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Electronic Properties of Multilayer Graphenes

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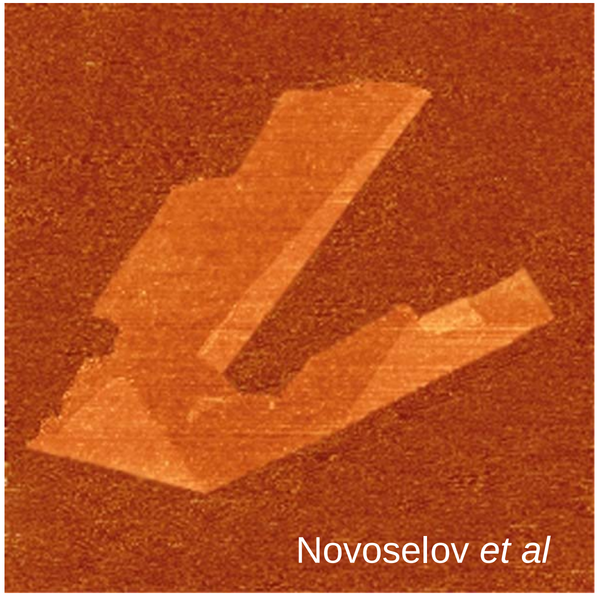
Electronic Properties of Multilayer Graphenes

Mikito Koshino

with Tsuneya Ando

Tokyo Institute of Technology

Monolayer to multilayers


Novoselov *et al*

Fabrication of single-layer graphene:

Novoselov *et al*.,Science **306**, 666 (2004).

Transport measurements:

Novoselov *et al*., Nature **438**, 197 (2005)

Y .Zhang *et al*., Nature **438**, 201 (2005)

Bilayer graphene:

Quantum Hall effect

Novoselov *et al*., Nature **438**, 197 (2005),

Novoselov *et al*., Nat. Phys. **2**,177 (2006).

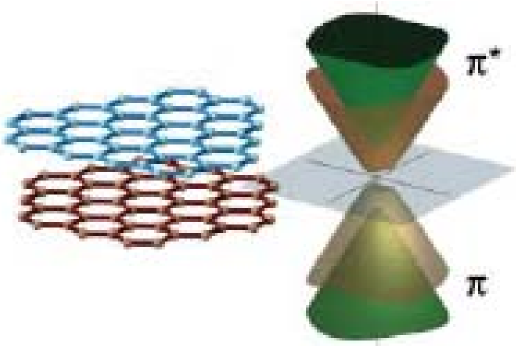
Cyclotron resonance

Henriksen *et al.* PRL **100**, 087403 (2008).

ARPES(<= 4lyrs, epitaxial):

T. Ohta *et al*., Science 313, **951** (2006);

T. Ohta *et al*., PRL **98,** 206802 (2007)



Ohta *et al*

Electronic structure of multilayer graphene

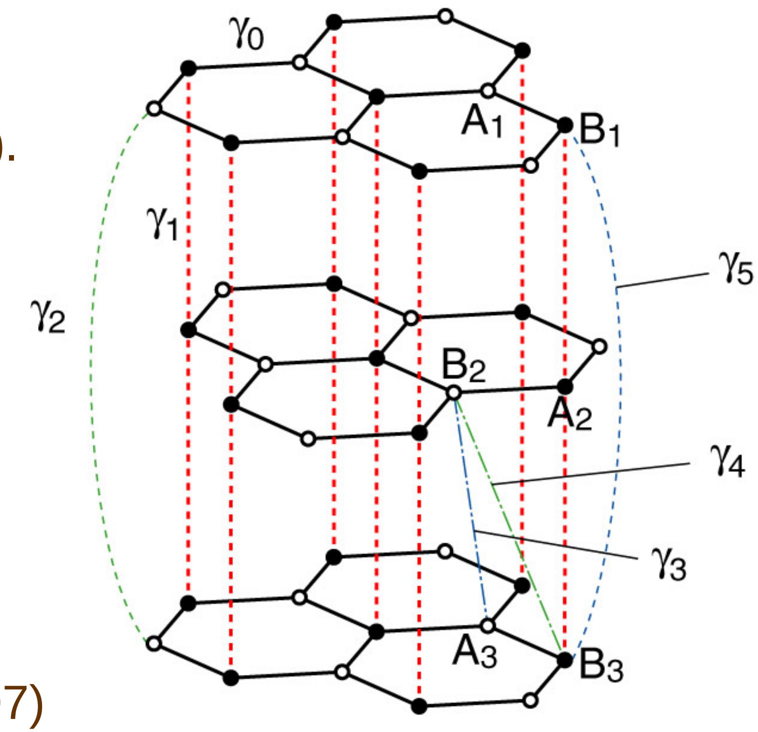
Electronic structure of 3D graphite

Wallace (1947), McClure (1957),
Slonczewski and Weiss (1958)
M. S. Dresselhaus and G. Dresselhaus(1965).

Few-layers graphene

McCann and Fal'ko, PRL **96**, 086805 (2006)
Latil *et al*. PRL **97**,036803 (2006)
Guinea *et al*. PRB **73** 245426 (2006)
Nilsson *et al*. PRL **97**, 266801 (2006).
Partoens and Peeters, PRB **75,** 193402 (2007)
Koshino and Ando, PRB **76**, 085425 (2007).

Atomic structure of graphite
(AB stacked)



Outlines

Electronic properties of multilayer graphenes (AB-stacked) systematically studied within the effective mass approximation.

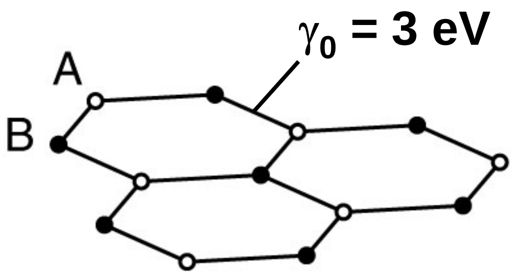
-- Decomposition into bilayer- and monolayer-type subsystems

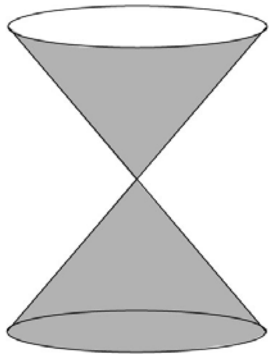
-- Orbital magnetization Koshino and Ando, PRB **76**, 085425 (2007)

-- Magneto-optical absoption Koshino and Ando, PRB **77**, 115313 (2008)

Effective Hamiltonian of graphenes

Monolayer graphene





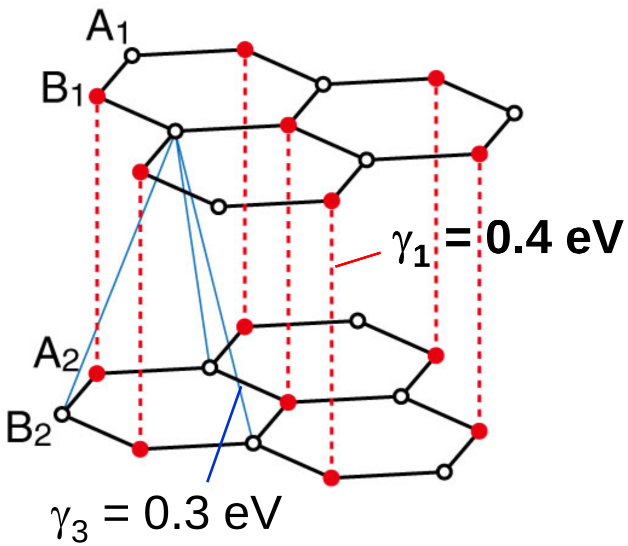
Effective Hamiltonian (around K)

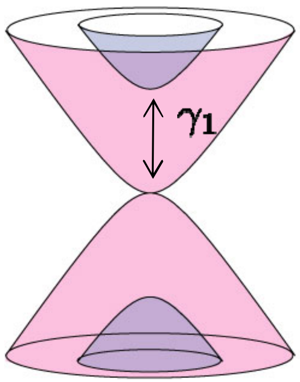
$$\begin{array}{cc} A & B \\ \end{array}$$
$$\mathcal{H} = \begin{pmatrix} 0 & vp_- \\ vp_+ & 0 \end{pmatrix} \qquad p_\pm = p_x \pm i p_y$$

Velocity: $v = \dfrac{\sqrt{3}a}{2\hbar}\gamma_0 \approx 1 \times 10^6 \mathrm{m/s}$

Bilayer graphene

McCann and Fal'ko, PRL **96**, 086805 (2006)





