Probing Electronic Interactions in Graphene by Optical Spectroscopy

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Columbia University. New York, NY

KITP Meeting

Topics

1. **Electron-electron interactions in single-layer graphene**

2. **Interlayer stacking effects in few-layer graphene**

   ABA and ABC stacking

Thanks to

Fai Mak

Joshua Lui

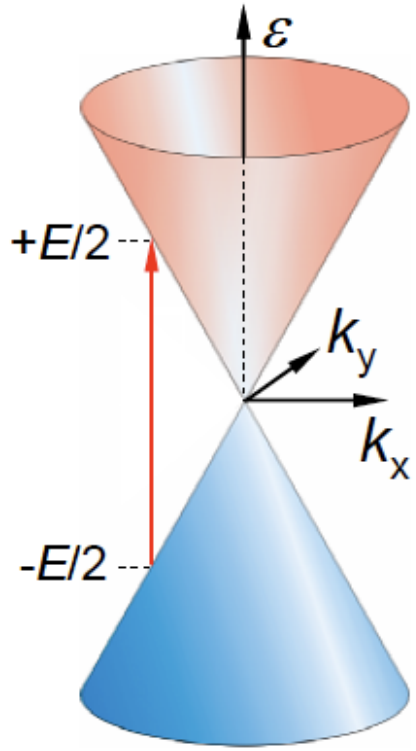
Zhiqiang Li

Prof. Jie Shan
(Case Western Reserve U.)

Steve Louie (UC Berkeley)
Theory: e-e interactions in SLG

Emmanuele Cappelluti
(Inst de Ciencia de Materiales, Madrid, Spain)
Theory: interlayer interactions

Interband Transitions: Optical Conductivity



Ando et al. J Phys. Soc. Jpn 71, 1310 (2002)
Gusynin et al., PRL 96, 256802 (2006)
Ryu et al., PRB 75, 205344 (2007)
Abergel et al., PRB 75, 155430 (2007)

Interband transitions for ideal linearly dispersing bands

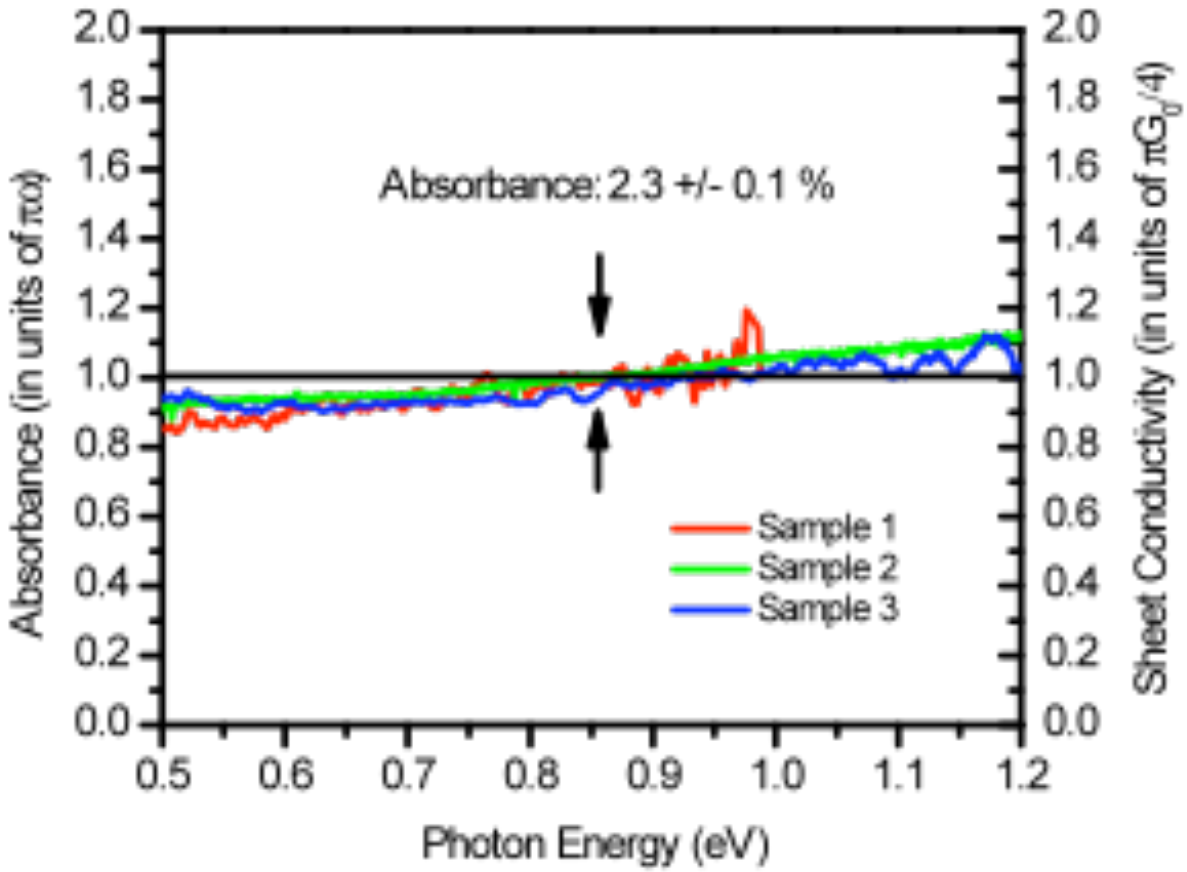
Optical (sheet) conductivity:

$$\sigma^{(1)}(\omega) = \frac{\pi e^2}{2h} = (\pi/4)\, G_0$$

Optical absorption:

$$A(\omega) = \frac{\pi e^2}{\hbar c} = \pi\alpha = \frac{\pi}{137} = 2.3\%$$

Graphene Optical Absorption/Conductivity



Universal behavior

Also Manchester group
[Science 320, 5881 (2008)]

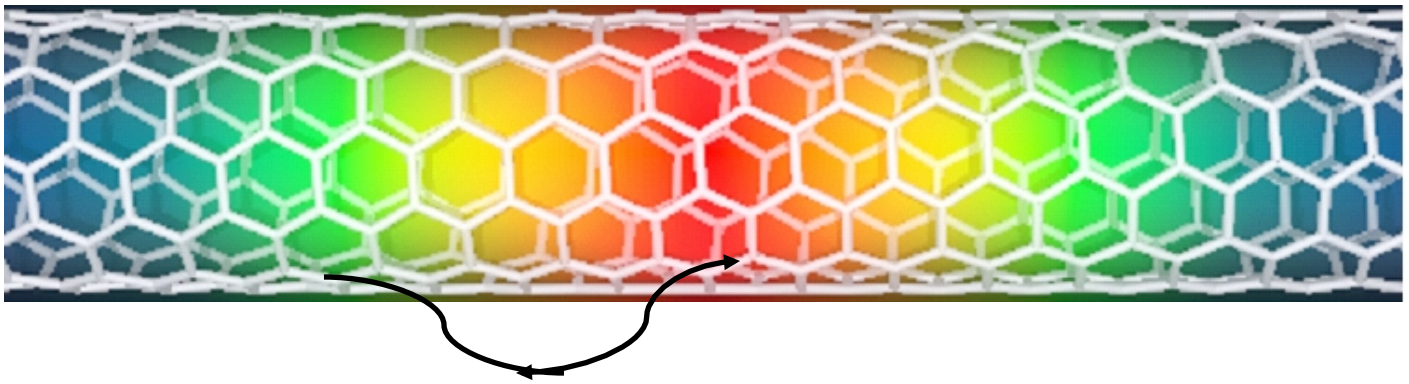
[Columbia grp, PRL 101, 196405 (2008)]

Role of Many-Body Effects

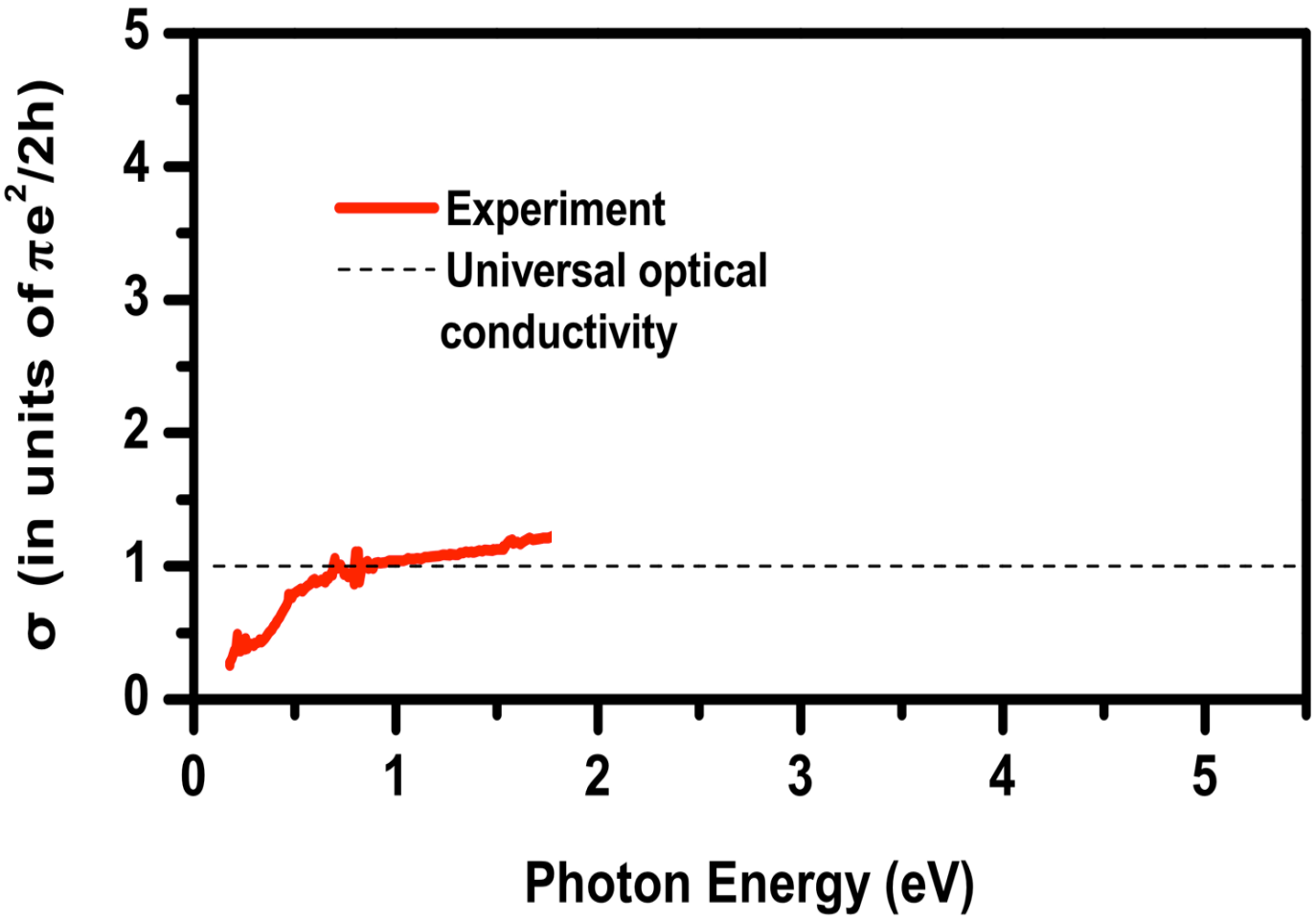
All optical processes involve electron and hole: Exciton formation?

