



UNIVERSITY OF  
**BATH**

**Graphene**  
2018  
June 26 - 29  
Dresden (Germany)

**MANCHESTER**  
1824  
The University of Manchester  
National Graphene Institute

*Superconductivity-induced features in the electronic Raman spectrum of monolayer graphene*

# Raman scattering minds the gap

**Aitor García-Ruiz**

**Supervisor: Marcin Mucha-Kruczynski**

**Collaborators: Joshua Thompson, Vladimir Falko**

Image from "Raman Spectroscopy in Graphene Related Systems" Ado Jorio et al.

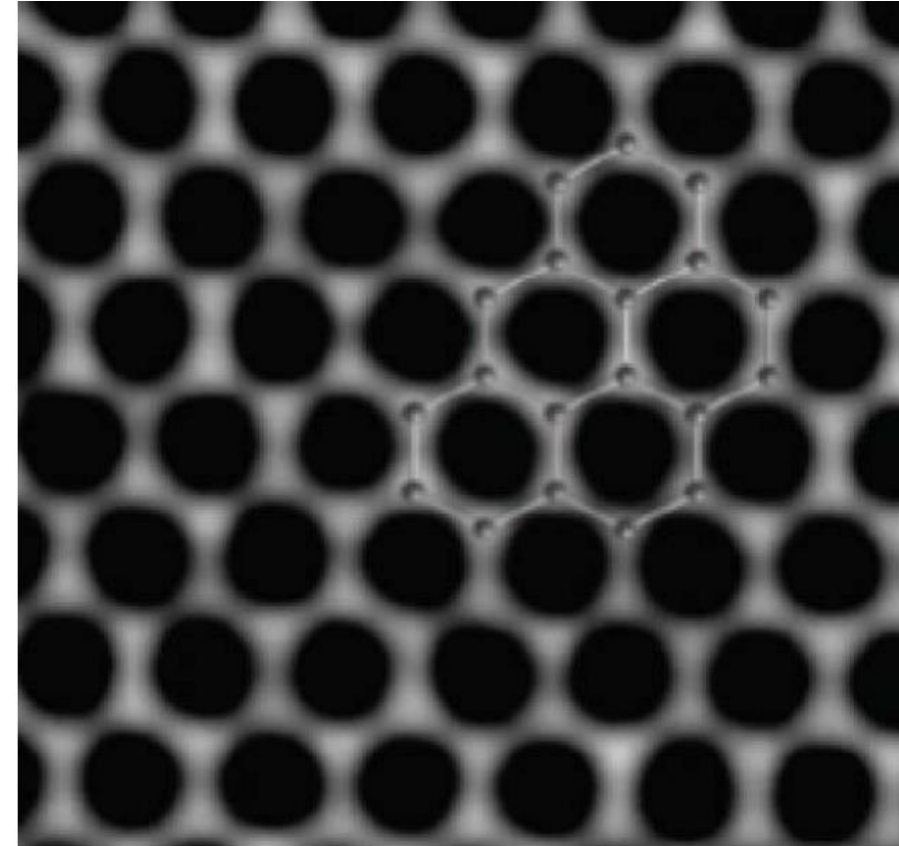
# Outline

- ERS in graphene
  - Graphene
  - Raman in graphene
  - Electronic Raman scattering in graphene
- ERS in superconducting graphene
  - Superconducting graphene
  - Electronic Raman scattering in superconducting graphene

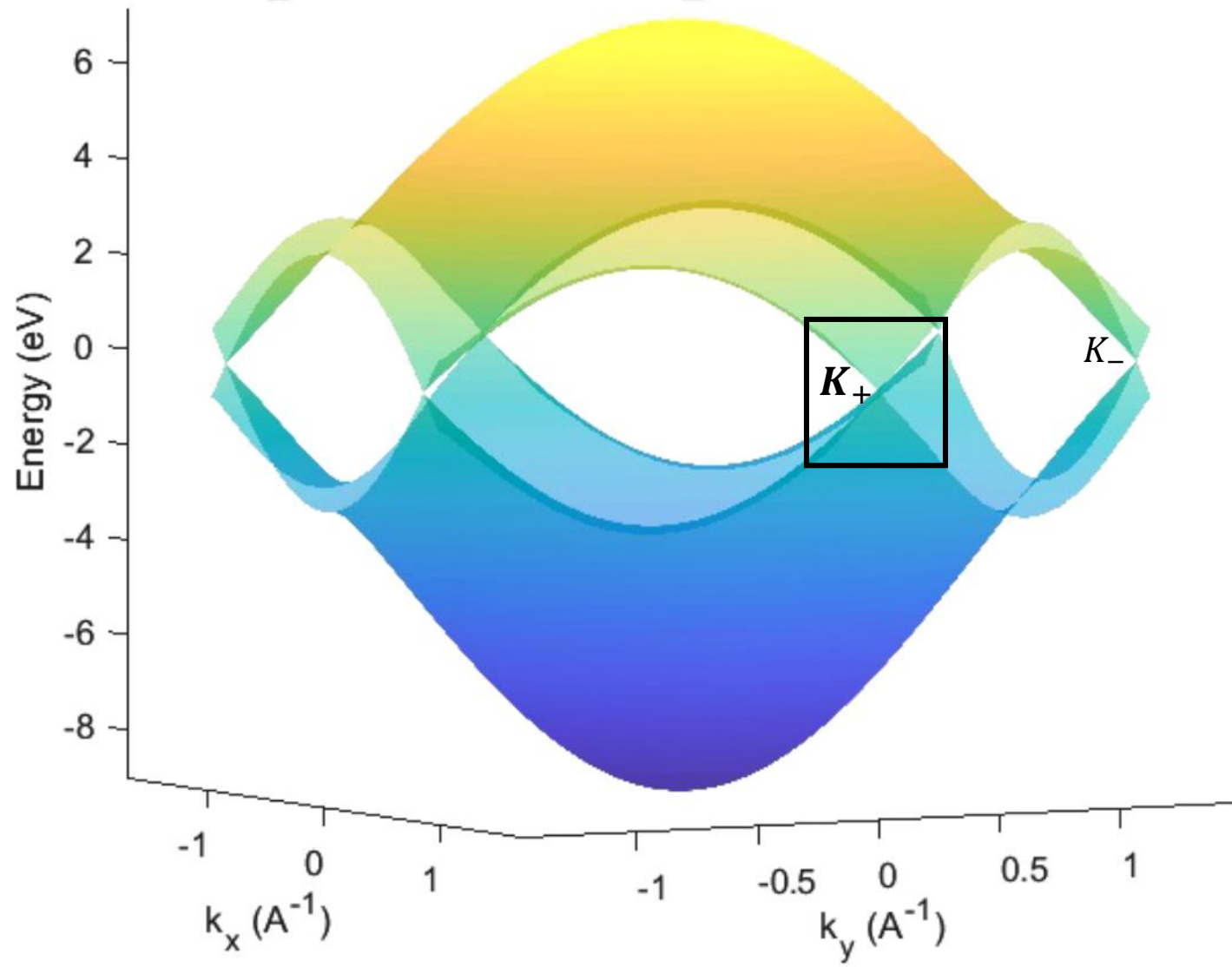


# Outline

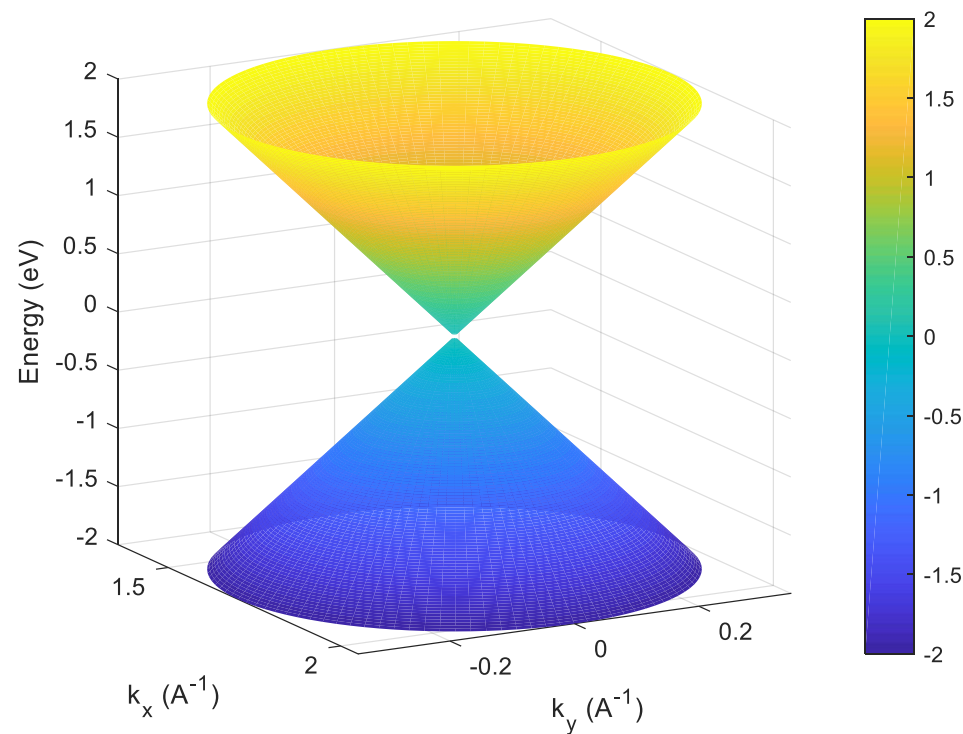
- **ERS in graphene**
  - Graphene
  - Raman Scattering in graphene
  - **Electronic Raman scattering in graphene**
- ERS in superconducting graphene
  - Superconducting graphene
  - Electronic Raman scattering in superconducting graphene



