Electronic phase transitions in charge-neutral graphene multilayers: Does graphene ever become graphite upon increasing thickness?

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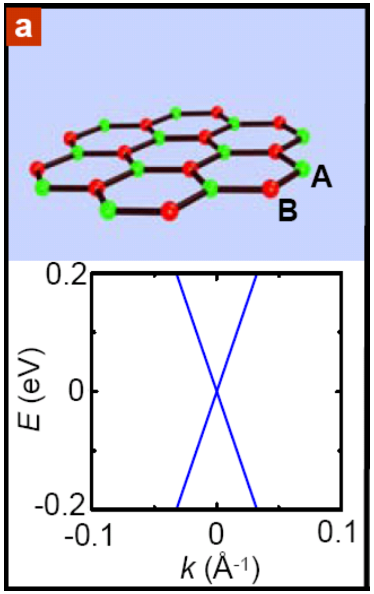
QSIT Quantum Science and Technology
National Centre of Competence in Research

FNSNF
Swiss National Science Foundation

GRAPHENE FLAGSHIP

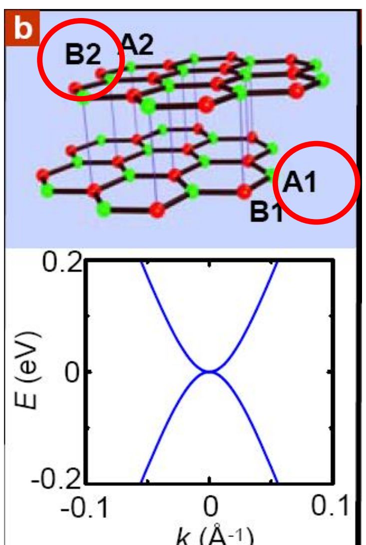
Single & bilayer graphene

In the absence of interactions



Monolayer = 1LG

$$H_{1L} = \begin{pmatrix} 0 & \hbar v_F(k_x - ik_y) \\ \hbar v_F(k_x + ik_y) & 0 \end{pmatrix}$$

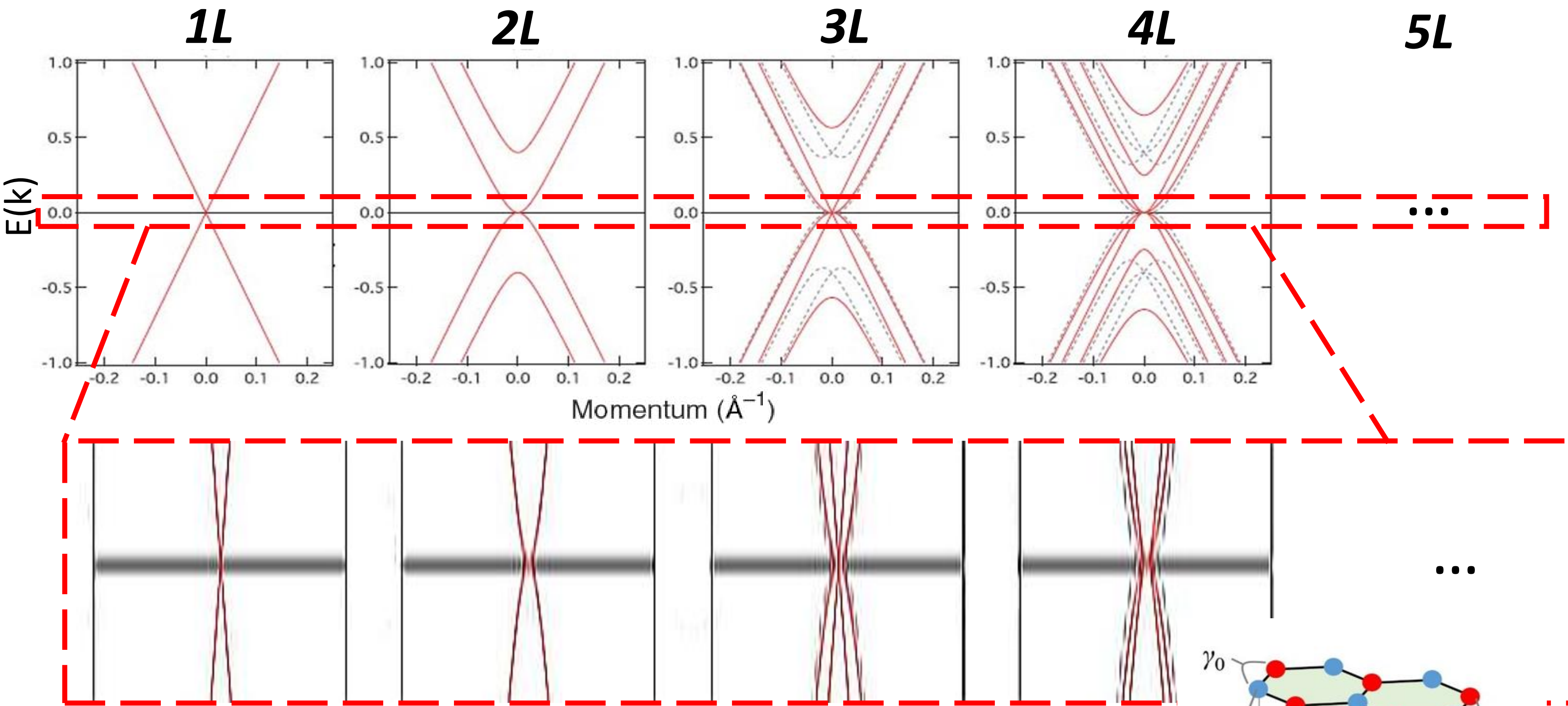


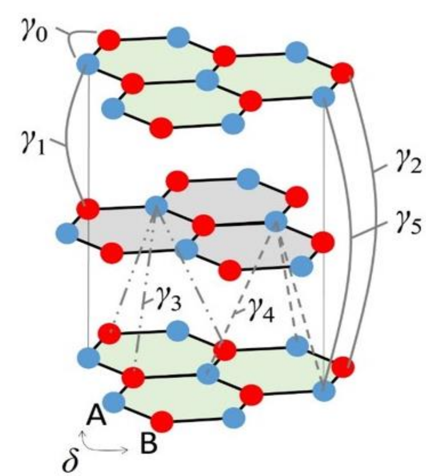
Bilayer = 2LG

$$H_{2L} = \begin{pmatrix} 0 & \frac{\hbar^2}{2m^*}(k_x - ik_y)^2 \\ \frac{\hbar^2}{2m^*}(k_x + ik_y)^2 & 0 \end{pmatrix}$$

Compare kinetic energy to interactions

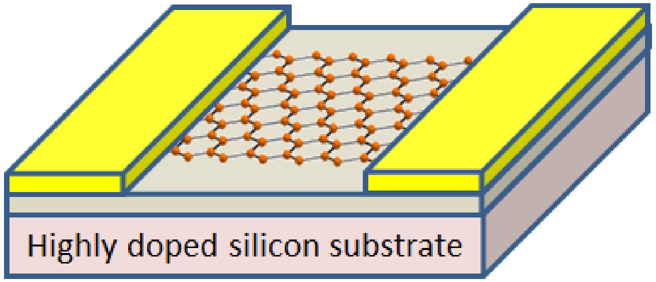
Search for interaction effects? zoom in on E=0 as close as possible



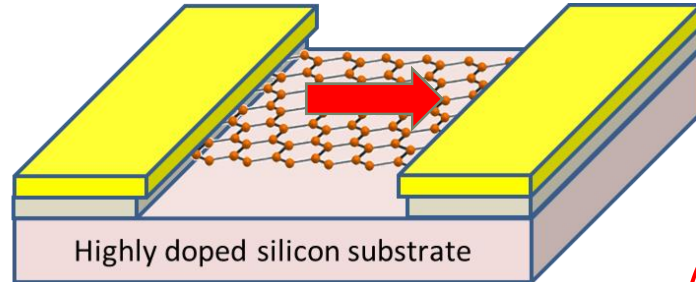


Today we will look at what happens for
Bernal stacked multilayers up to 8LG

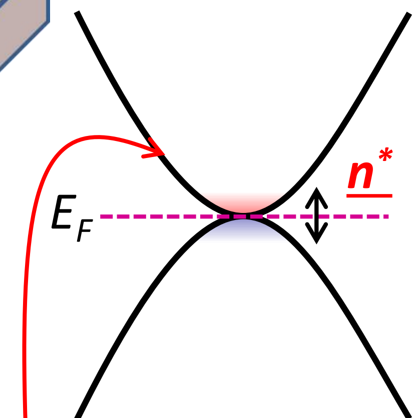
Approaching the Dirac point in Suspended graphene

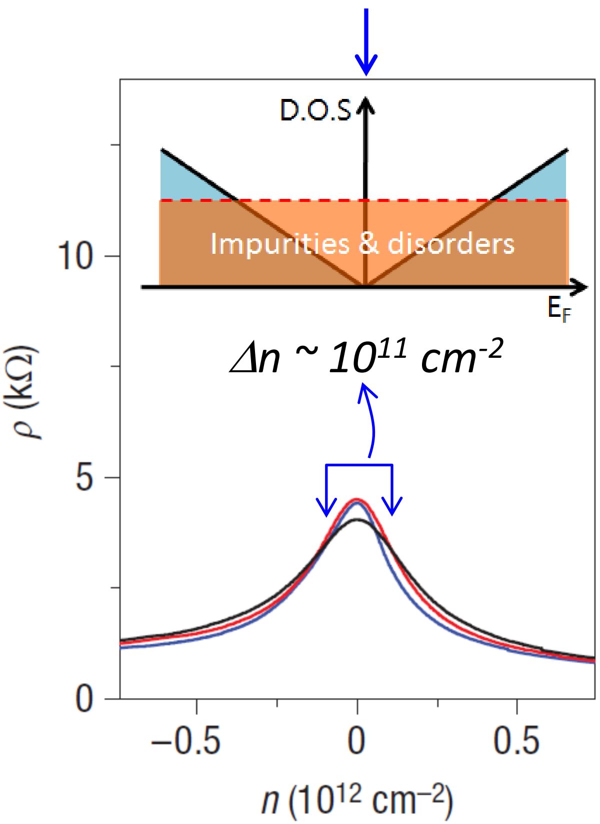


On substrate

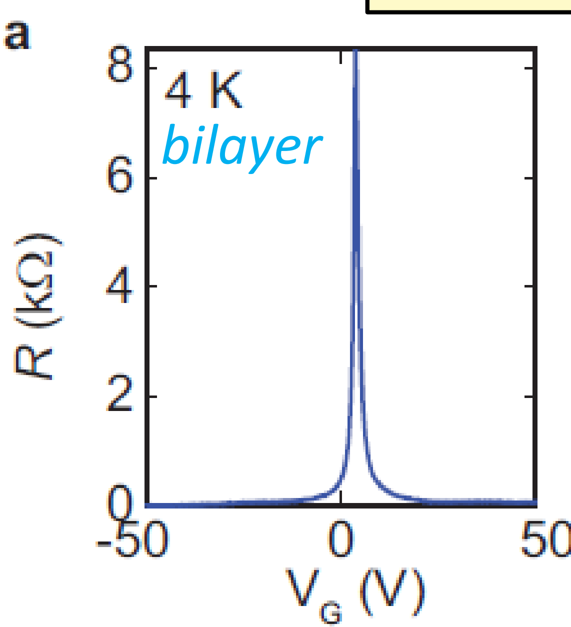


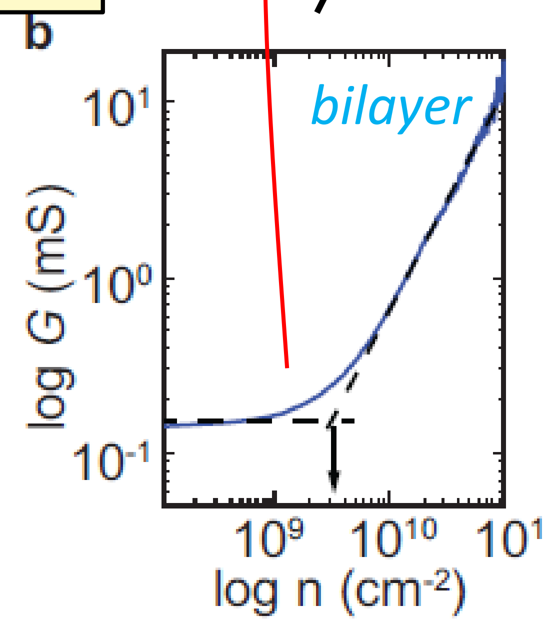
Suspended





$\Delta n \sim 10^{9}\ cm^{-2}$





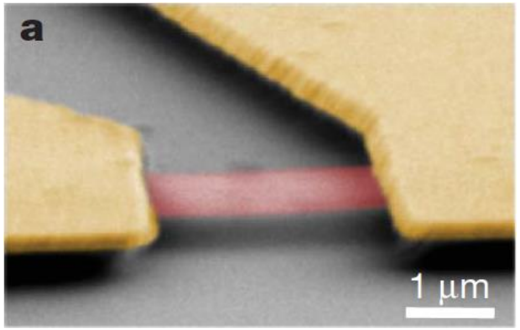
Δn approaching $10^8\ cm^{-2}$ for $T < 1K$

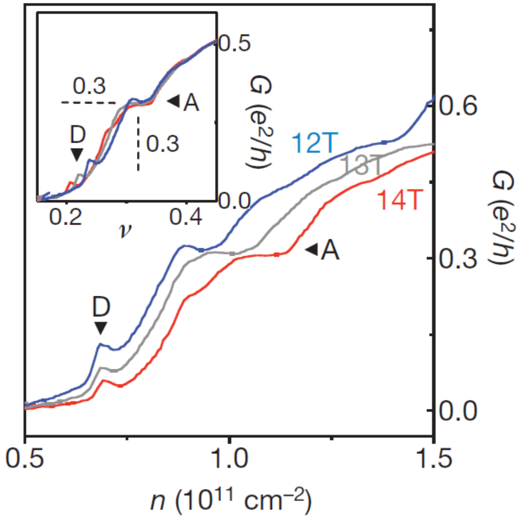
Early days of suspended Graphene

Under magnetic field

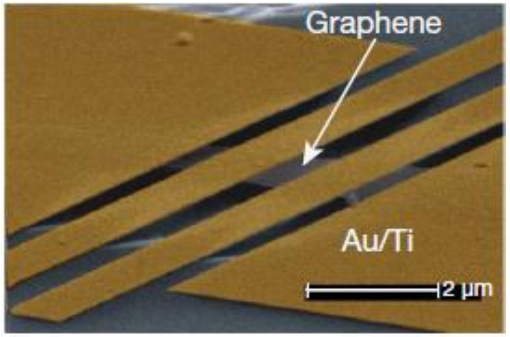
FQHE at $\nu = 1/3$ in monolayer graphene