Dominant in semiconducting carbon nanotube

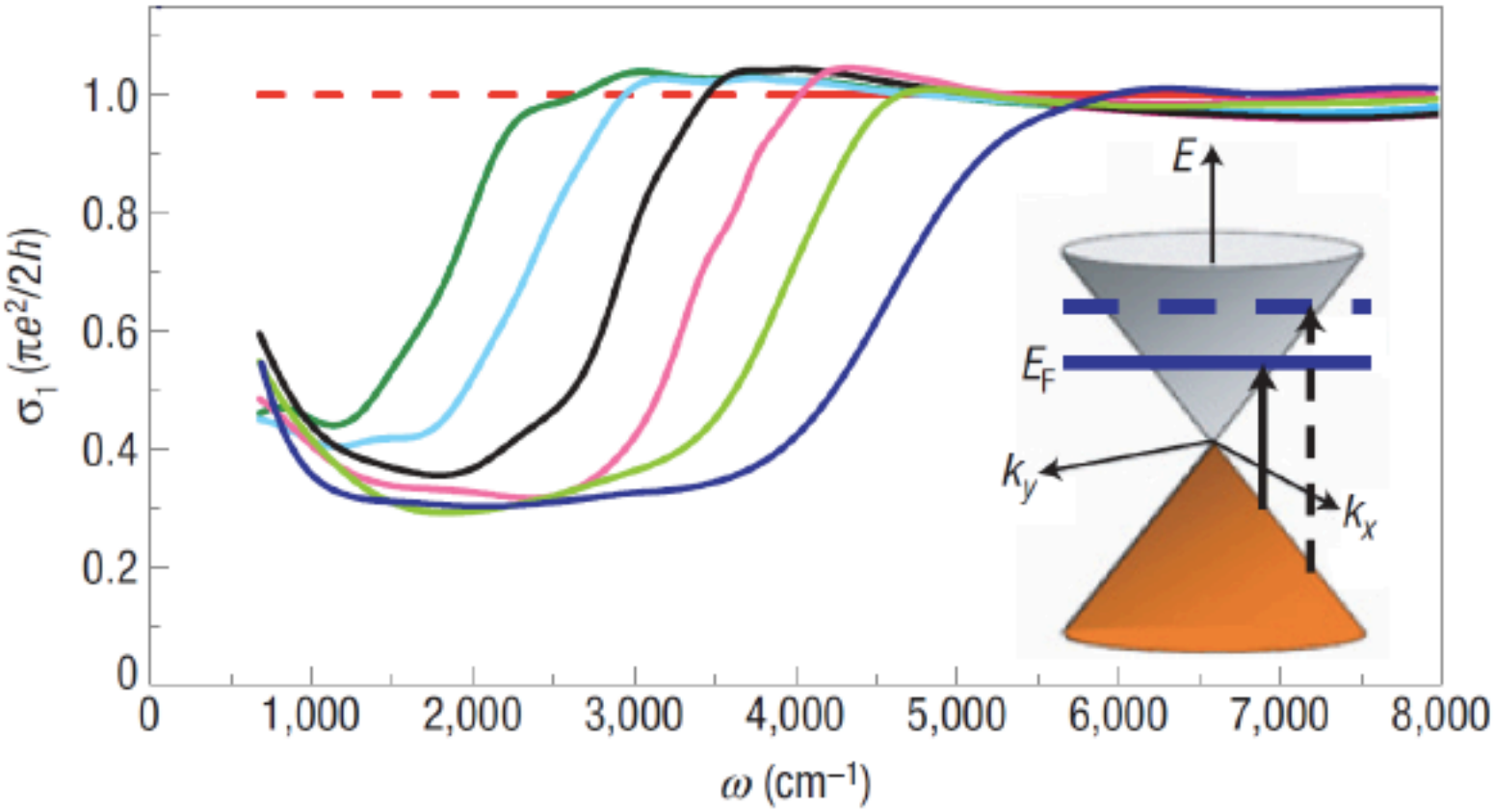
Significant even in metallic nanotubes



Graphene Absorption Spectrum

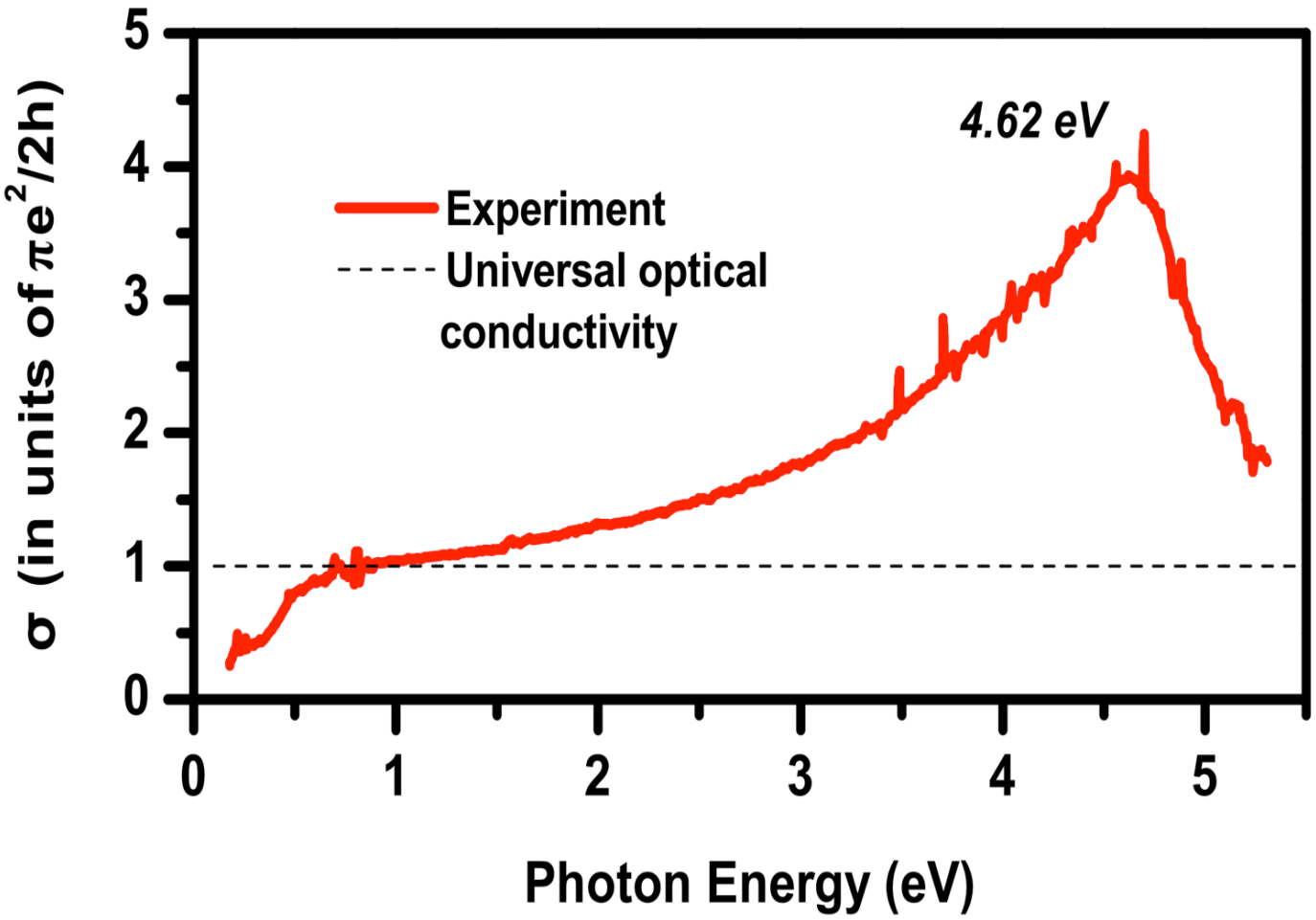


Tunable Graphene Absorption by Electrostatic Gating

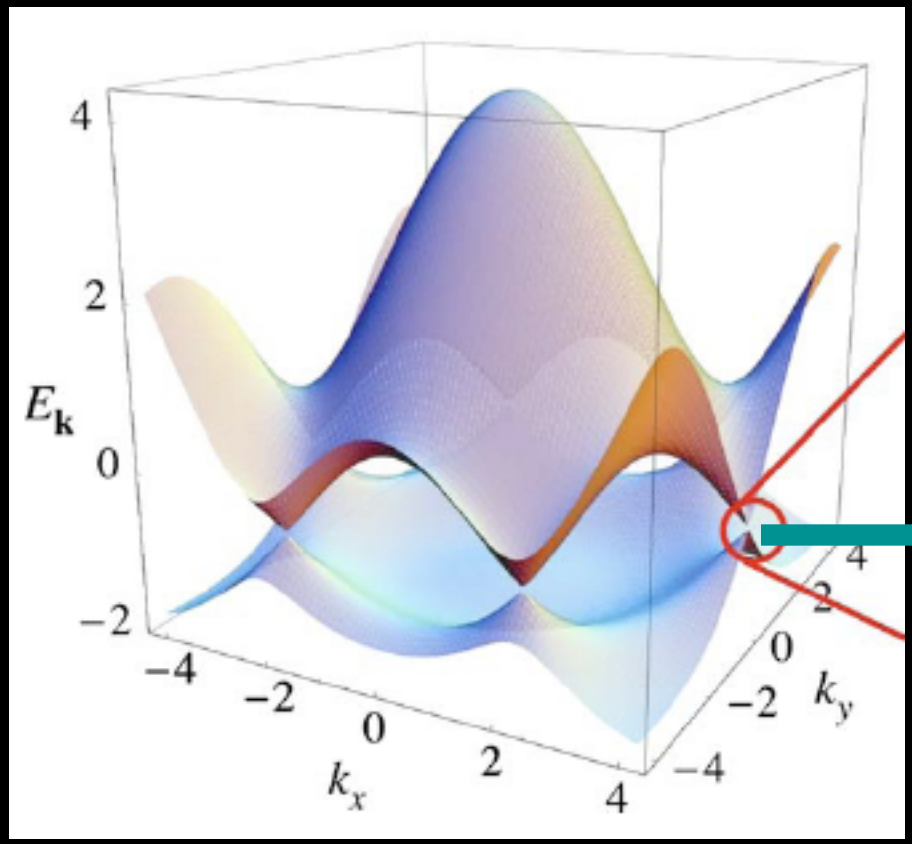


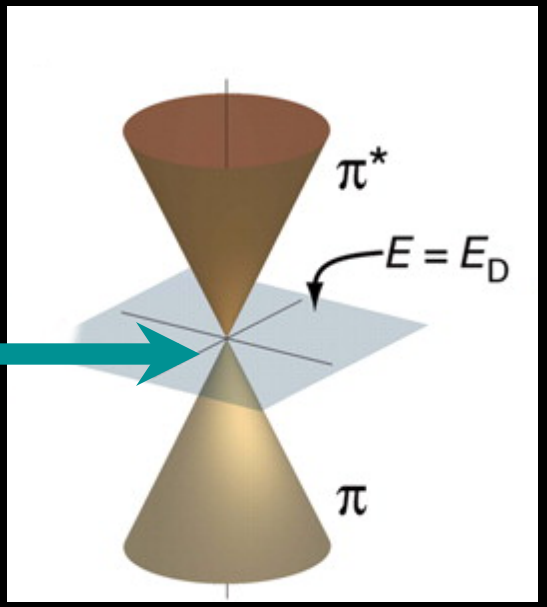
D. Basov et al., UC San Diego
Also: Feng Wang, UC Berkeley