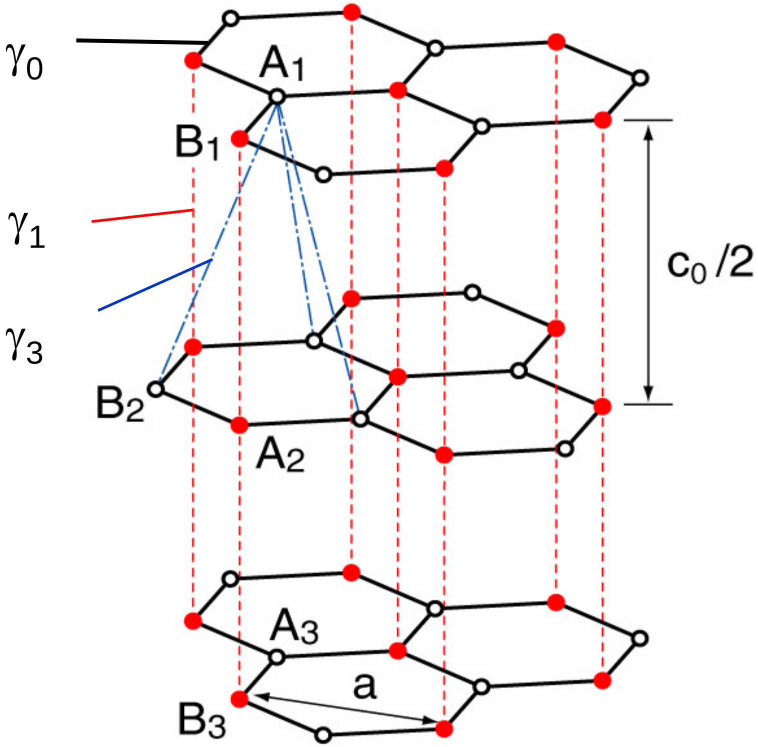
$$\mathcal{H} = \begin{array}{c} \begin{array}{cccc} A_1 & B_1 & A_2 & B_2 \end{array} \\ \begin{pmatrix} 0 & vp_- & 0 & v'p_+ \\ vp_+ & 0 & \gamma_1 & 0 \\ 0 & \gamma_1 & 0 & vp_- \\ v'p_- & 0 & vp_+ & 0 \end{pmatrix} \end{array}$$

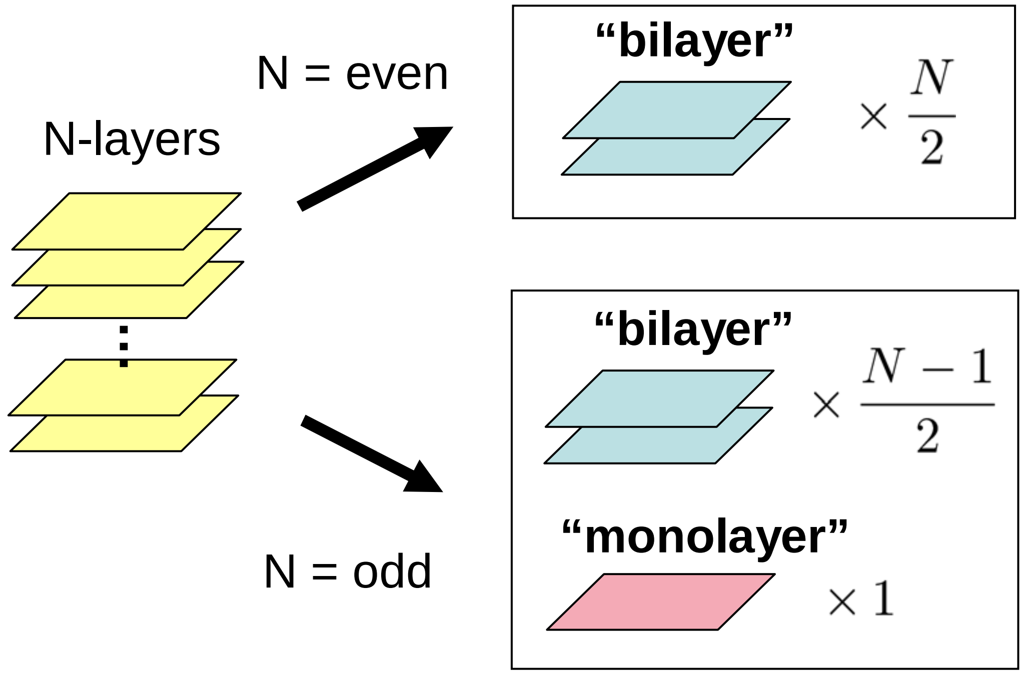
Band mass $m^* \sim 0.033 m_0$

$v' \propto \gamma_3$

Graphene multilayers (N>2)

Hamiltonian of multilayer graphene can be **decomposed into** effective **monolayer-** and **bilayer-like** subsystems





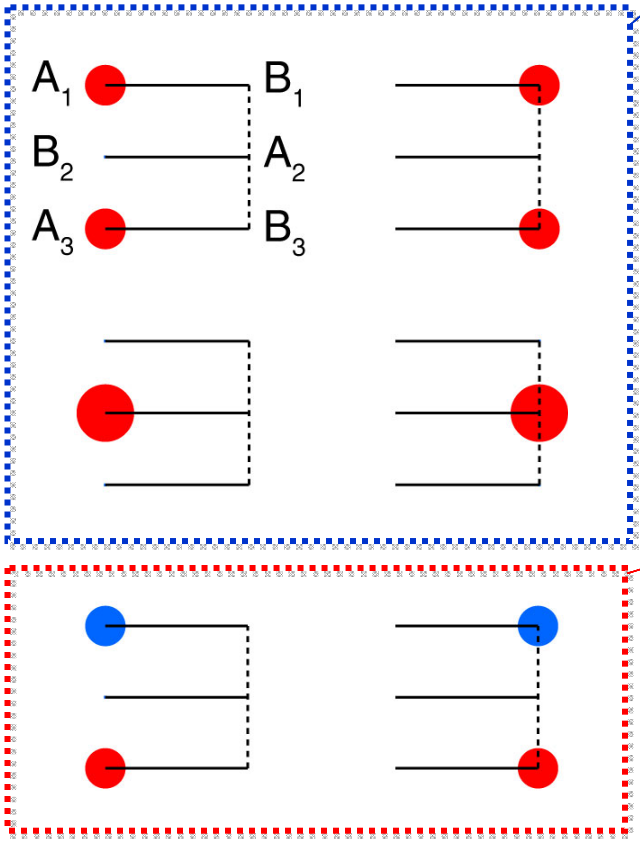
Koshino and Ando, PRB **76**, 085425 (2007)

Cf. Partoens and Peeters, PRB **75,** 193402 (2007)

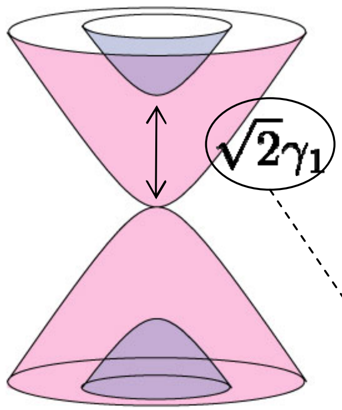
Example: 3-layers

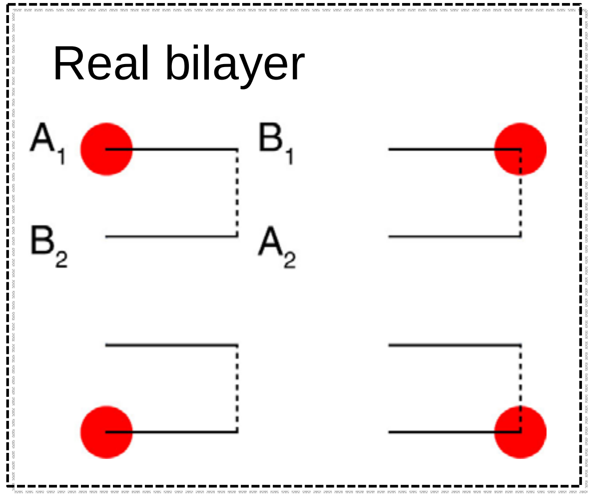
M. Koshino and T. Ando, PRB **76**, 085425 (2007)
M. Koshino and E. McCann, unpublished

Rearrange the basis



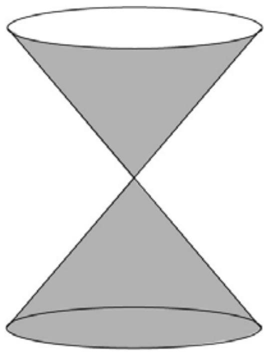
bilayer-type

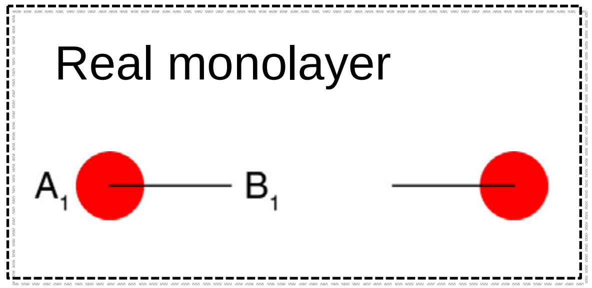




Effective inter-layer coupling (multipled by $\sqrt{2}$)

