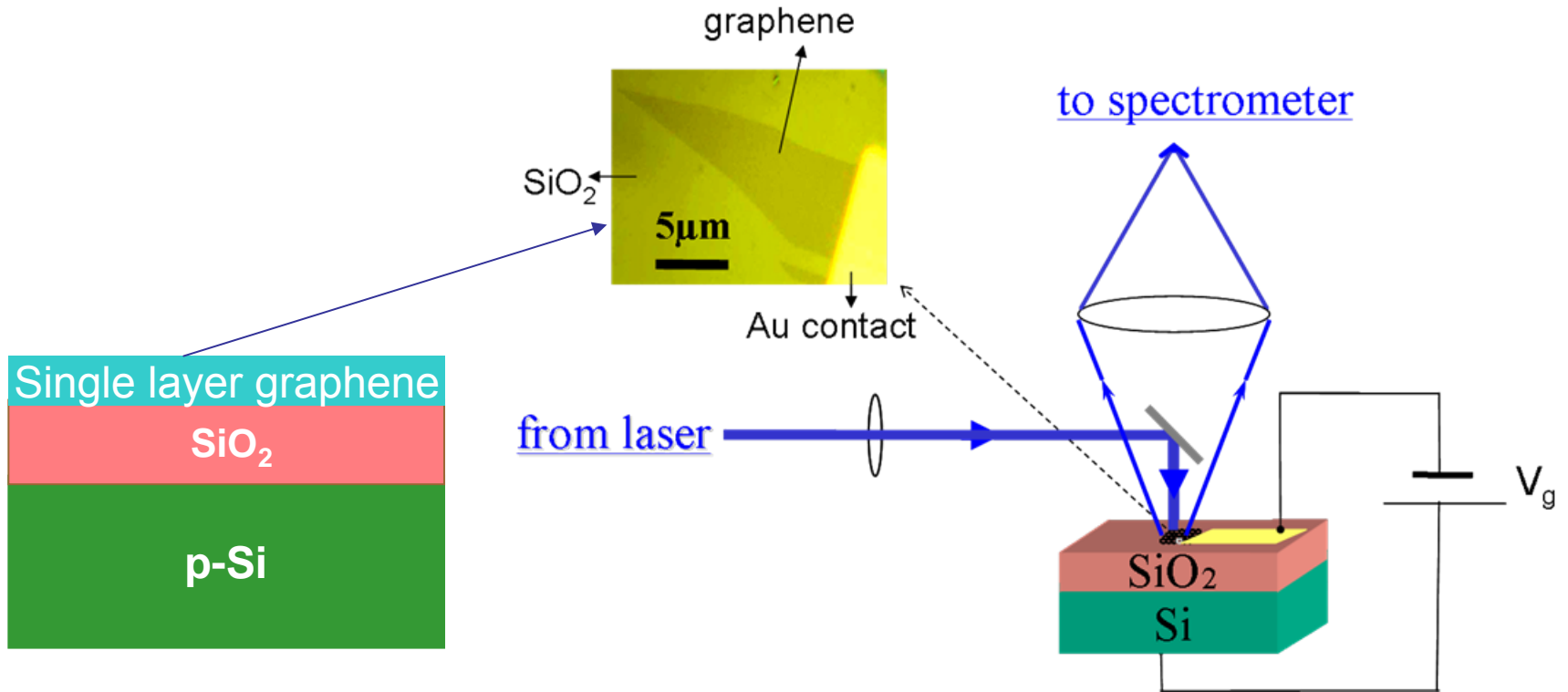


Kirill Bolotin



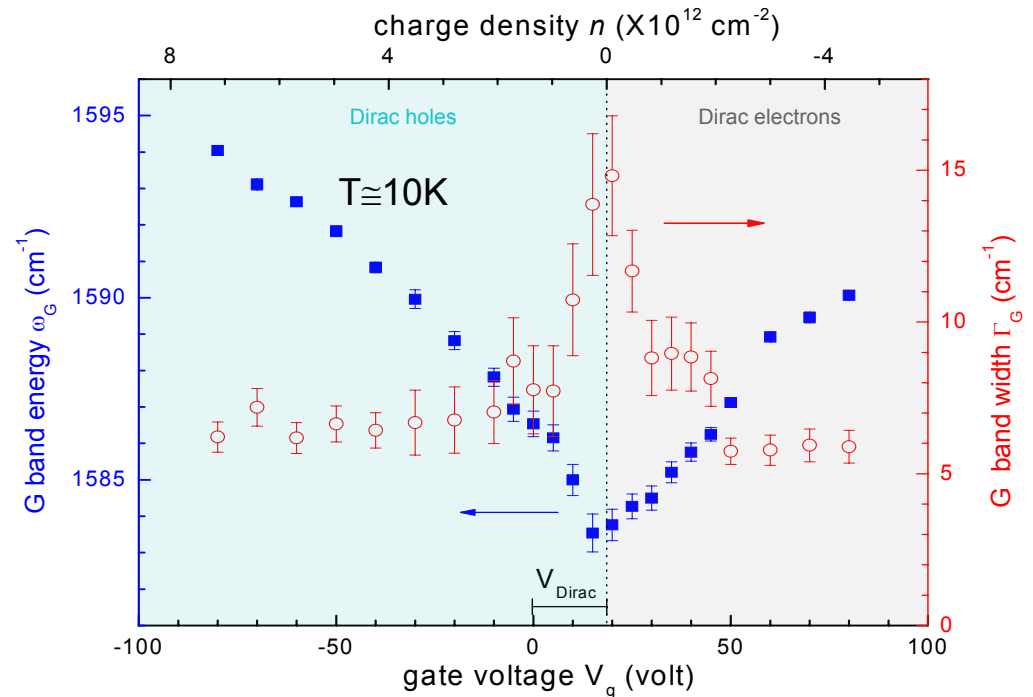
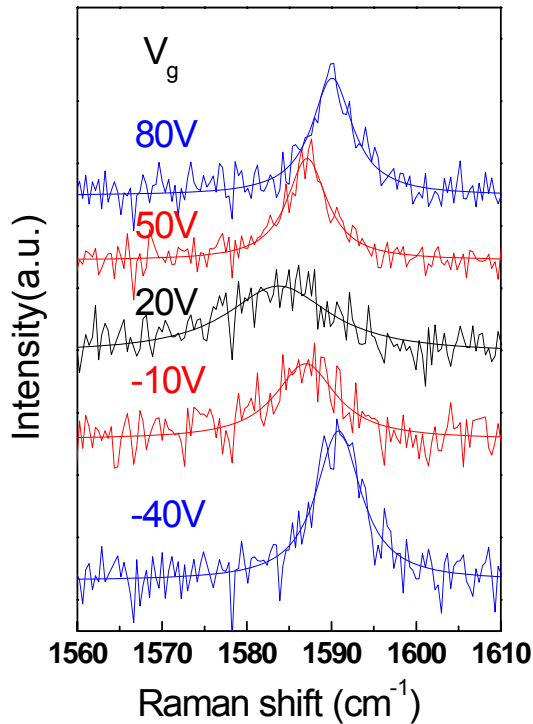
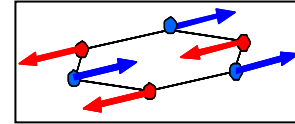
Erik Henriksen