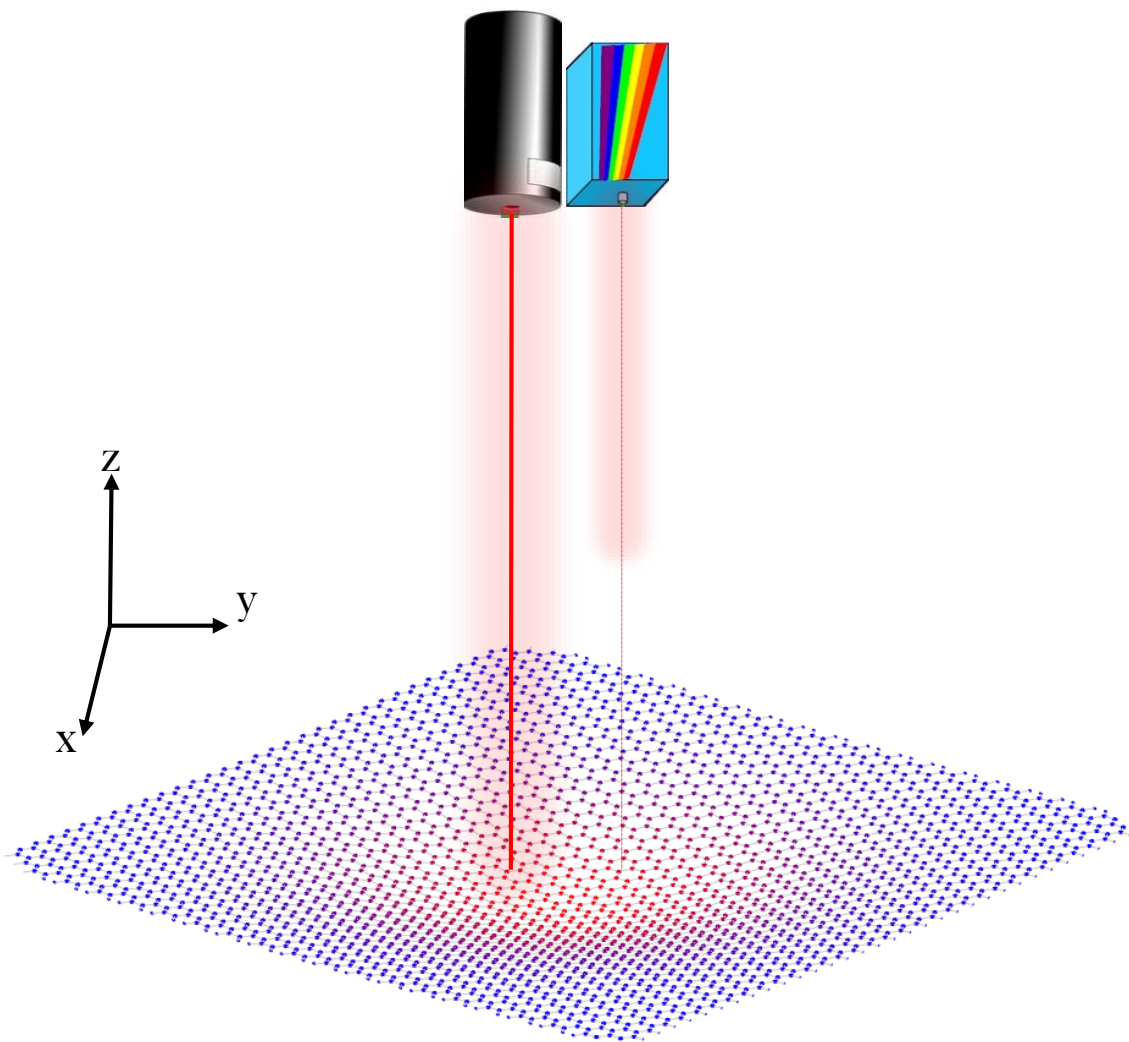
# Graphene: dispersion relation



Linear dispersion  $\mathcal{H}_{gr}^{lin}$

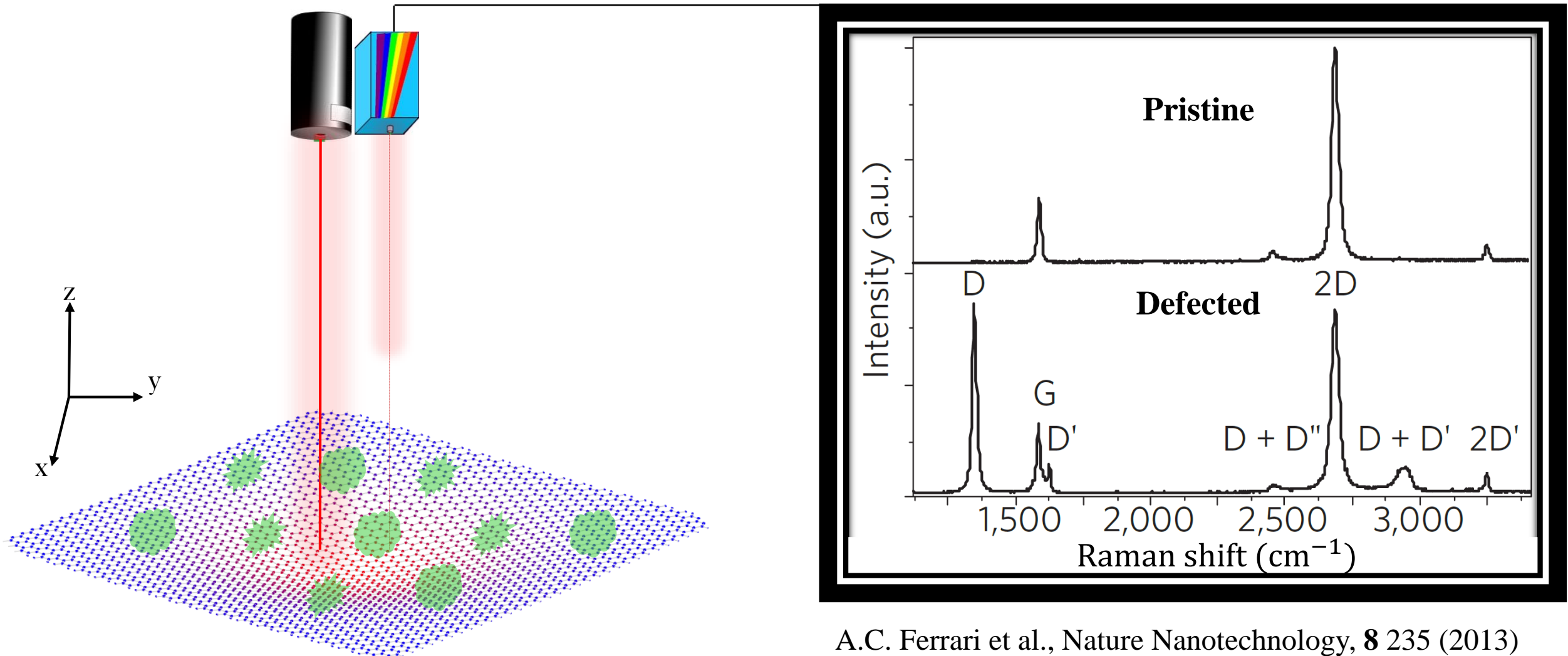


# Raman scattering in graphene

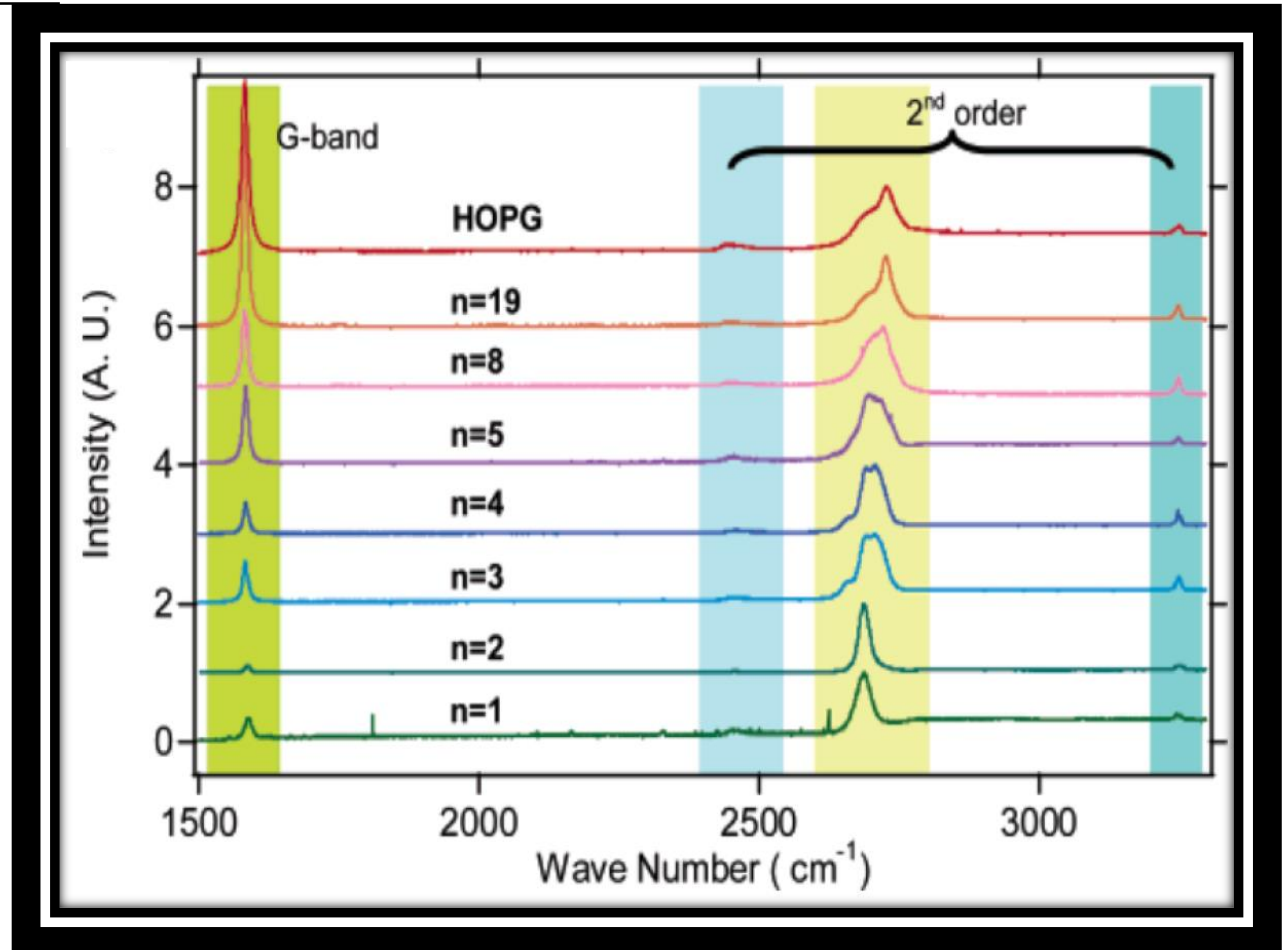
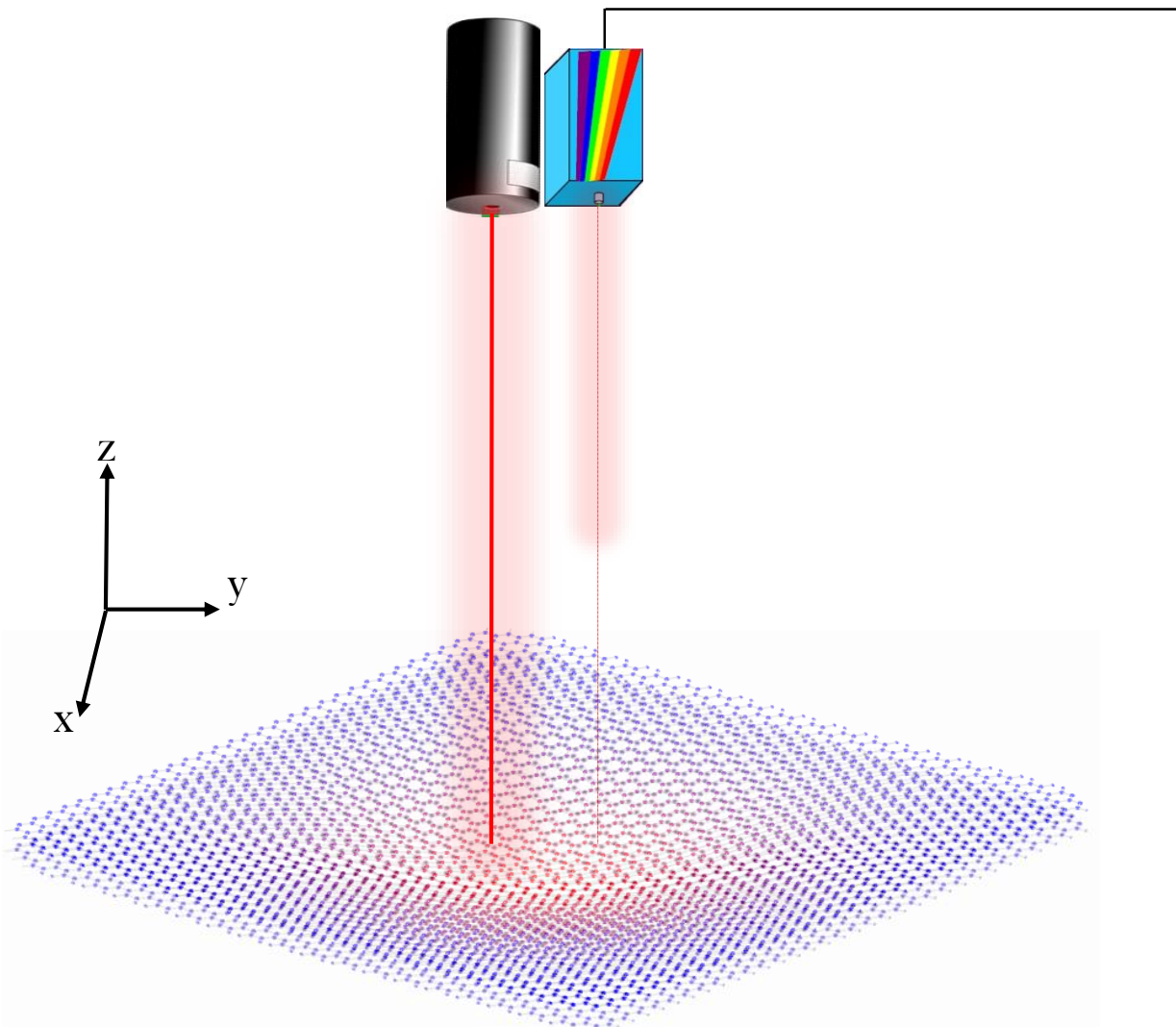