monolayer-type



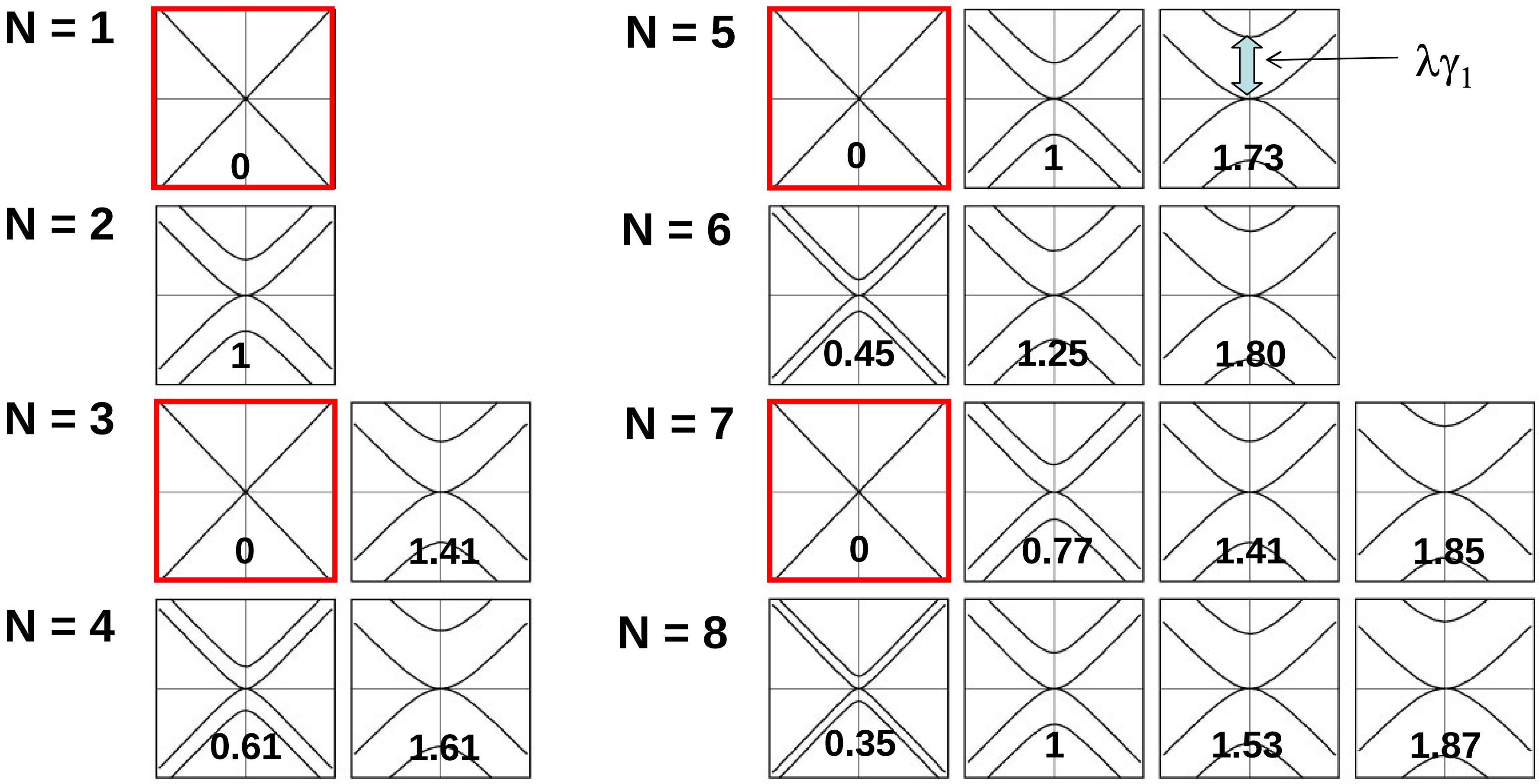


Examples (N=1 to 8)

Effective
inter-layer coupling:

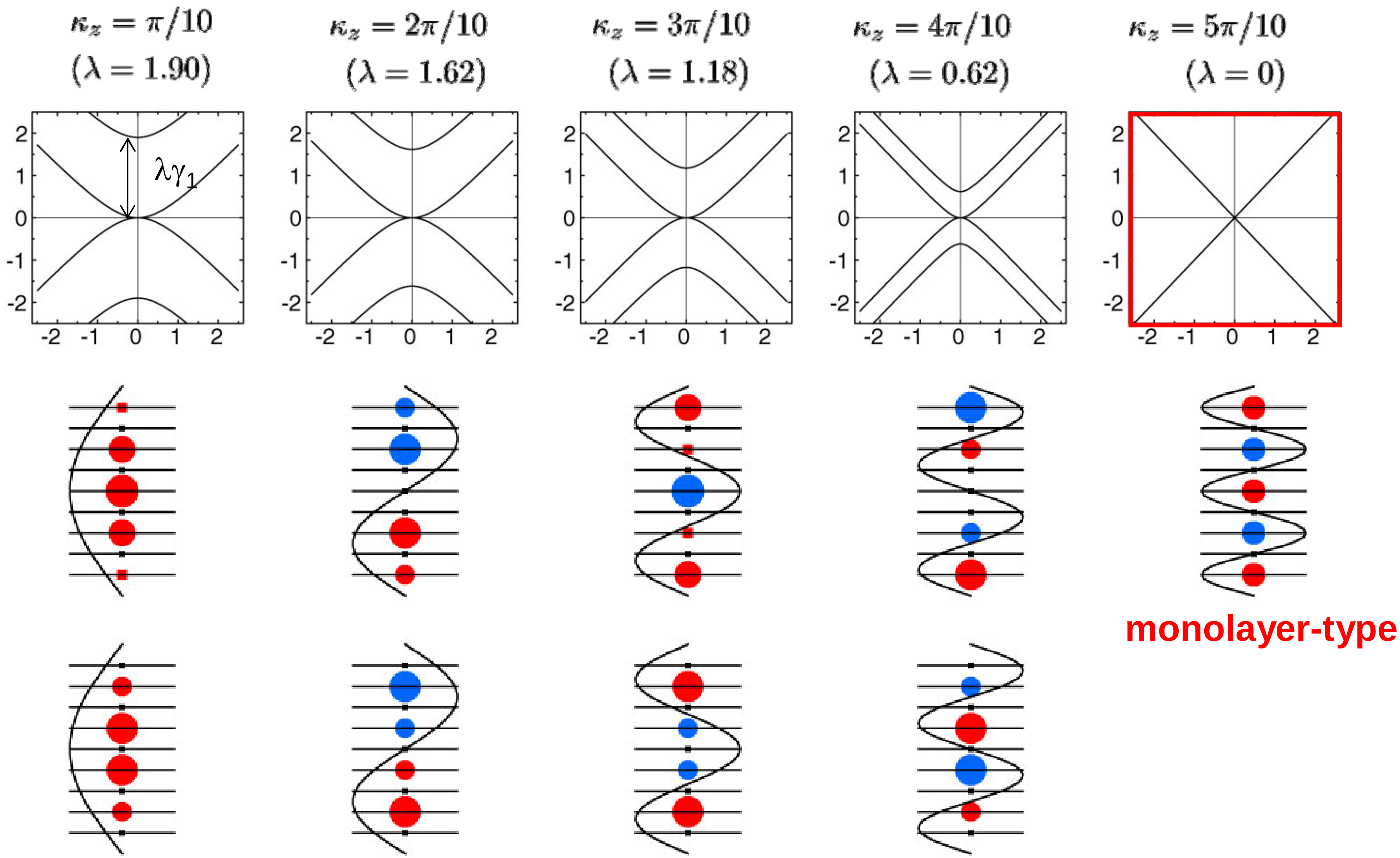
$$\lambda = 2\cos\frac{m\pi}{N+1}$$

m = 1, 2, ... , [N/2]



Example: 9 layers

$$\boxed{\lambda = 2\cos\kappa_z} \qquad \kappa_z = \frac{\pi}{10}, \frac{2\pi}{10}, \cdots, \frac{5\pi}{10}$$



Magneto-optical measurements

Epitaxial thin graphite film

Sadowski *et al*., PRL. 97, 266405 (2006)

Monolayer graphene

Jiang *et al*., PRL. 98, 197403 (2007).
Daecon *et al*., PRB 76, 081406R (2007).

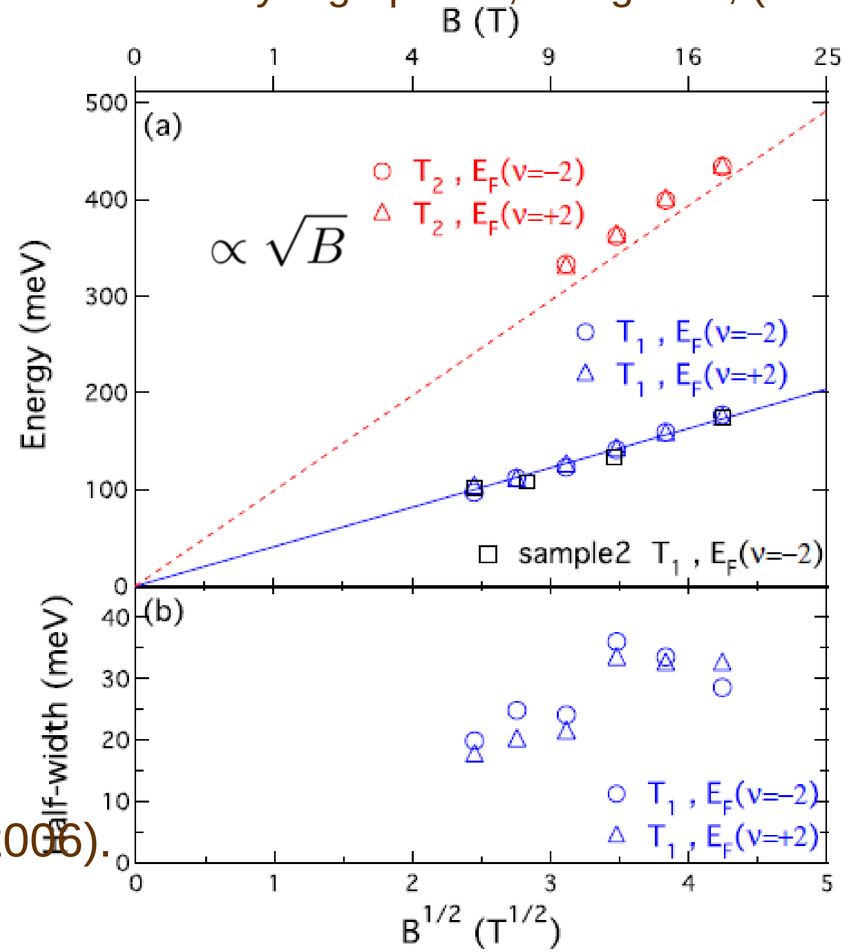
Bilayer graphene

Henriksen *et al.* PRL 100, 087403 (2008).