# Raman Scattering in Gated Single Layer Graphene



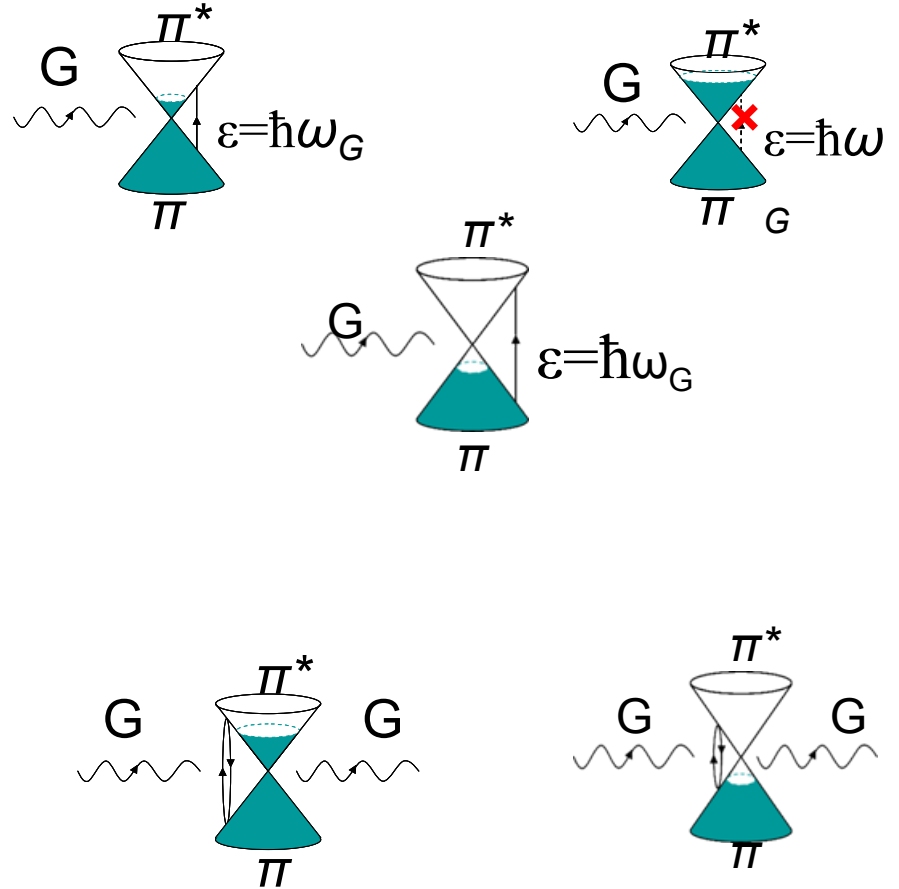
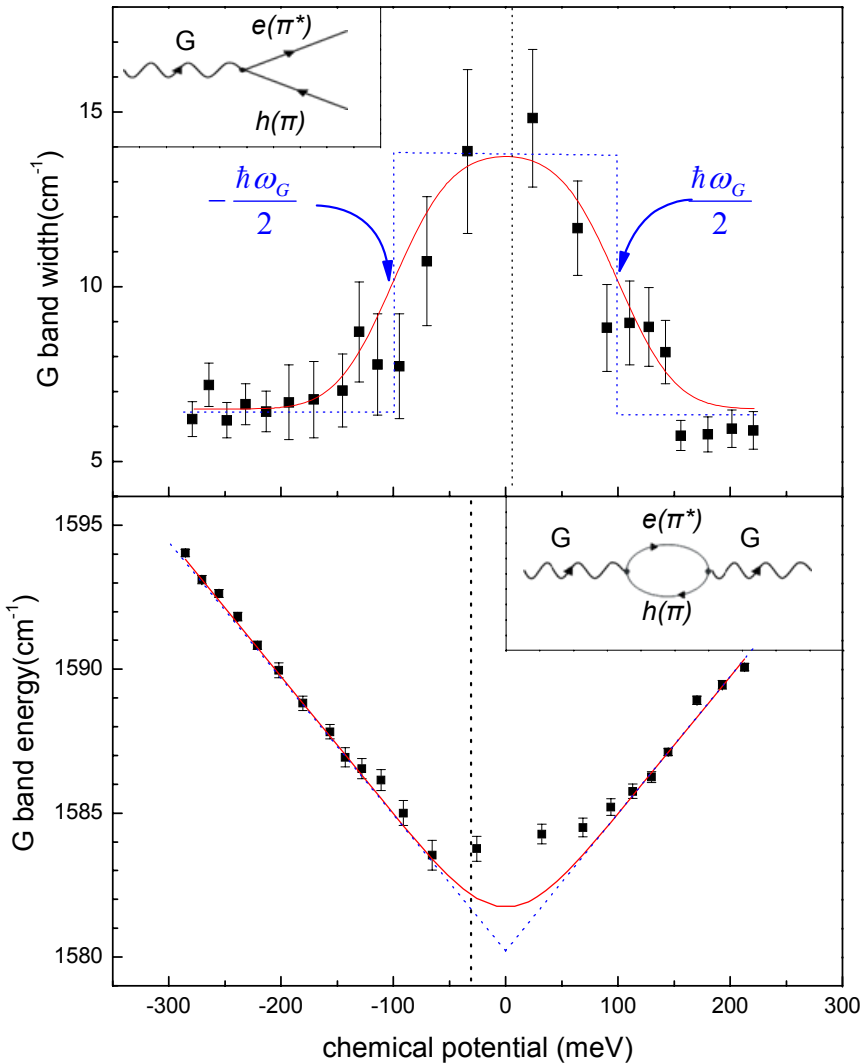
# Raman Scattering in Gated Single Layer Graphene

G band vibration of graphene

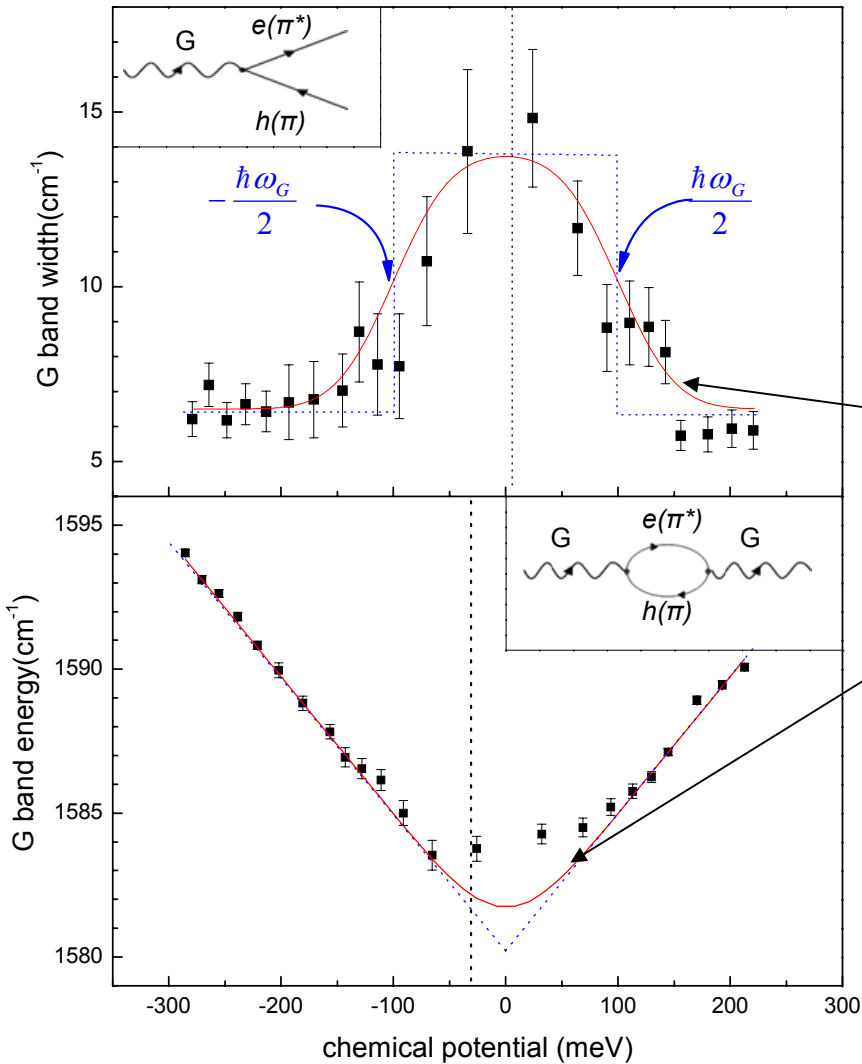


Jun Yan, Yuanbo Zhang, P. K, and A.P., submitted

# Electron-Phonon Coupling



# Electron-Phonon Coupling



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

**Non-uniformity**

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

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

# Electric-Field-Effect in Graphene: Evaluation of Electron-phonon Coupling

## Electron-Phonon Coupling and Raman Spectroscopy in Graphene

A. H. Castro Neto

*Department of Physics, Boston University, 590 Commonwealth Avenue, Boston, MA 02215, USA*

Francisco Guinea

*Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco E28049 Madrid, Spain*

We show that the electron-phonon coupling in graphene, in contrast with the non-relativistic two-dimensional electron gas, leads to shifts in the phonon frequencies that are non-trivial functions of the electronic density. These shifts can be measured directly in Raman spectroscopy. We show that depending whether the chemical potential is smaller (larger) than half of the phonon frequency, the frequency shift can be negative (positive) relative to the neutral case (when the chemical potential is at the Dirac point), respectively. We show that the use of the static response function to calculate these shifts is incorrect and leads always to phonon softening. In samples with many layers, we find a shift proportional to the carrier concentration, and a splitting of the phonon frequencies if the charge is not homogeneously distributed. We also discuss the effects of edges in the problem.