# Raman scattering in graphene: defects

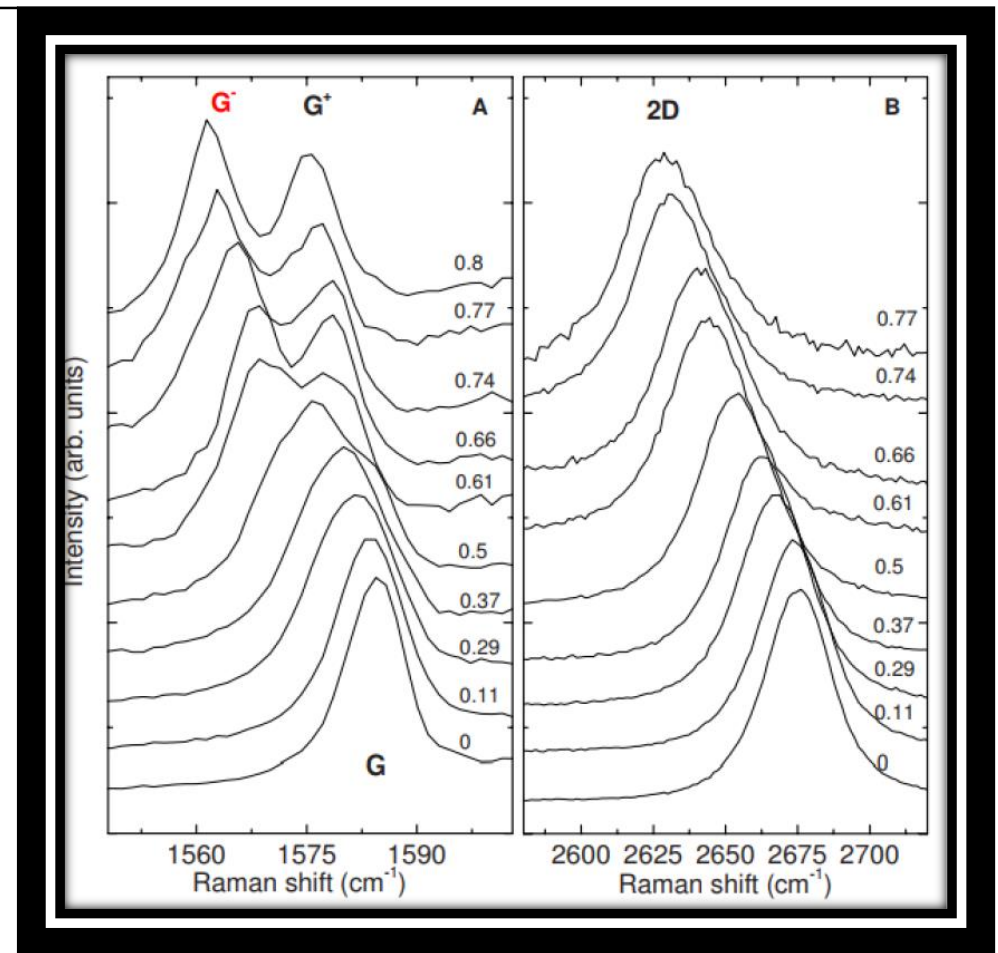
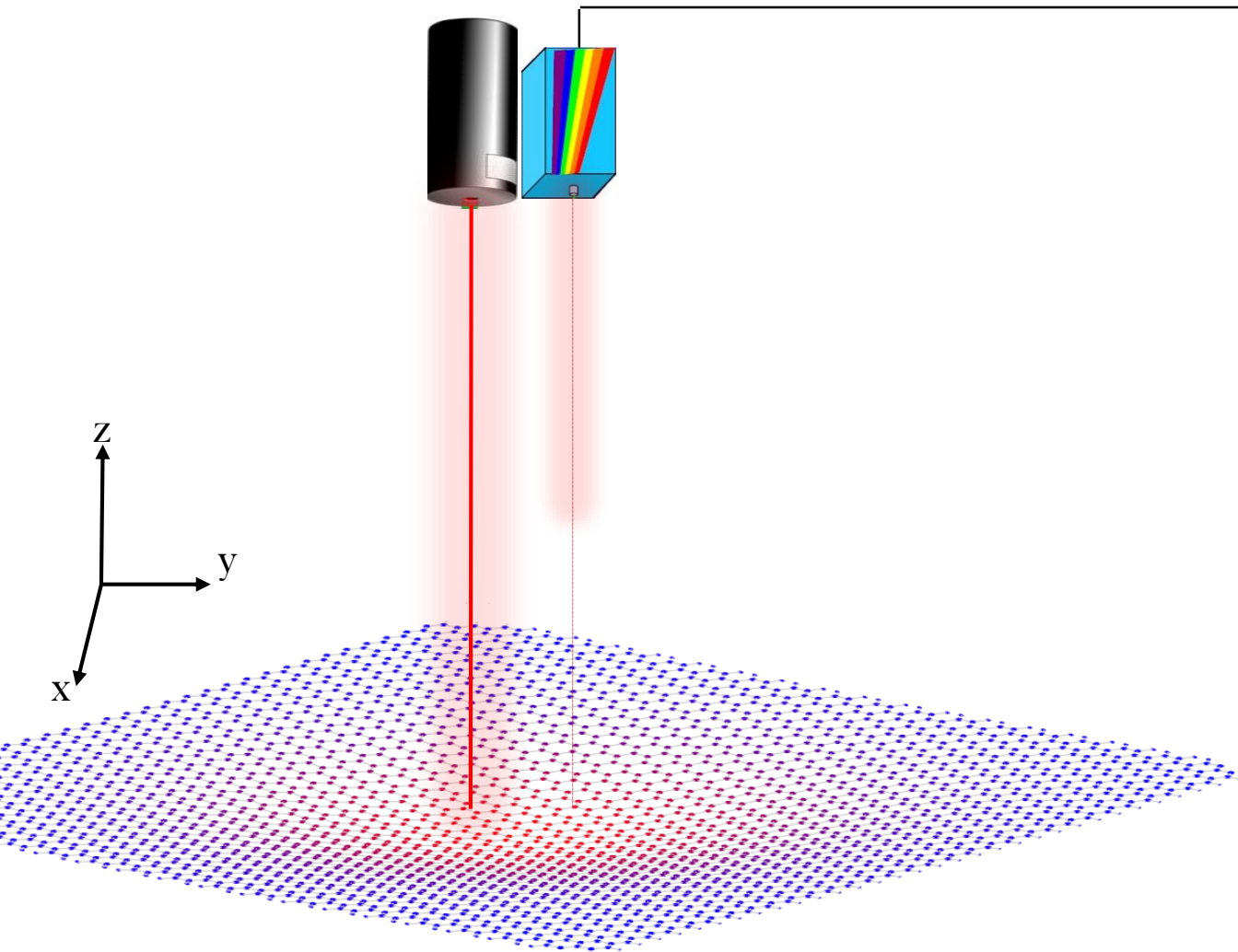


# Raman scattering in graphene: layers



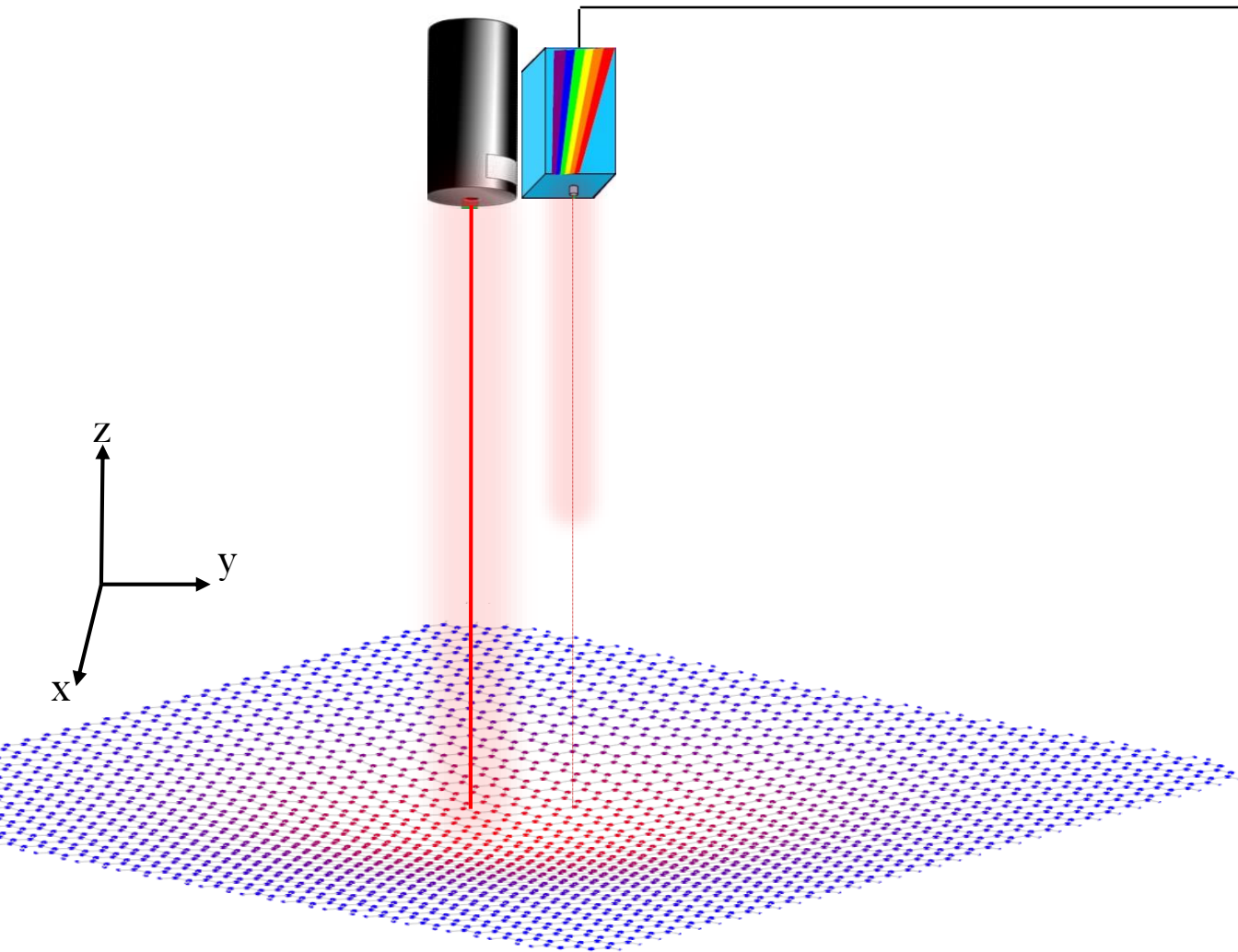
A. Gupta, et al., Nano Lett. **6**, 12, 2667-2673 (2006)