Theories on optical properties of graphene

T. Ando *et al*., JPSJ. 71, 1318 (2002).
Gusynin and Sharapov, PRB 73, 245411 (2006).
Abergel *et al*., PRB 75, 155430 (2007).

Monolayer graphene, Jiang et al, (2007)



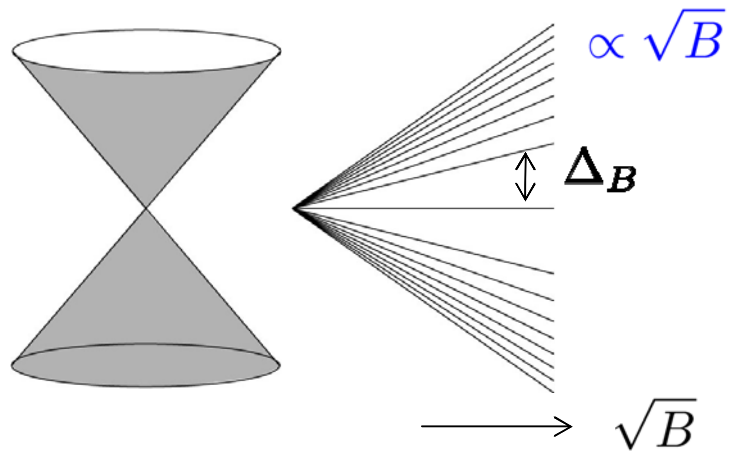
Landau level of graphene

Monolayer graphene

McClure, Phys. Rev. **104**, 666 (1956)

$$\boxed{\begin{aligned}\varepsilon_{n\pm} &= \pm\Delta_B\sqrt{n} \\ \Delta_B &= \sqrt{2\hbar v^2 eB}\end{aligned}} \approx 36\sqrt{B(\mathrm{T})}\ \mathrm{meV}$$

spacing btw. n=0 and 1



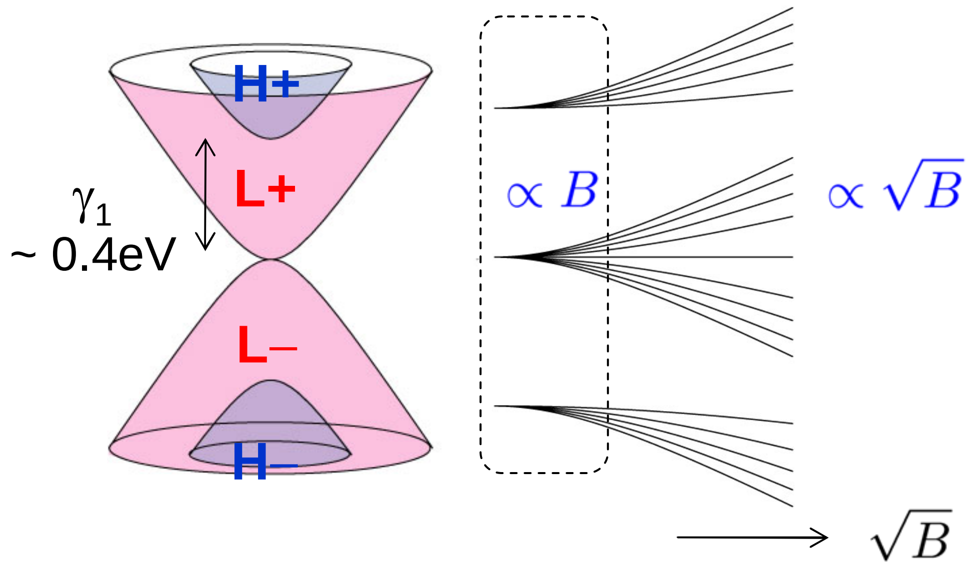
Bilayer graphene

H, L (higher/ lower subband)
+, – (electron / hole band)

Landau levels

$\propto B \quad (\Delta_B \ll \gamma_1)$

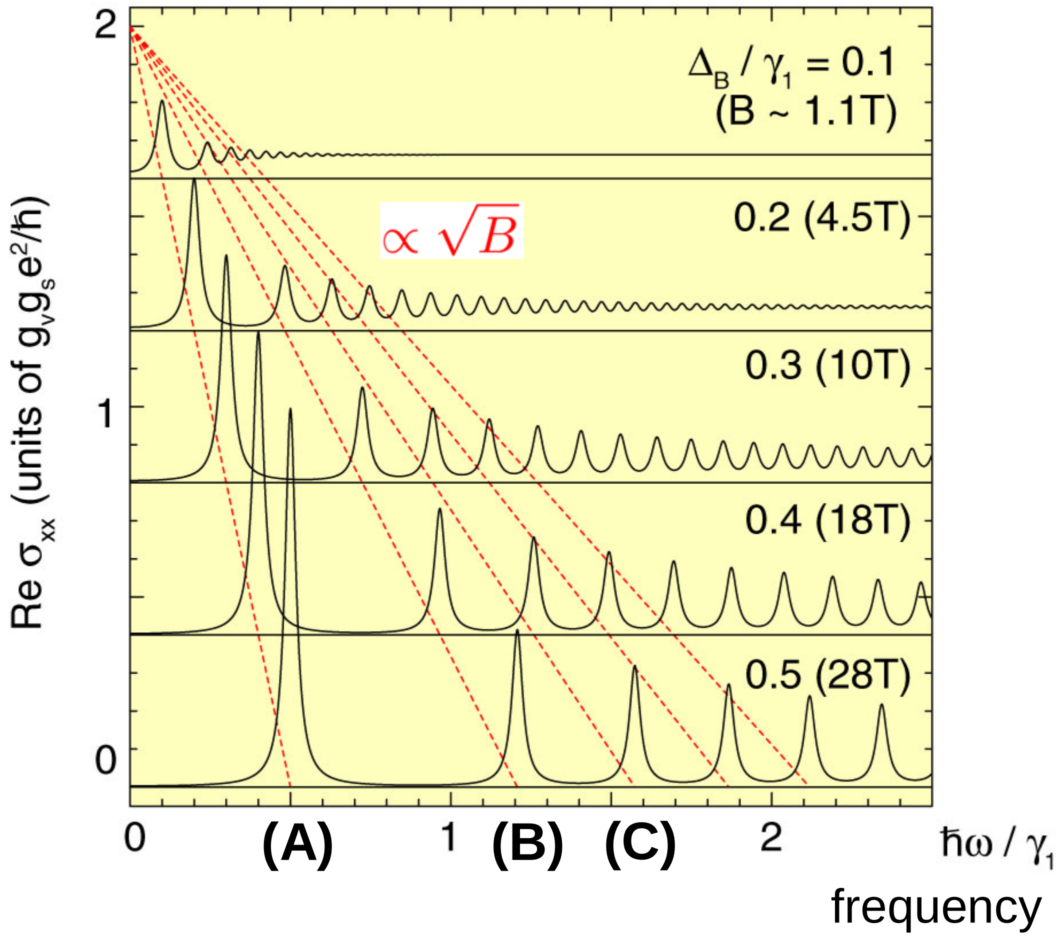
$\propto \sqrt{B} \quad (\Delta_B > \gamma_1)$

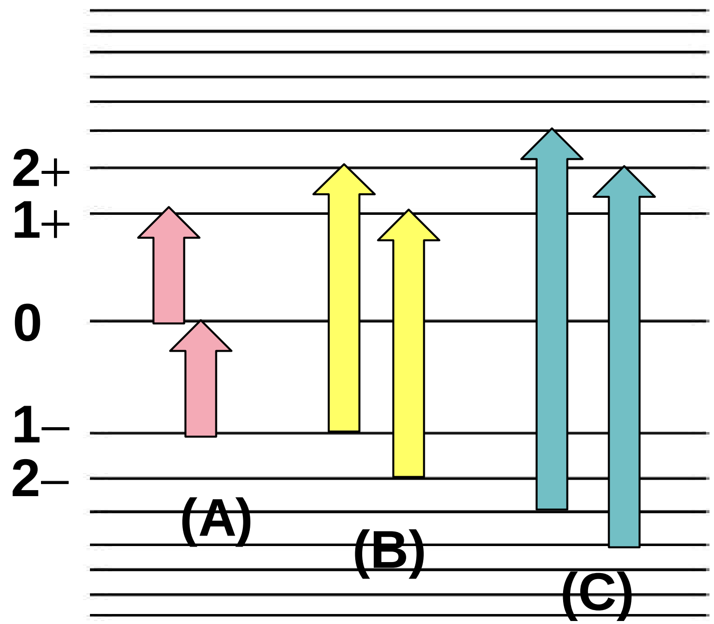


Magneto-optical absorption (monolayer)