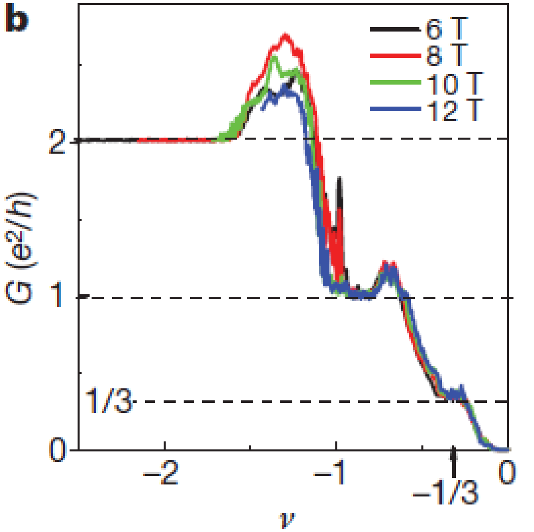
Kim's group





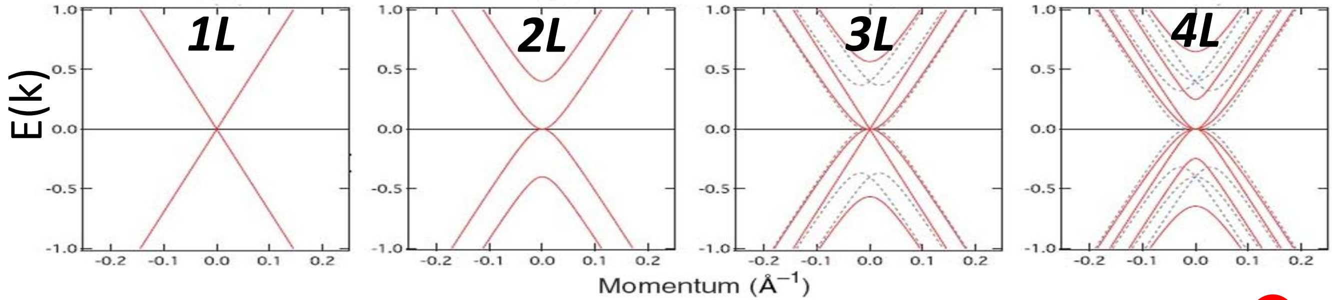
Andrei's group

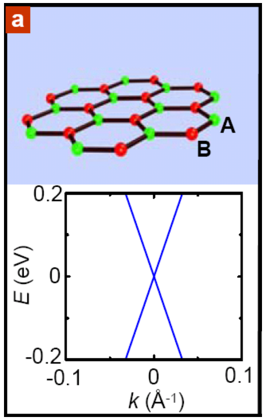




Nature 2009

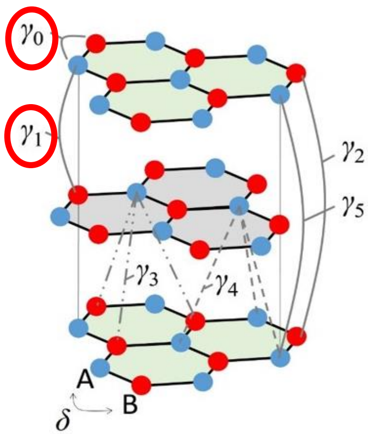
"Ideal" single-particle mono & bilayer at B=0

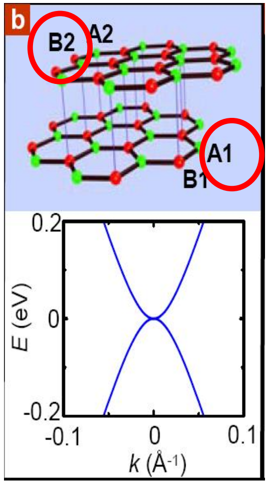




Monolayer = 1LG

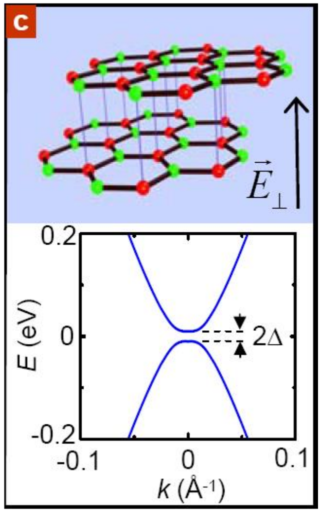
$$H_{1L} = \begin{pmatrix} 0 & \hbar v_F (k_x - i k_y) \\ \hbar v_F (k_x + i k_y) & 0 \end{pmatrix}$$





Bilayer = 2LG

$$H_{2L} = \begin{pmatrix} \Delta & \frac{\hbar^2}{2m^*}(k_x - i k_y)^2 \\ \frac{\hbar^2}{2m^*}(k_x + i k_y)^2 & -\Delta \end{pmatrix}$$

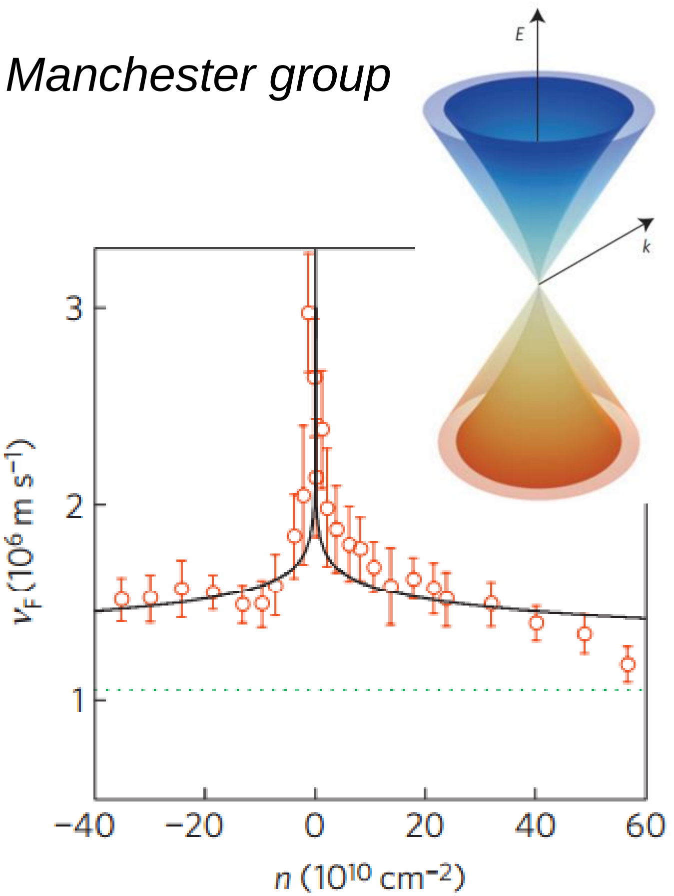


Interaction effects at B=0

Monolayer

Renormalization of Fermi velocity

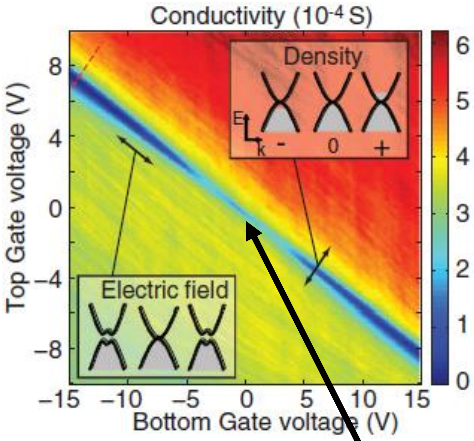
Manchester group



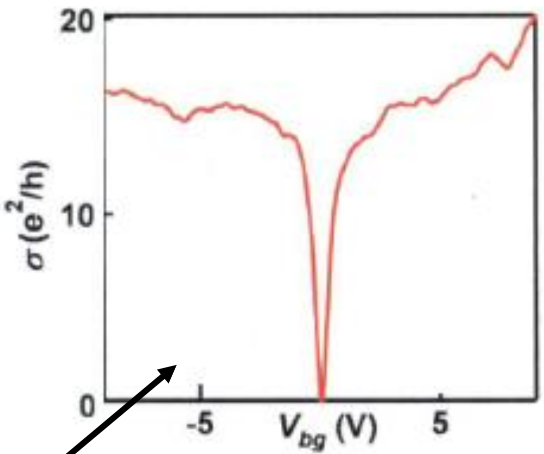
Nat. Phys. 2011

Bilayer

Insulating bilayer @ charge neutrality



Science 2011



PNAS 2012

Suppressed conductance at charge neutrality

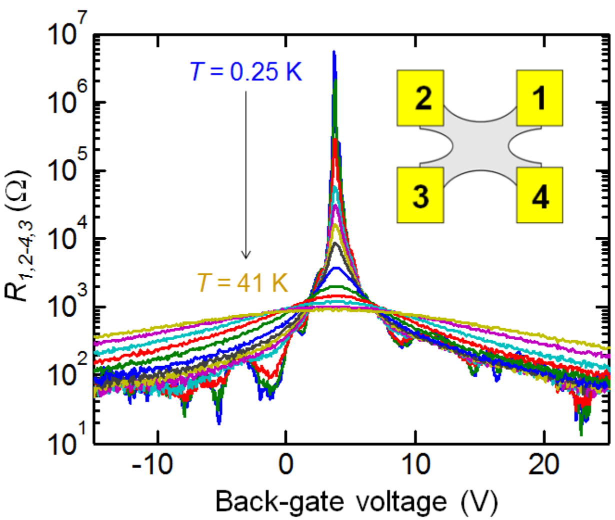
All the action happens for $n < 10^{10}$ cm^{-2}

Symmetry broken gapped state in bilayers

What does it really mean?

$$H_{2L} = \frac{\hbar^2}{2m^*}\begin{pmatrix} \Delta_{s,v} & (k_x - ik_y)^2 \\ (k_x + ik_y)^2 & -\Delta_{s,v} \end{pmatrix}$$

Phase transition
a gap opens because of e-e interactions:
The bilayer becomes an insulator



Nano Lett 2013

- *$\Delta \neq 0$ occurs spontaneously due to e-e interactions*
- *Broken symmetry state with Δ as order parameter*
- *Sign of Δ depends on valley and spin*
- *Exchange energy: in general there is no electric field between the two layers*

…and then:

virtually all the world started working on graphene on hBN and forgot about suspended graphene….

…BUT…

…Graphene on hBN is NOT Graphene…

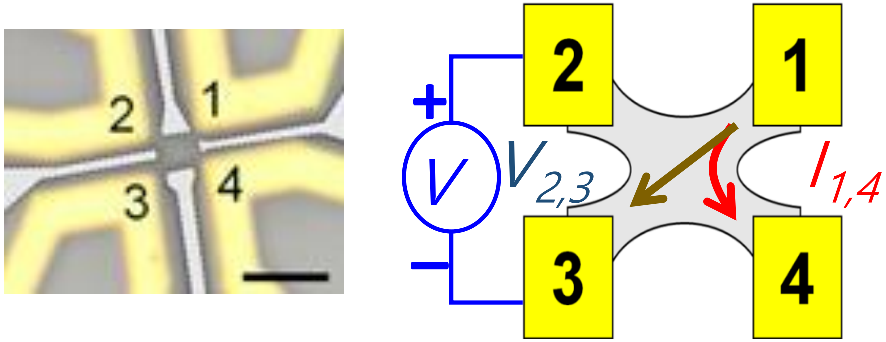
&

…Graphene on any substrate does not reach the quality of suspended Graphene!

So:

We continued working on suspended graphene!