# Electric-Field-Effect in Graphene: Evaluation of Electron-phonon Coupling

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Vol. 75, No. 12, December, 2006, 124701  
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## Anomaly of Optical Phonon in Monolayer Graphene

Tsuneya ANDO

*Department of Physics, Tokyo Institute of Technology,  
2-12-1 Ookayama, Meguro-ku, Tokyo 152-8551*

(Received August 17, 2006; accepted October 3, 2006; published November 27, 2006)

The frequency shift and broadening of long-wavelength optical phonons due to interactions with electrons are calculated in a monolayer graphene as a function of the electron density. The broadening disappears and the frequency shift exhibits a logarithmic singularity when the Fermi energy is half of the energy of the optical phonon. The shift increases in proportion to the Fermi energy for sufficiently high electron density.

# Electric-Field-Effect in Graphene: Evaluation of Electron-phonon Coupling

## Non-adiabatic Kohn-anomaly in a doped graphene monolayer

Michele Lazzeri and Francesco Mauri

*IMPMC, Universités Paris 6 et 7, CNRS, IPGP, 140 rue de Lourmel, 75015 Paris, France*

(Dated: November 28, 2006)

We compute, from first-principles, the frequency of the  $E_{2g}$ ,  $\Gamma$  phonon (Raman  $G$  band) of graphene, as a function of the charge doping. Calculations are done using i) the *adiabatic* Born-Oppenheimer approximation and ii) time-dependent perturbation theory to explore *dynamic* effects beyond this approximation. The two approaches provide very different results. While, the *adiabatic* phonon frequency weakly depends on the doping, the *dynamic* one rapidly varies because of a Kohn anomaly. The *adiabatic* approximation is considered valid in most materials. Here, we show that doped graphene is a spectacular example where this approximation miserably fails.

# Spectroscopy of Electrons and Phonons in Graphene Structures

## Raman Scattering

- Electric-field effect in graphene
- Electron-phonon coupling

## Inelastic Light Scattering

- Dirac fermion excitations
- Graphene hetero-layers

# Spectroscopy of Electrons and Phonons in Graphene Structures

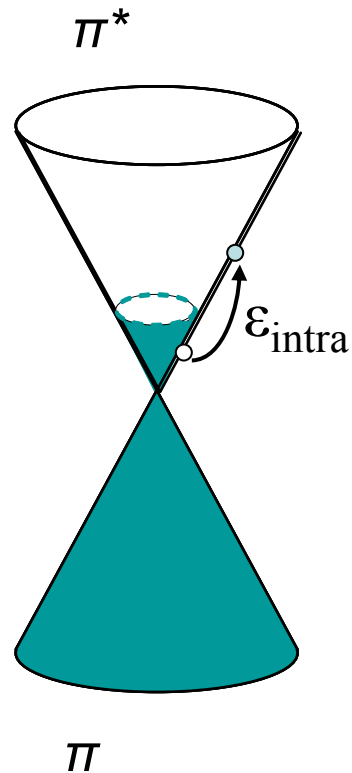
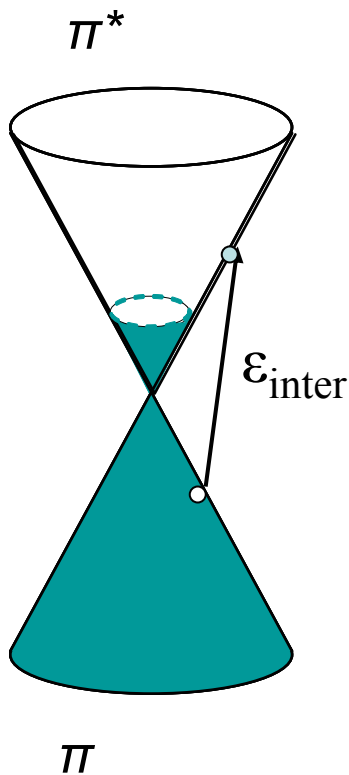
## Inelastic Light Scattering

### Studies of Dirac fermion excitations

- \* Collective and single particle modes:
  - \* plasmons, intraband, interband, etc
- \* Excitations of quantum Hall fluids
  - \* Landau level excitations (CR in absorption)
  - \* spin waves
  - \* magnetorotons

# Dirac Fermions: particle-hole excitations

n-type

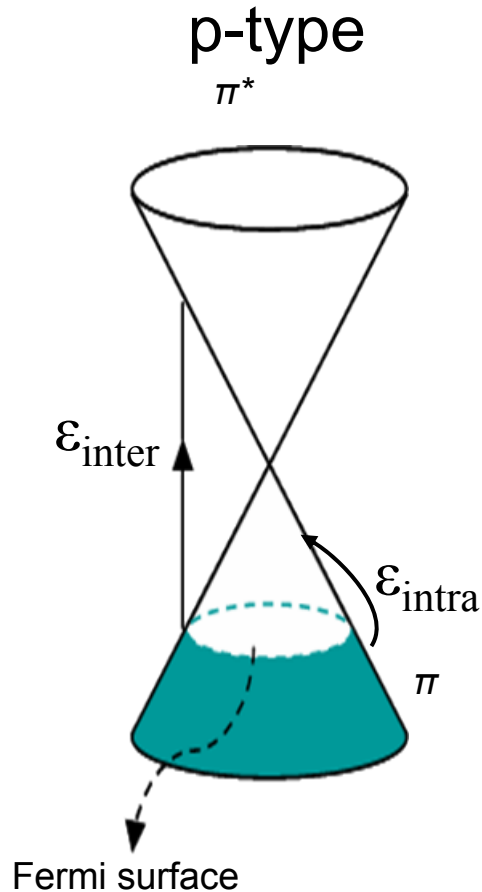


$\pi \rightarrow \pi^*$  transitions  
are allowed  
(*interband excitations*)

$\pi^* \rightarrow \pi^*$  transitions  
are allowed  
(intraband excitations)

$\pi \rightarrow \pi$  transitions  
are forbidden  
(intraband excitations)

# Dirac Fermions: particle-hole excitations



$\pi \rightarrow \pi^*$  transitions  
are allowed  
(*interband excitations*)

$\pi \rightarrow \pi$  transitions  
are allowed  
(intraband excitations)

$\pi^* \rightarrow \pi^*$  transitions  
do not exist  
(intraband excitations)

# Excitations of Dirac Fermions: RPA Theory

## Dynamical polarization of graphene at finite doping

B. Wunsch<sup>1,2</sup>, T. Stauber<sup>2</sup>, F. Sols<sup>1</sup>, and F. Guinea<sup>2</sup>

<sup>1</sup> *Departamento de Física de Materiales, Facultad de Ciencias Físicas,  
Universidad Complutense de Madrid, E-28040 Madrid, Spain.*

<sup>2</sup> *Instituto de Ciencia de Materiales de Madrid, CSIC, Cantoblanco, E-28049 Madrid, Spain.*  
(Dated: October 23, 2006)

The polarization of graphene is calculated exactly within the random phase approximation for arbitrary frequency, wave vector, and doping. At finite doping, the static susceptibility saturates to a constant value for low momenta. At  $q = 2\mu/\hbar v_F$  it has a discontinuity only in the second derivative. In the presence of a charged impurity this results in Friedel oscillations which decay with same power law as the Thomas Fermi contribution, the latter being always dominant. The spin density oscillations in the presence of a magnetic impurity are also calculated. The dynamical polarization for low  $q$  and arbitrary  $\omega$  is employed to calculate the dispersion relation and the decay rate of plasmons and acoustic phonons as a function of doping. The low screening of graphene, combined with the absence of a gap, leads to a significant stiffening of the longitudinal acoustic lattice vibrations.