Optical transmission

$$T \approx 1 - \frac{4\pi}{c} \mathrm{Re}\, \sigma_{xx}(\omega)$$

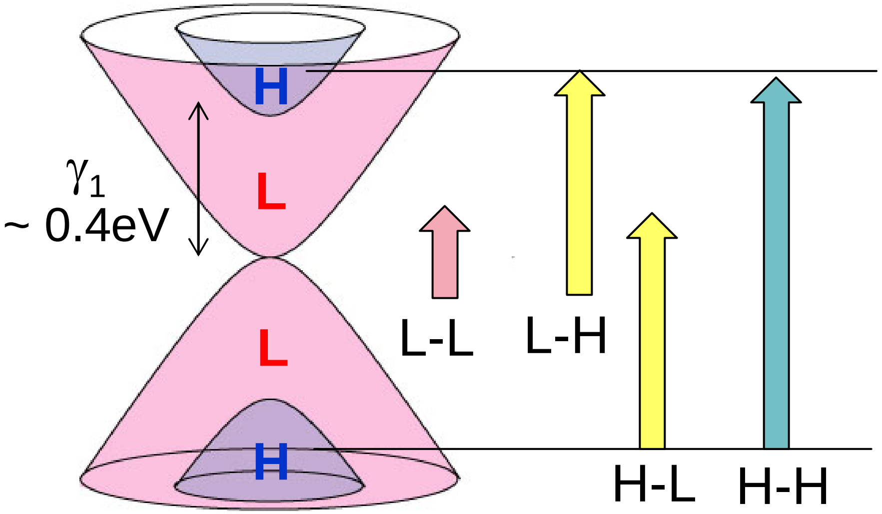




Transition energies $\propto \sqrt{B}$

Experiment:
Jiang *et al*, PRL**98** 197403 (2007)

Magneto-optical absorption (bilayer)



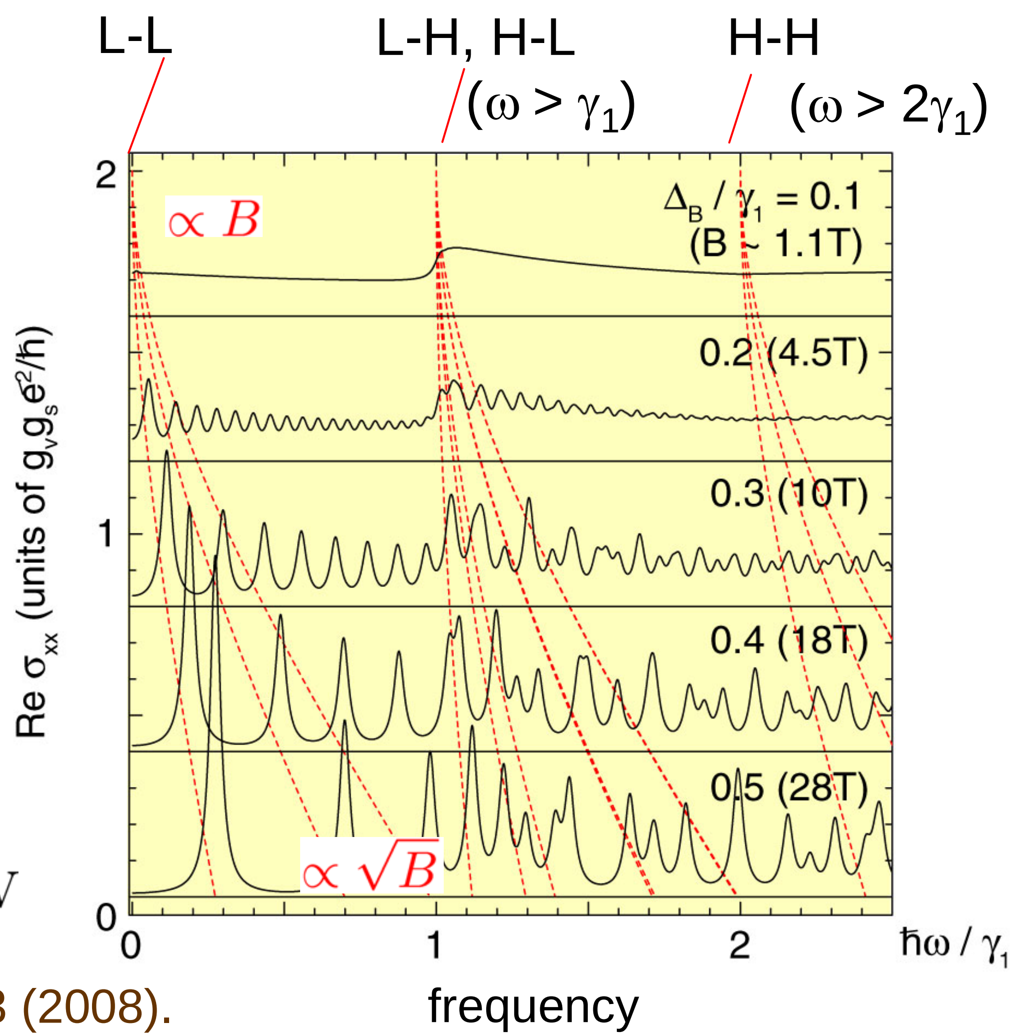
Transition energies

$\propto B \quad (\Delta_B < \gamma_1)$

$\propto \sqrt{B} \quad (\Delta_B > \gamma_1)$

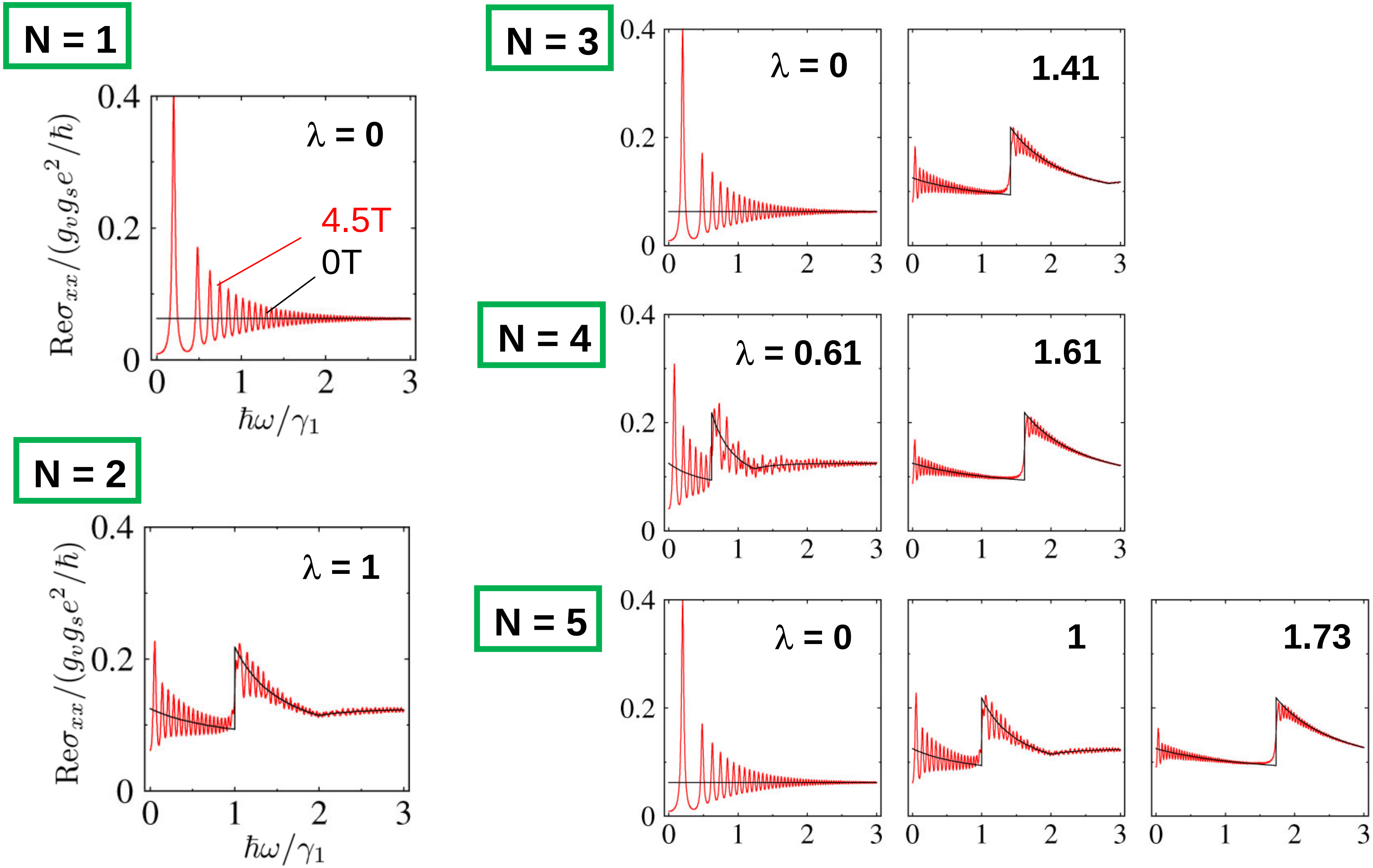
$$\Delta_B \approx 36\sqrt{B(\mathrm{T})}\ \mathrm{meV} \qquad \gamma_1 \approx 0.4\ \mathrm{meV}$$