“Multi” devices: Multi-terminal and/or Multi-layer devices



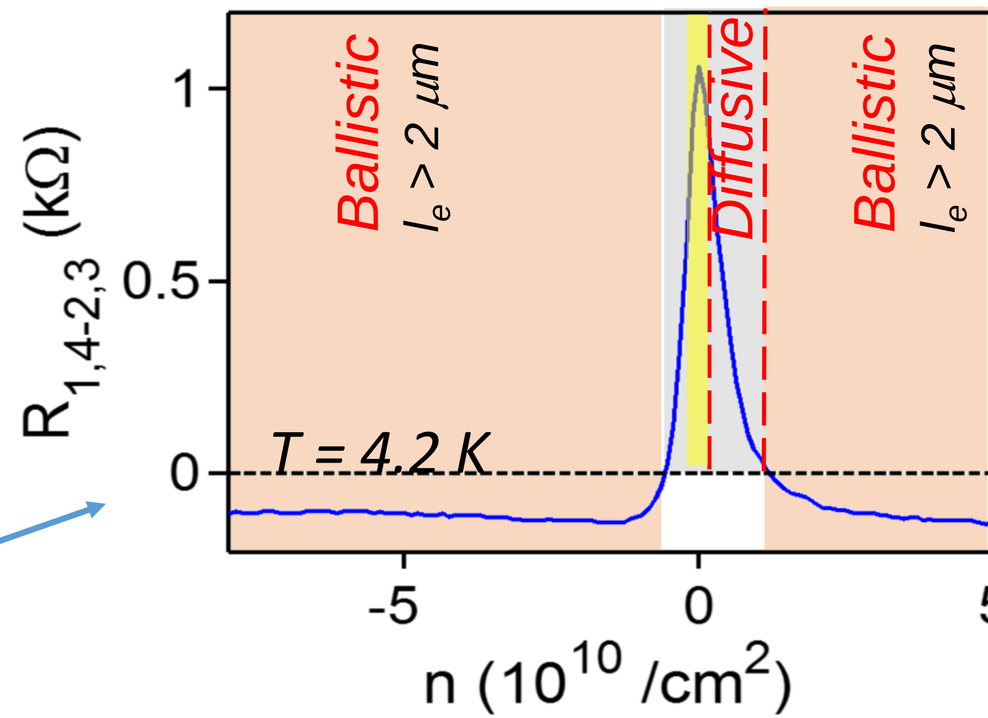
Suspended multilayer devices of very high quality

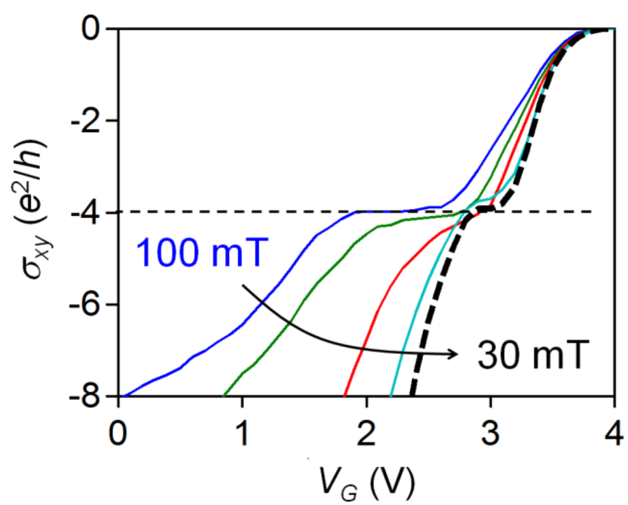
- *Ballistic transport*
For $n > 10^{10}$ cm^{-2}

- *Quantum Hall plateaus*
starting from 300 Gauss

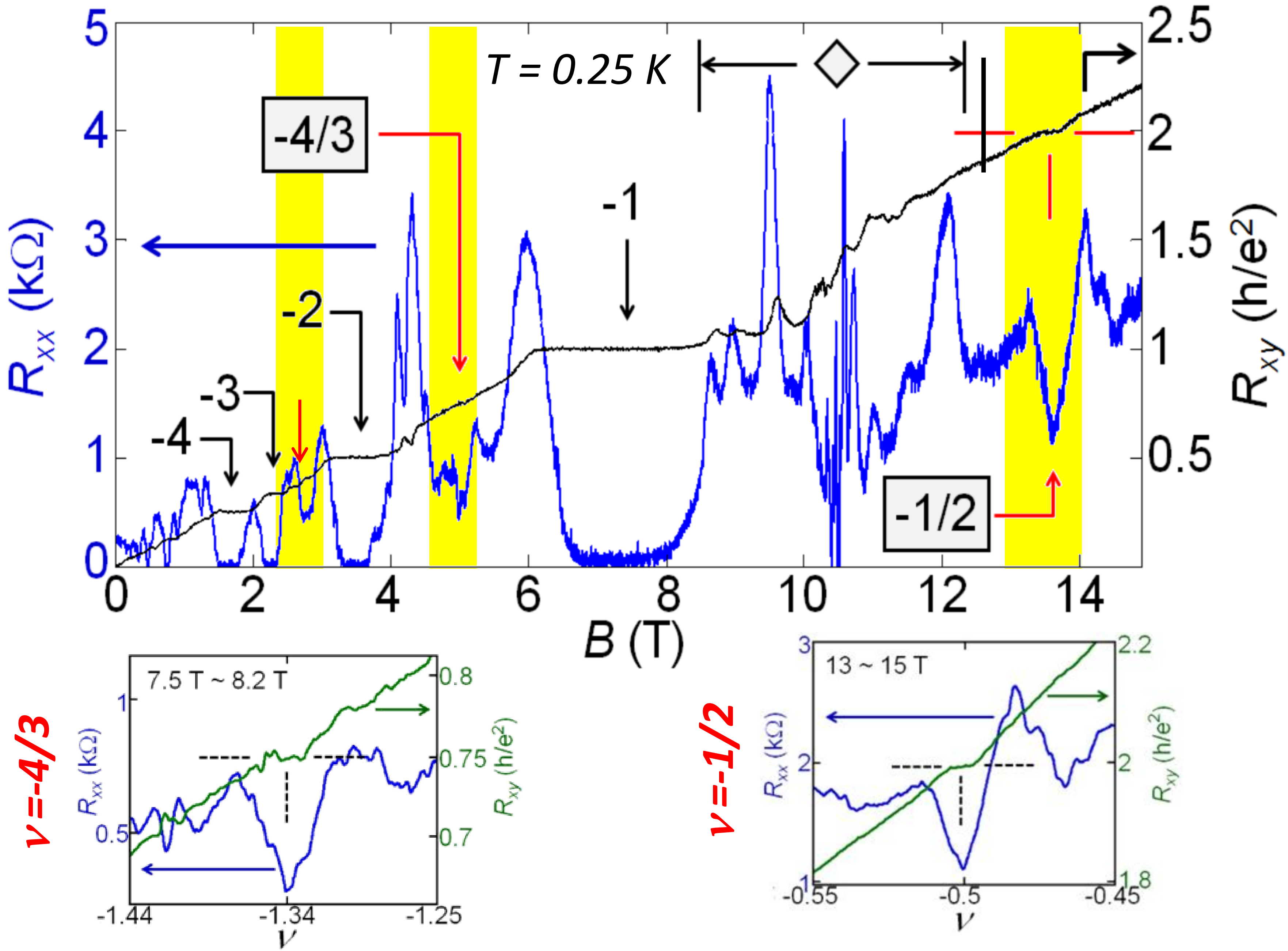
- *Observed*
even-denominator FQHE

Bilayer Graphene

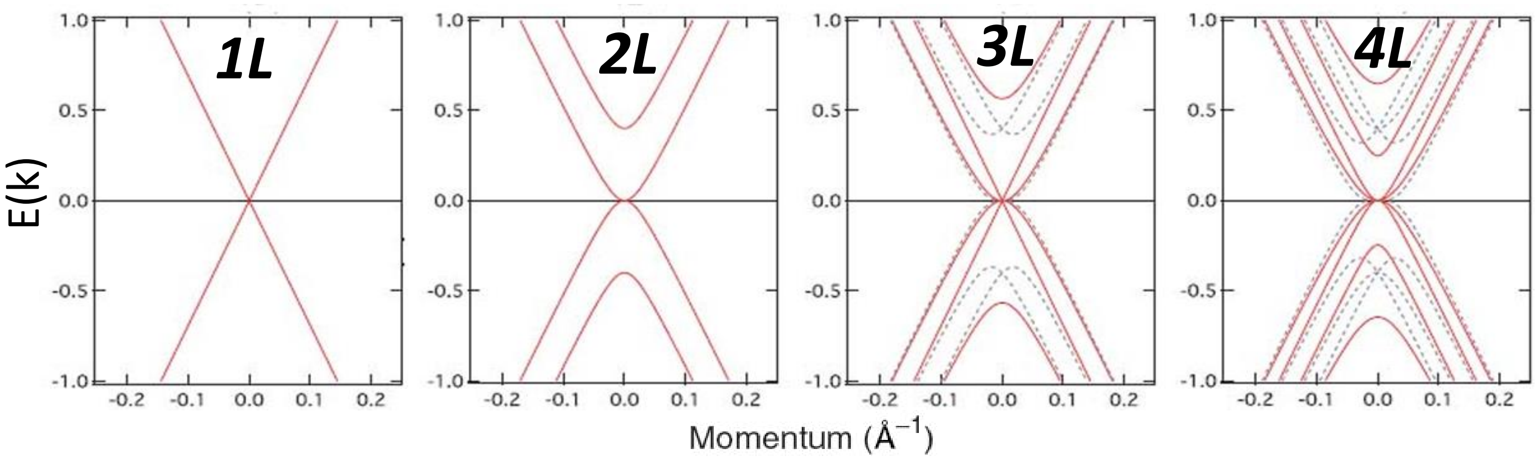




Even denominator FQHE: first time not in GaAs-2DEGs



Identifying Multilayers from Low-B integer QHE



Dirac Band Landau Levels

$$\varepsilon_N = \pm v_F \sqrt{2e\hbar BN}$$

E=0 level: 4 x Degenerate

Bilayer Band Landau Levels

$$\varepsilon_N = \pm \hbar\omega_c \sqrt{N(N-1)}$$

E=0 level: 8 x Degenerate

Shared by valence and conduction band

Ex. Bernal trilayer:

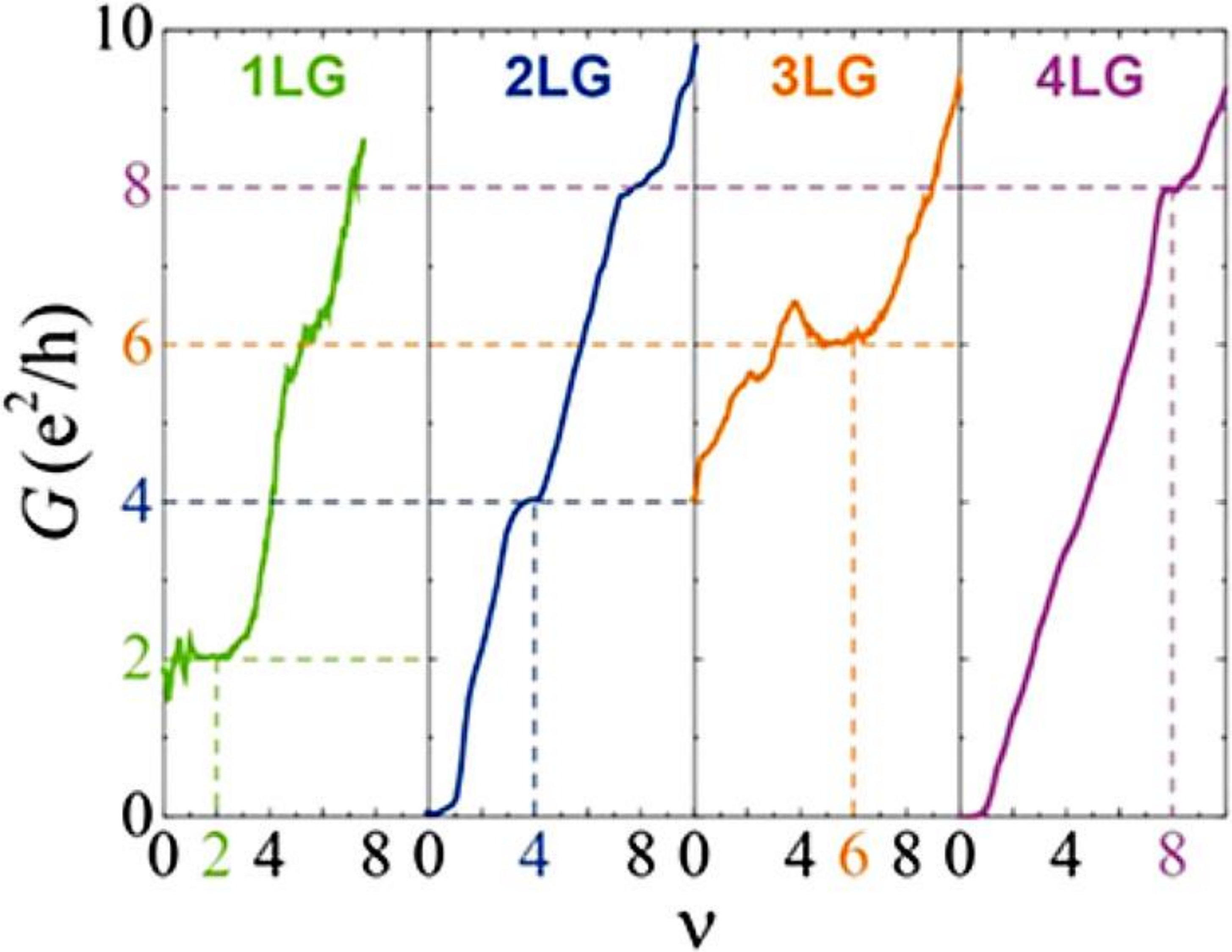
1 x Dirac + 1 x Bilayer bands ----- *E=0 level: 12 x Degenerate*

First low-B plateaus appearing at filling factor:

ν = - 6 (valence band) & ν = + 6 (conduction band)