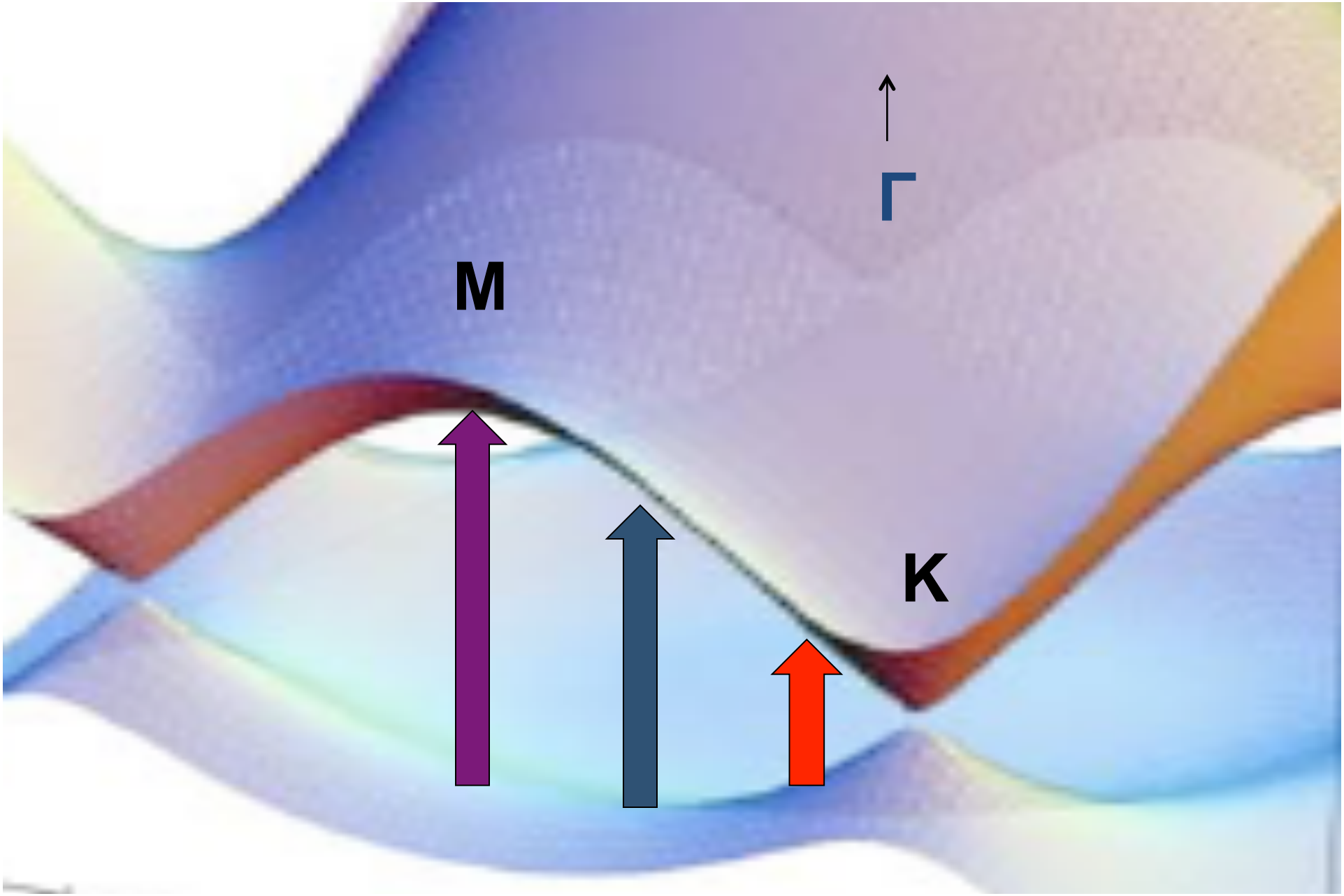
Graphene Absorption Spectrum



Graphene Electronic Structure



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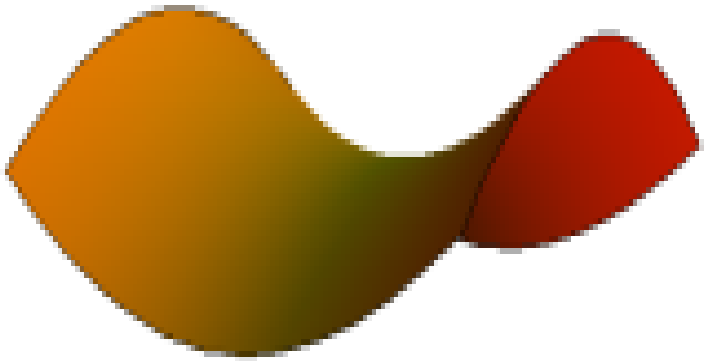
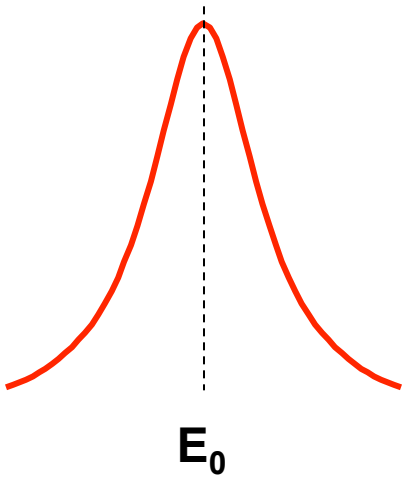
Absorption at Saddle Point (no e-h interactions)