Dyamical conductivity



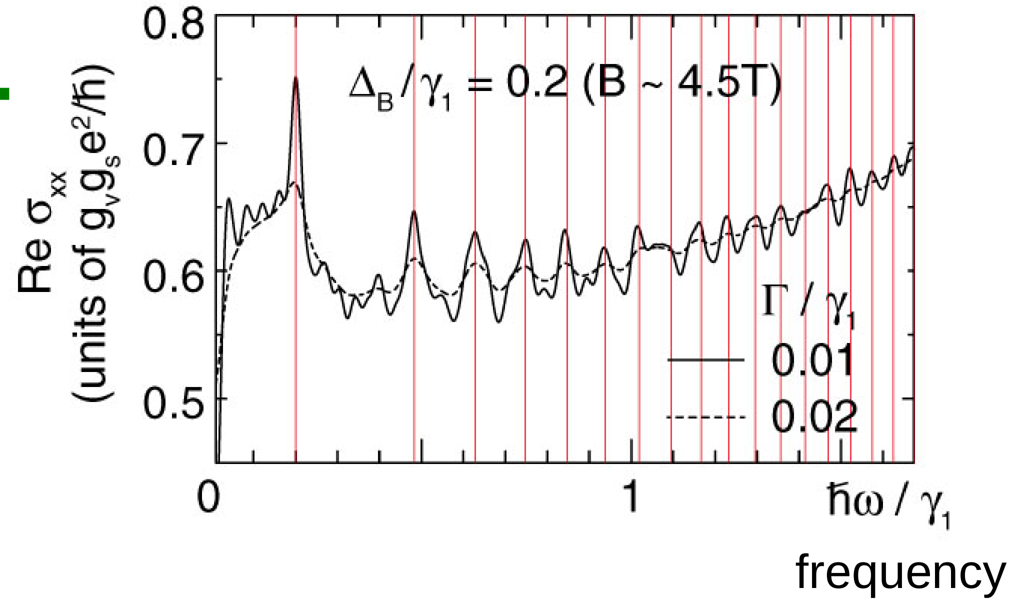
Exp.: Henriksen *et al.* PRL **100**, 087403 (2008).

Magneto-optical absorption (multilayers)

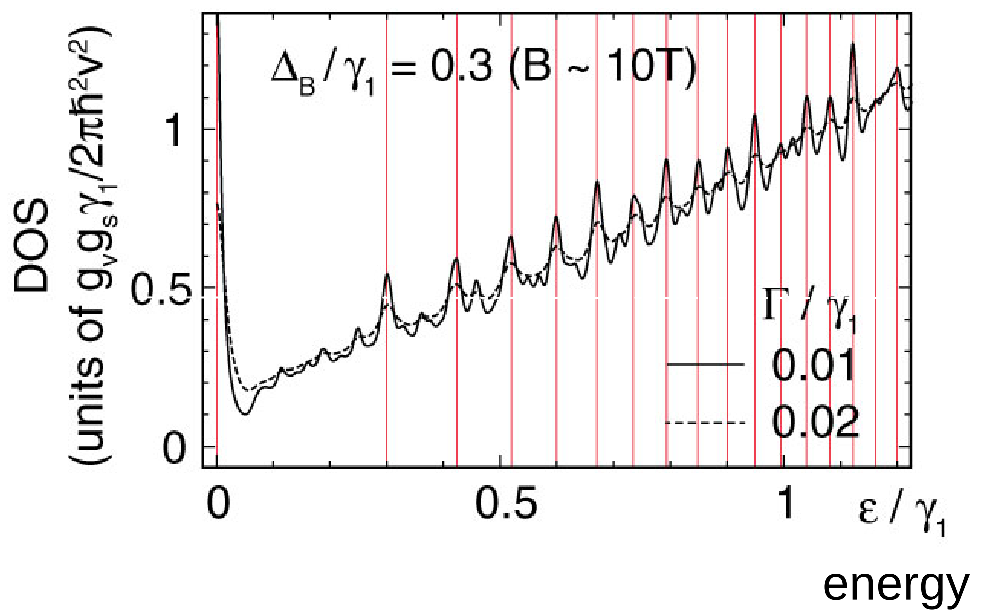


Compound of multilayer graphenes

Magneto-optical absorption
avaraged over N =1 to 20



DOS on the top layer
avaraged over N =1 to 20



Monolayer-like signals still survives
(contributed by "effective" monolayer-like subbands)

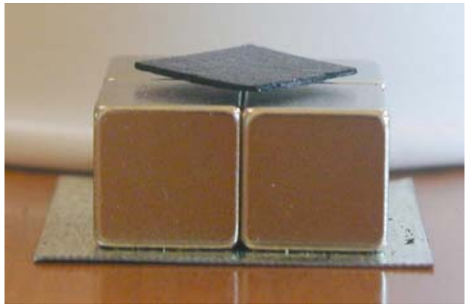
Orbital magnetism of graphene

3D graphite: Strong diamagnetism

	$-\chi$ (x 10^{-6}) in SI units
Water	8.8
Gold	34
Bismuth	170
Pyrolytic graphite $\perp$	450
Pyrolytic graphite //	85

Simon *et al.* (2001)

Levitating pyrolytic graphite
(At room temperature)

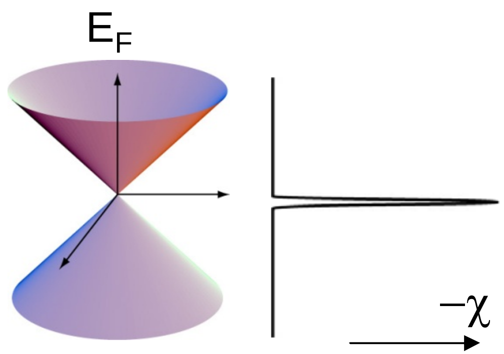


S. Q. Field

Monolayer graphene (Ideal, T→0)

$$\chi(\varepsilon_F) = -\frac{2e^2v^2}{3\pi}\,\delta(\varepsilon_F)$$

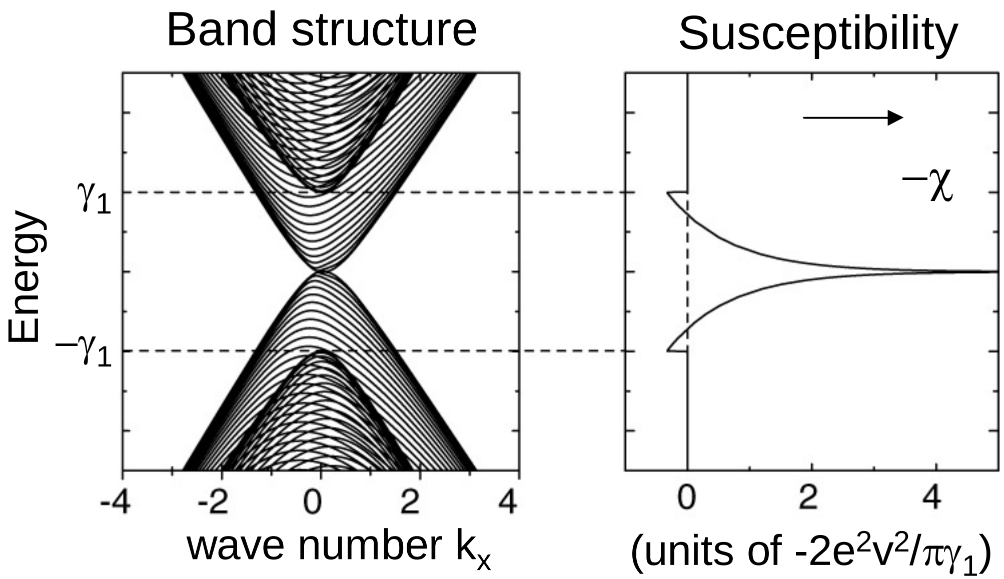
Diamagnetic singularity
at the Dirac point



McClure, Phys. Rev. **104**, 666 (1956); Phys. Rev. **108**, 612 (1957).

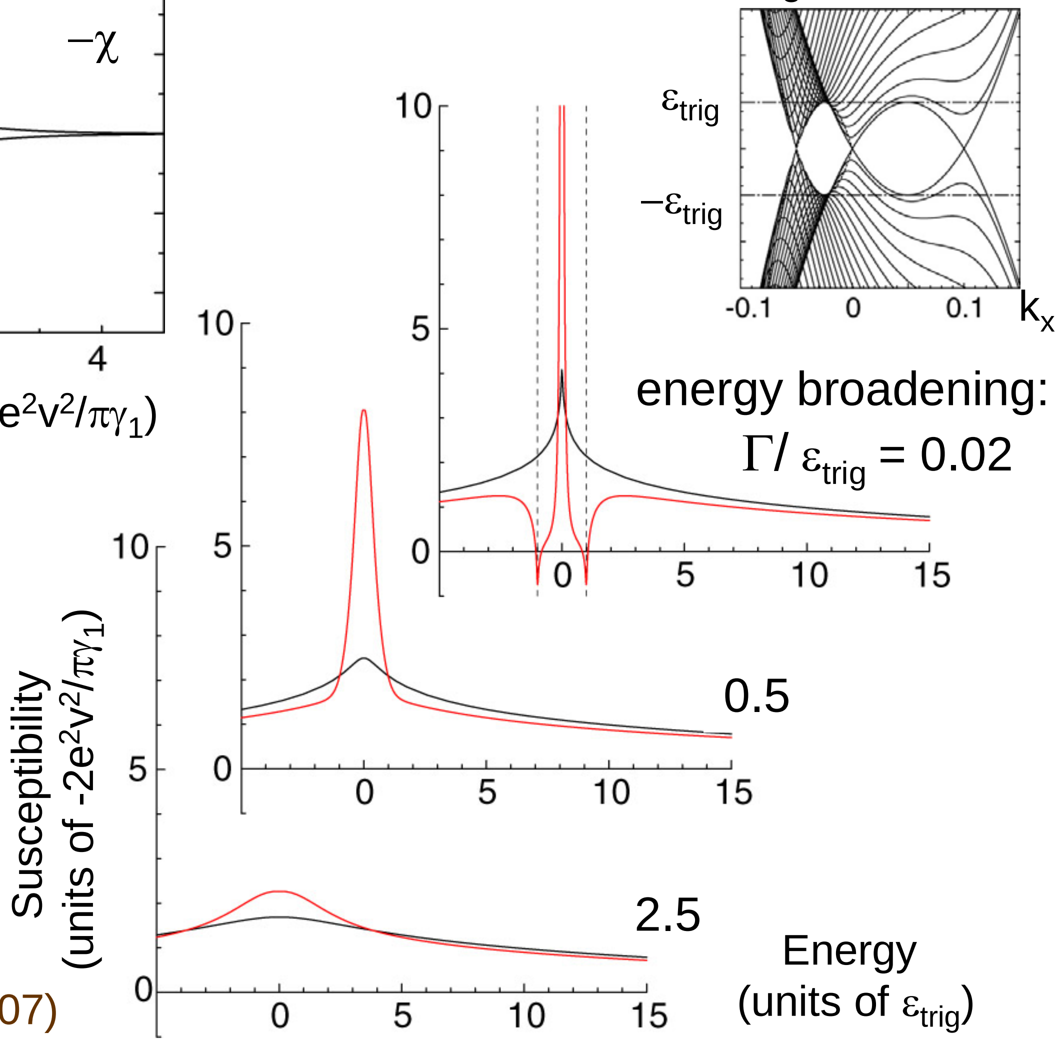
Magnetism of 3D graphite: Sharma et al, PRB **9**, 2467 (1974), Safran et al, PRB **20**, 4889 (1979). Blinowski et al J. Phys. Paris **45**, 545 (1984),
Intercalated graphite: Saito and Kamimura, PRB **33**, 7218 (1986).