PACS numbers: 63.20.-e, 73.20.Mf, 73.21.-b

# Excitations of Dirac Fermions: RPA Theory

## Dielectric function, screening, and plasmons in 2D graphene

E. H. Hwang and S. Das Sarma

*Condensed Matter Theory Center, Department of Physics, University of Maryland, College Park, Maryland 20742-4111*

(October 4, 2006)

The dynamical dielectric function of two dimensional graphene at arbitrary wave vector  $q$  and frequency  $\omega$ ,  $\epsilon(q, \omega)$ , is calculated in the self-consistent field approximation. The results are used to find the dispersion of the plasmon mode and the electrostatic screening of the Coulomb interaction in 2D graphene layer within the random phase approximation. At long wavelengths ( $q \rightarrow 0$ ) the plasmon dispersion shows the local classical behavior  $\omega_{cl} = \omega_0 \sqrt{q}$ , but the density dependence of the plasma frequency ( $\omega_0 \propto n^{1/4}$ ) is different from the usual 2D electron system ( $\omega_0 \propto n^{1/2}$ ). The wave vector dependent plasmon dispersion and the static screening function show very different behavior than the usual 2D case.



# Excitations of Dirac Fermions: RPA Theory

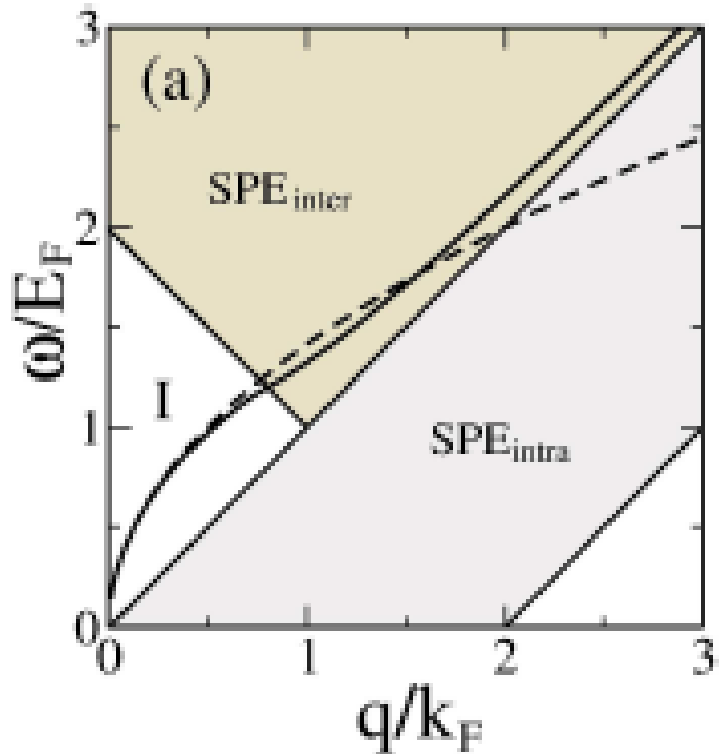
## Collective excitations of Dirac electrons in a graphene layer with spin-orbit interaction

X.F. Wang and Tapash Chakraborty

*Department of Physics and Astronomy, The University of Manitoba, Winnipeg, Canada, R3T 2N2*

Coulomb screening and excitation spectra of electrons in a graphene layer with spin-orbit interaction (SOI) is studied in the random phase approximation. The SOI opens a gap between the valence and conduction bands of the semi-metal Dirac system and reshapes the bottom and top of the bands. As a result, we have observed a dramatic change in the long-wavelength dielectric function of the system. An undamped plasmon mode emerges from the inter-band electron-hole excitation continuum edge and vanishes or merges with a Landau damped mode on the edge of the intra-band electron-hole continuum. The characteristic collective excitation of the Dirac gas is recovered in a system with a high carrier density.

# Excitations of Dirac Fermions: RPA Theory

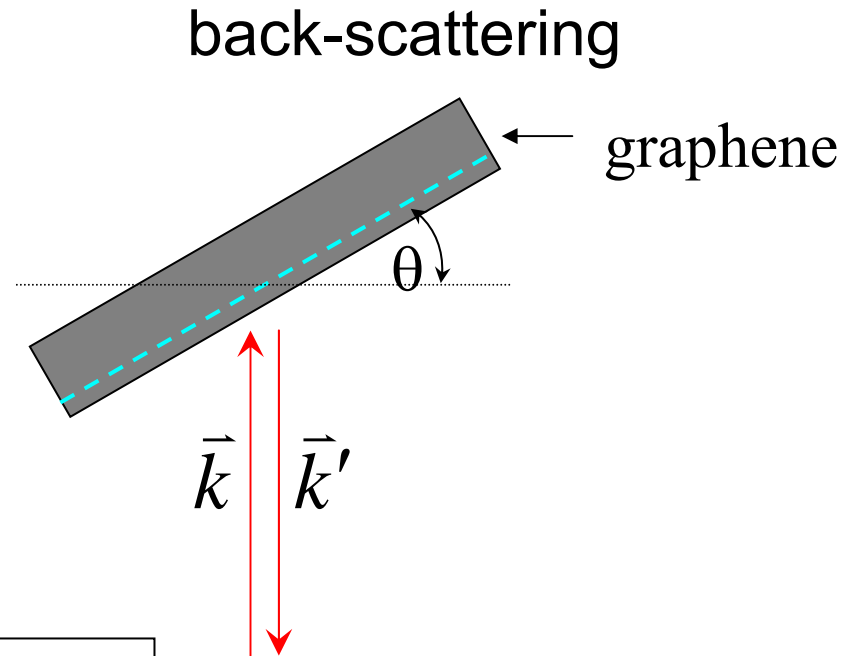
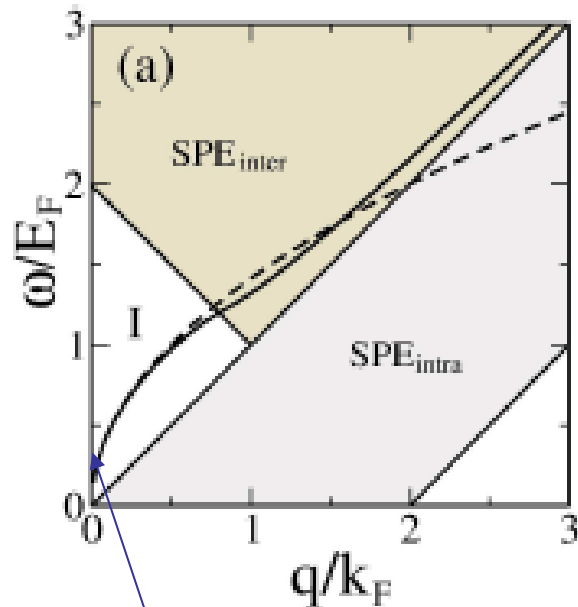


(a) Plasmon mode dispersion in 2D graphene (solid thick line) calculated within RPA. Thin solid lines represent the boundaries of the single particle excitation (SPE) Landau damping regime for intra- and inter-band electron-hole excitations.

After E.H. Wang and S. Das Sarma

# Excitations of Dirac Fermions: inelastic light scattering

wavevector transfer



$$\vec{q}_{\parallel} = \vec{k}_{\parallel} - \vec{k}'_{\parallel} = \frac{4\pi}{\lambda_{laser}} \sin \theta$$

# Excitations of Dirac Fermions: the light scattering toolbox

## selection rules

**For collective modes and single particle excitations (SPE)**

## light scattering mechanisms

**Resonant processes enhance the light scattering intensities of the dilute 2D systems**

# Excitations of Dirac Fermions: the light scattering toolbox

## light scattering mechanisms

Plasmons: **q-independent** electro-optic processes are forbidden under the inversion symmetry of the honeycomb lattice

Plasmons: **q-dependent** Coulomb interaction processes are allowed and could be significant.