# Raman scattering in graphene: strains





# Raman scattering in graphene: strains



**Are there  
purely  
electronic  
excitations?**

# ERS in graphene: the picture

— Virtual state

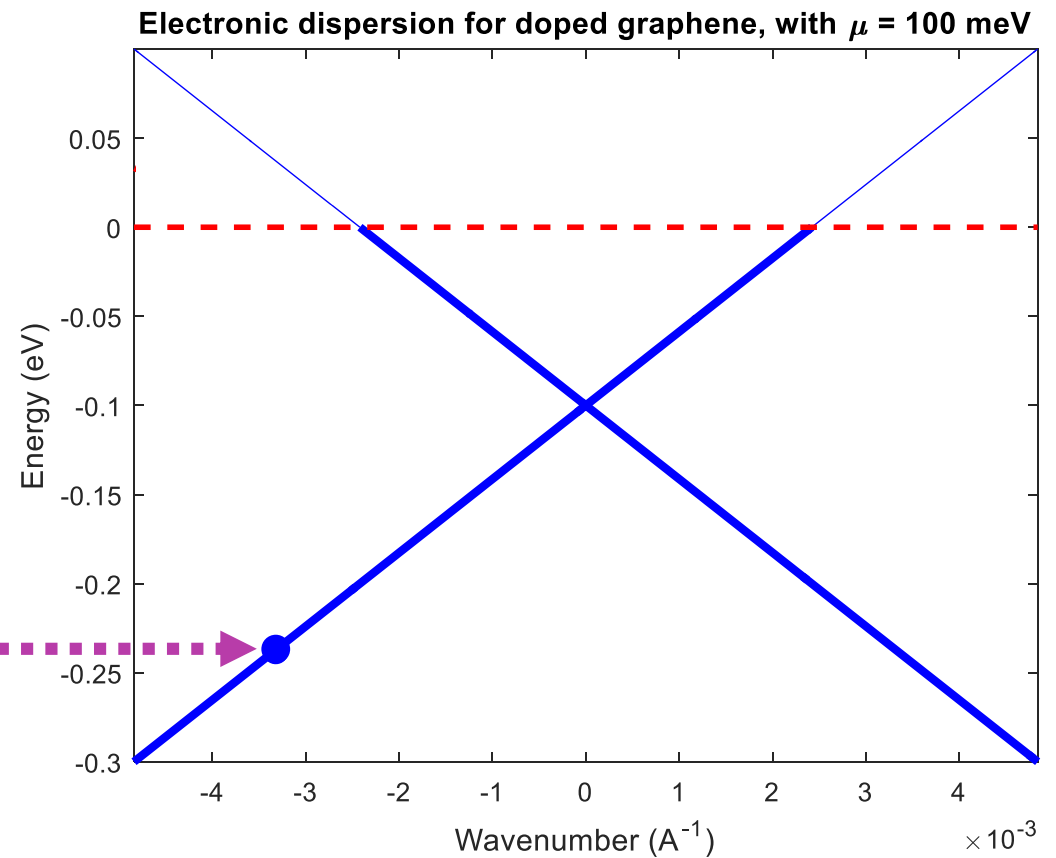
Incoming  $\gamma$



Virtual  $e^-$



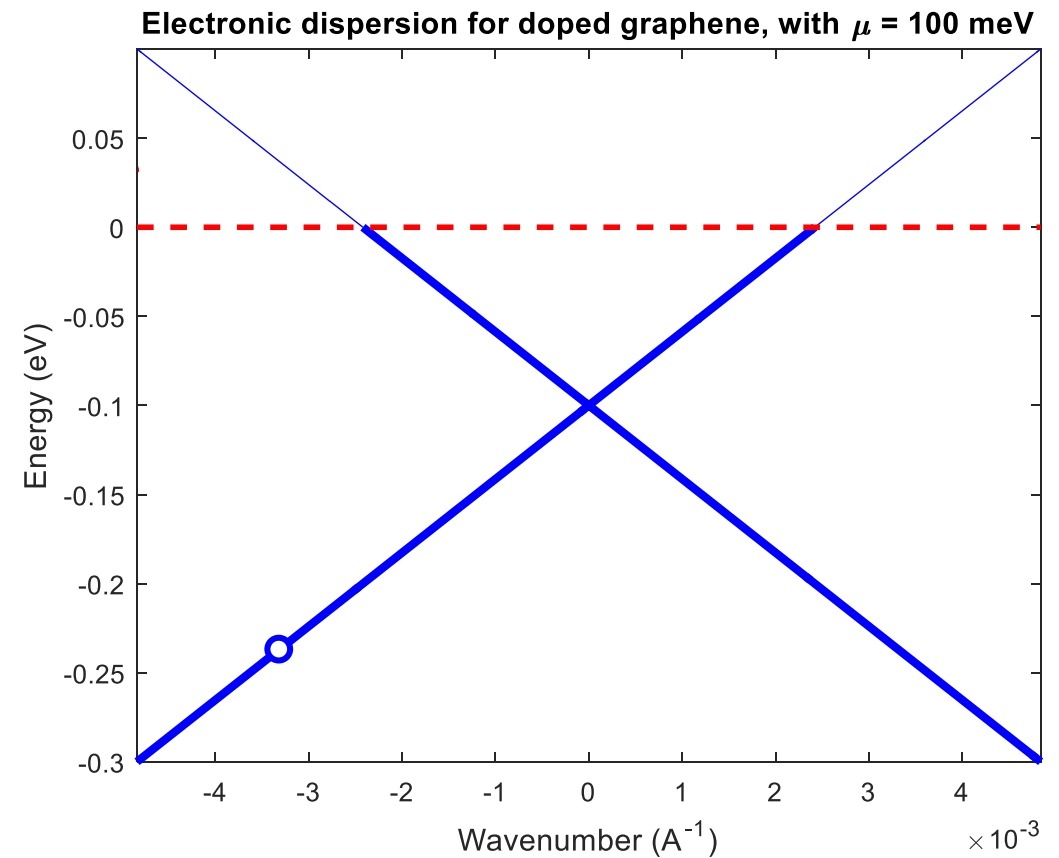
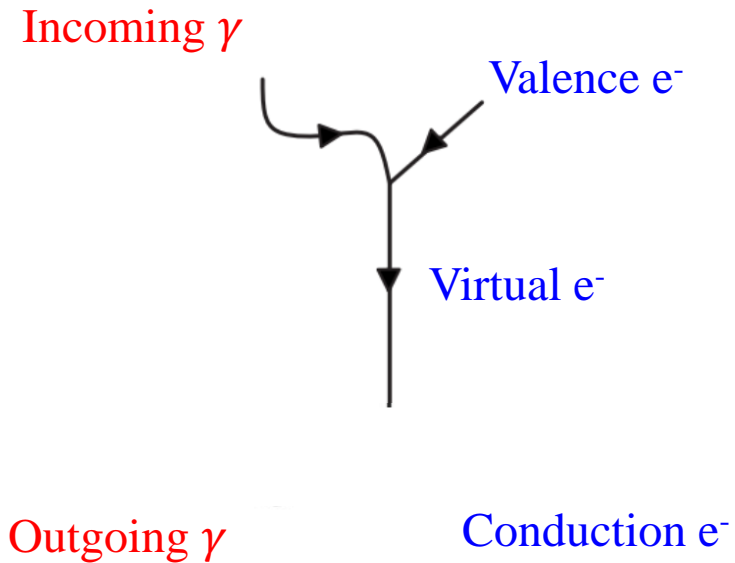
incoming





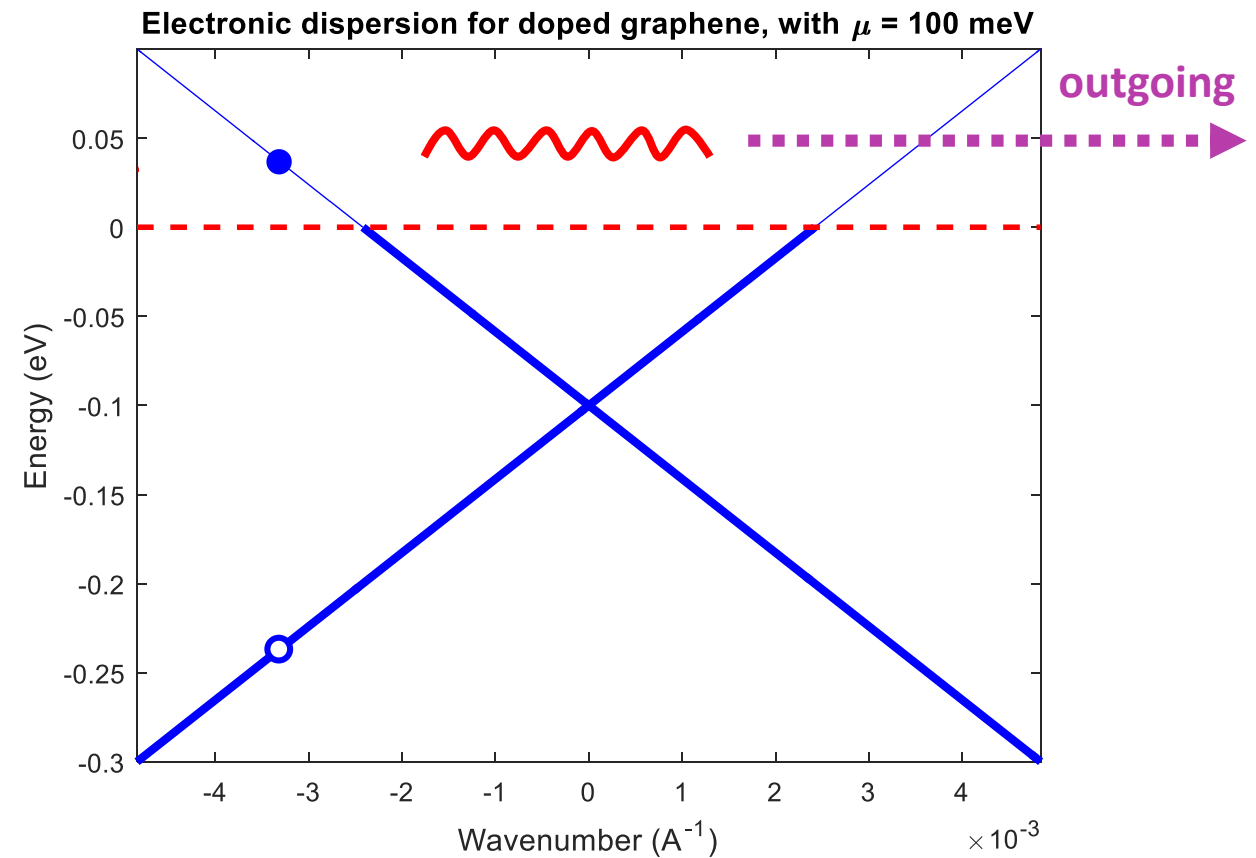
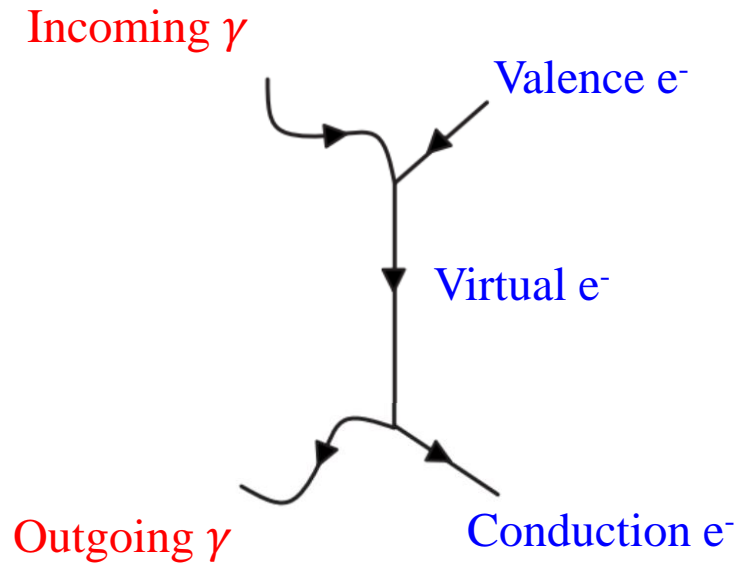
# ERS in graphene: the picture

—●— Virtual state



# ERS in graphene: the picture

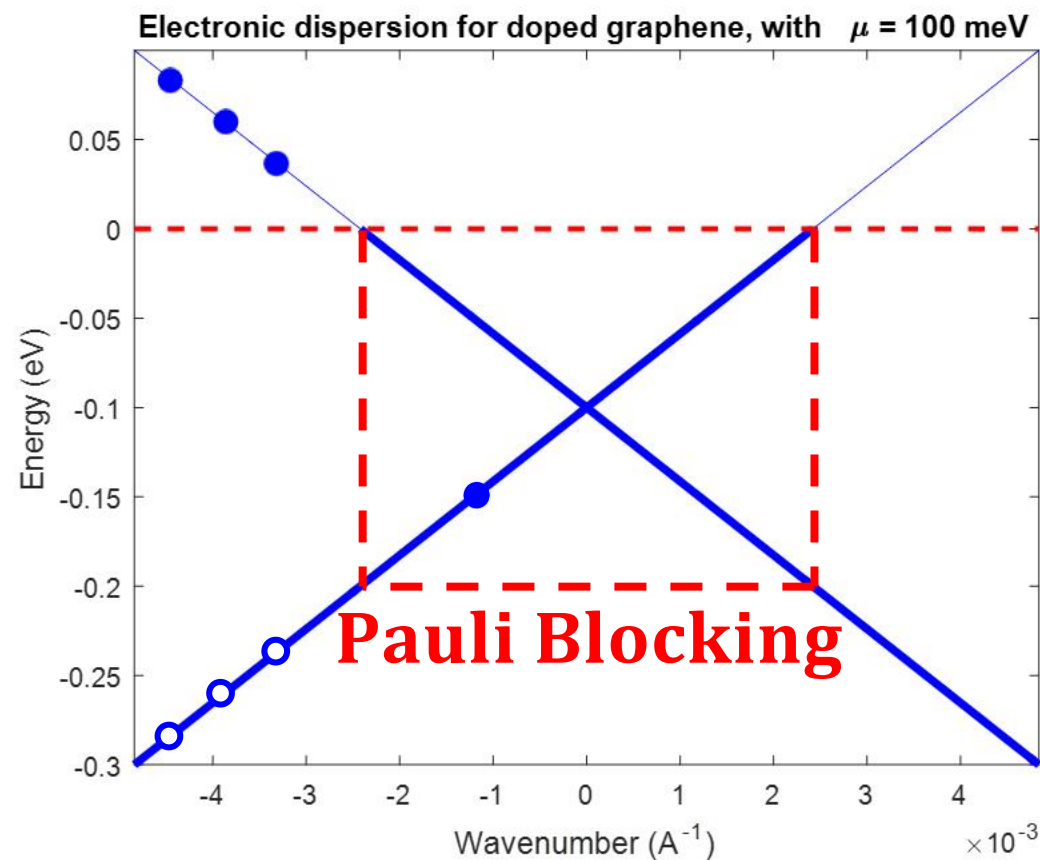
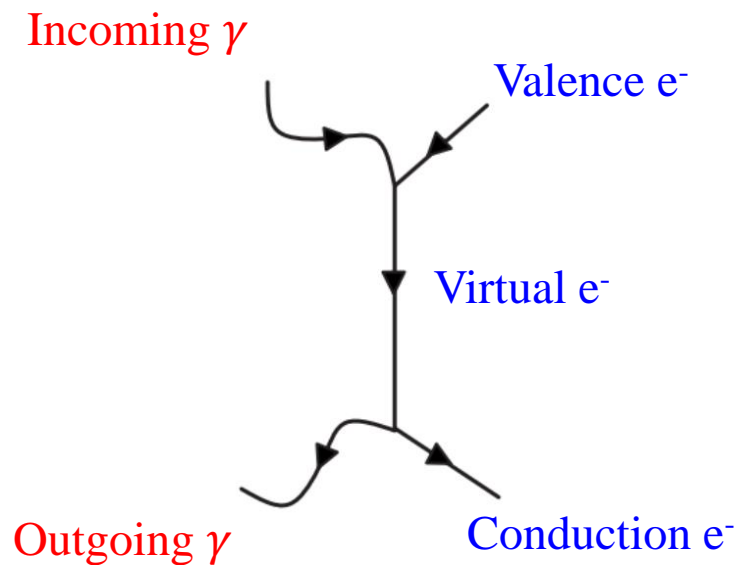
— Virtual state