Magnetism of bilayer graphene



Susceptibility around E=0

(With / Without γ_3)



Without trigonal warping

$$\chi(\varepsilon_F) \propto \ln|\varepsilon_F/\gamma_1|$$

Safran, PRB **30**, 421 (1984)

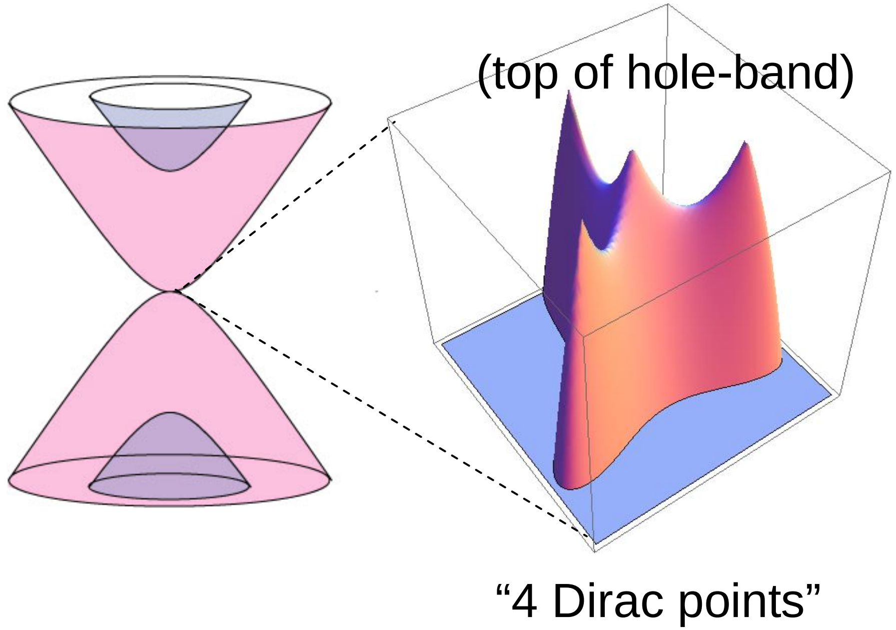
With warping (γ_3)

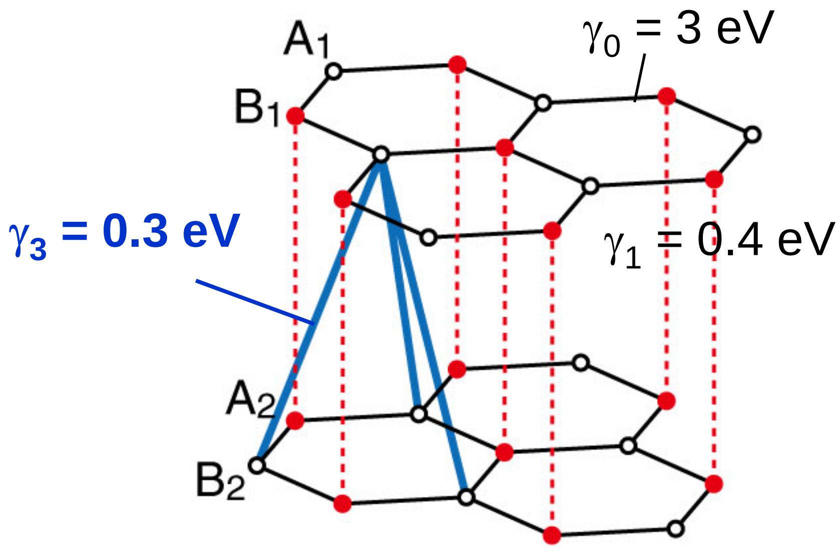
δ-function at E = 0

Koshino and Ando, PRB **76**, 085425 (2007)

Trigonal warping in the bilayer band

Fine structure around E=0





Energy scale

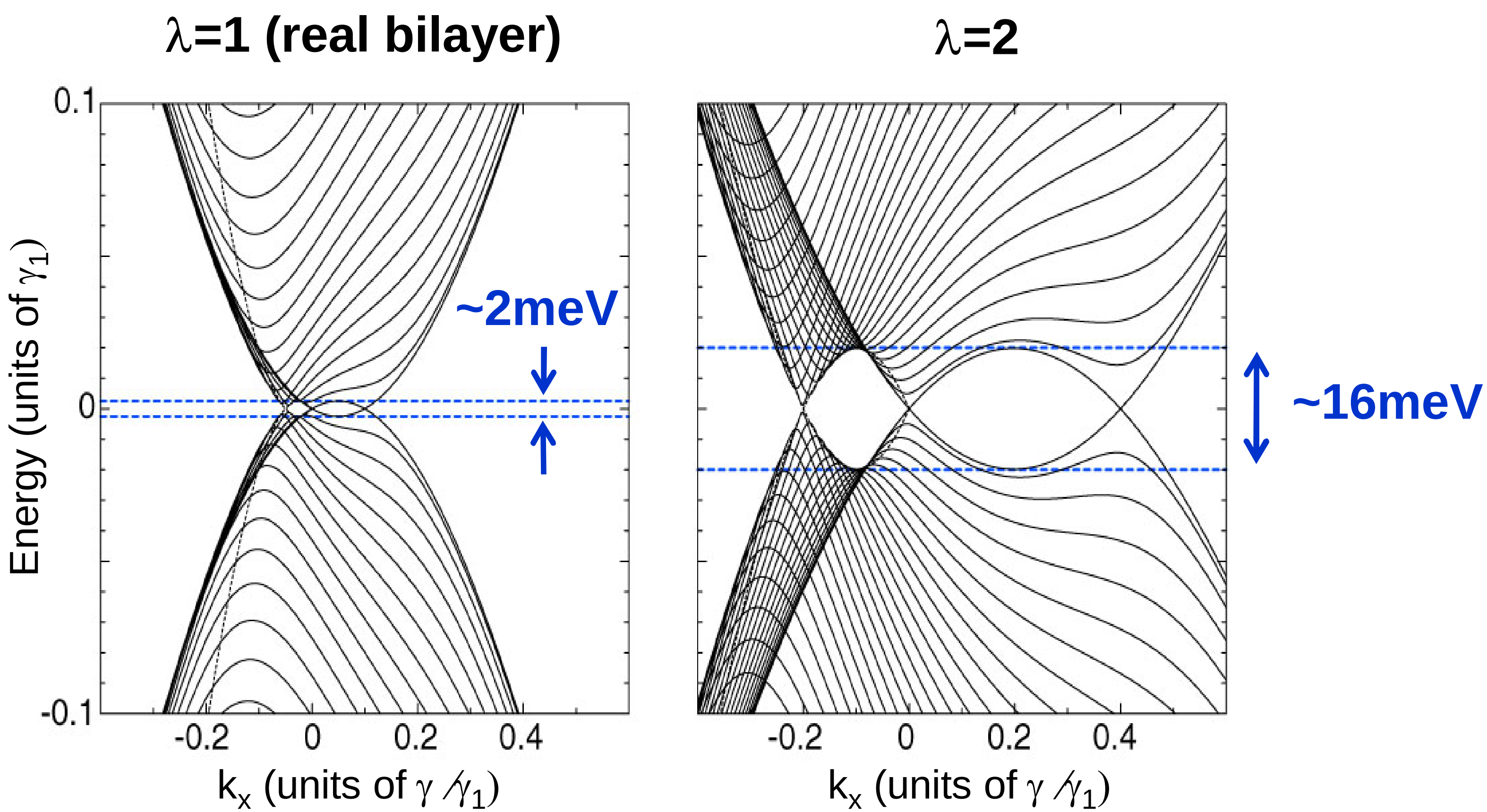
$$\varepsilon_{\mathrm{trig}} = \left(\frac{\gamma_3}{\gamma_0}\right)^2 \gamma_1 \sim 1\mathrm{meV}$$

McCann and Fal'ko,
PRL **96**, 086805 (2006)