Plasmons and SPE: the fermion density fluctuation mechanisms are likely to be weak in graphene

# 'Impact' of High Magnetic Fields



# Excitations of Dirac Fermions: inter-Landau level transitions

## Excitations from Filled Landau Levels in Graphene

A. Iyengar<sup>1</sup>, Jianhui Wang<sup>1</sup>, H. A. Fertig<sup>1</sup>, L. Brey<sup>2</sup>

<sup>1</sup>*Department of Physics, Indiana University, Bloomington, IN 47405 and*

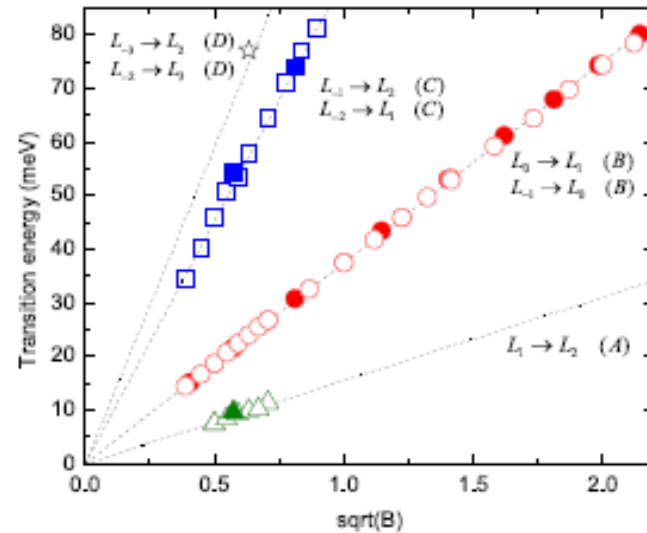
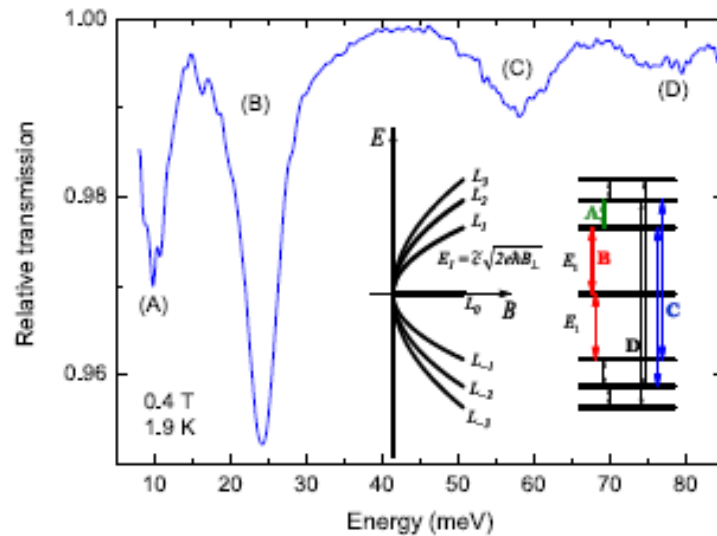
<sup>2</sup>*Instituto de Ciencia de Materiales de Madrid (CSIC), Cantoblanco, 28049 Madrid, Spain*

(Dated: August 17, 2006)

We consider graphene in a strong perpendicular magnetic field at zero temperature with an integral number of filled Landau levels and study the dispersion of single particle-hole excitations. We first analyze the two-body problem of a single Dirac electron and hole in a magnetic field interacting via Coulomb forces. We then turn to the many-body problem, where particle-hole symmetry and the existence of two valleys lead to a number of effects peculiar to graphene. We find that the coupling together of a large number of low-lying excitations leads to strong many-body corrections, which could be observed in inelastic light scattering or optical absorption. We also discuss in detail how the appearance of different branches in the exciton dispersion is sensitive to the number of filled spin and valley sublevels.