# ERS in graphene: the picture

— Virtual state



# ERS in graphene: the theory

Light-matter interaction

$$\vec{P}$$

$$\downarrow$$

$$\vec{p} - \frac{e}{c} \vec{A}$$

$\mathcal{H}_{gr}^{lin}$



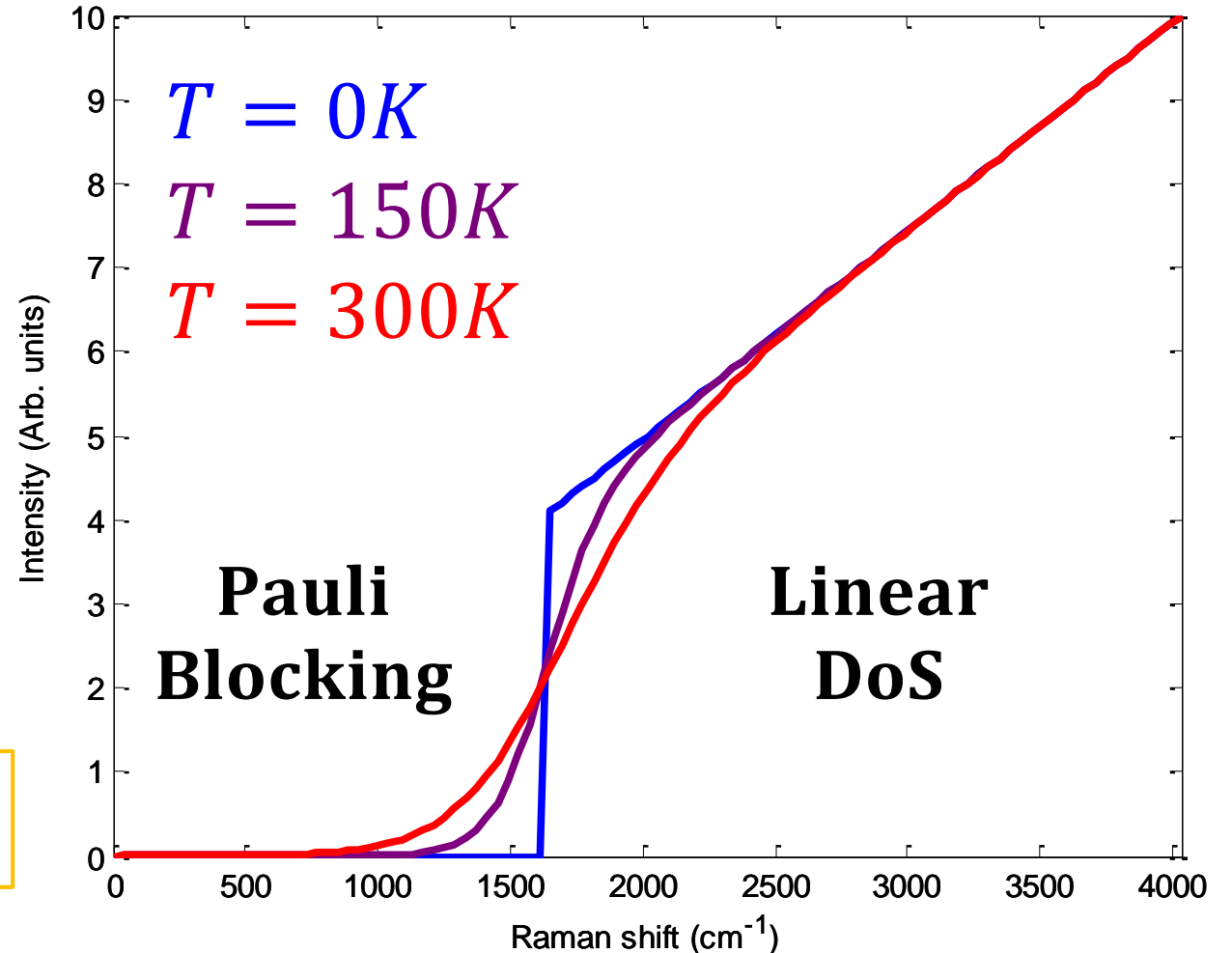
$$w(\omega) = \int \frac{dp}{2\pi\hbar^3} \left| \langle c | \mathcal{V}_{lin} | v \rangle \right|^2 \delta(\epsilon_c - \epsilon_v + \omega)$$

O. Kashuba and V. I. Fal'ko, Phys. Rev. B **80**, 241404 (2009)

$$w(\omega) = \text{Polarization factor (XY)} \cdot \text{Linear DoS} \cdot \text{Pauli Blocking}$$

XY – configuration

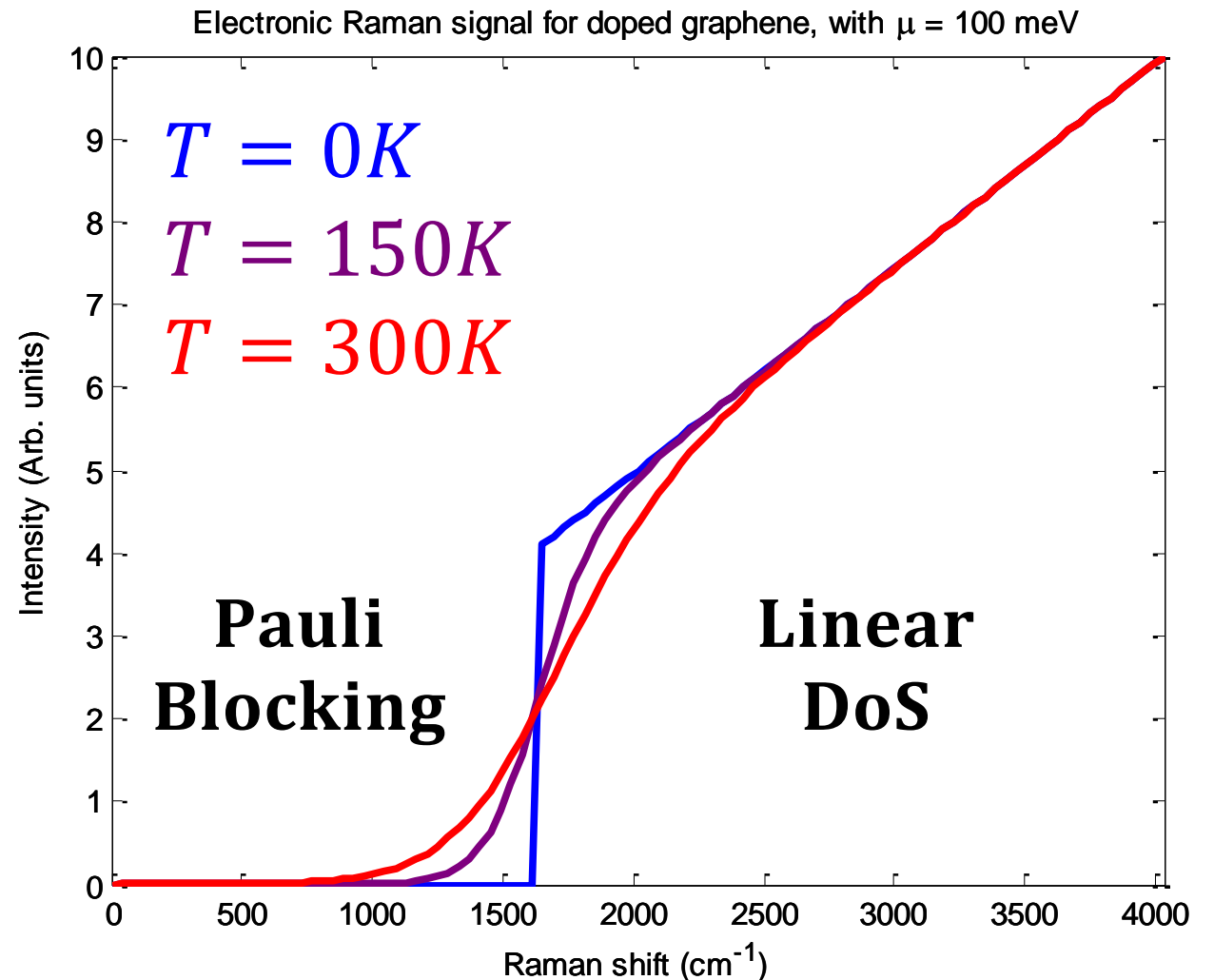
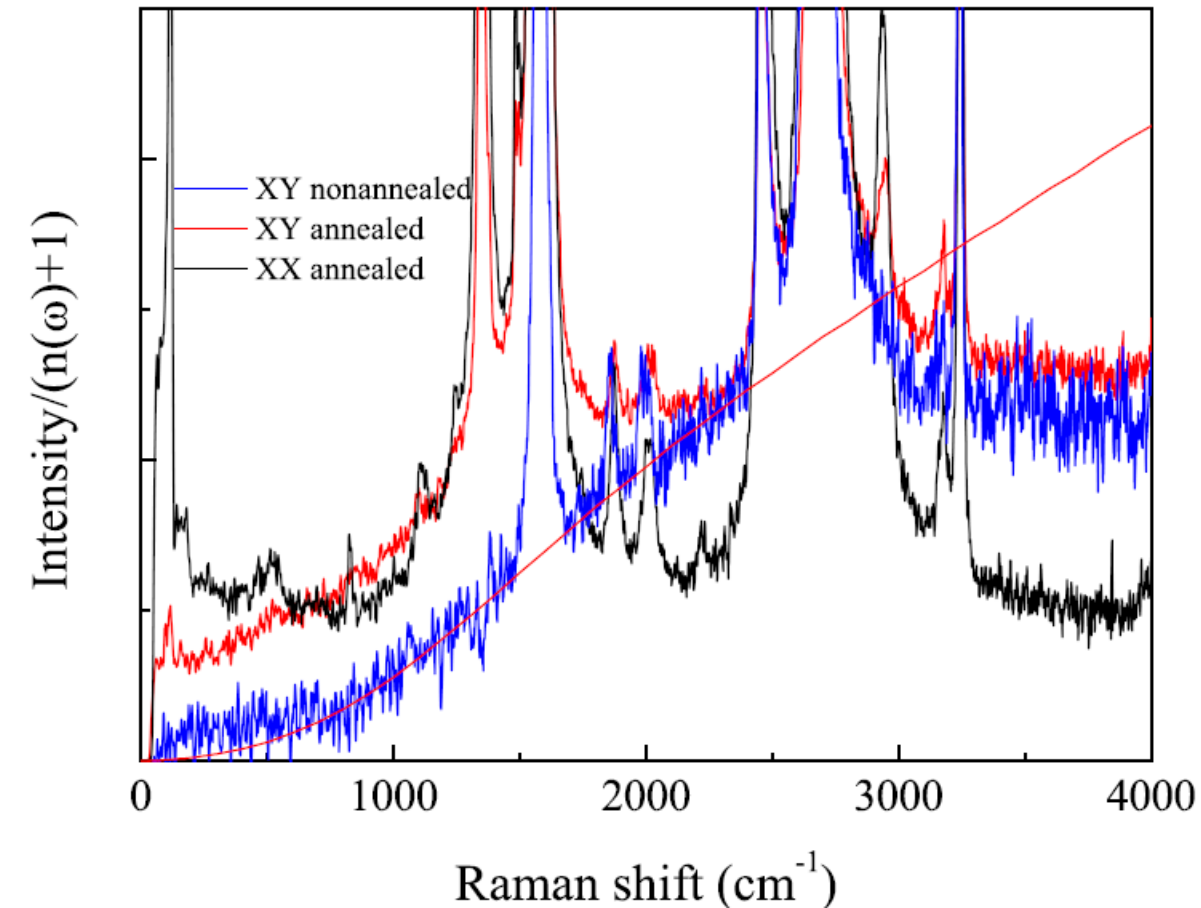
Electronic Raman signal for doped graphene, with  $\mu = 100$  meV