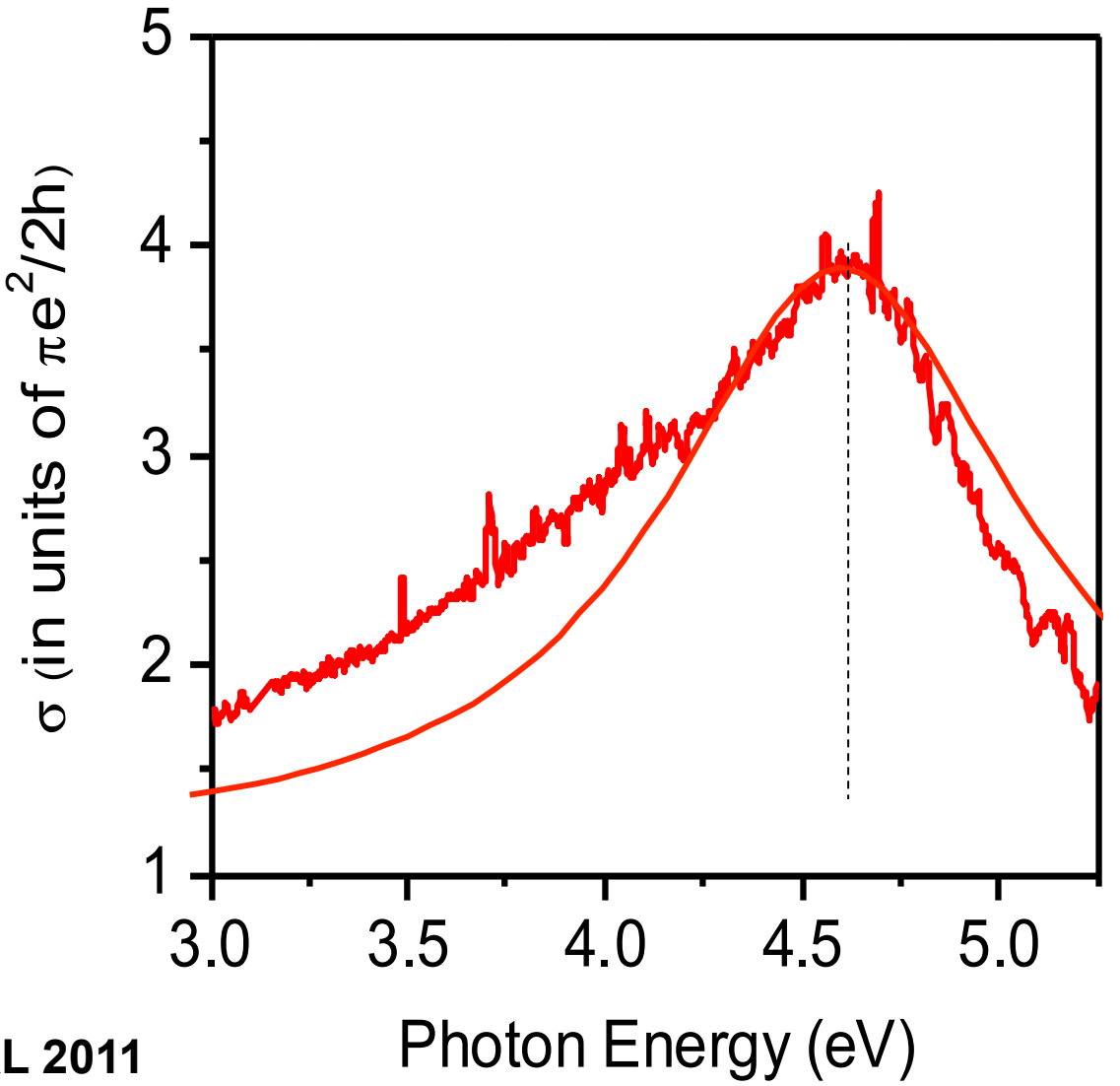


$$DOS_{SP}^{2D}(\omega) \propto -\ln\left|1-\frac{\omega}{E_0}\right|$$



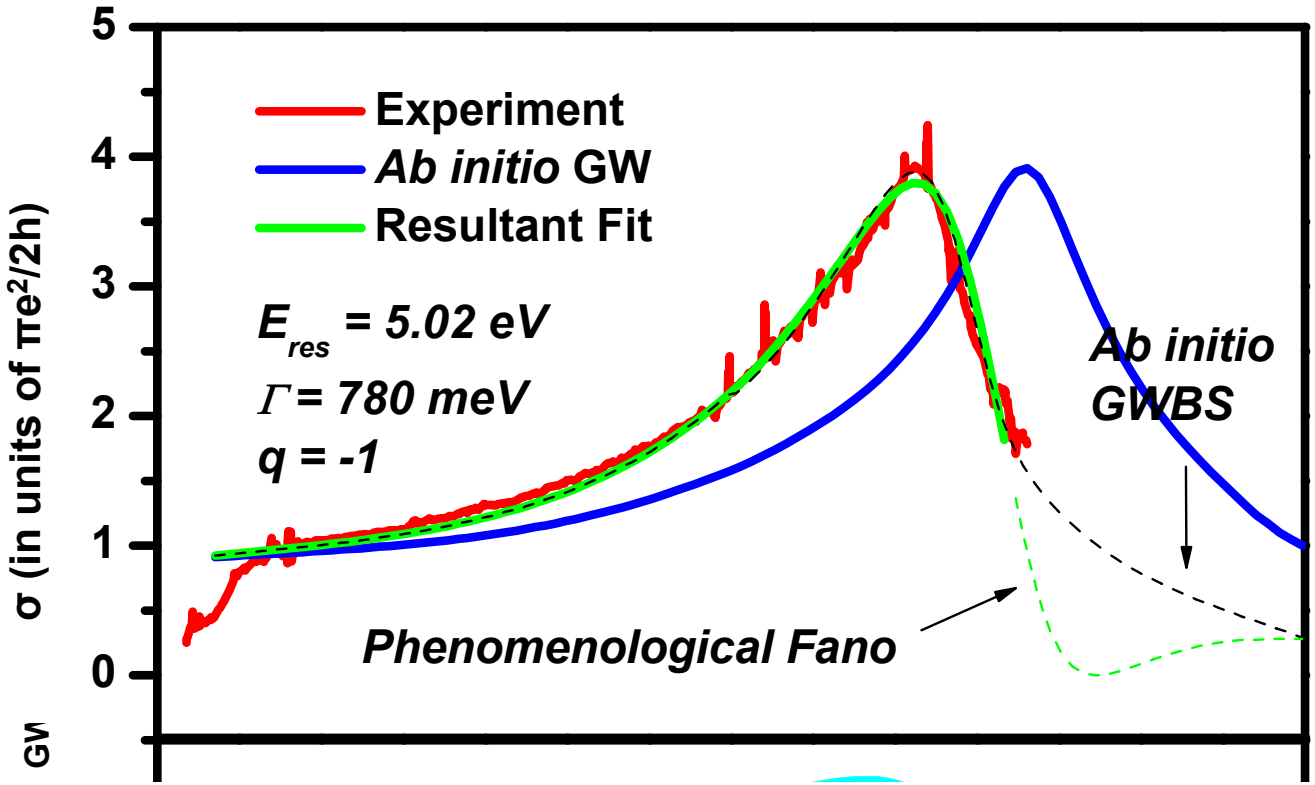
Symmetric line shape near the saddle point

Absorption Peak in Graphene



Heinz group, PRL 2011
Also Manchester, Stuttgart groups

Excitonic Effects at the Saddle Point



Yang et al. PRL 103, 186802 (2009)

Mak et al. PRL 106, 046401 (2011)

Existence of strong e-h interactions

Quasiparticle lifetime near the M-point ~ 10 fs

Summary 1

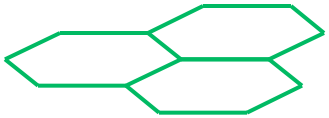
SINGLE-LAYER GRAPHENE: e-e interactions

- Major increase in absorption above πα for higher photon energies, rising to close to 10% at 4.6 eV
- Peak is associated with saddle-point at M-point in BZ.
- *Position and asymmetry lineshape require strong excitonic correlations:*

  Saddle-point exciton -- discrete state embedded in continuum

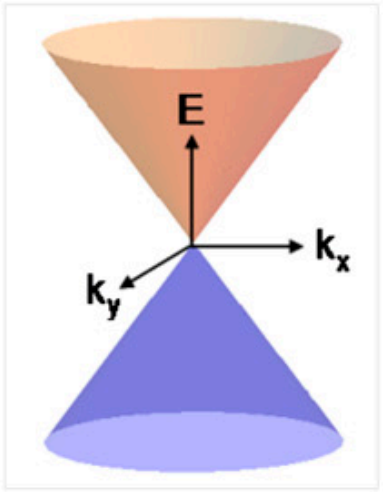
  Fano absorption profile

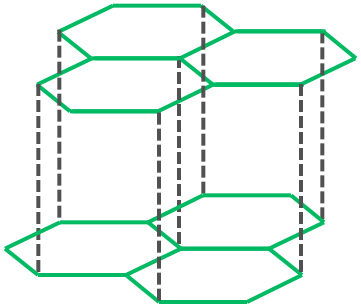
Changing the Low-Energy Band Structure by Layer Stacking