It works -- but often requires multiterminal devices

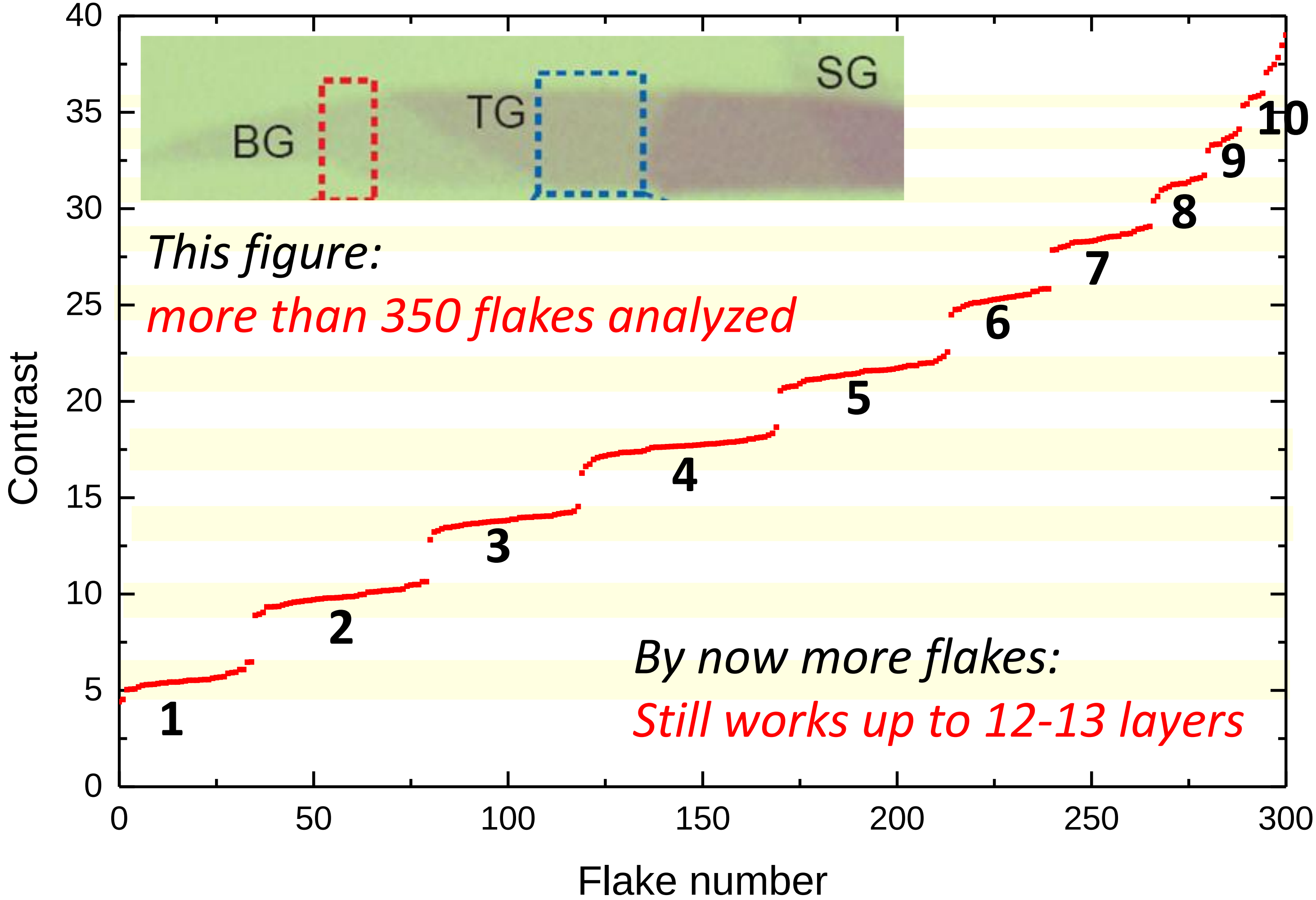
At low B look for the first plateau appearing



Successfully identified up to 8LG so far

Thickness determination from careful contrast analysis

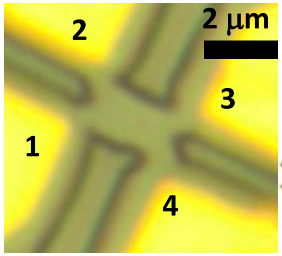
Graphene multilayers on Si/SiO_2



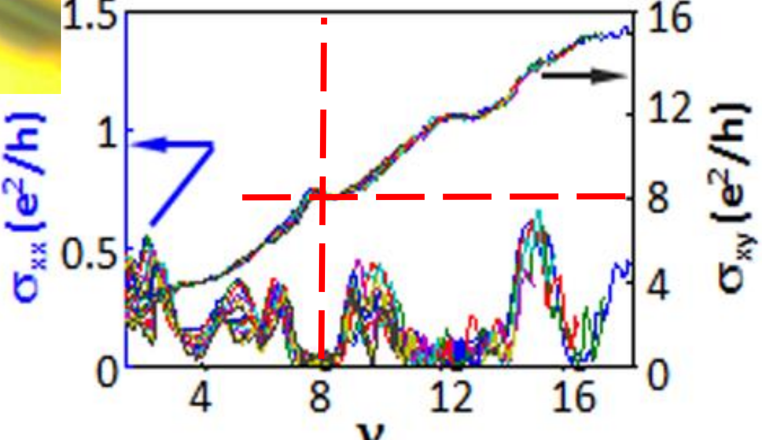
Tell you thickness but does not tell you stacking:

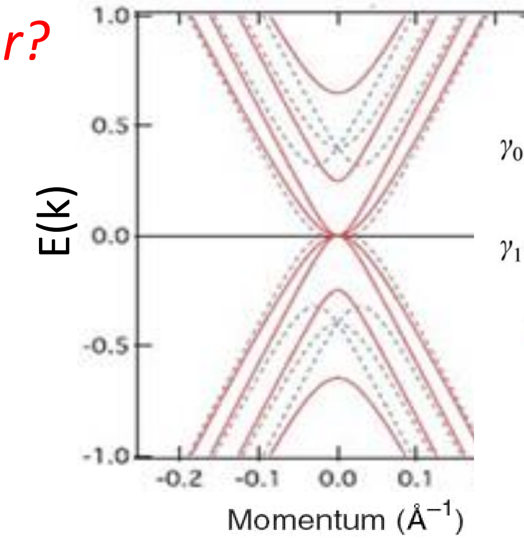
if natural graphite is used to exfoliate Bernal predominates

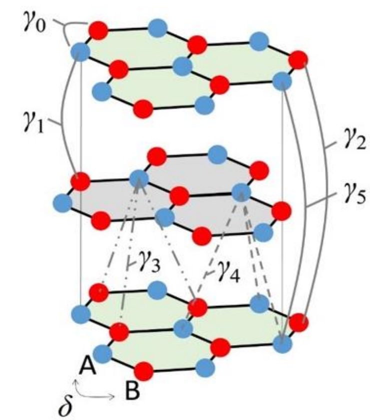
OK…But is there anything interesting in thicker multilayers?



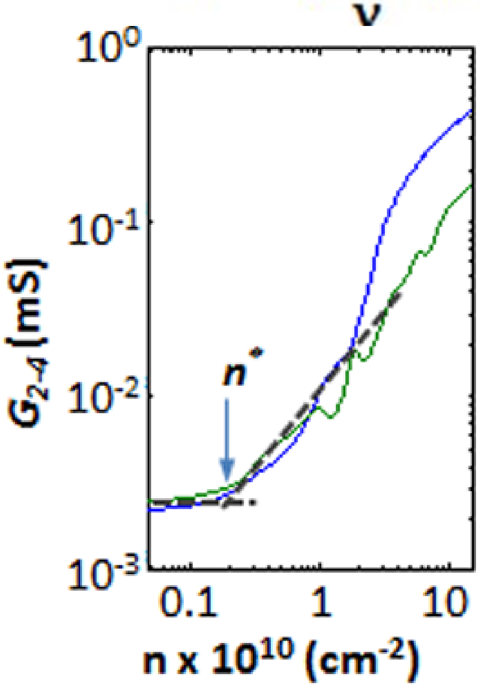
4LG: - Zero-gap semiconductor?
- Semi-metal?



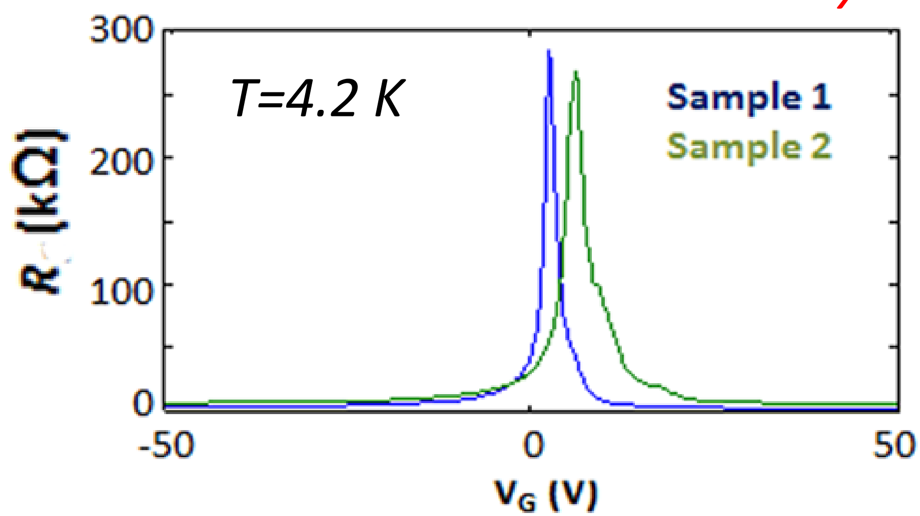




Very narrow Dirac peak
(n^* ~ 3 10^9 cm^{-2} at 4.2 K)

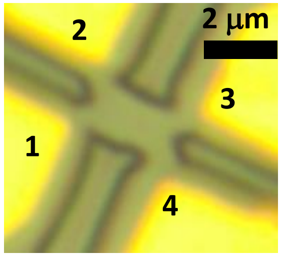


Highly resistive @ T=4.2 K:
50 x more resistive than bilayers



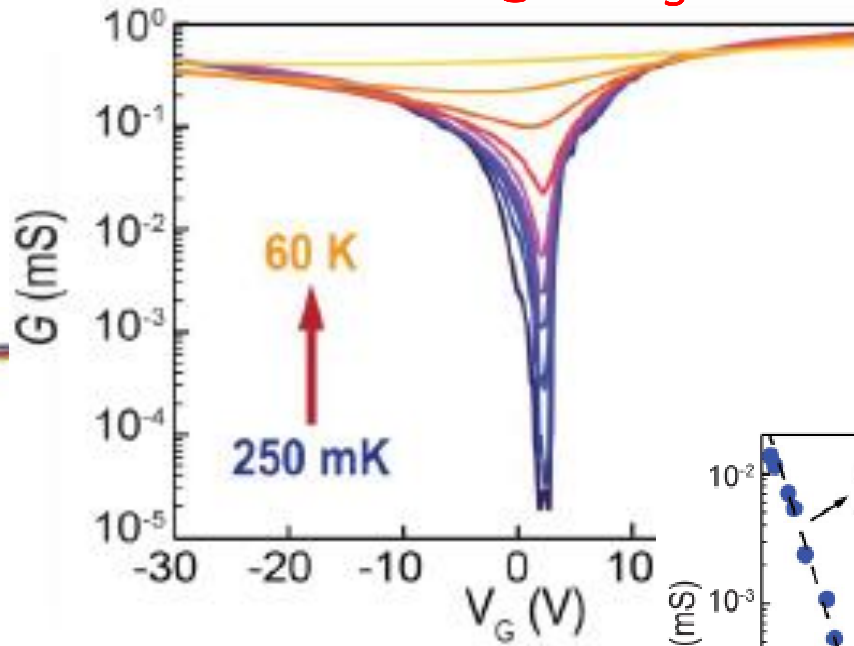
…Yes: there is!

Bernal 4LG are insulators --- more insulating than bilayers



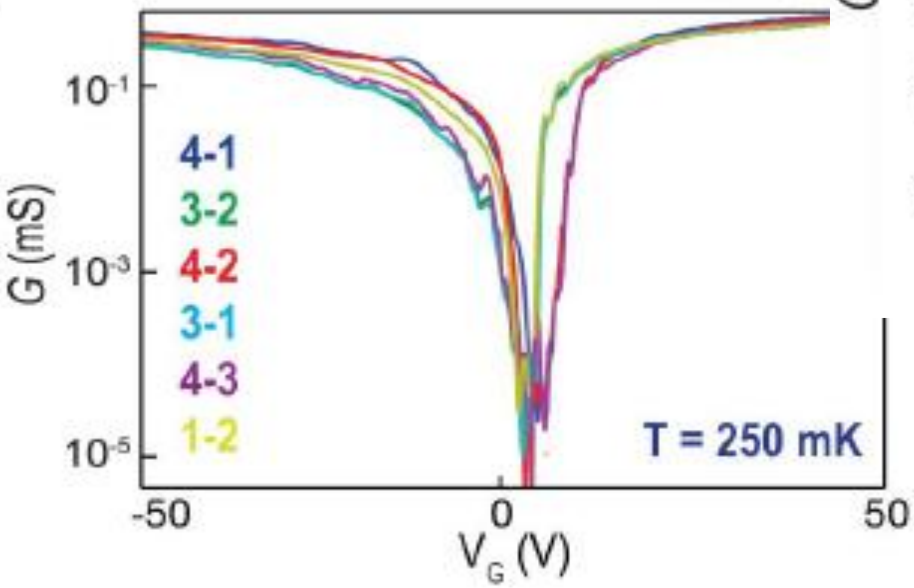
Thermally activated conductance

@ charge neutrality



Uniform:

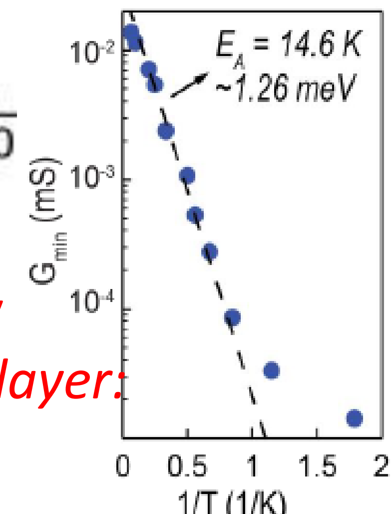
seen in all pair of contacts



Activation energy

Comparable to bilayer:

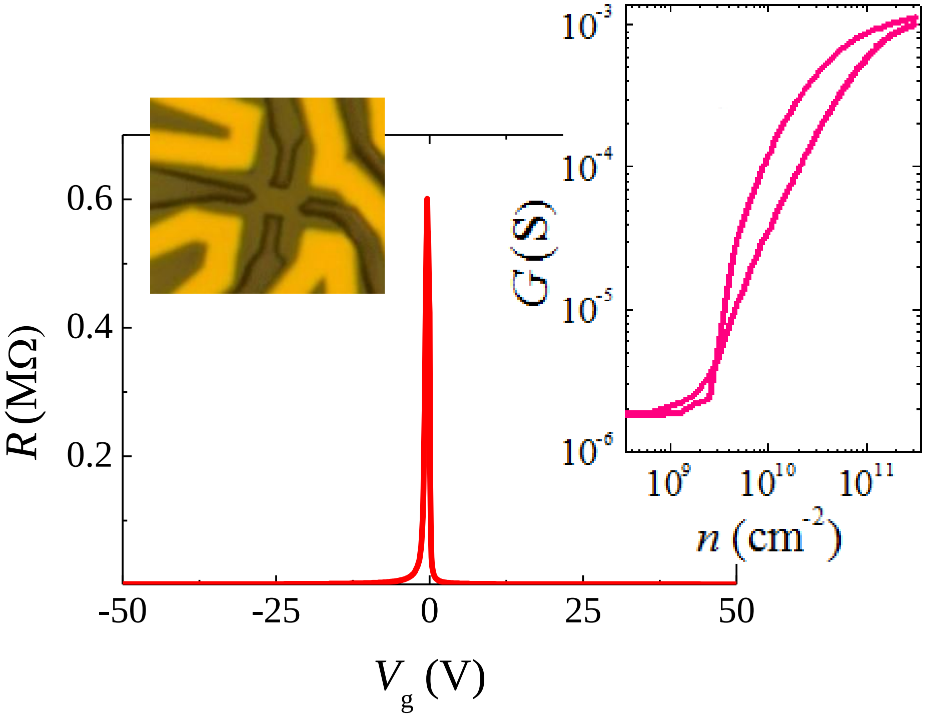
….strange



Increasing thickness makes the behavior and deviate more from that of graphite….is this a coincidence?