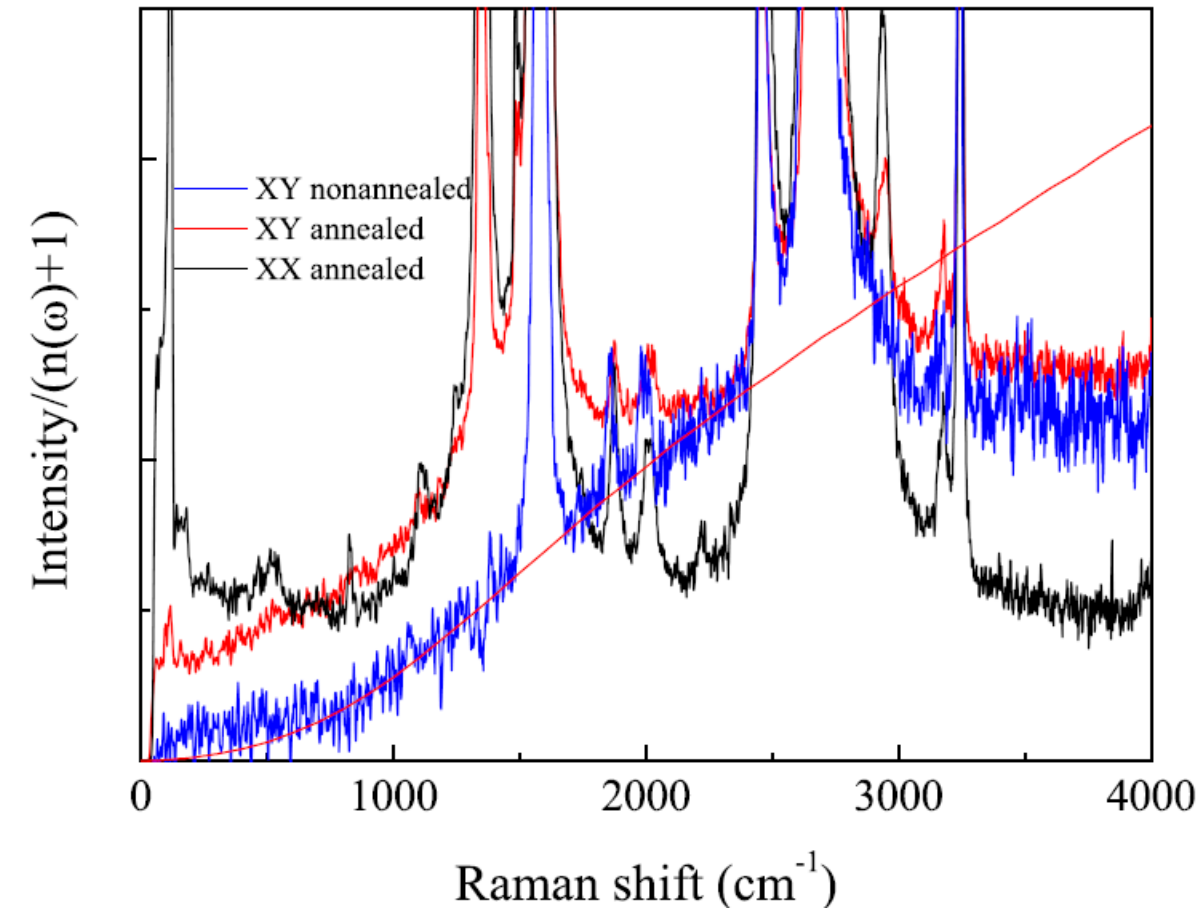


# ERS in graphene: the experiment

XY – configuration

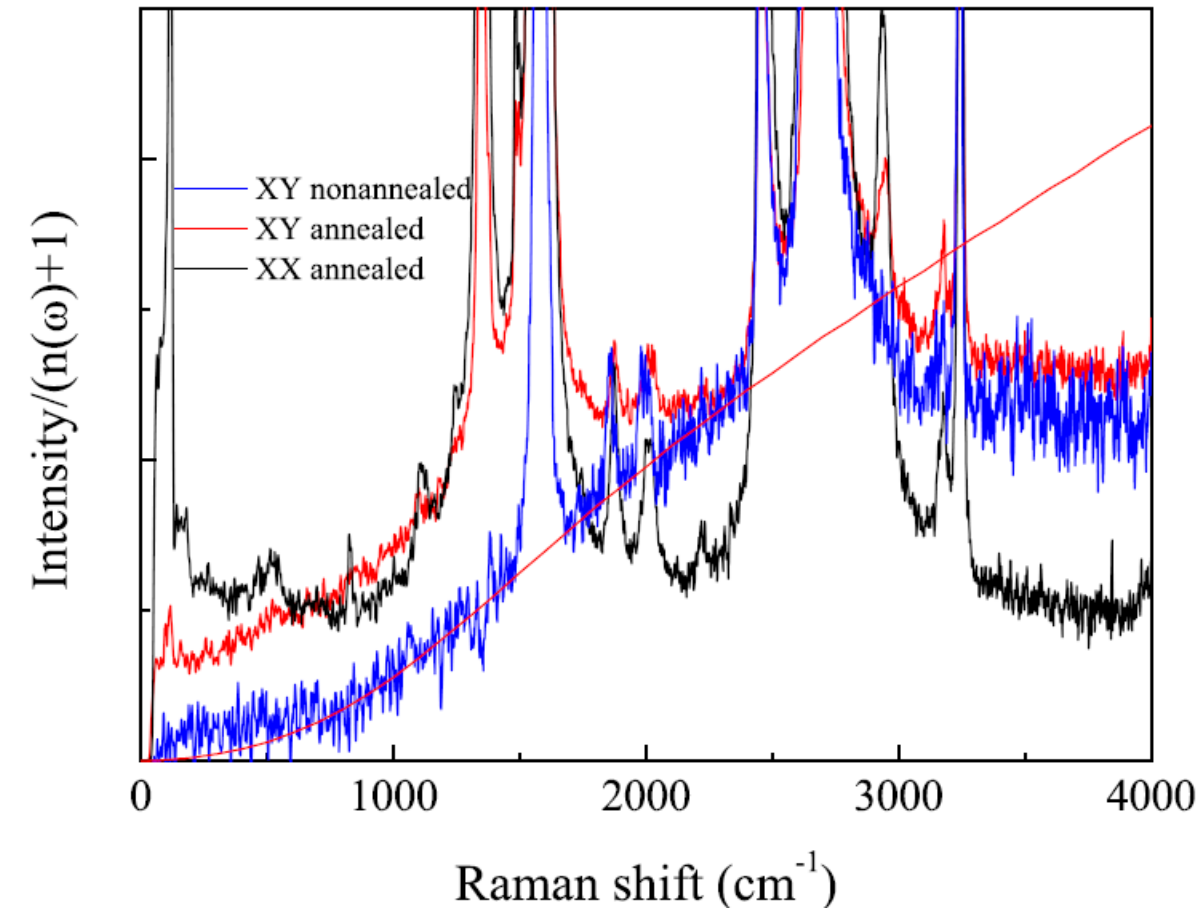


# ERS in graphene: the experiment



“ Thus one can see that electronic Raman scattering at zero magnetic field is a sensitive tool for probing the low-energy electronic structure and pseudospin symmetry in pure and doped graphitic structures, and it has a potential for studying gapped structures formed by different methods.

# ERS in graphene: the experiment



“  
m i n d s

Raman scattering  
the

gap

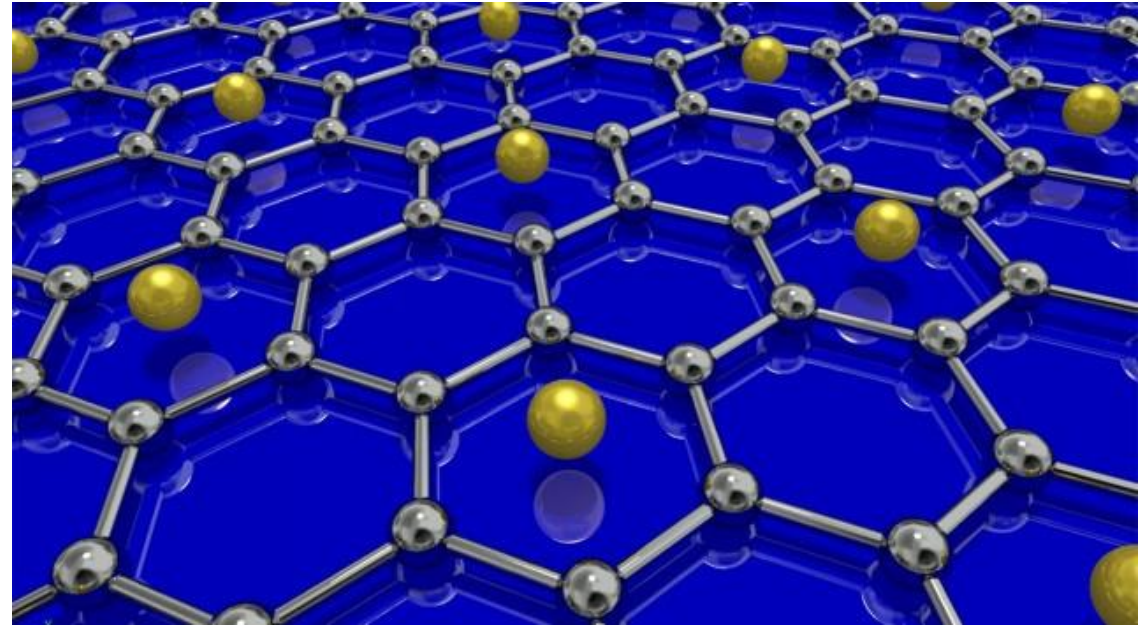
”

**What if we induce superconductivity?**

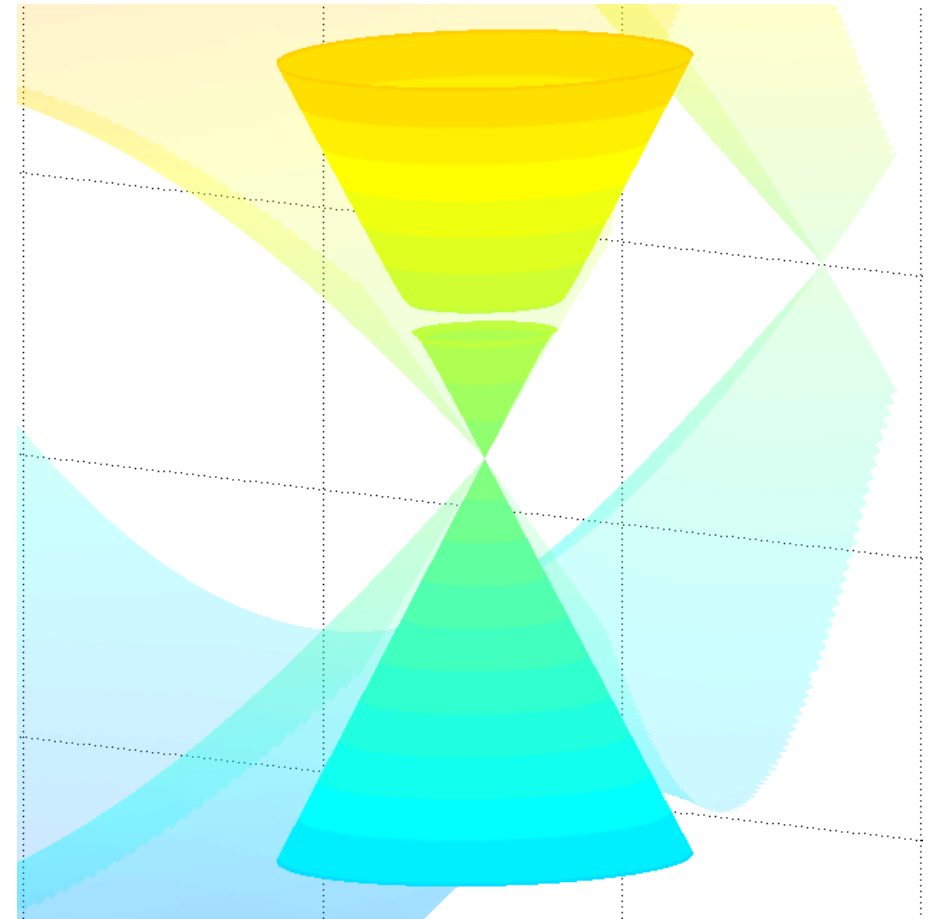
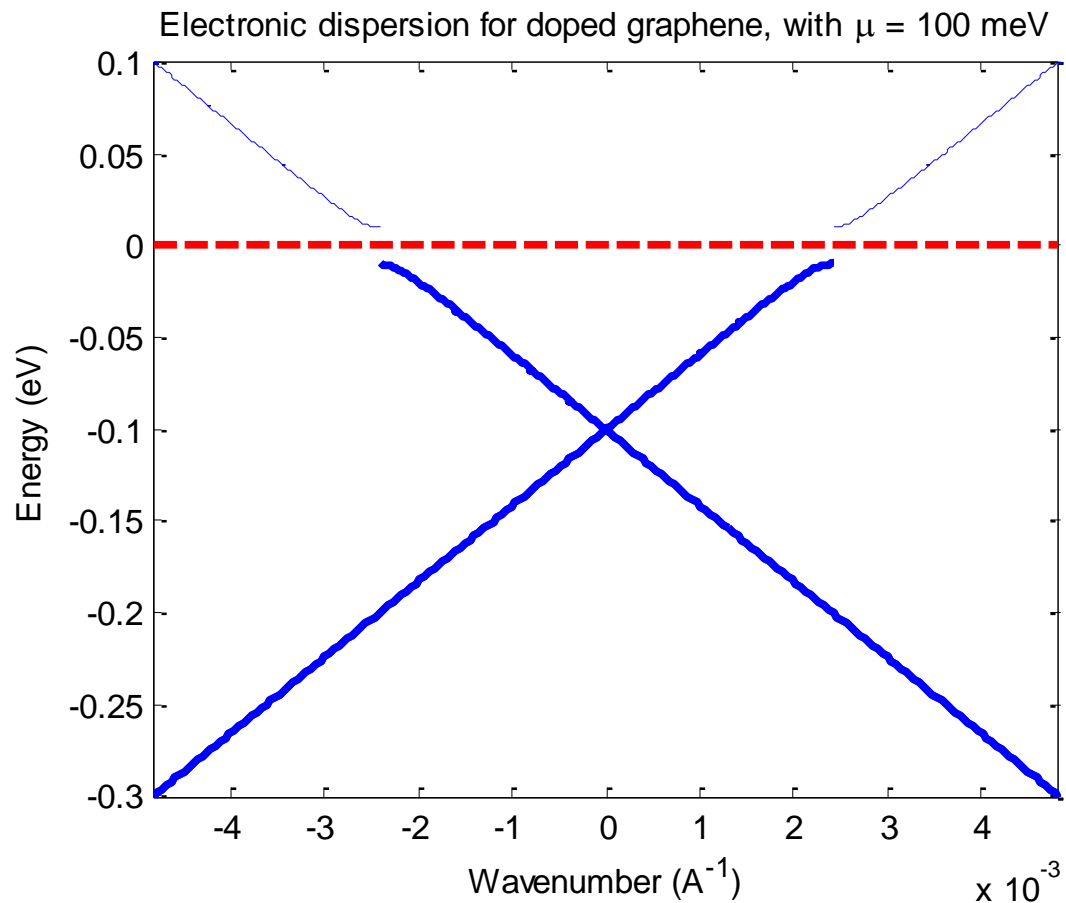