Graphene Monolayer

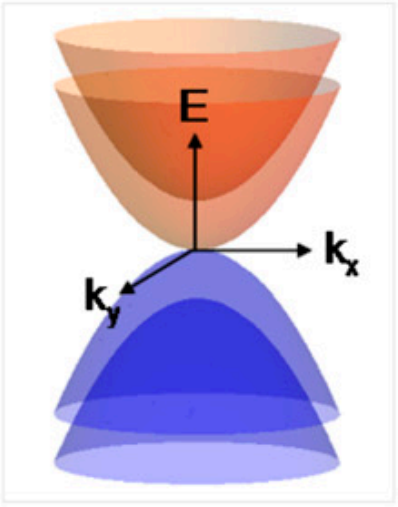
Chiral massless fermions



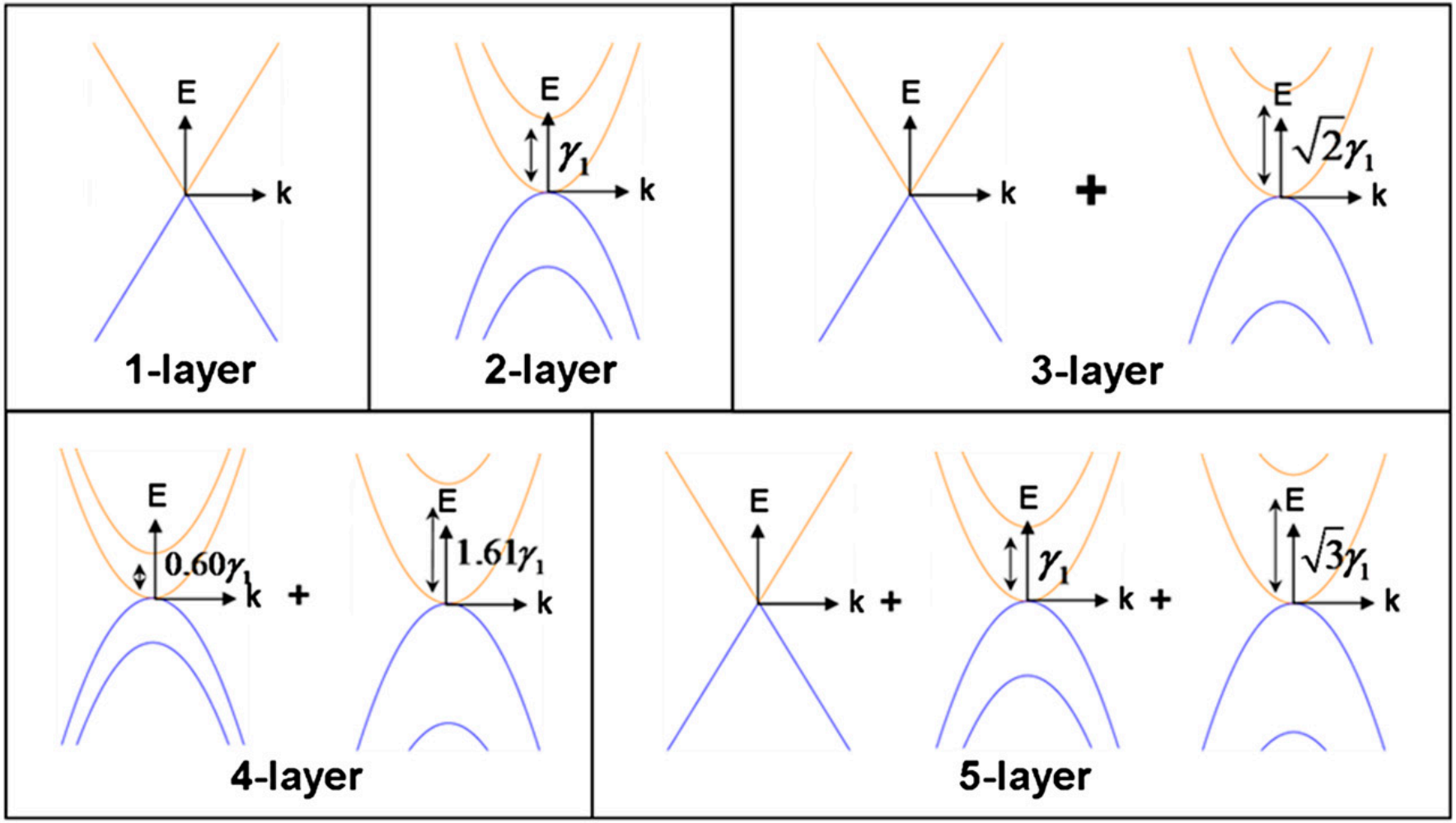


Graphene AB Bilayer

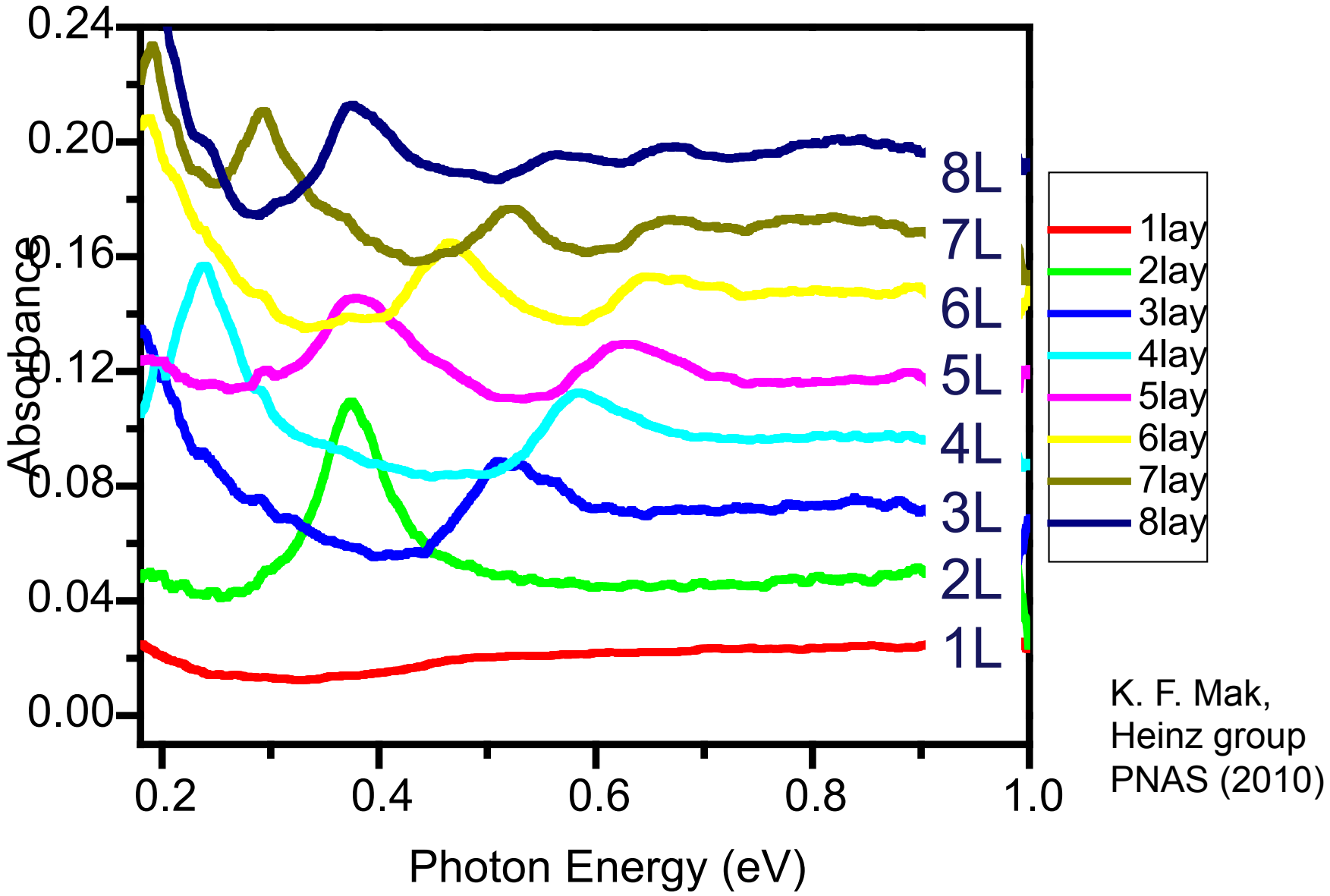
Chiral massive fermions



Few-layer Bernal-stacked graphene

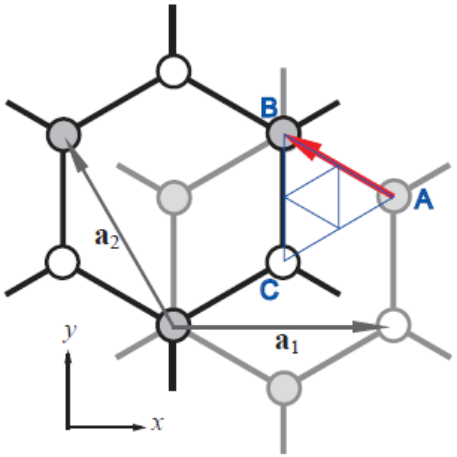


Few-Layer Graphene Electronic Absorption Spectra

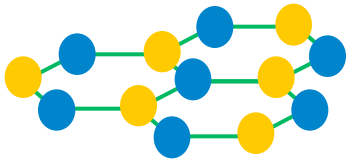


K. F. Mak,
Heinz group
PNAS (2010)

Stacking order in few-layer graphene

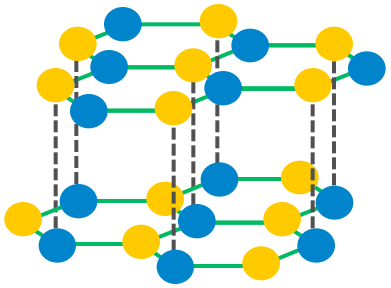


1-Layer

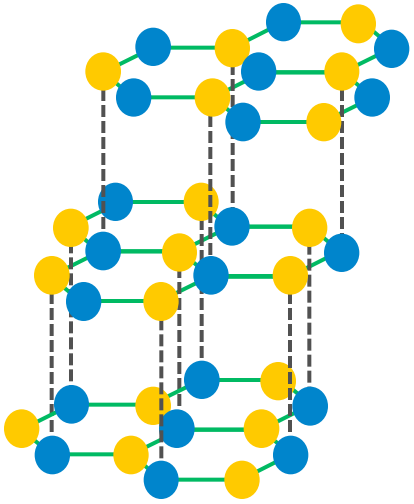


AB = AC

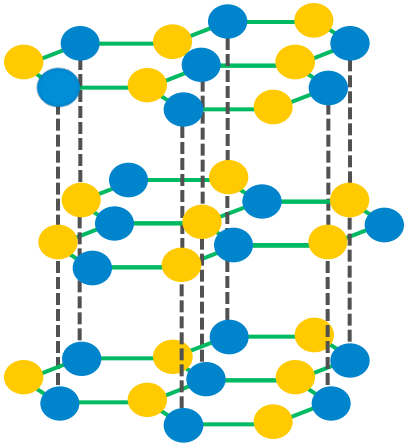
2-Layer



3-Layer



ABC



ABA

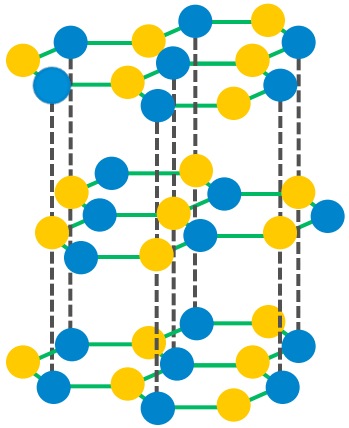
S. Latil and L. henrard, PRL 036803 (2006)

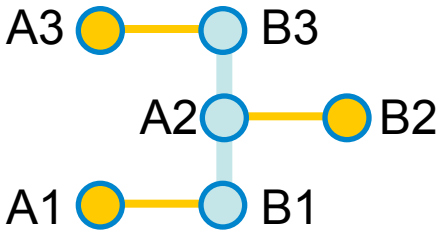
M. Aoki and H. Amawashi, Solid State Commun. 142, 123 (2007)

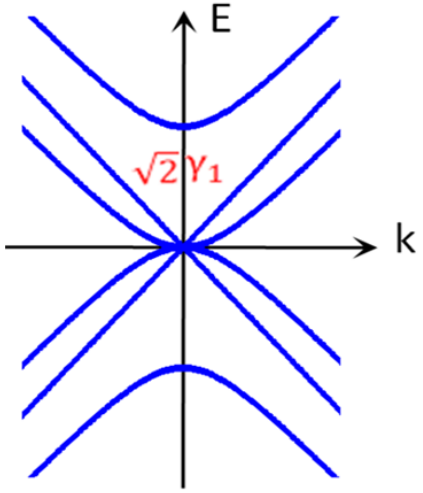
Trilayer graphene

ABA

Mirror symmetry



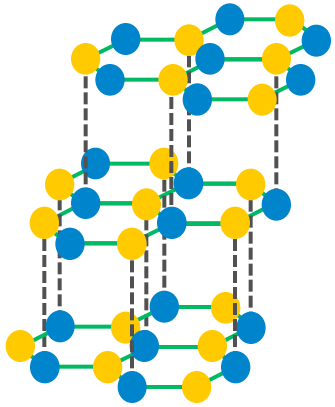


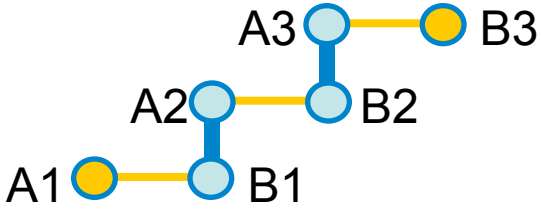


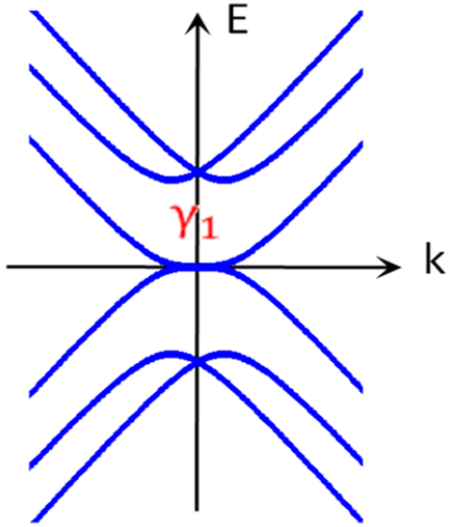
(b) ABC

ABC

Inversion symmetry





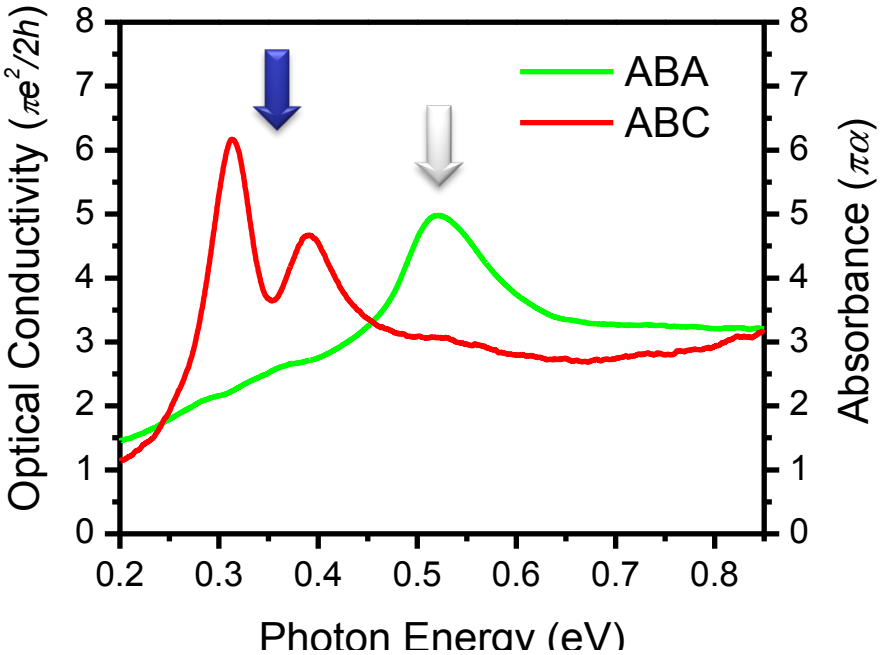


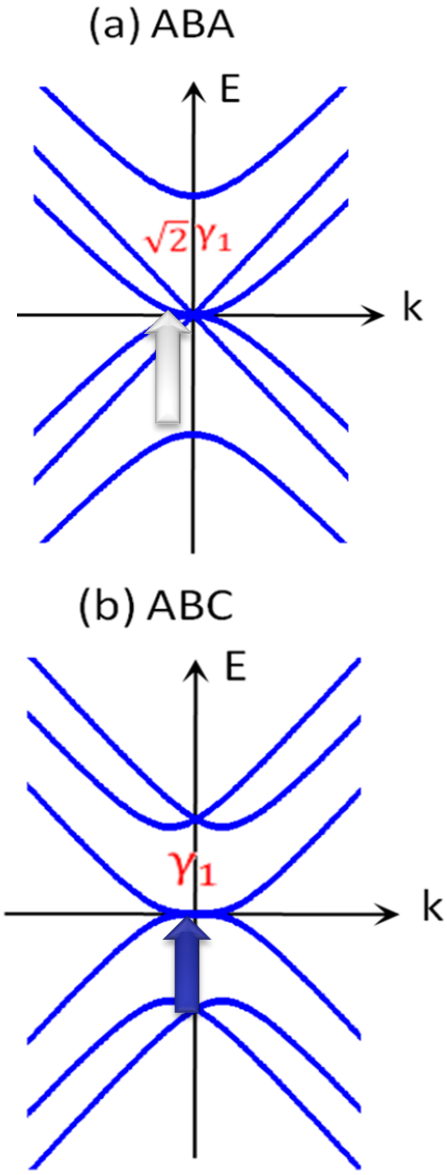
M. Aoki and H. Amawashi, Solid State Commun. 142, 123-127 (2007)

Trilayer graphene

- Characterization of stacking order by infrared and Raman spectroscopy
- Controlling the electronic structure by electrical gating – inducing a band gap