Let's check Bernal-stacked 6LG

- *Very sharp peak:* $\delta n \sim 2\ 10^{9}\ cm^{-2}$;



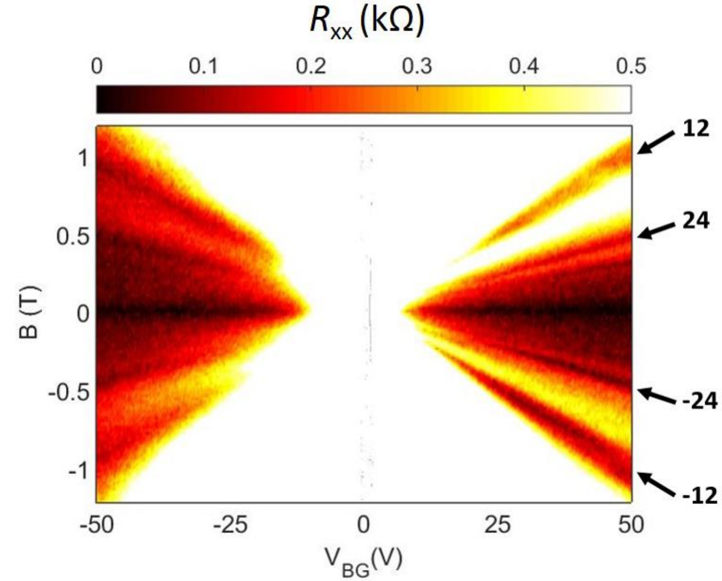
High resistance:

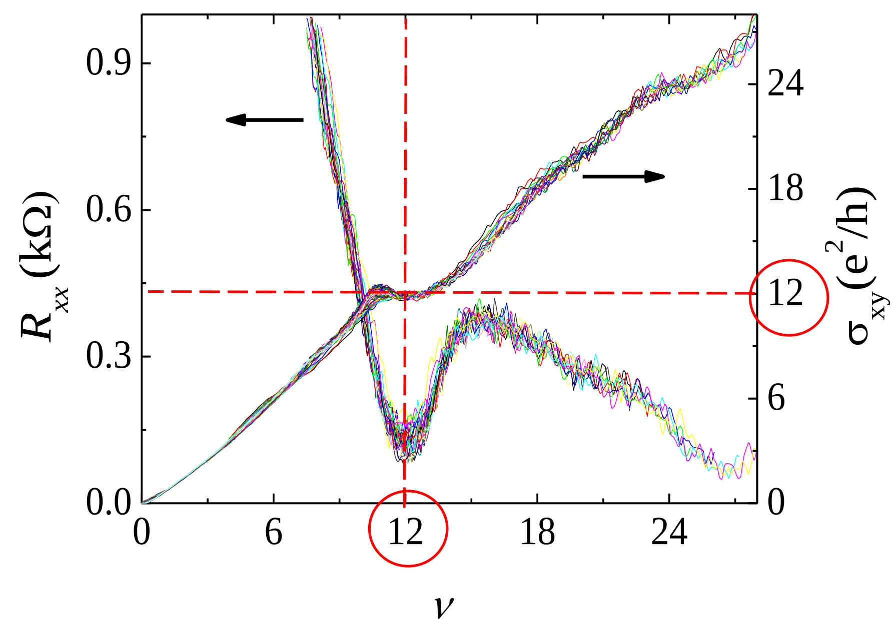
@ charge neutrality and T= 4.2 K

R=0.6 MΩ

- *At low B*

Dominant plateau in σ_{xy} at 12 e^2/h





6L Graphene also has high resistance at CNP

It does not stop…Bernal-stacked 8LG

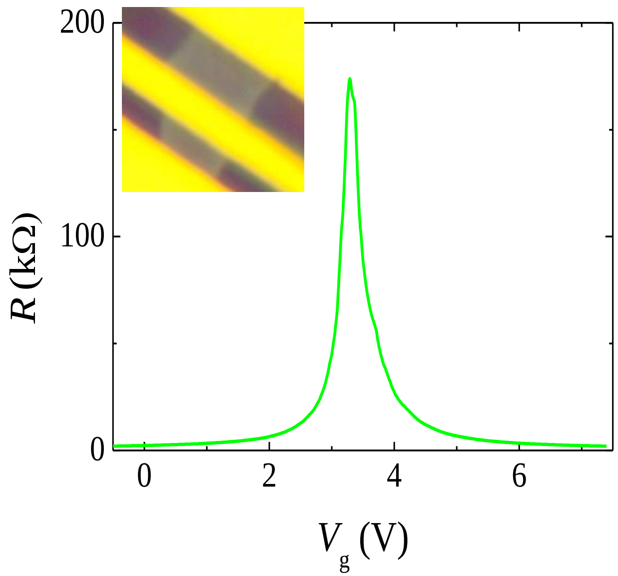
At low B

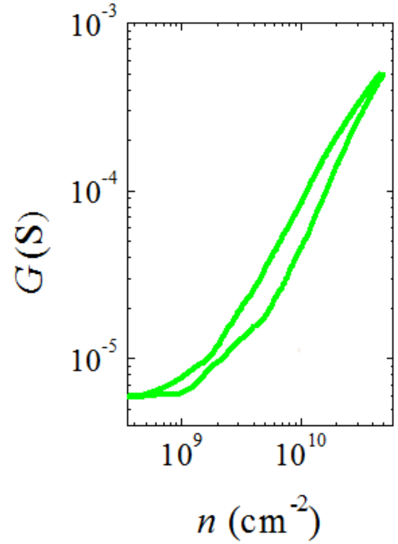
Dominant plateau

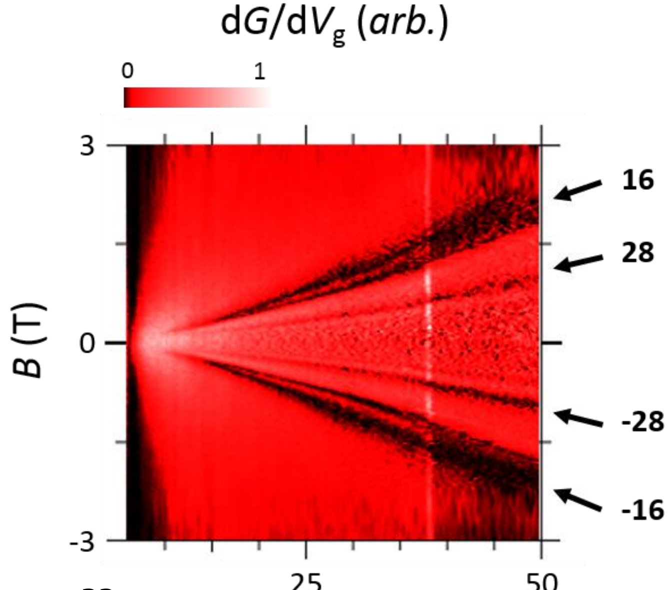
σ_{xy} at 16 e^2/h

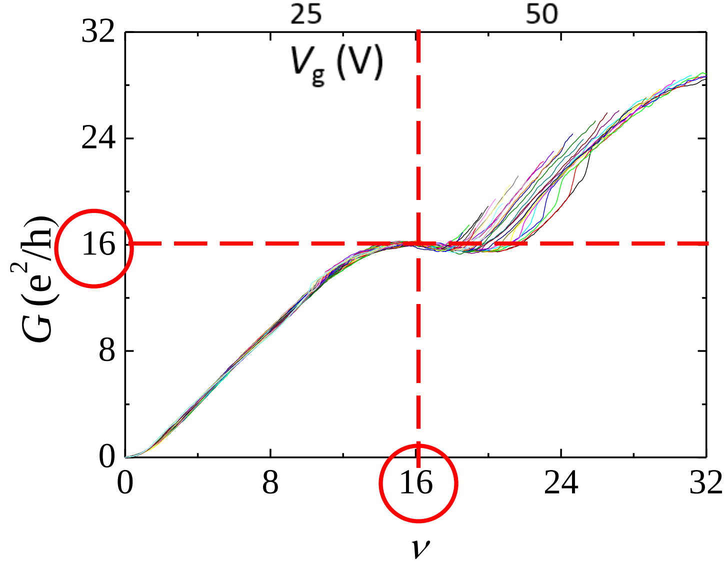
In this case only 2-terminal device

- *Again: very sharp peak, $\delta n \sim 2\ 10^9\ cm^{-2}$;*









High resistance:

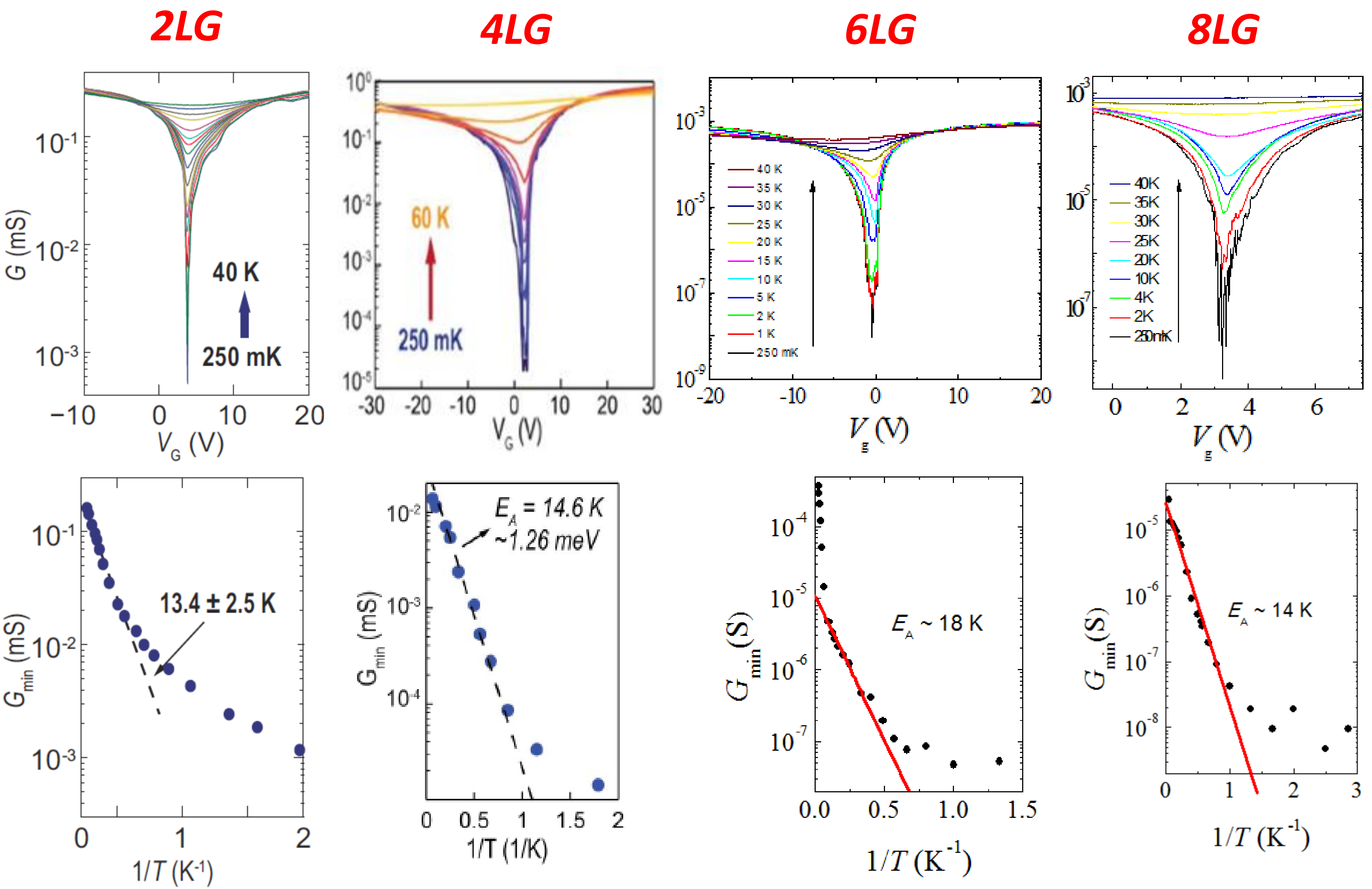
@charge neurality & T= 4.2 K

R=0.2 MΩ

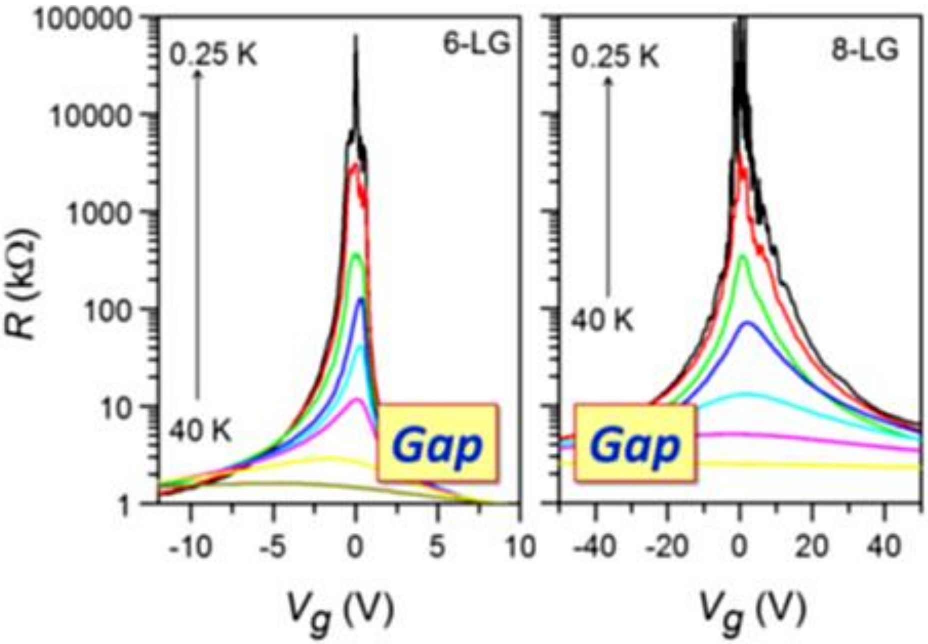
Square resistance ~ 350 kW

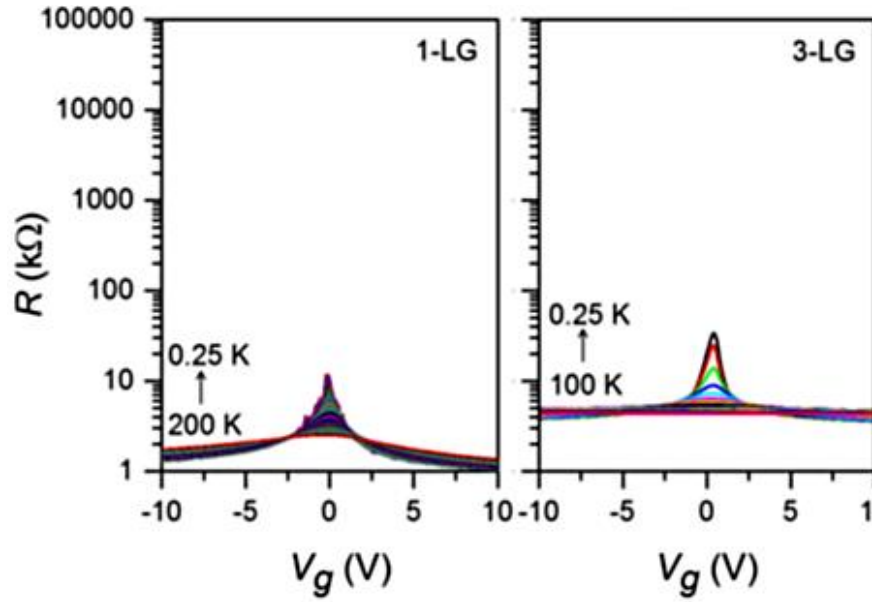
Same as in 4LG and 6LG

Resistance temperature dependence



What about odd multilayers? Even-Odd !





We are onto something…but what?

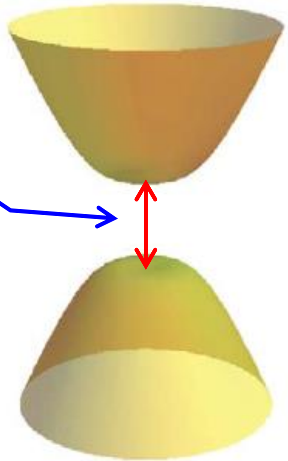
Let's not forget: Even-Odd !

Δ —— a self-consistent mean-field potential explains insulating bilayer

$+\Delta$

$-\Delta$

opens a gap at CNP



$$H = \begin{pmatrix} +\Delta & (k_x - ik_y)^2 \\ (k_x + ik_y)^2 & -\Delta \end{pmatrix}$$

Let's generalize this idea to thicker multilayers:

Δ —— a self-consistent mean-field staggered potential

Nat. Commun. **6** 6419 (2015)
2D Mater. **3**, 045014 (2016)

Ex.: 4-layer graphene

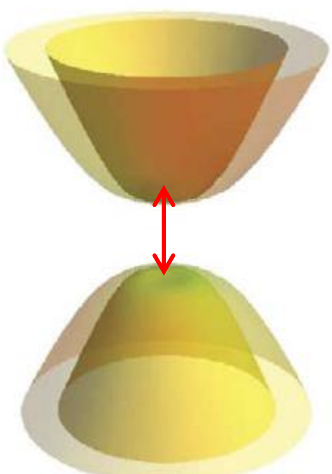
Staggered potential:
opens a gap @ CNP in 4LG

$+\Delta$

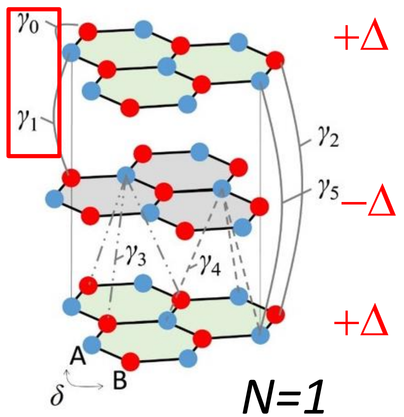
$-\Delta$

$+\Delta$

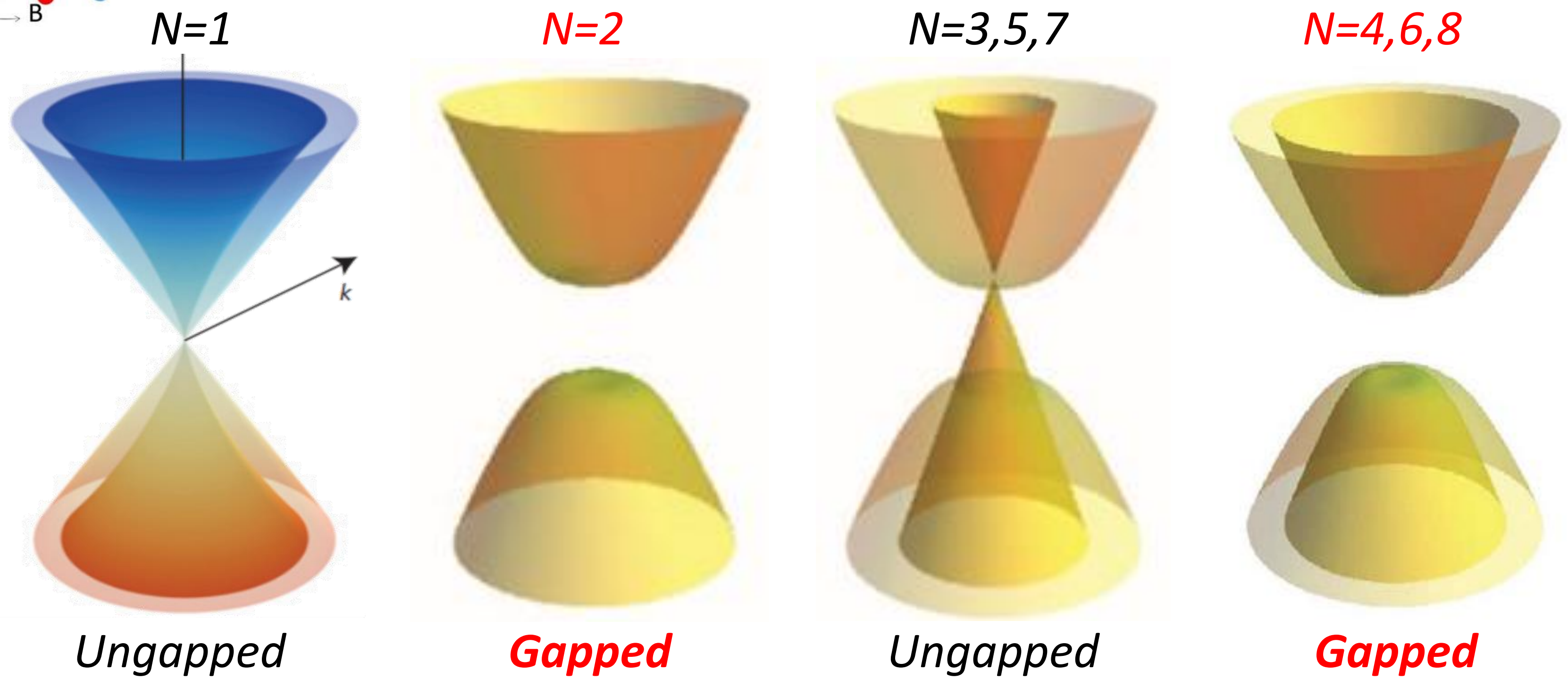
$-\Delta$



Staggered potential: it works for all thicknesses!



- *All even multilayers:*
  fully gapped
- *All odd multilayers:*
  parabolic bands gap out, but Dirac band doesn't



Nat. Commun. ***6*** *6419 (2015)*
2D Mater. ***3****, 045014 (2016)*

Insulating state and Even-Odd effect explained!

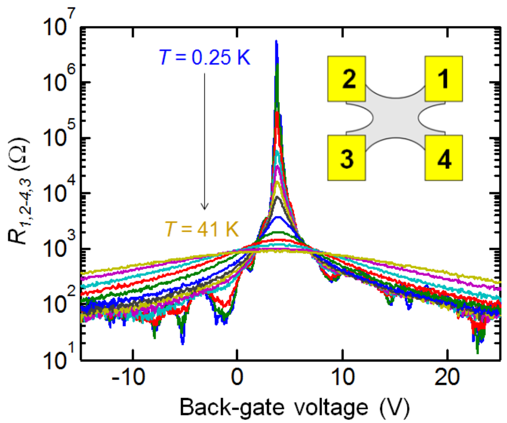
Are we done ? *Not really*

- *Do we have any direct evidence that e-e interactions are doing anything in odd layers?* *No !*

- *Mean-field staggered potential:*

There must be a phase transition at which the staggered potential (which is the order parameter) appears. What kind of phase transition?

– *Quantum phase transition at T=0 as a function of n?*
– *Finite temperature phase transition at T_c? What is T_c?*



- *How large is the staggered potential? Is it the same for all thicknesses?*

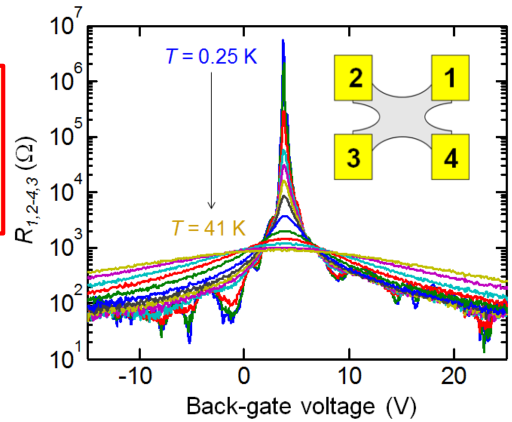
- *At which thickness do we recover the behavior of graphite?*

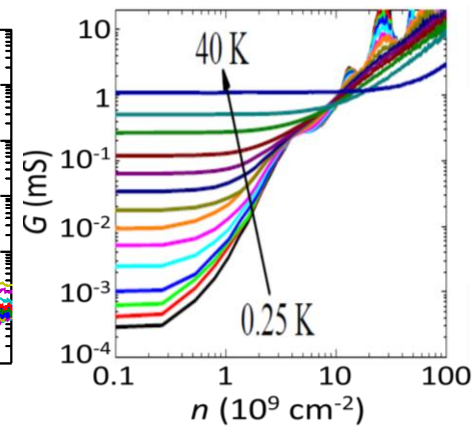
Probing the density of states from the width of the Dirac peak

Beyond R(T) measurements

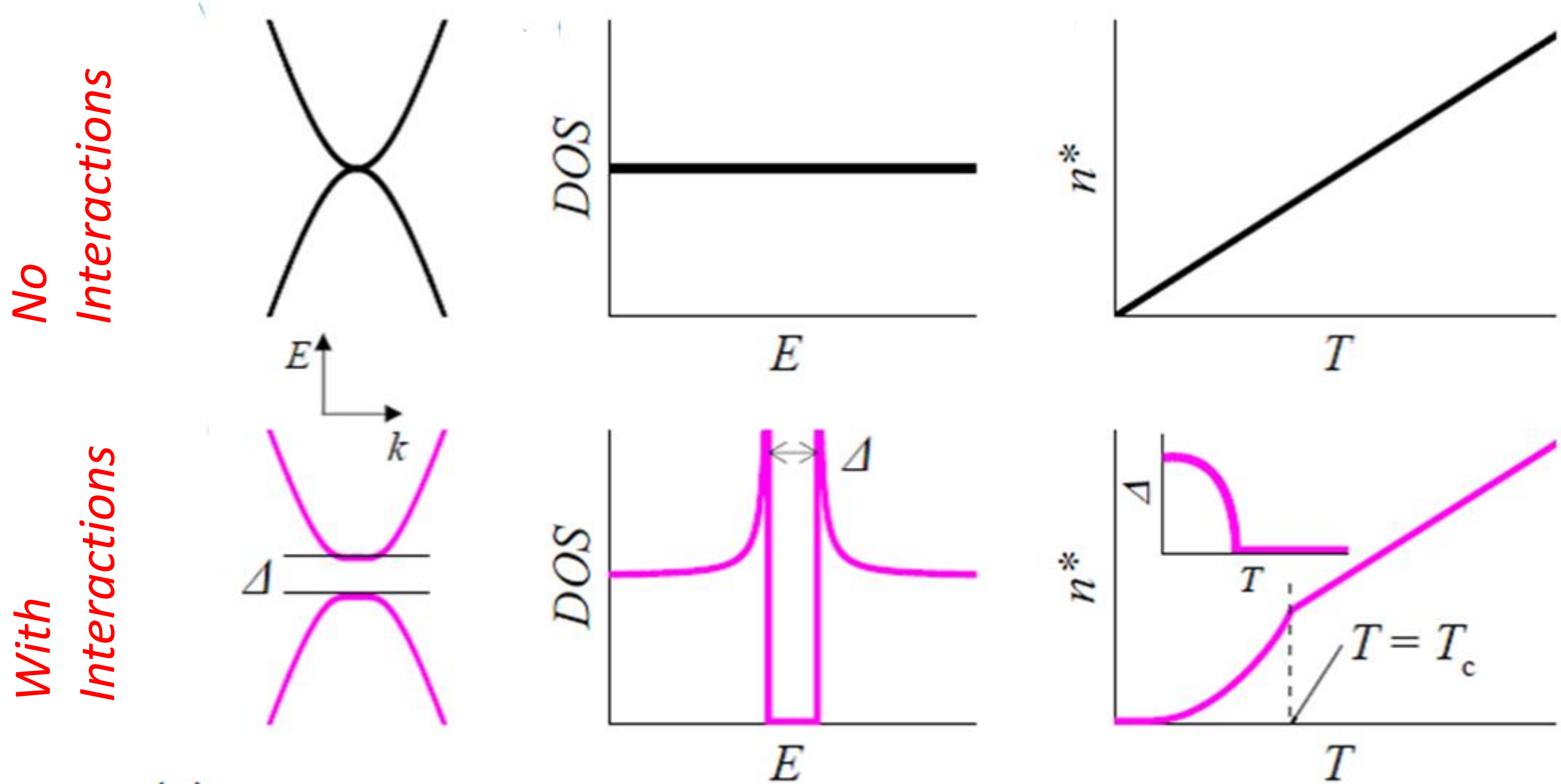
$$\boxed{n^*(T) = \int_0^\infty g(E,T) \times f(E,T)\,dE}$$