# Excitations of Dirac Fermions: inter-Landau level optical absorption

M. Sadowski et al, PRL 12/2006

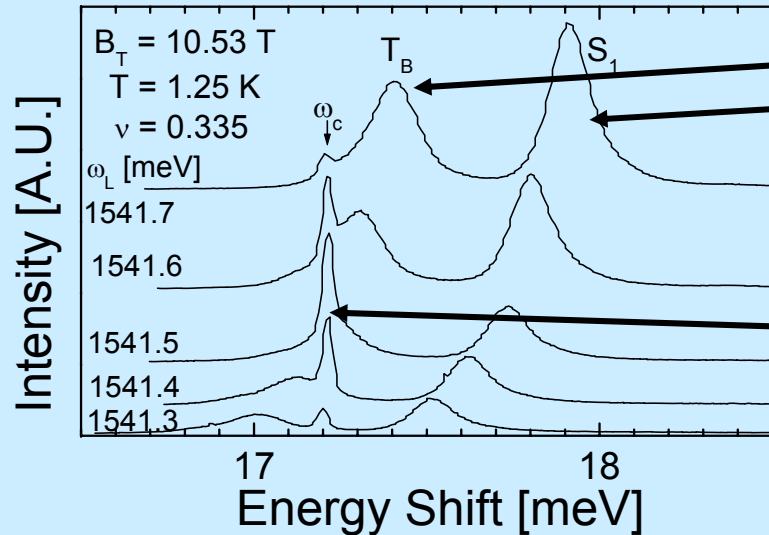


Single-layer measurements of the  
Columbia-NHMFL collaboration



# Inter-Landau-level Light Scattering in GaAs Quantum Wells

## The GaAs benchmark

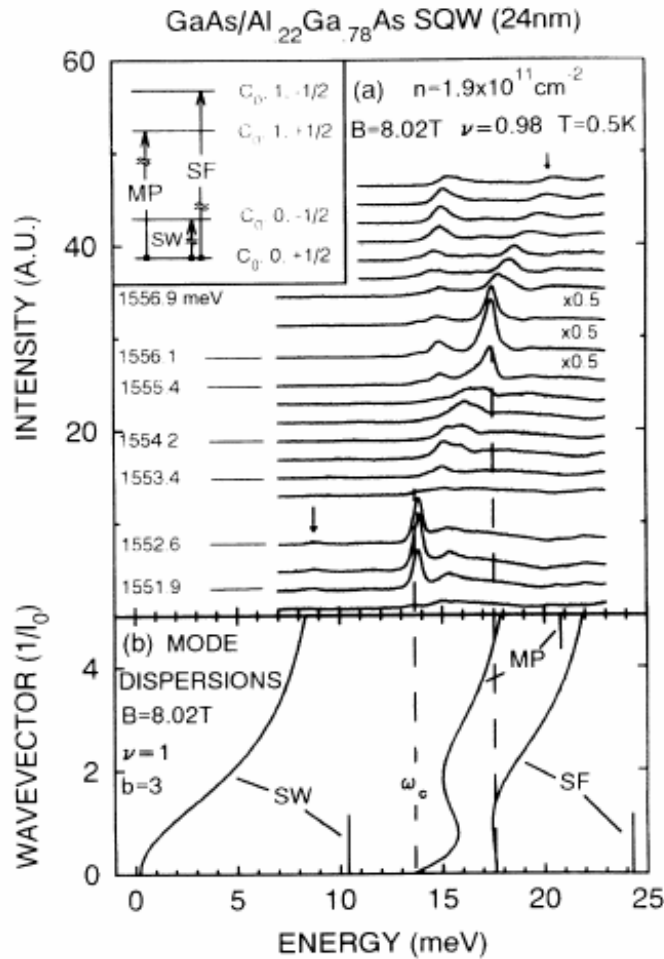


optical recombination  
at the fundamental gap  $E_G$

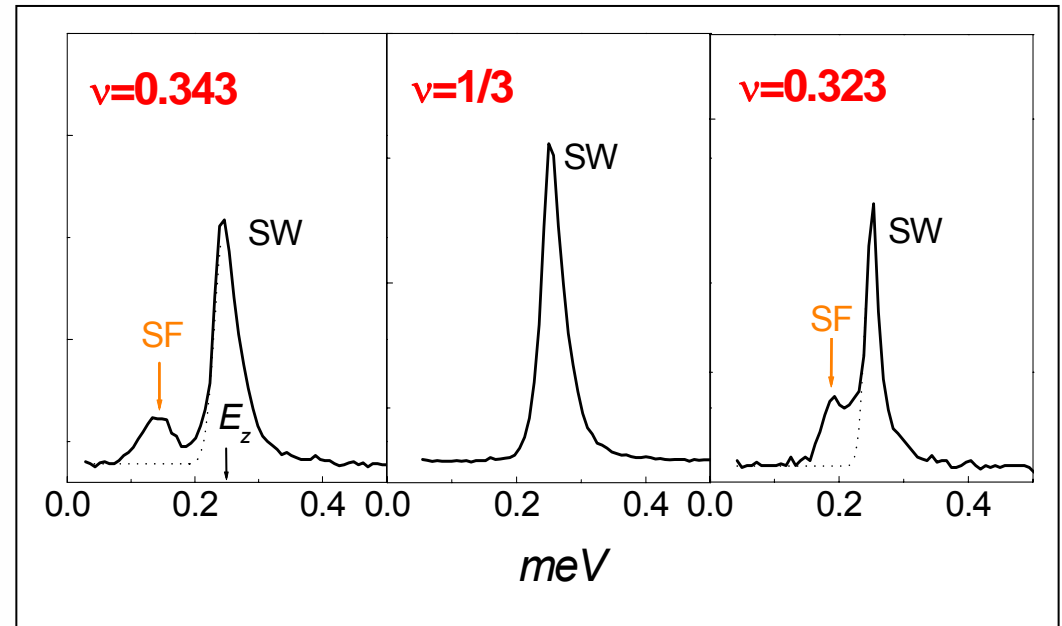
'cyclotron' mode of  
electrons at energy  $\omega_c$

# Light Scattering by Spin excitations in GaAs quantum wells

## The GaAs benchmark



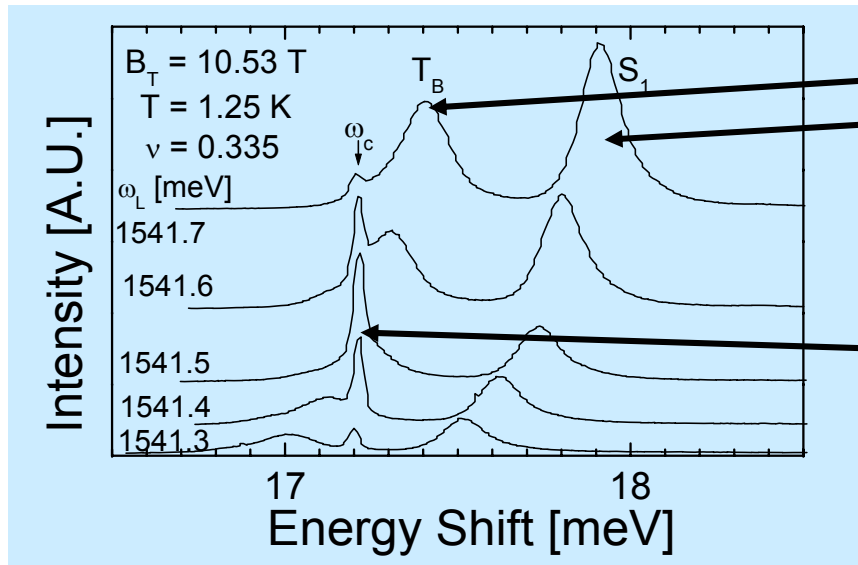
PRL (1992)



Y. Gallais et al, PRL (2006)

# Resonant Light Scattering in GaAs

These studies are enabled by strong resonant enhancements of the scattering cross-sections

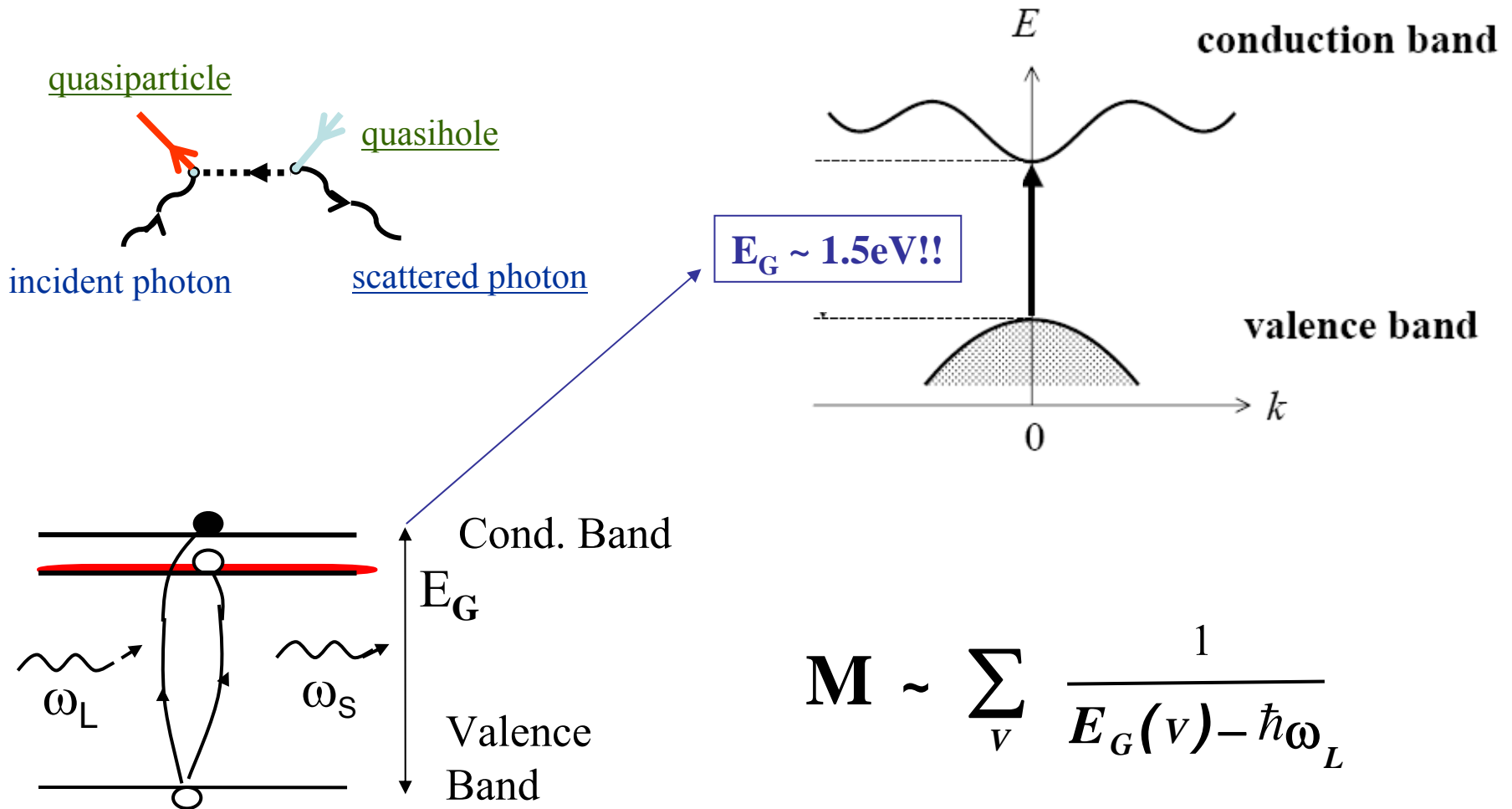


optical recombination  
at the fundamental gap  $E_G$

'cyclotron' mode of  
electrons at energy  $\omega_c$

incident photon energies need to overlap optical transitions of the GaAs quantum well