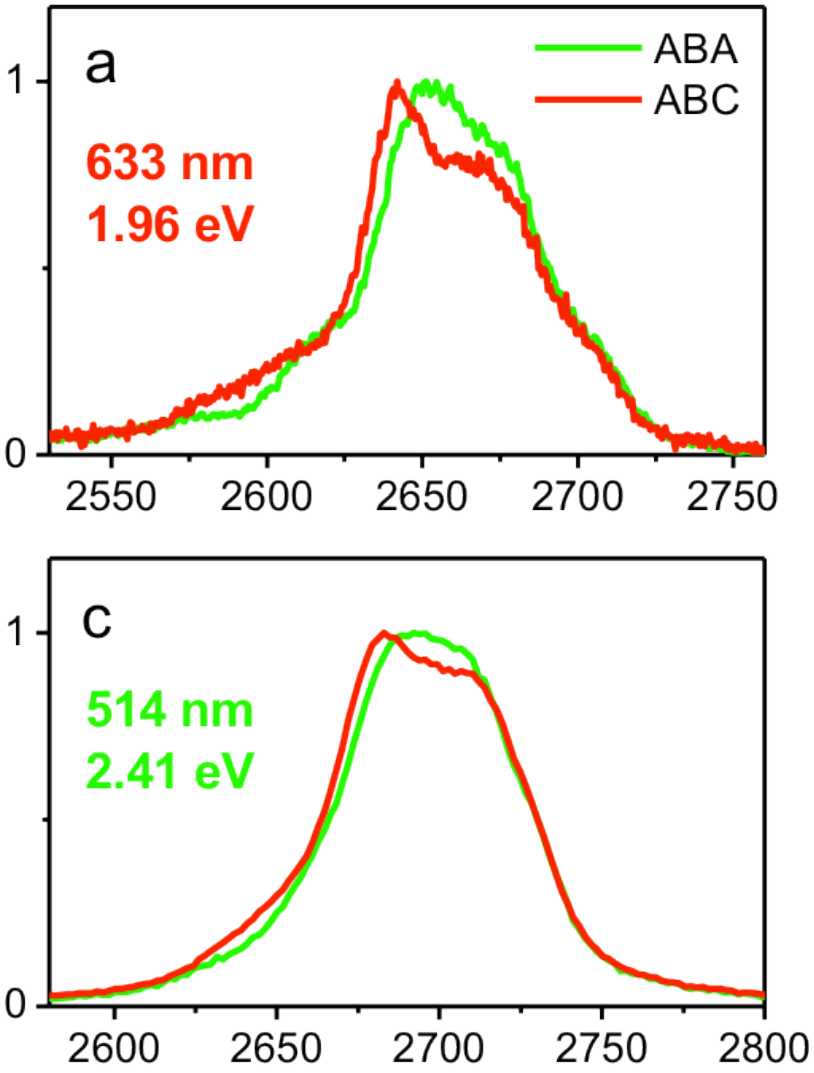
Infrared absorption of trilayer graphene

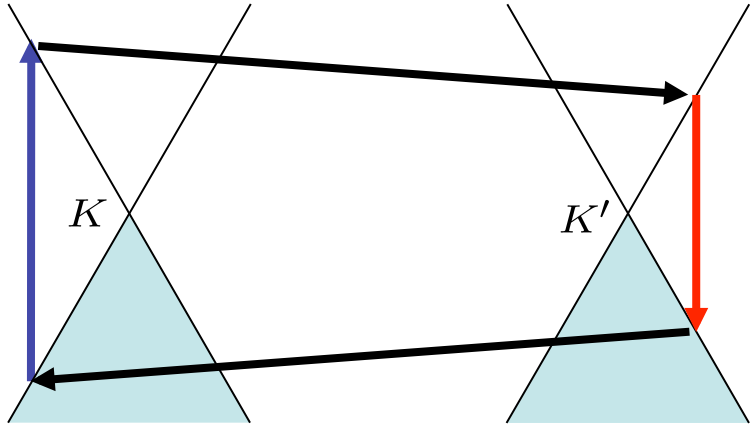




C. H. Lui, TFH *et al, Nano Lett.*, *11,* 164–169 (2011)

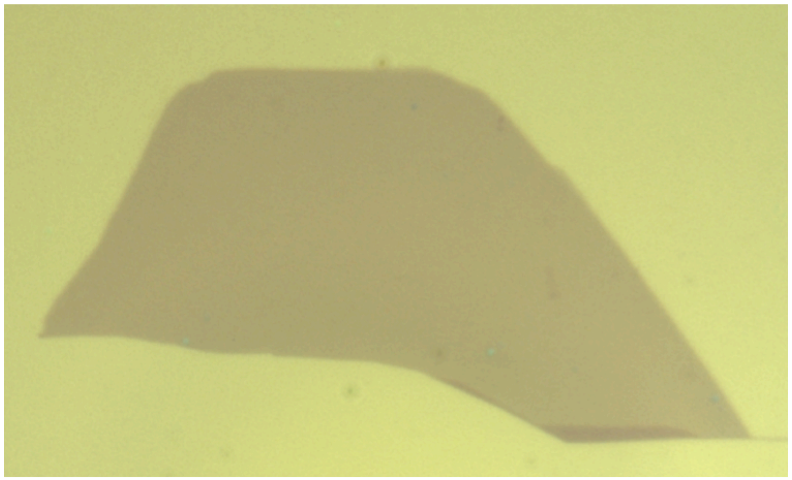
Raman 2D/G' spectra

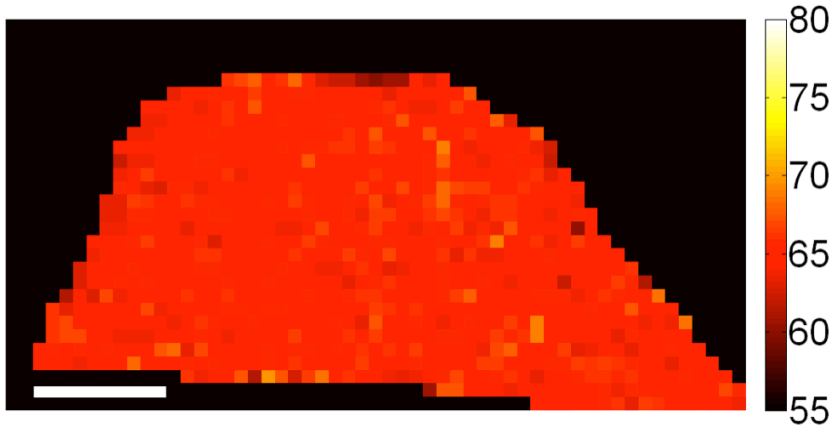


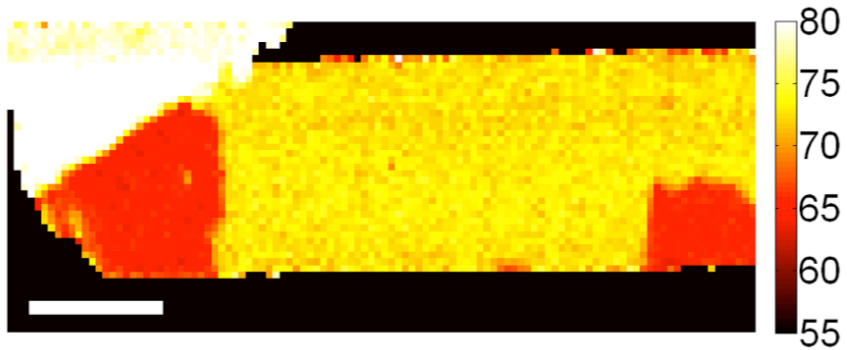


C. H. Lui, TFH *et a.l, Nano Lett.*, *11,* 164–169 (2011)

Raman images of 2D linewidth







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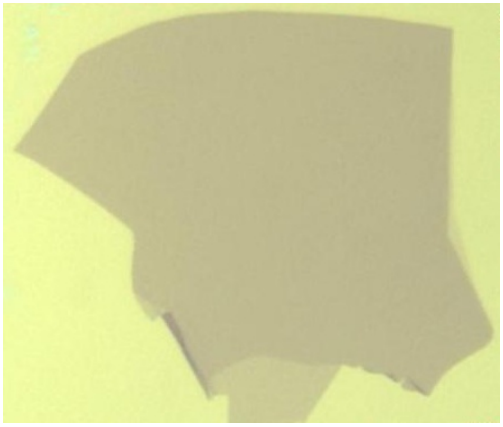
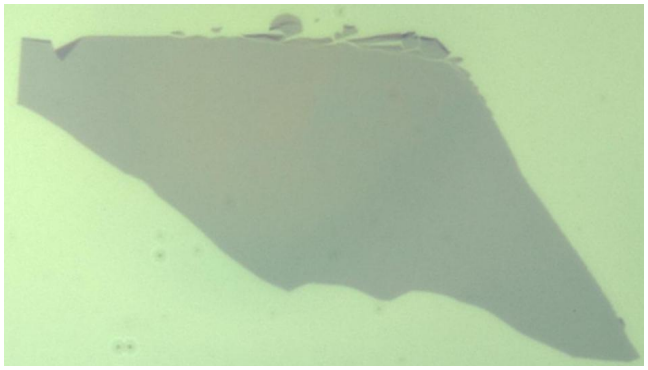
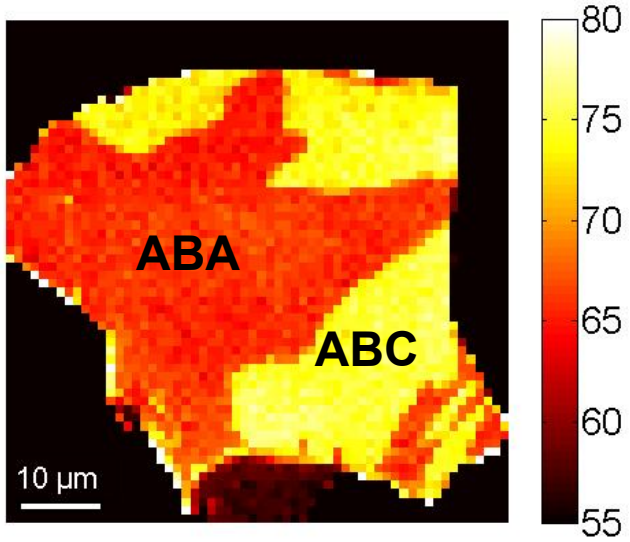
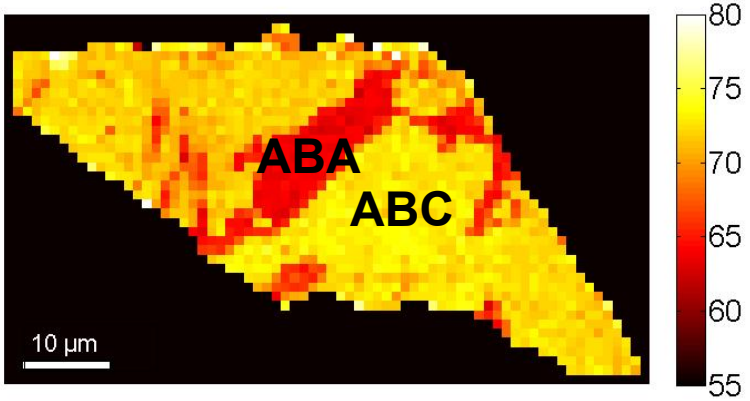