Density of states



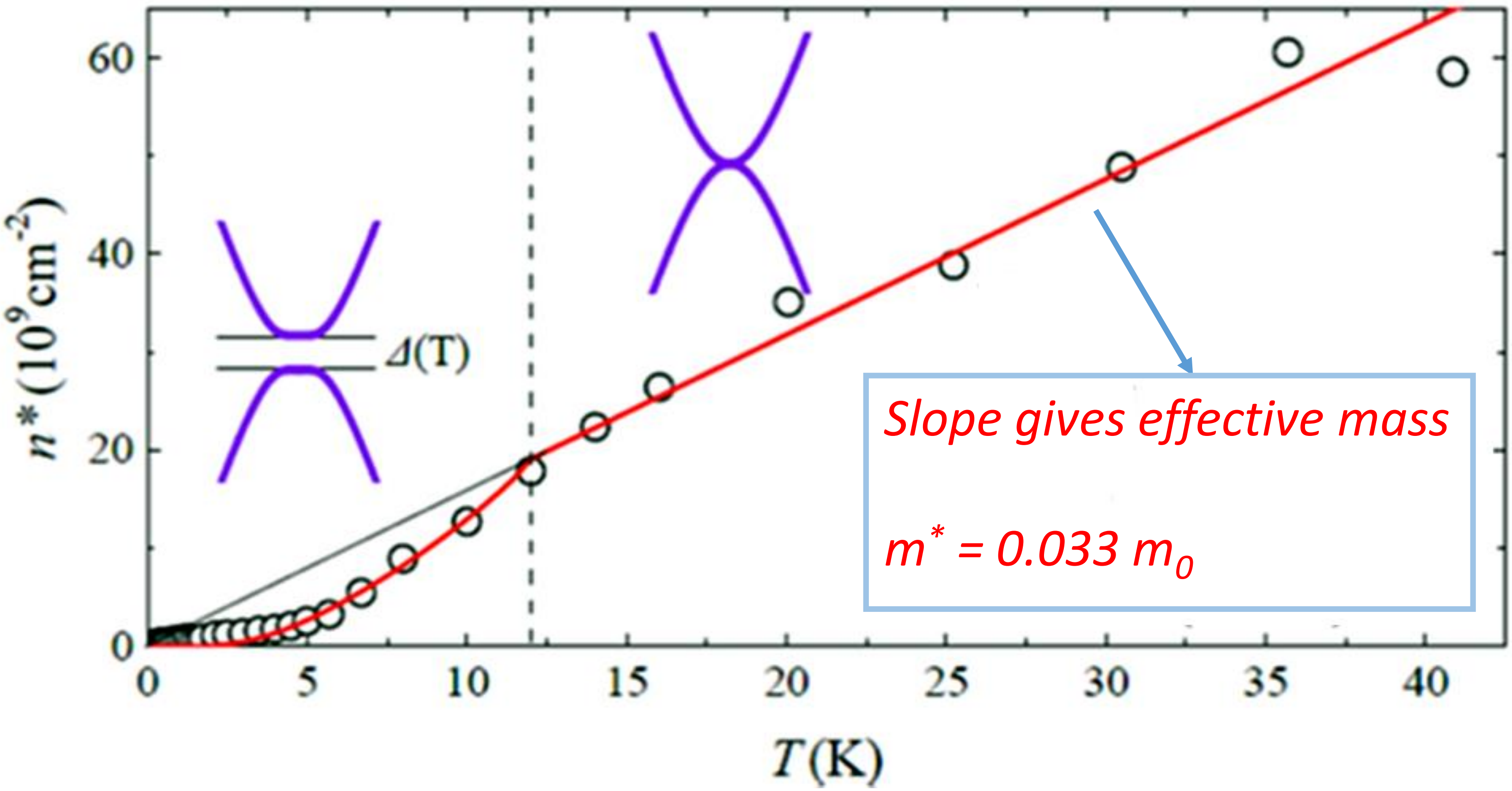


Ex.: Bilayer Graphene



It works!

Second-order phase transition at finite T: Δ goes to zero at T_c=12 K

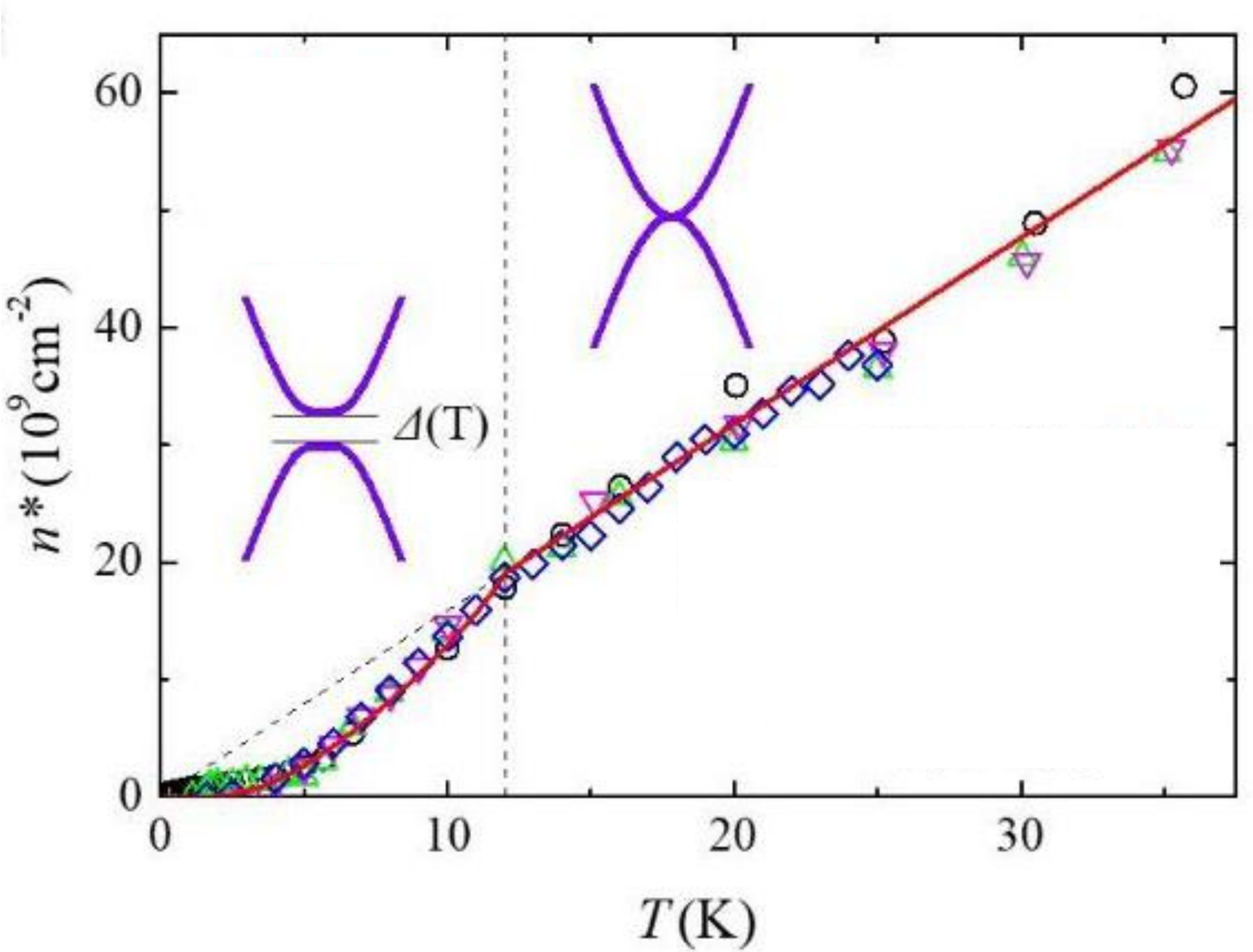


Mean-field T-dependence of the gap Δ(T)