# Resonant Light Scattering in GaAs



# Resonant Light Scattering in Graphene

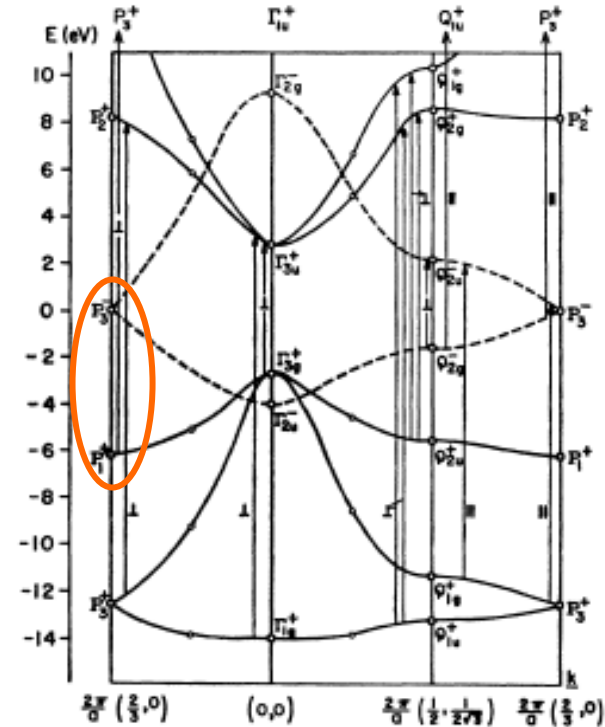
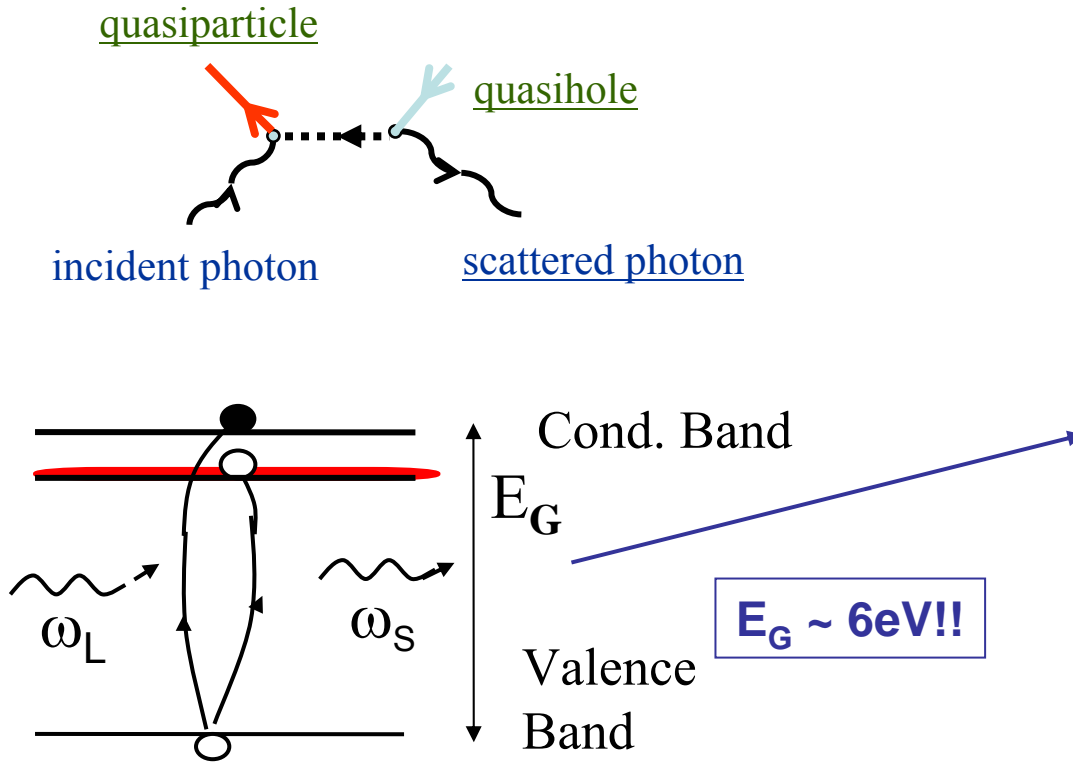
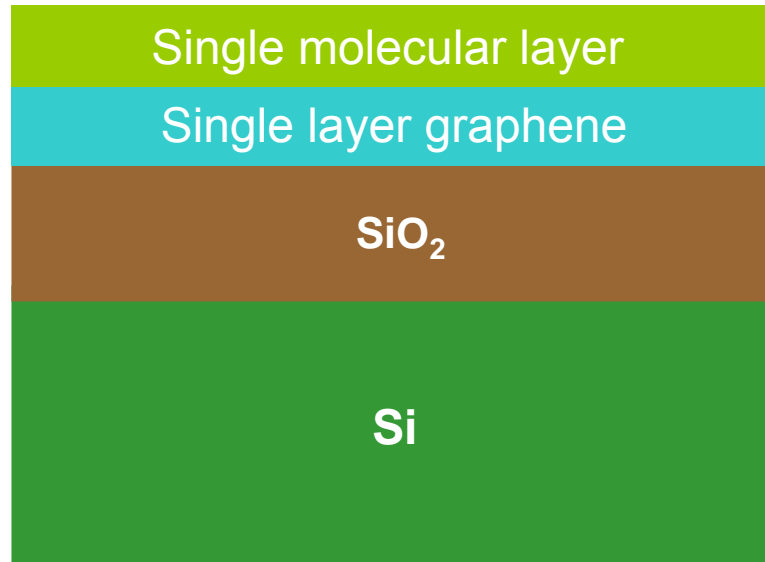


FIG. 8. Band structure of two-dimensional graphite after Bassani and Pastori (Ref. 10).  $\sigma$  and  $\pi$  bands are indicated by continuous and broken lines, respectively. The allowed transitions at critical points are indicated for both polarizations.

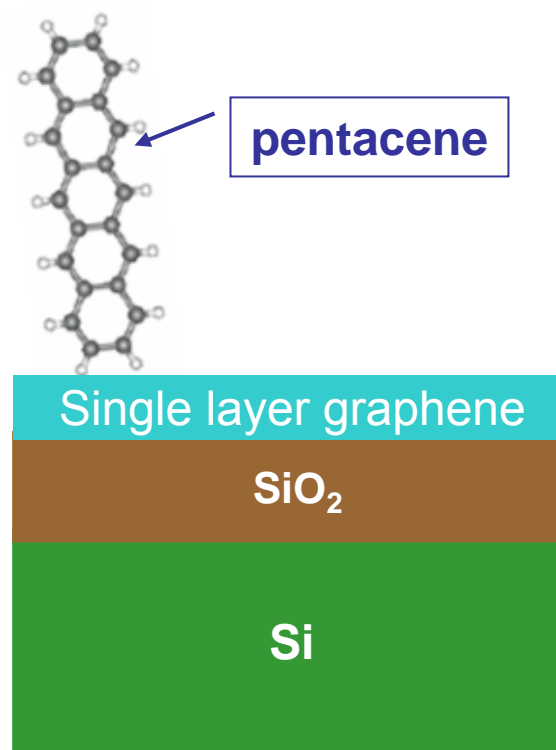
F. Bassani and G. Pastori (1967)

**This resonance is not accessible with current laser sources**

# Spectroscopy of Graphene Hetero-layers



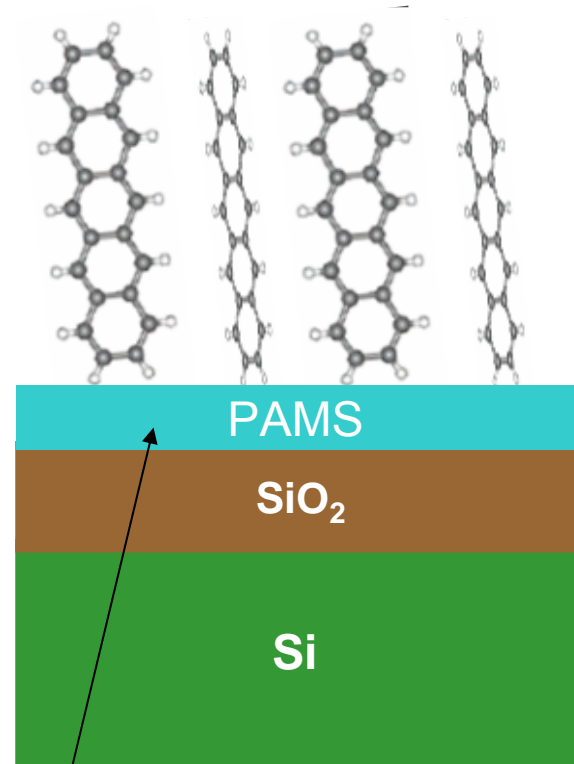
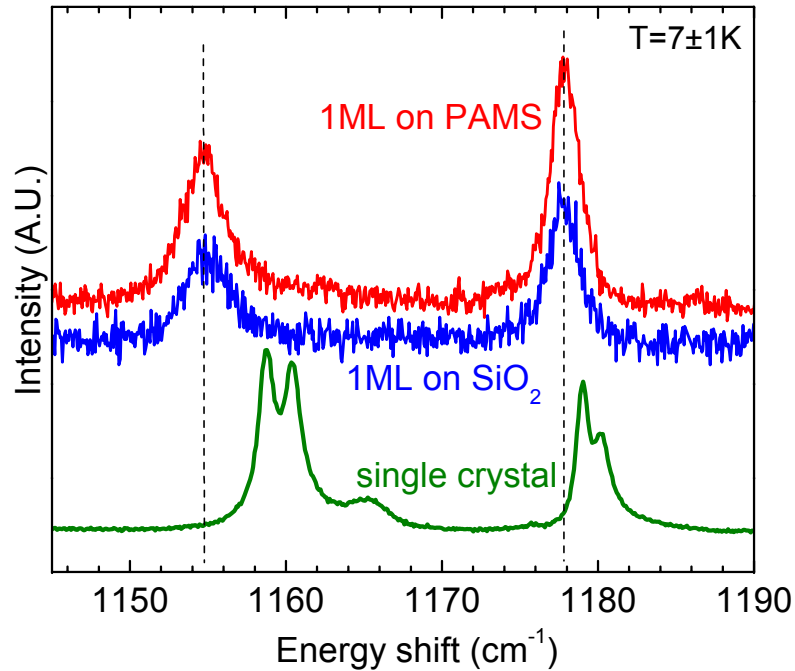
# Spectroscopy of Graphene Hetero-layers