C. H. Lui, TFH *et a.l, Nano Lett.*, *11,* 164–169 (2011)

Statistics

Trilayer graphene

- total sample number: 45
- Purely ABA samples: 26
- Purely ABC samples: 0
- Mixed-stacking samples: 19
- ABA/ABC area ratio: **85:15**

Graphite (X-ray diffraction studies)

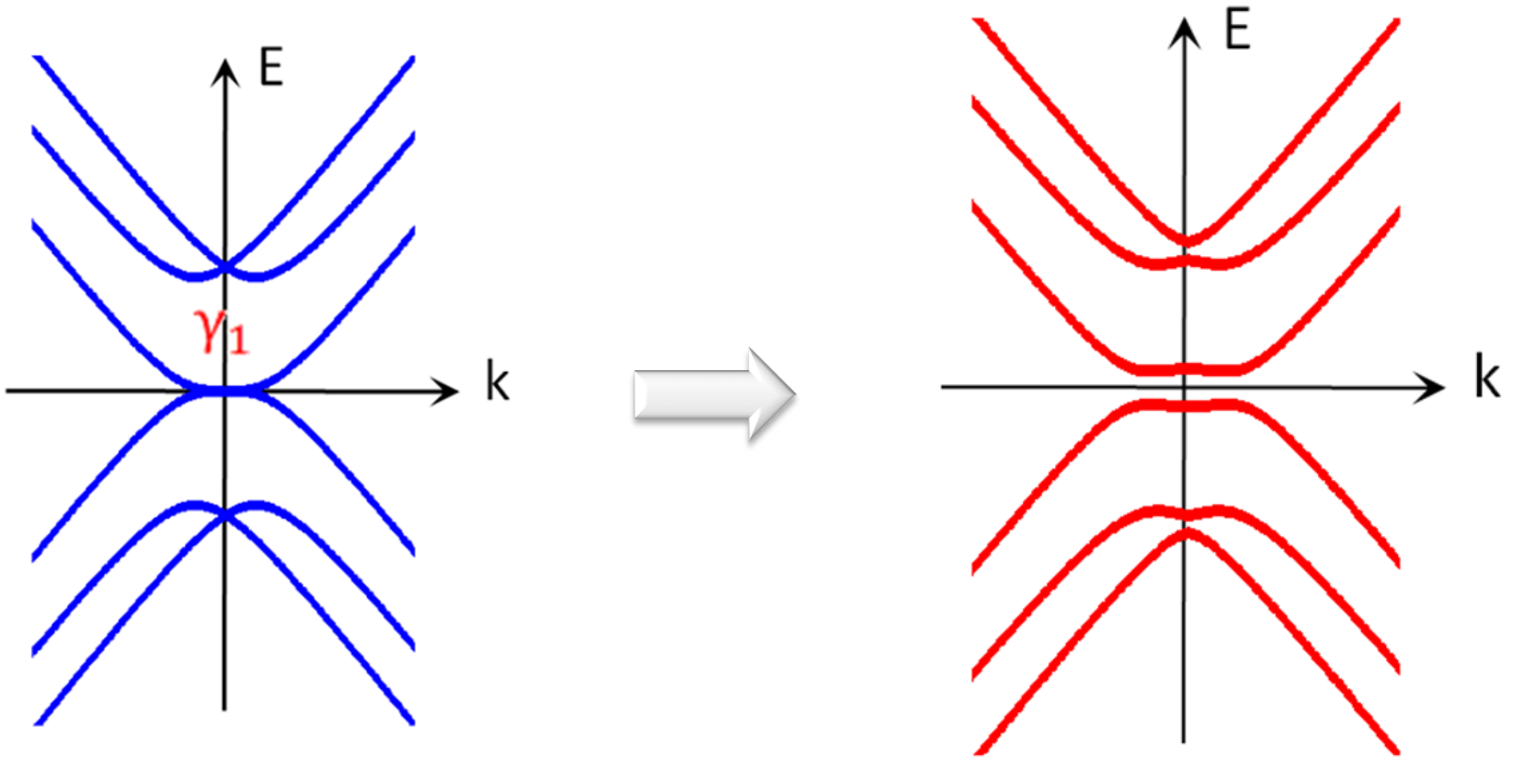
- **14% of rhombohedral (ABC) structure;**
- 6% of disordered structure;
- 80% of Bernal (ABA) structure.

1. Lipson, H., and Stokes, A. R., Proc. Roy. Soc., A, **181**, 101 (1942).
2. Rooksby, H. P. and E. G. Steward, E. G., Nature **159**, 638 (1947)

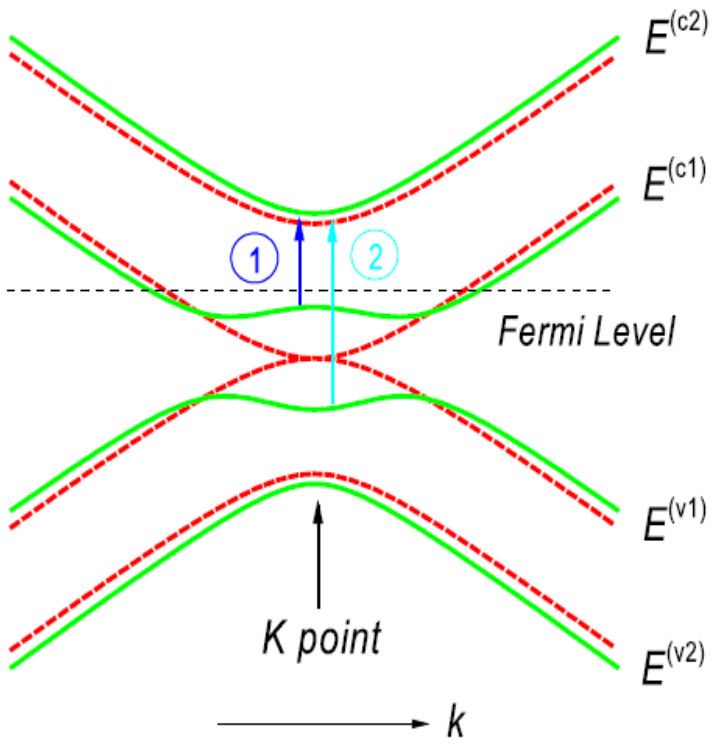
Graphene trilayers inherit the structure of graphite.

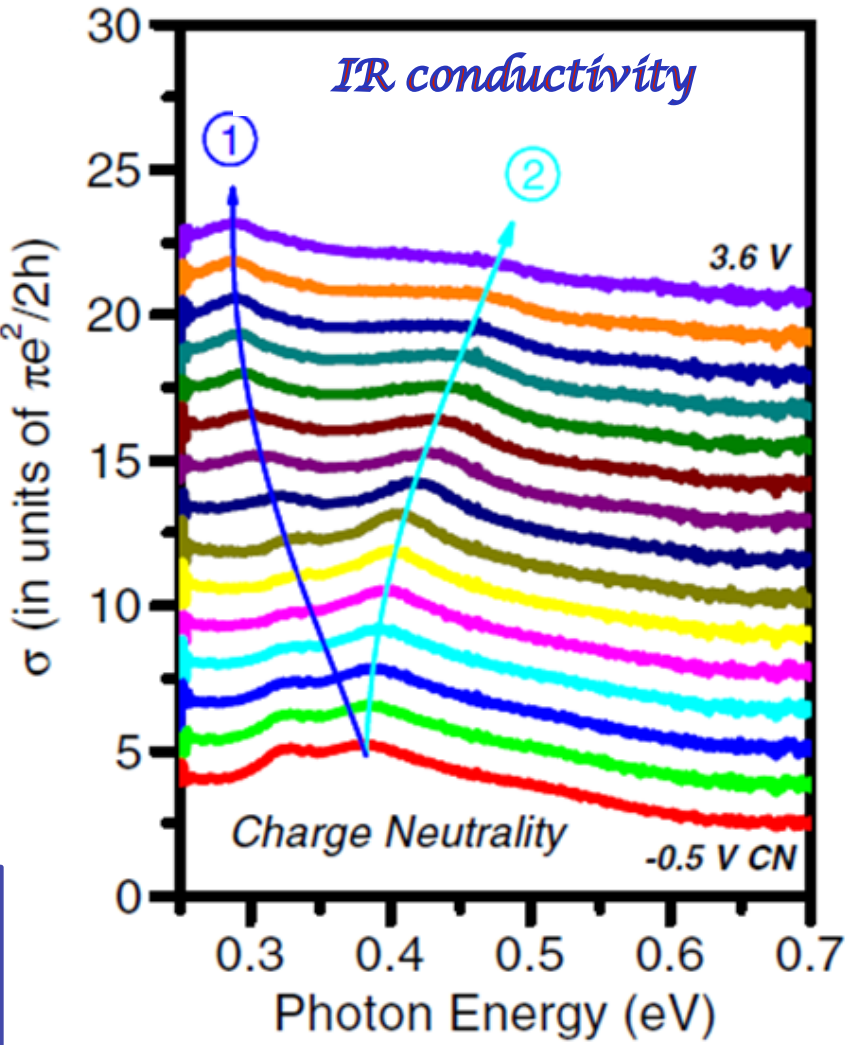
C. H. Lui *et al, Nano Lett.*, *11,* 164–169 (2011)

Controlling the Electronic Structure of Trilayer Graphene by Applied E-Fields



Probing the bilayer band gap by IR





[K. F. Mak *et al*, PRL 102, 256405 (2009).]

[Y. B. Zhang *et al*, Nature 459, 820 (2009)]

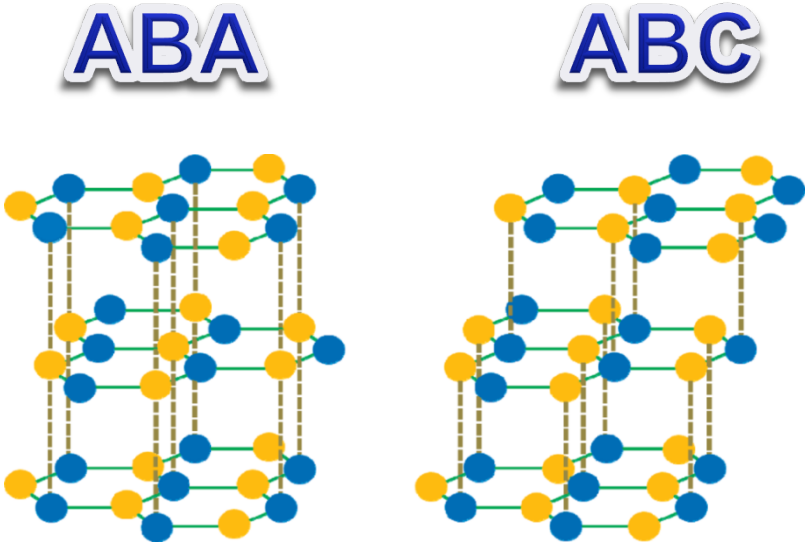
[A.B. Kuzmenko *et al*, PRB 80, 165406 (2009).]