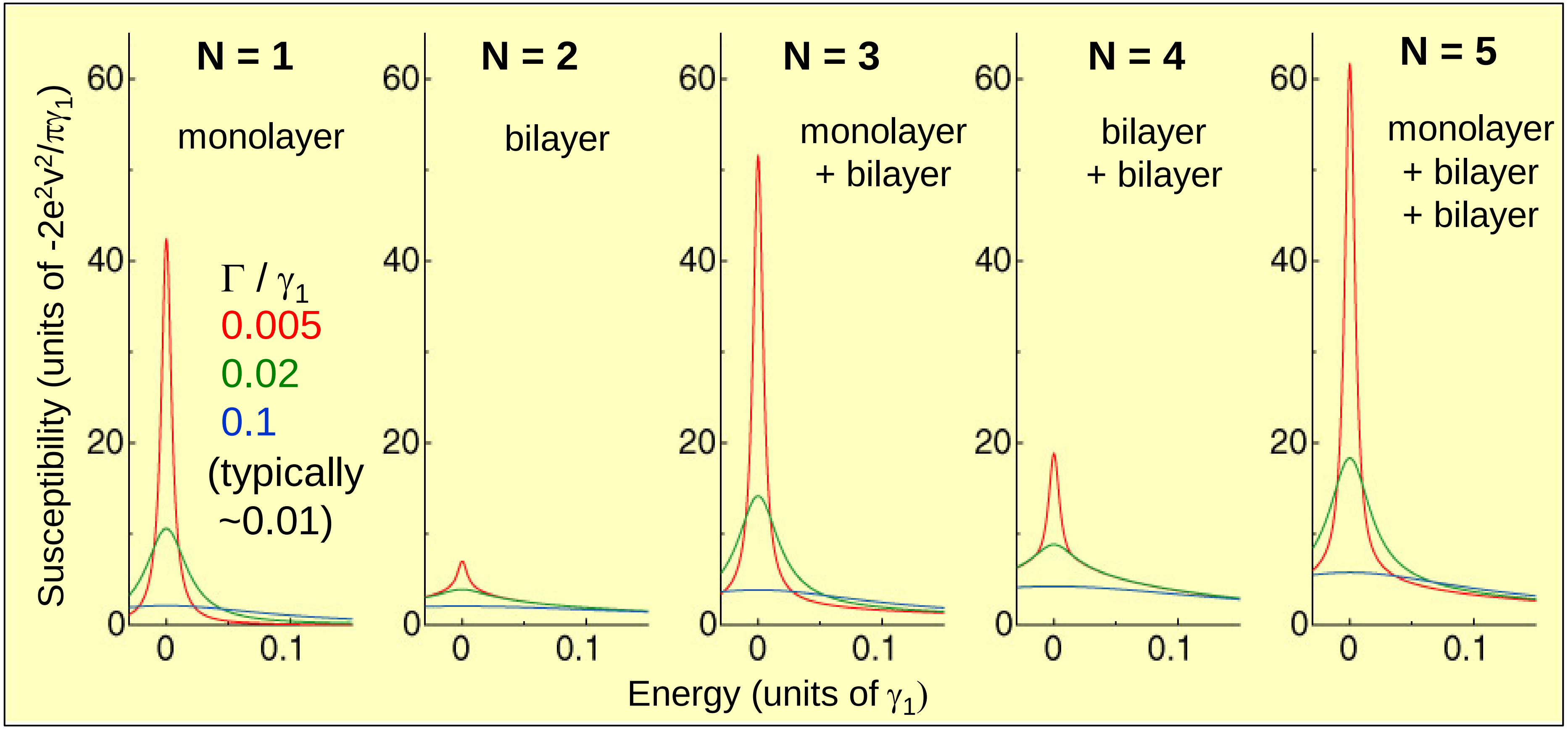
Real bilayer		Effective "bilayer"
γ_0	→	γ_0
γ_1 , γ_3	→	$\lambda\,\gamma_1$, $\lambda\,\gamma_3$
$\varepsilon_{\mathrm{trig}}$	→	$\lambda^3\ \varepsilon_{\mathrm{trig}}$

Trigonal warping in effective bilayer bands

Energy scale of the trigonal warping $\varepsilon_{\mathrm{trig}} = \left(\frac{\lambda\gamma_3}{\gamma_0}\right)^2 \lambda\gamma_1$ $\propto \lambda^3$



Susceptibility of multilayer graphenes



Koshino and Ando,
PRB **76**, 085425 (2007)

Monolayer-type **(odd N only):** δ-function

Bilayer-type: Log tail + δ-function (trigonal warping)

Summary

Electronic properties of AB-stacked multilayer-graphenes studied in the effective mass approximation.

Multilayer = bilayers + monolayer (odd N)

--- Characterized by single parameter λ (0 to 2)

--- Trigonal warping $\propto \lambda^3$

Magneto-optical absoption

--- Crossover from linear B to sqrt B

Orbital magnetization

--- Singularity at zero energy in monolayer graphene

--- Singularity from trigonal warping

Koshino and Ando, PRB **76**, 085425 (2007); PRB **77**, 115313 (2008)

Magnetization formula

Magnetization

$$M = -\left(\frac{\partial F}{\partial B}\right)_N = -\left(\frac{\partial \Omega}{\partial B}\right)_\mu$$

F : Free energy
N : Electron number

Ω : Thermodynamic potential
μ : Chemical potential

Susceptibility

$$\chi = \left.\frac{\partial M}{\partial B}\right|_{B=0}$$

Linear response formula Fukuyama, Prog. Theor. Phys. **45**, 704 (1971)

$$\chi = -\frac{1}{2\pi L^2}\frac{e^2}{\hbar^2}\mathrm{Im}\int_{-\infty}^{\infty} d\varepsilon f(\varepsilon)\mathrm{Tr}\left(G\mathcal{H}_x G\mathcal{H}_y G\mathcal{H}_x G\mathcal{H}_y\right)$$

$$\mathcal{H}_i = \frac{\partial \mathcal{H}(\vec{k})}{\partial k_i}, \quad G(\varepsilon) = \frac{1}{\varepsilon - \mathcal{H} + i\delta}$$

Derived from $B(x) = B\cos qx$
with q → 0 and B → 0

Hamiltonian of graphene multilayer

k.p Hamiltonian

$$\mathcal{H} = \begin{array}{c} \begin{array}{cccccc} A_1 & B_1 & A_2 & B_2 & A_3 & B_3 \end{array} \\ \begin{pmatrix} \boxed{\begin{matrix} H_0 & V \\ V^\dagger & H_0 \end{matrix}} & V^\dagger & & \\ V & H_0 & V & \\ & V^\dagger & H_0 & V^\dagger \\ & & & \ddots \end{pmatrix} \end{array}$$

bilayer

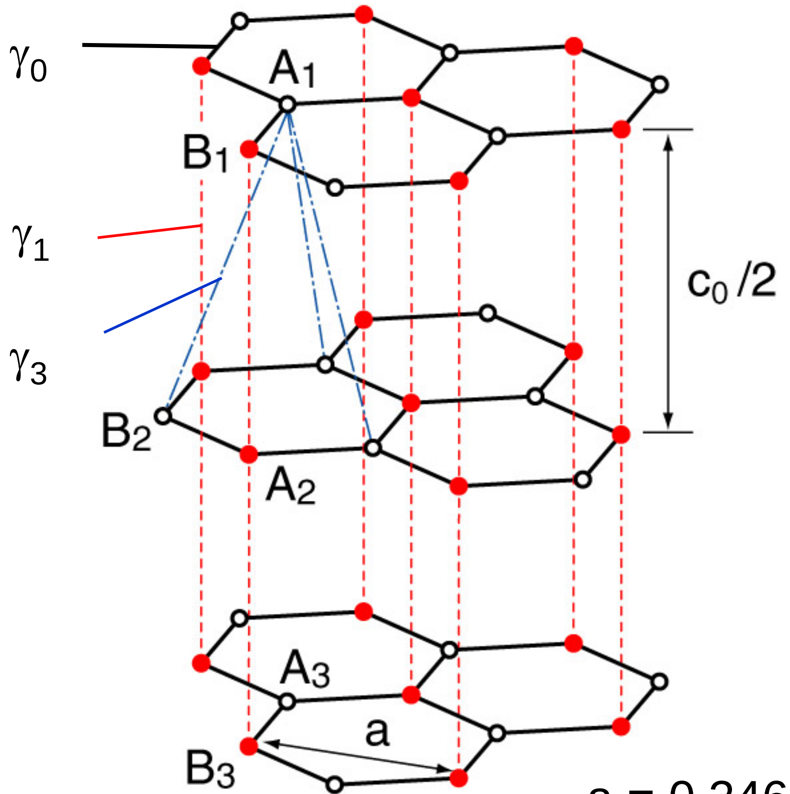
Intra-layer (= monolayer)

$$H_0 = -\begin{pmatrix} 0 & vp_- \\ vp_+ & 0 \end{pmatrix} \qquad p_\pm = p_x \pm ip_y$$

$$v = \frac{\sqrt{3}a}{2\hbar}\gamma_0$$

Inter-layer

$$V = -\begin{pmatrix} 0 & v'p_+ \\ \gamma_1 & 0 \end{pmatrix} \qquad v' = \frac{\sqrt{3}a}{2\hbar}\gamma_3$$



a = 0.246nm

k.p in monolayer:
Ajiki and Ando, JPSJ. **62**, 1255 (1993)
Kane and Mele, PRL **78**, 1932 (1997).