# Outline

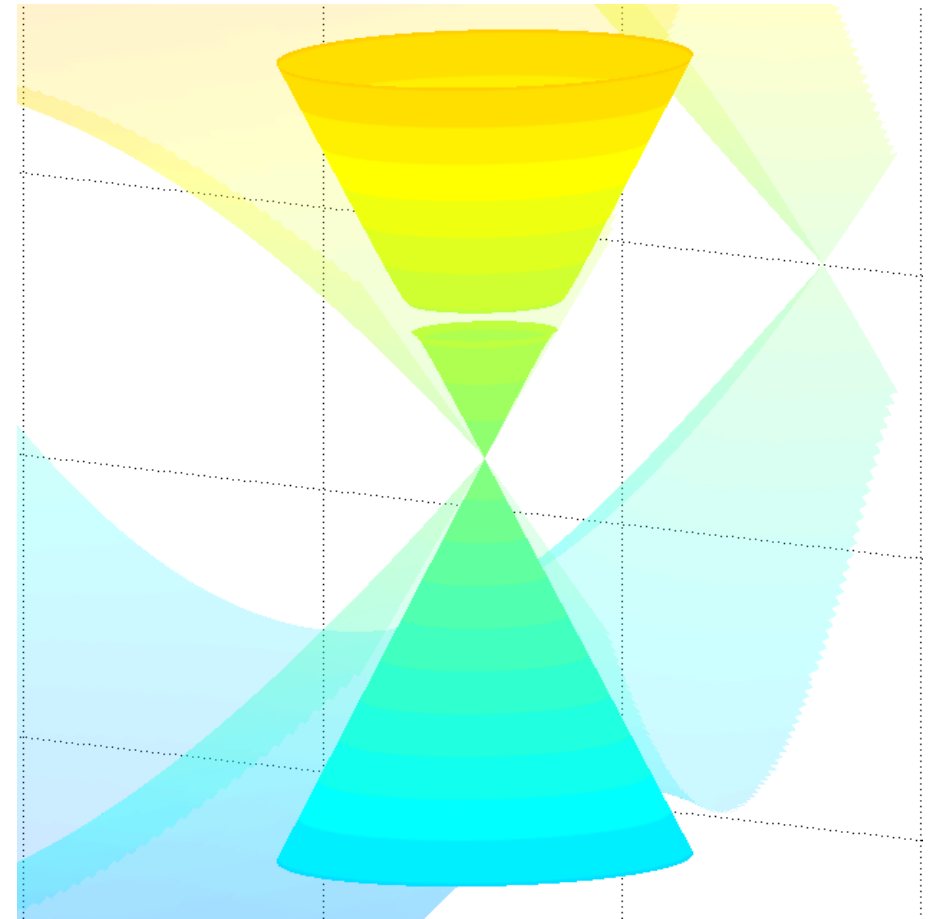
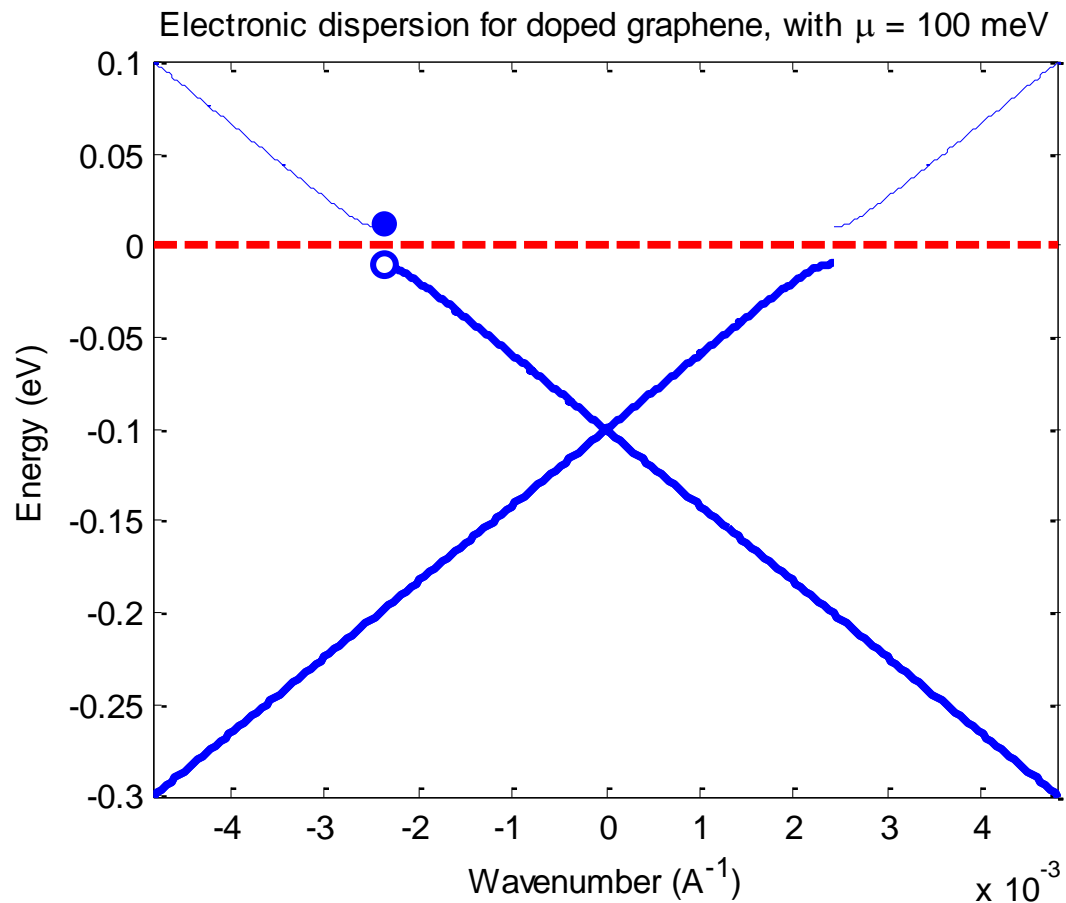
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  - Graphene
  - Raman Scattering in graphene
  - Electronic Raman scattering in graphene
- **ERS in superconducting graphene**
  - Superconducting graphene
  - **Electronic Raman scattering in superconducting graphene**



# ERS in superconducting graphene: the picture



# ERS in superconducting graphene: the picture



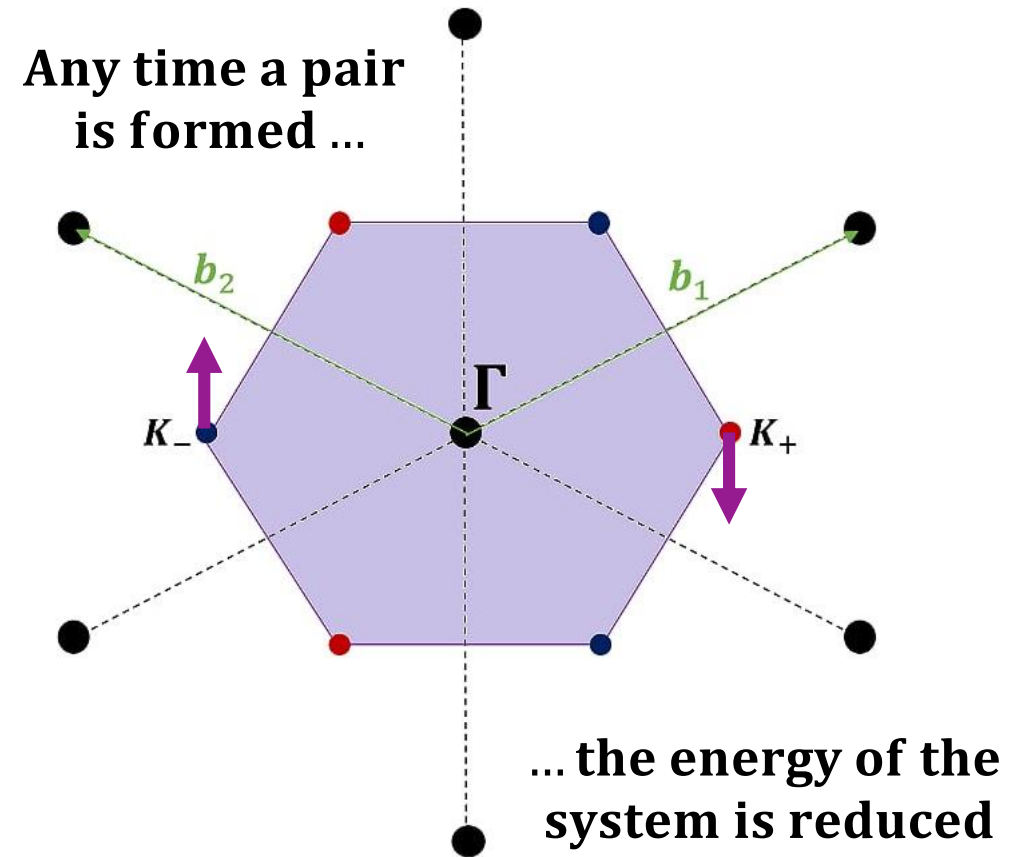
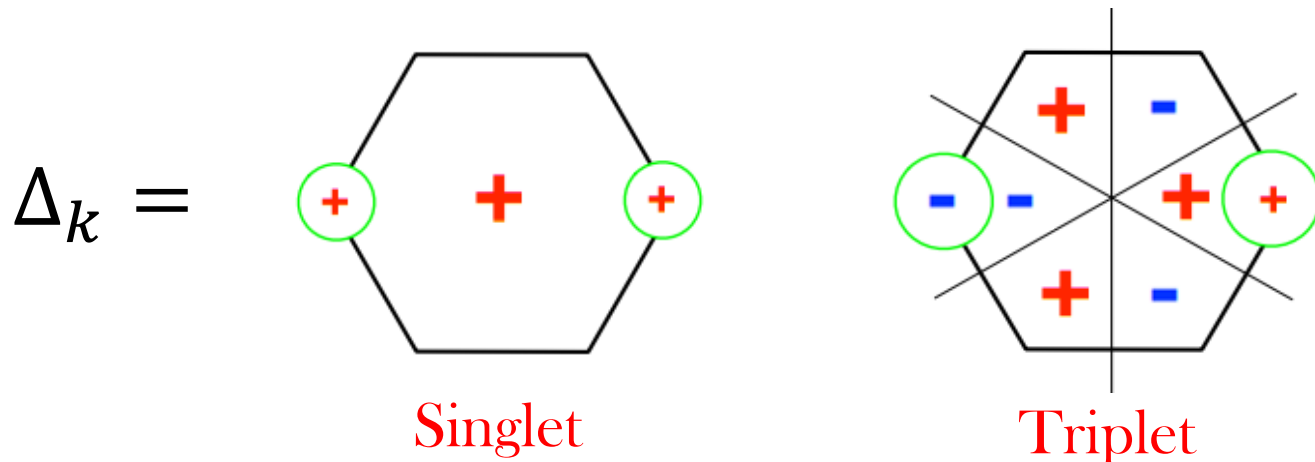


# ERS in superconducting graphene: the theory

$$\Psi_k = \boxed{\text{Spin}} \otimes \text{momentum}$$

## Order parameter

$$\Delta_k \equiv \langle \text{GS} | \Psi_k | \text{GS} \rangle$$



# ERS in superconducting graphene: the theory

Light-matter  
interaction

$$\vec{P}$$

$$\downarrow$$

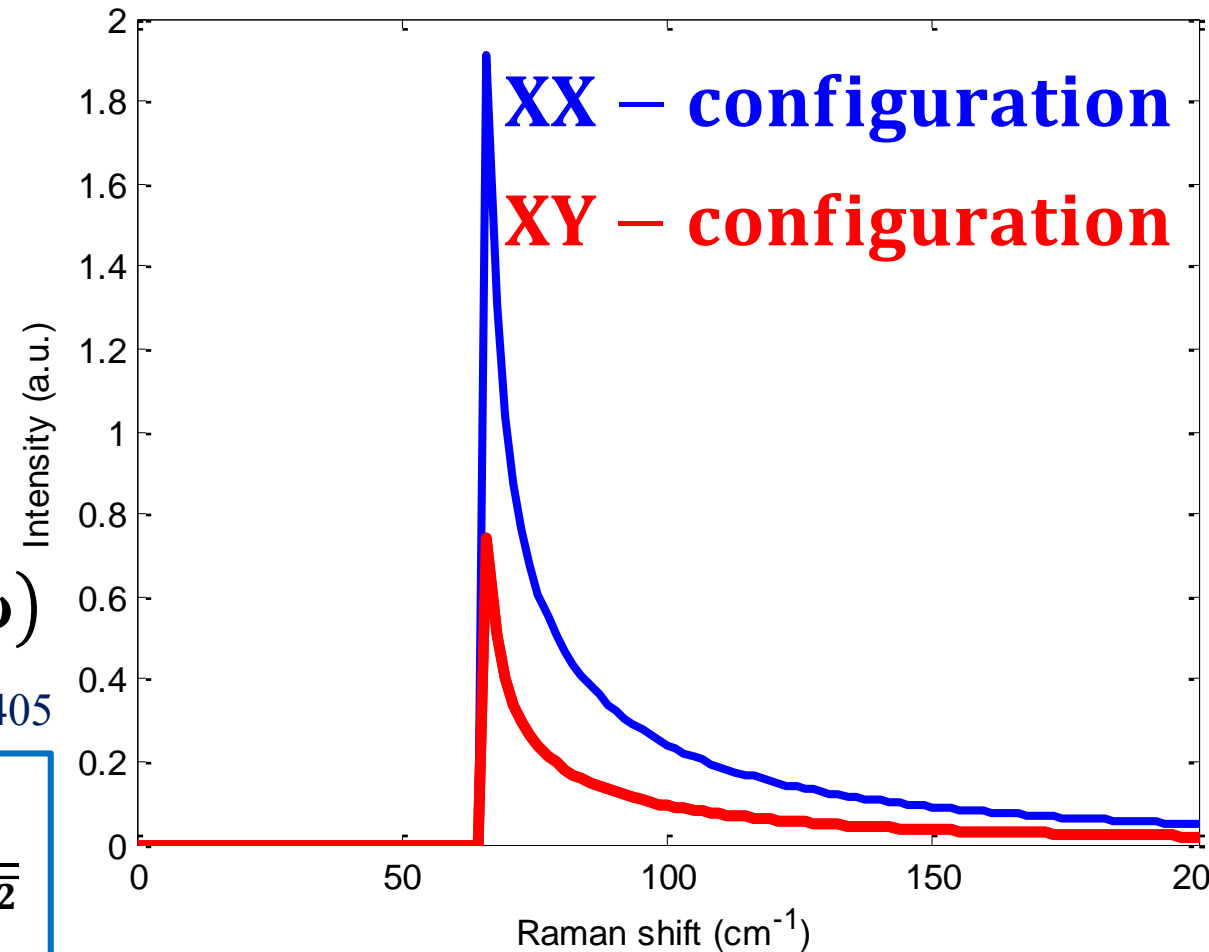
$$\vec{p} - \frac{e}{c} \vec{A}$$

$\mathcal{H}_{gr}^{lin}$

$$w(\omega) = \int \frac{dp}{2\pi\hbar^3} \left| \langle \mathbf{c}_2 | \mathcal{V}_{lin} | \mathbf{c}_1 \rangle \right|^2 \delta(\epsilon_f - \epsilon_i + \omega)$$