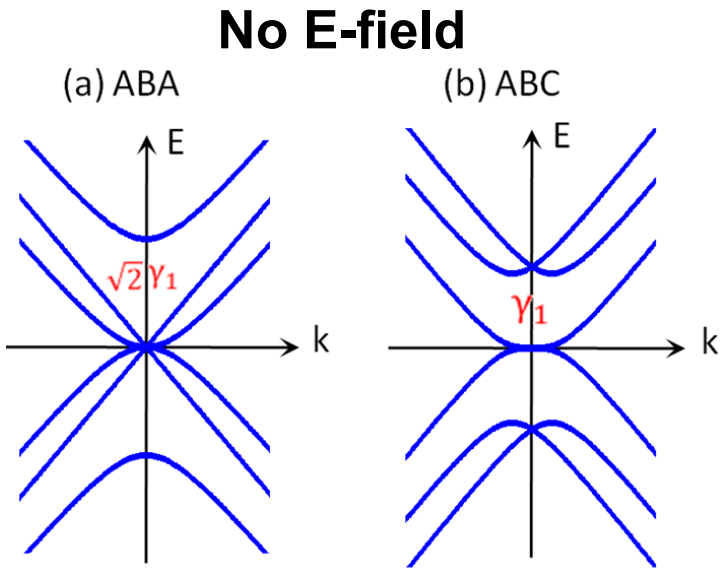
Is the induction of a tunable band gap possible in **trilayer** graphene?

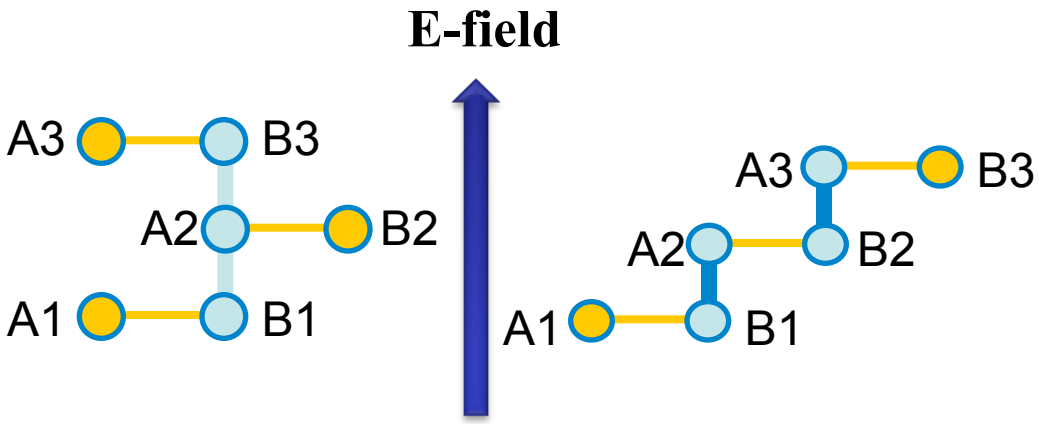
Yes ! But it depends on the stacking order.

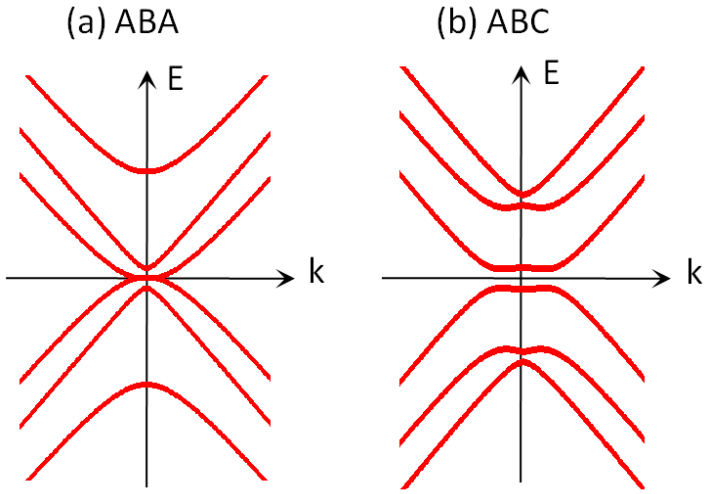
Guinea, Castro-Neto & Peres, Aoki et al., Koshino and McCann, MacDonald, Avetisyan et al,…

Electric-field effect in trilayer graphene









Guinea, Castro-Neto & Peres, Aoki et al., Koshino and McCann, MacDonald, Avetisyan et al,…

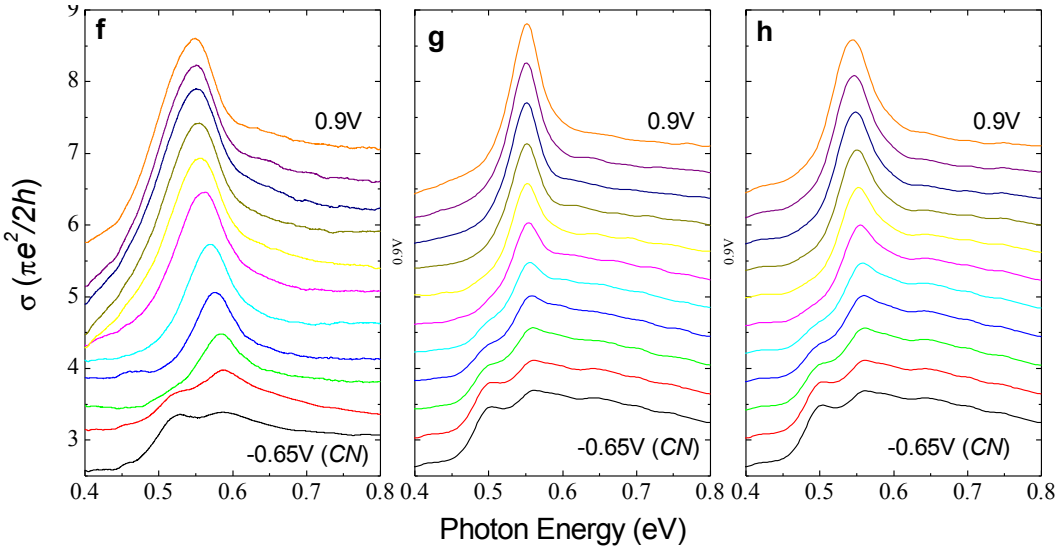
Electric-field effect in trilayer graphene

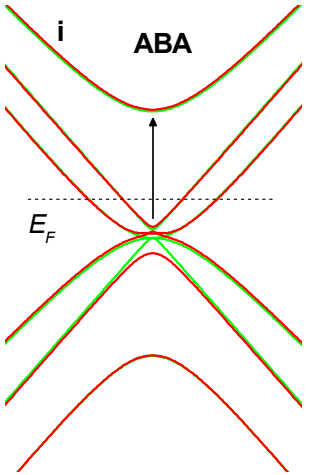
Expt.

Field-modified band structure

Unchanged band structure

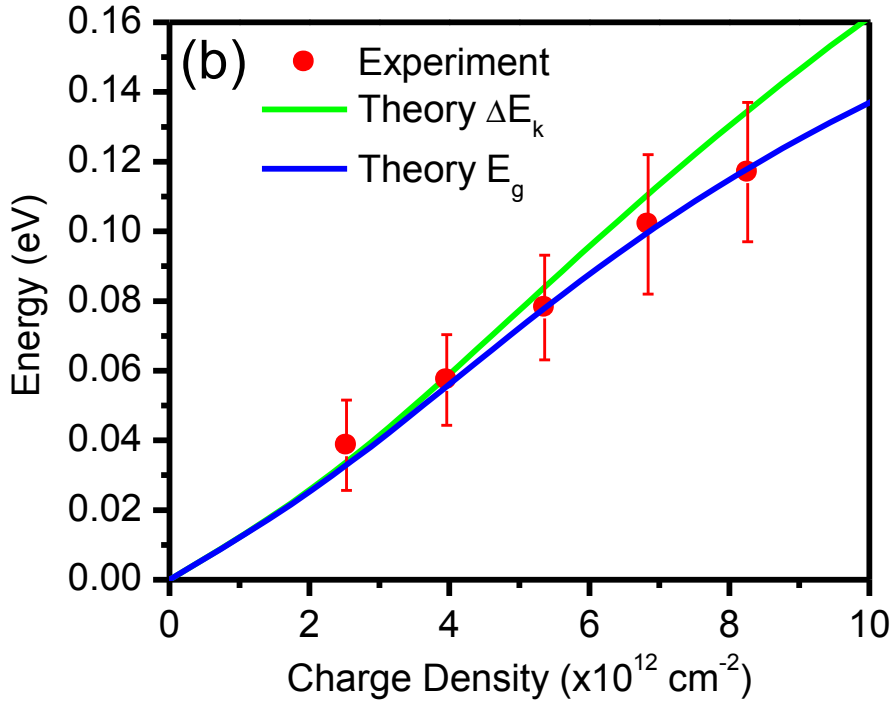
ABA

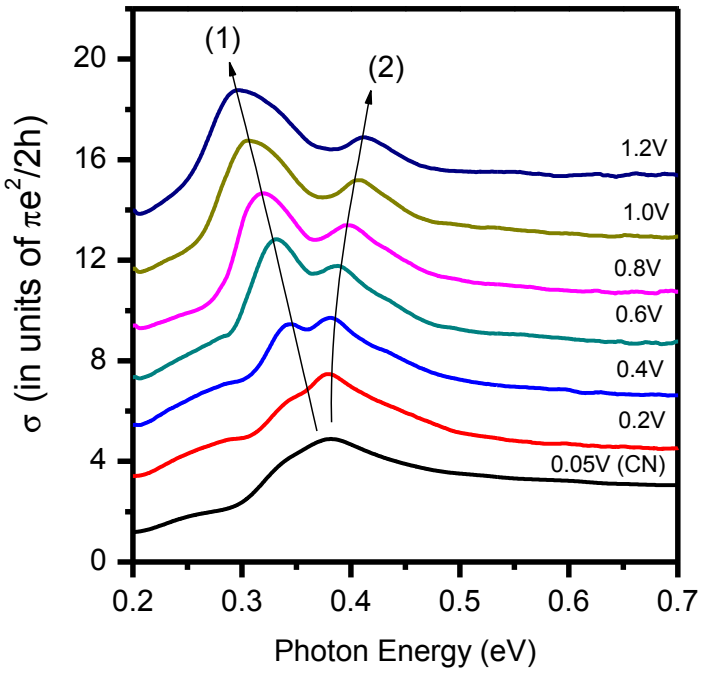




Nature Physics **7**, 944-947 (2011)

Induced band gap in ABC trilayer





Nature Phys. **7**, 944-947 (2011)

Also: Related transport measurements: Lau et al., Nature Phys. (2011).

Summary

SINGLE-LAYER GRAPHENE: e-e interactions

- Major increase in absorption above πα for higher photon energies, rising to close to 10% at 4.6 eV
- Peak is associated with saddle-point at M-point in BZ.
- *Position and asymmetry lineshape require strong excitonic correlations:*

  Saddle-point exciton -- discrete state embedded in continuum

  Fano absorption profile

TRILAYER GRAPHENE: stacking-dependent interlayer interactions

- Identification of ABA (Bernal) and ABC (rhombohedral) trilayers
  - IR conductivity
  - Raman 2D/G'
- ABA and ABC trilayers exhibit completely different low-energy band structure, as revealed in IR conductivity
- E-field induced band gap exceeding100 meV in ABC trilayers; no band gap is found in ABA trilayers.