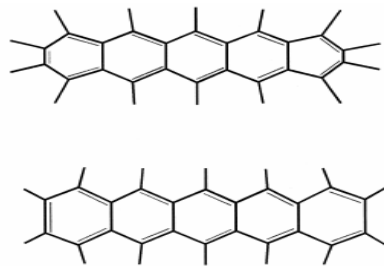
Pentacene, for example, has a huge optical resonance at  $\sim 2\text{eV}$  (red light)

# Resonant Raman Scattering in Pentacene Monolayers

## C-H in-plane bending modes



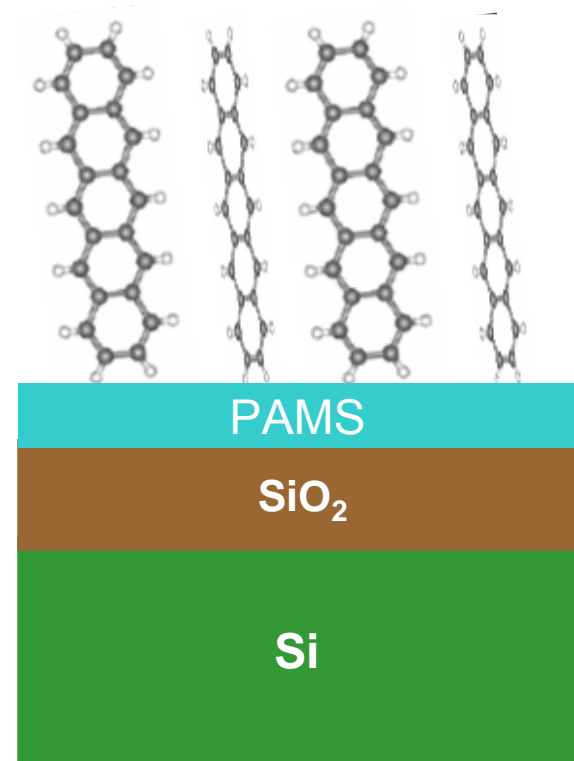
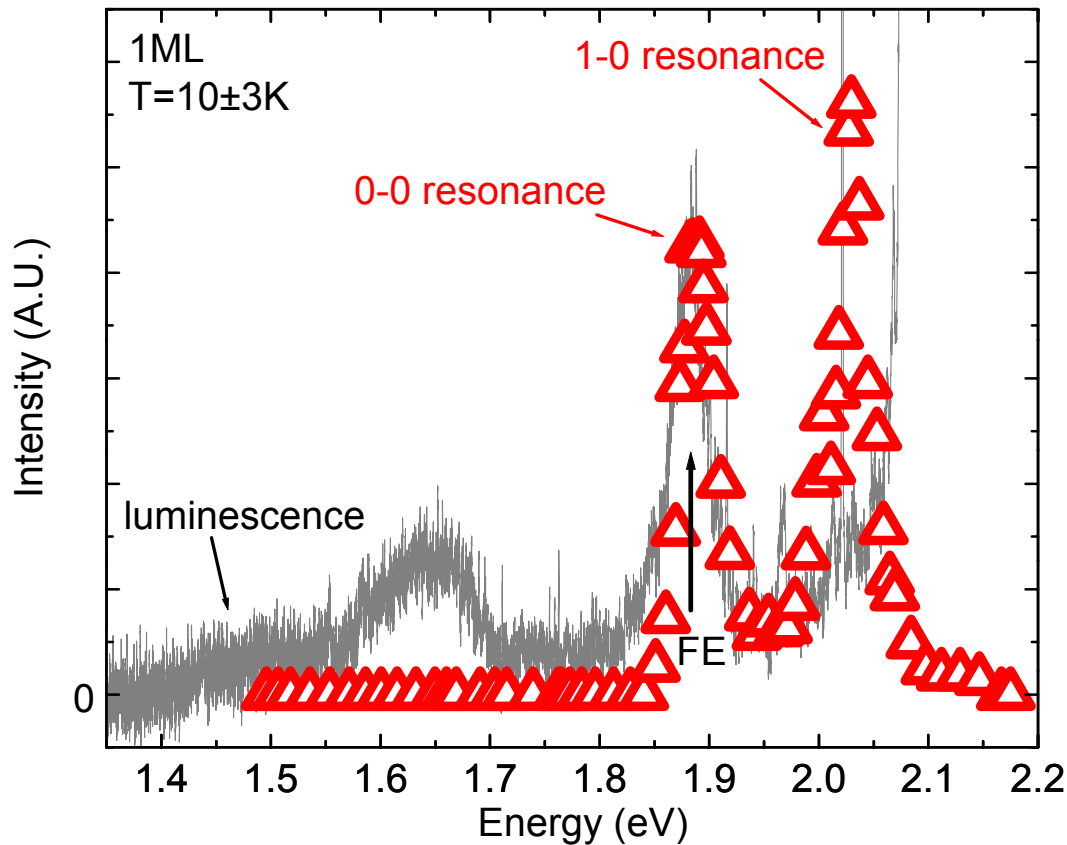
PAMS: poly alpha methyl styrene



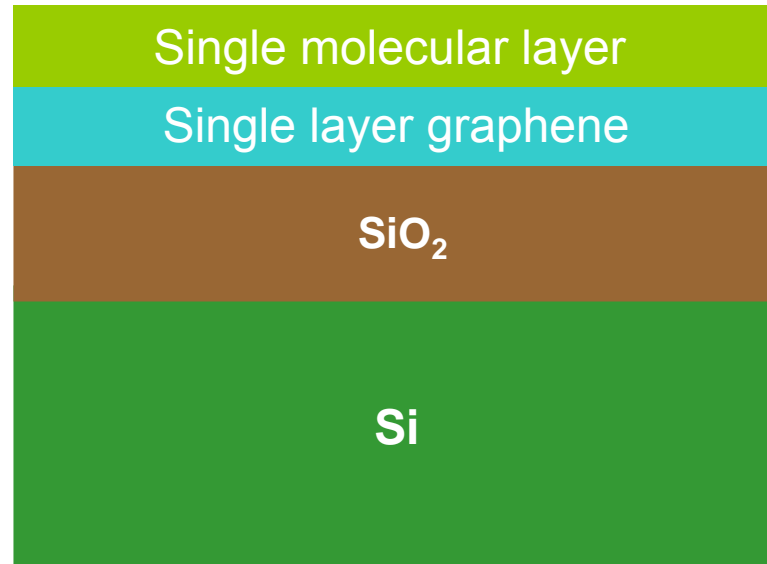


# Pentacene monolayers on PAMS: C-H in-plane bending modes

## Resonance Enhancement of Raman Scattering



# Spectroscopy of Graphene Hetero-layers



optical resonances of molecular  
overlayers will enable  
Raman scattering measurements