A. García-Ruiz, M. Mucha-Kruczyński, and V. I. Fal'ko, PRB **97**, 155405

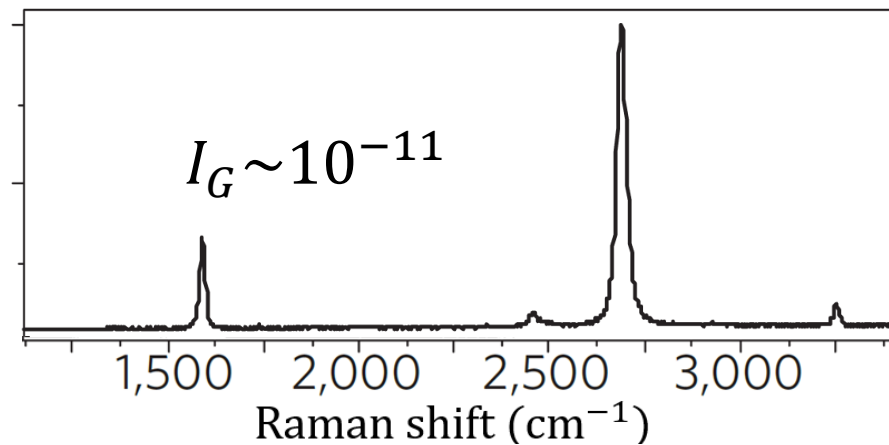
$$w(\omega) = \frac{8\hbar e^4 v^2 \mu}{\Omega^2} \left( \left( \frac{\mu}{\Omega^2} - \frac{\gamma_n}{2\gamma_0^2} \right)^2 \mathbb{E}'_s + \frac{\mu^2}{2\Omega^4} \mathbb{E}'_o \right) \frac{4\Delta}{\omega \sqrt{\omega^2 - 4\Delta^2}}$$



# ERS in superconducting graphene: the theory

**The challenge: Is this feature measurable?**

$$\text{Quantum efficiency} = \int d\omega \int d(\text{scattered angles}) w(\omega) \sim 10^{-14}$$



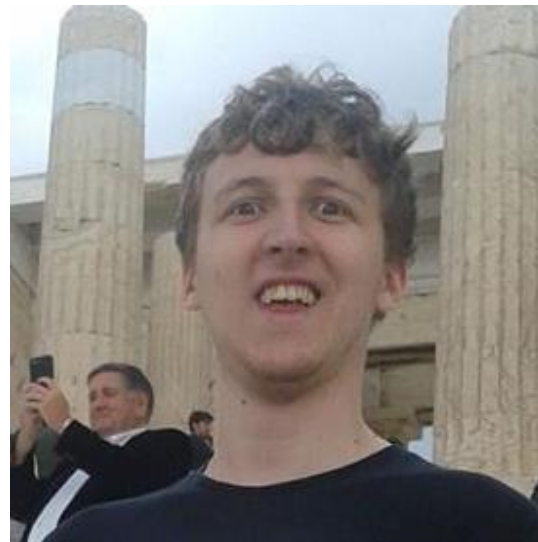




Marcin Mucha-Kruczynski



Stephen Clark



Joshua Thompson



Vladimir Falko

**#148**



*Superconductivity-induced features in the electronic Raman spectrum of monolayer graphene*

# Raman scattering minds the gap

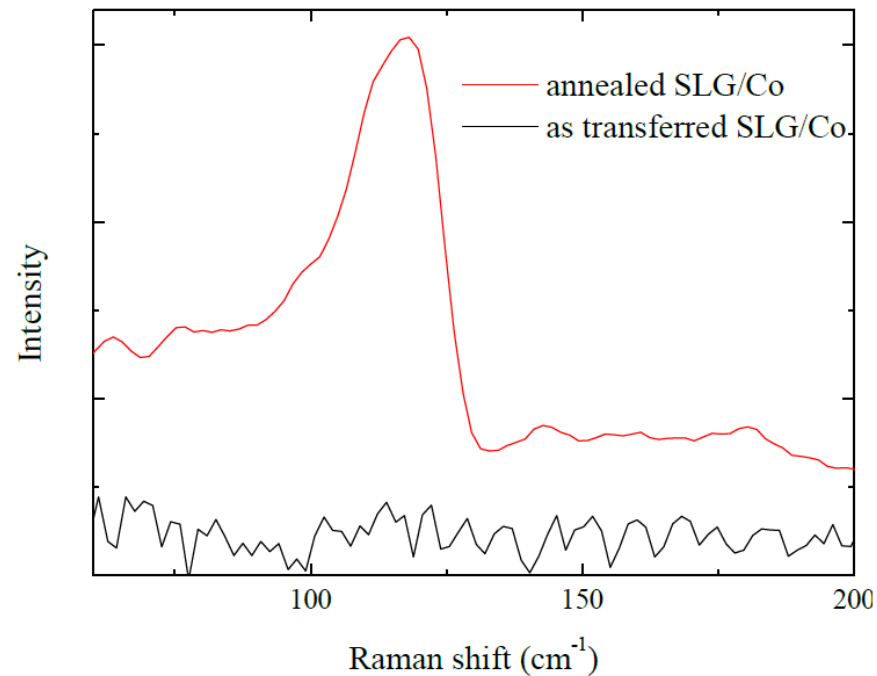




# Extra slide: peak at $117\text{cm}^{-1}$

Phys. Rev. B **91**, 195435 (2015)

[...] the *XX* spectrum of the annealed sample demonstrates a surprising appearance of the narrow intense band at  $117\text{ cm}^{-1}$  (see Fig. S3 in [30]). Its line shape is very asymmetric, showing the well-known Breit-Wigner-Fano (BWF) interference between the phonon and continuum [33]. This implies the **appearance of the defect-induced low-energy electronic excitations, perhaps of the intraband type discussed in [16,17]**.



Supplementary information

The heat treatment results in a strong intensity increase of the D mode at  $1346\text{ cm}^{-1}$  and an appearance of asymmetric low-frequency peak at  $117\text{ cm}^{-1}$ . This implies a growth of the defect density leading to selection rules violation and Raman activity of layer-breathing mode at  $117\text{ cm}^{-1}$ .



# ERS in superconducting graphene: the theory

Light-matter interaction

$$\vec{P} \rightarrow \vec{p} - \frac{e}{c} \vec{A}$$

$\mathcal{H}_{gr}^{lin}$

$$\downarrow$$

$$\mathcal{V}_{lin}$$

$\mathcal{H}_{gr}^{TW}$

$$\downarrow$$

$$\mathcal{V}_{TW}$$

$\mathcal{H}_{gr}^{nnn}$

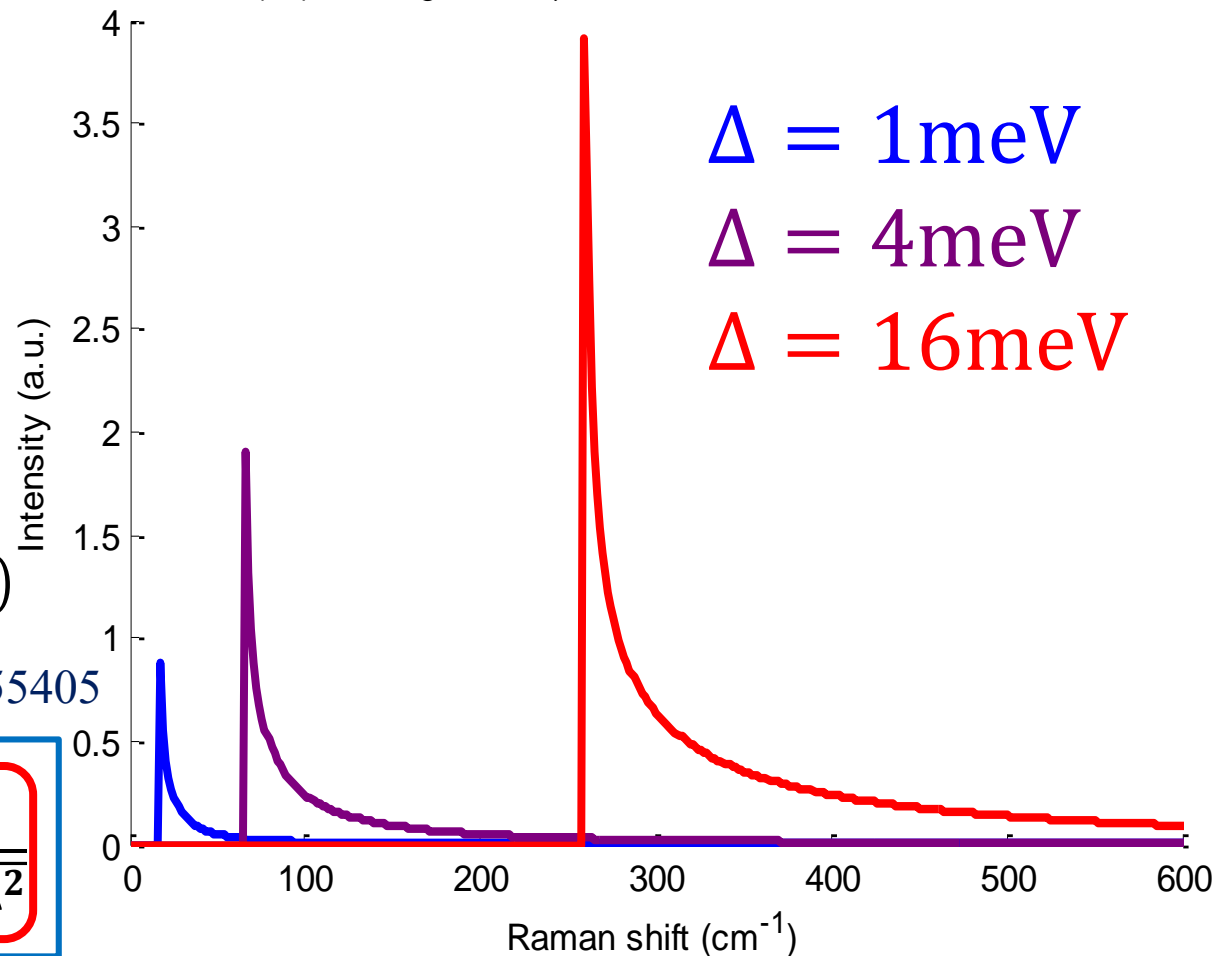
$$\downarrow$$

$$\mathcal{V}_{nnn}$$

$$w(\omega) = \int \frac{dp}{2\pi\hbar^3} \left| \langle f | \mathcal{V}_{lin} + \mathcal{V}_{TW} + \mathcal{V}_{nnn} | i \rangle \right|^2 \delta(\epsilon_f - \epsilon_i + \omega)$$

A. García-Ruiz, M. Mucha-Kruczyński, and V. I. Fal'ko, PRB 97, 155405

$$w(\omega) = \frac{8\hbar e^4 v^2 \mu}{\Omega^2} \left( \left( \frac{\mu}{\Omega^2} - \frac{\gamma_n}{2\gamma_0^2} \right)^2 \mathbb{E}'_s + \frac{\mu^2}{2\Omega^4} \mathbb{E}'_o \right) \frac{4\Delta}{\omega\sqrt{\omega^2 - 4\Delta^2}}$$



# ERS in superconducting graphene: the theory

Light-matter  
interaction

$$\vec{P} \rightarrow \vec{p} - \frac{e}{c} \vec{A}$$

$\mathcal{H}_{gr}^{lin}$

$$\downarrow$$

$$\mathcal{V}_{lin}$$

$\mathcal{H}_{gr}^{TW}$

$$\downarrow$$

$$\mathcal{V}_{TW}$$

$\mathcal{H}_{gr}^{nnn}$

$$\downarrow$$

$$\mathcal{V}_{nnn}$$

$$w(\omega) = \int \frac{dp}{2\pi\hbar^3} \left| \langle f | \mathcal{V}_{lin} + \mathcal{V}_{TW} + \mathcal{V}_{nnn} | i \rangle \right|^2 \delta(\epsilon_f - \epsilon_i + \omega)$$

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