$$\Delta_{T<T_c} = \Delta_0 tanh\left(1.74\sqrt{\frac{T_c}{T}-1}\right)$$

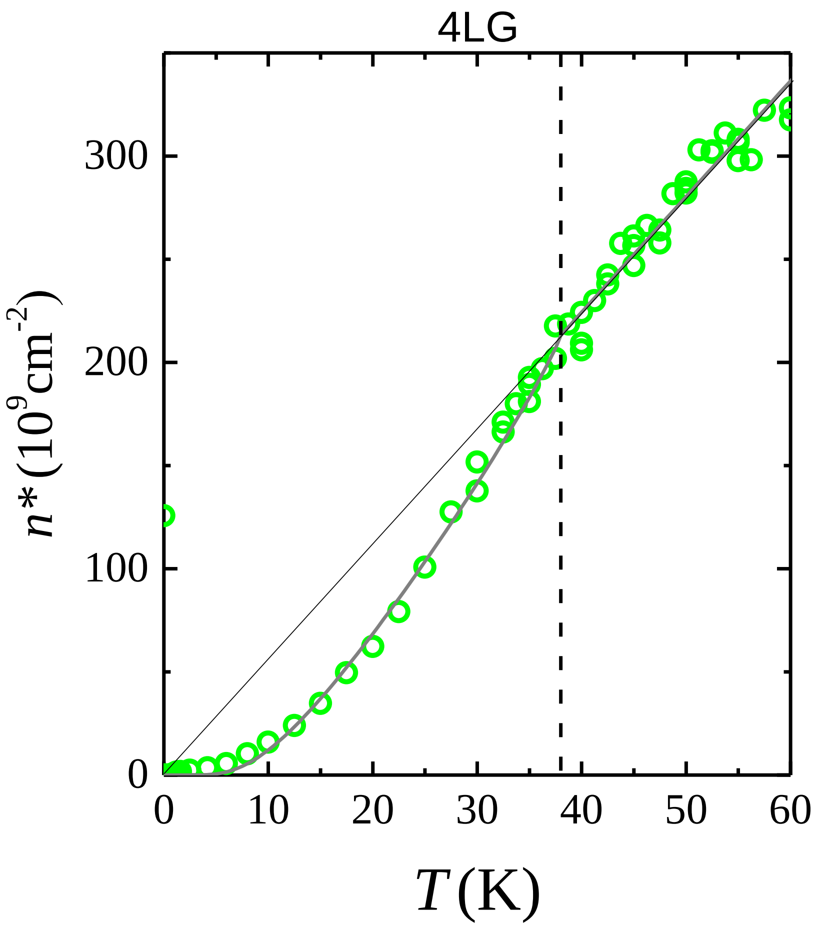
$$\Delta_{T>T_c} = 0$$

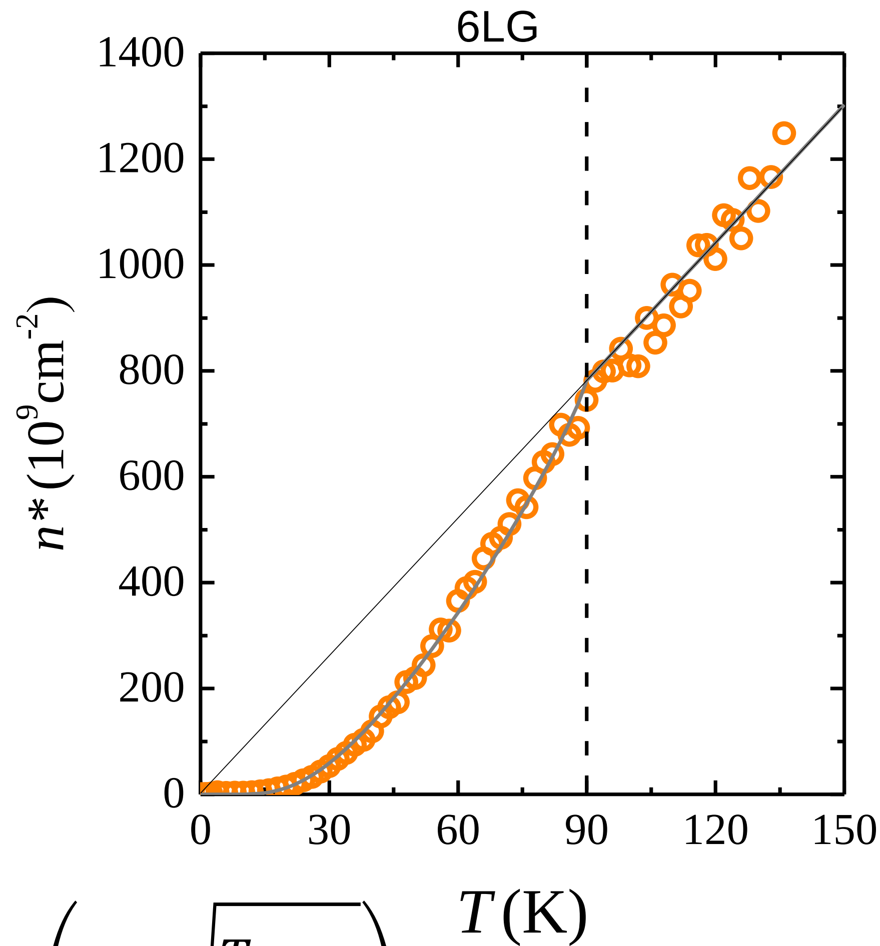
Very Reproducible



Not only for bilayer, also for 4LG, 6LG,…

And look at the values of T_c…





$$\Delta_{T<T_c} = \Delta_0 tanh\left(1.74\sqrt{\frac{T_c}{T}-1}\right)$$

$$\Delta_{T>T_c} = 0$$

4LG

T_C = 38 K

$\Delta(0)$ = 60 K

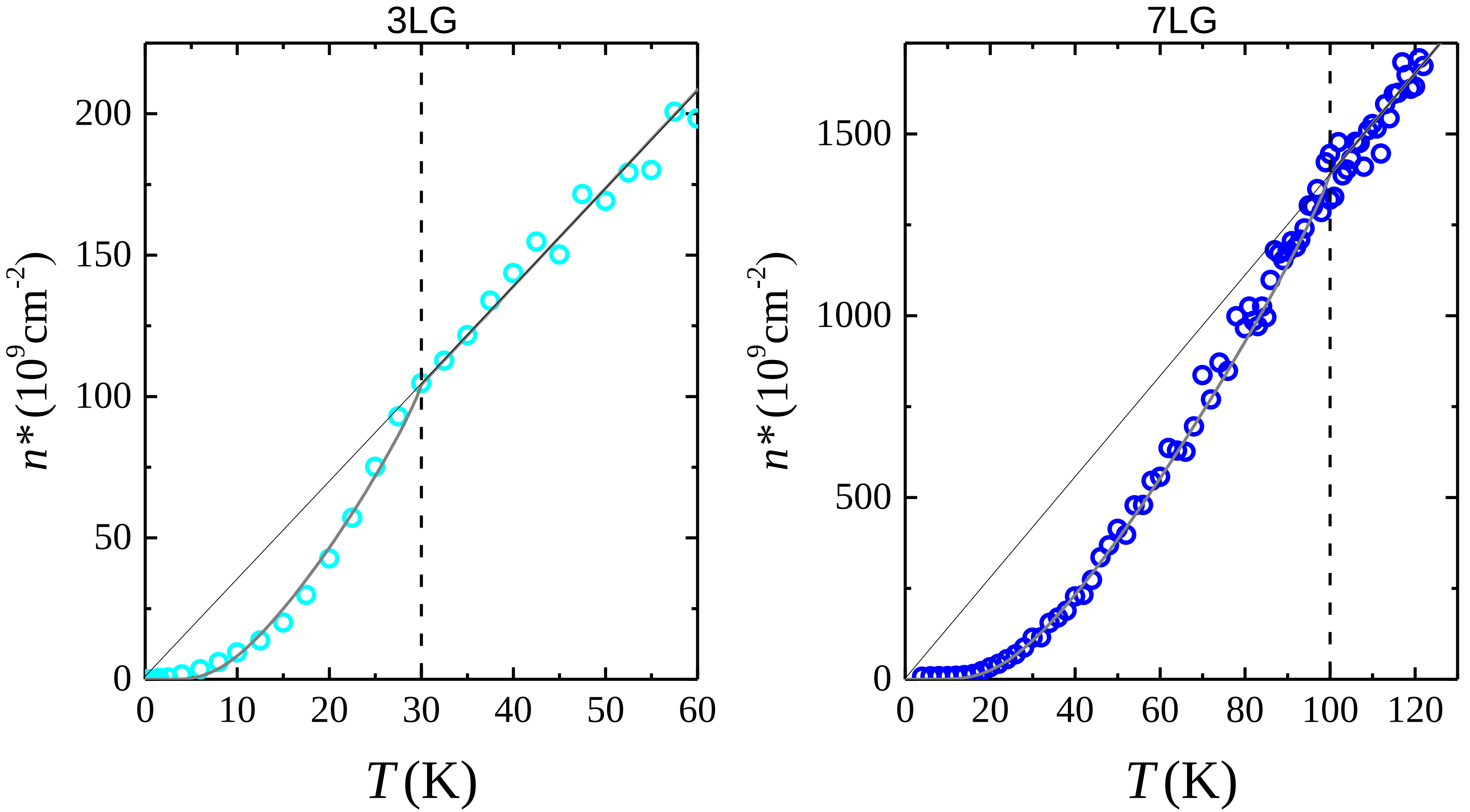
6LG

T_C = 90 K

$\Delta(0)$ = 150 K

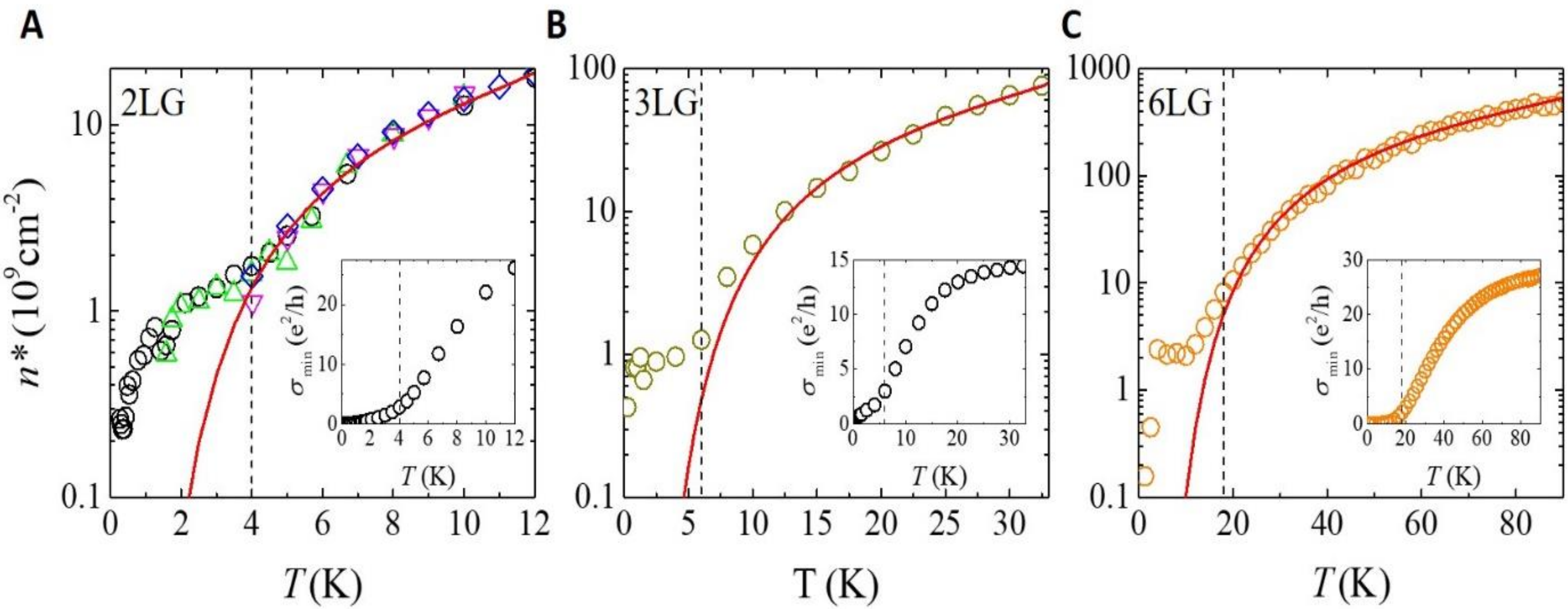
…and it works for odd layers as well: 3LG, 5LG, 7LG

In this density range the DOS of the quadratic bands is ~ 100 x the DOS of the Dirac band



Direct demonstration of e-e interaction effects in odd Bernal-stacked multilayers

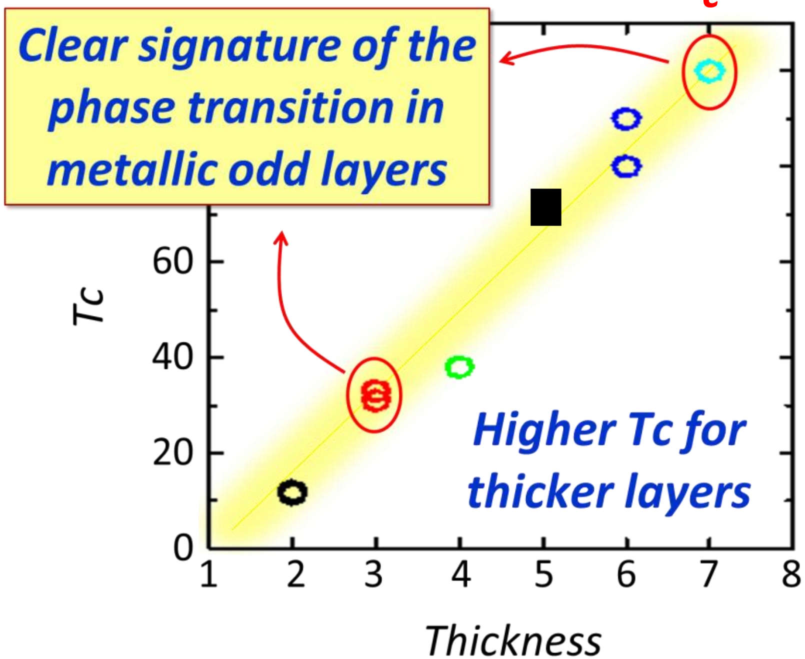
Looks good also on log scale



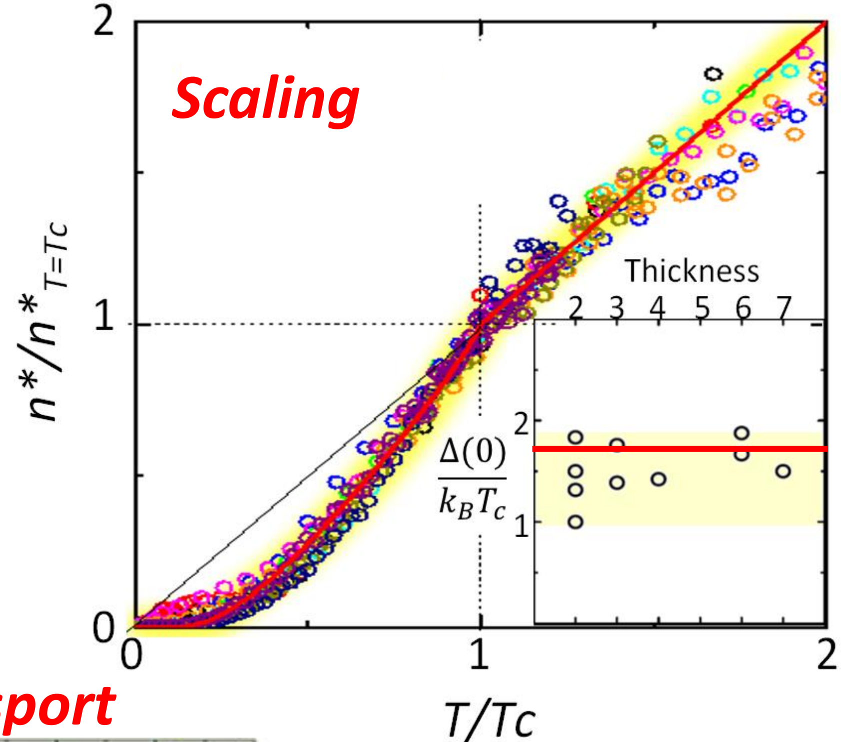
Methods break when G < e^2/h

Systematics

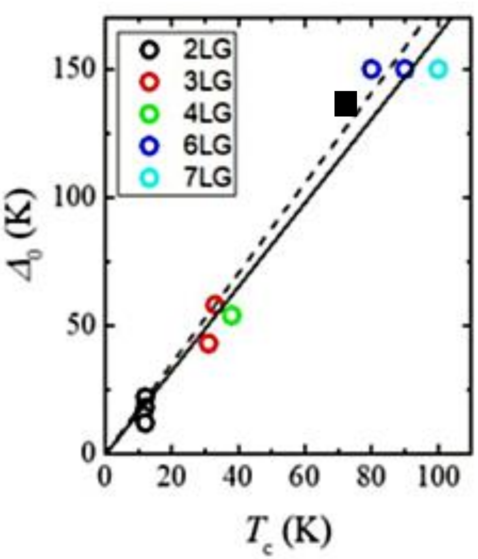
T_c



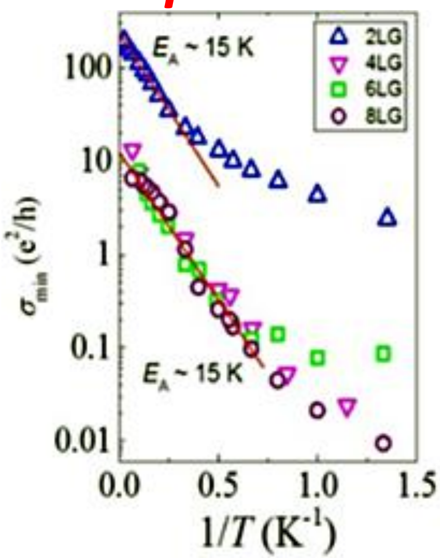
Scaling



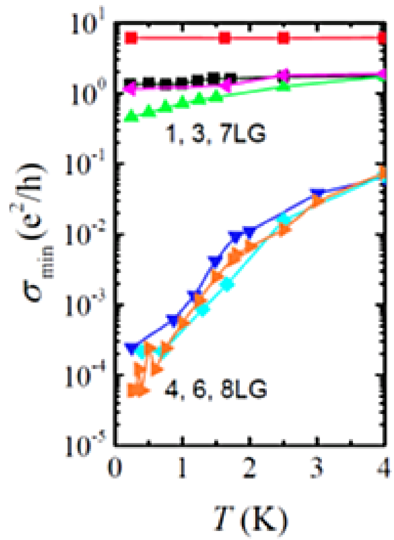
Gap



Transport

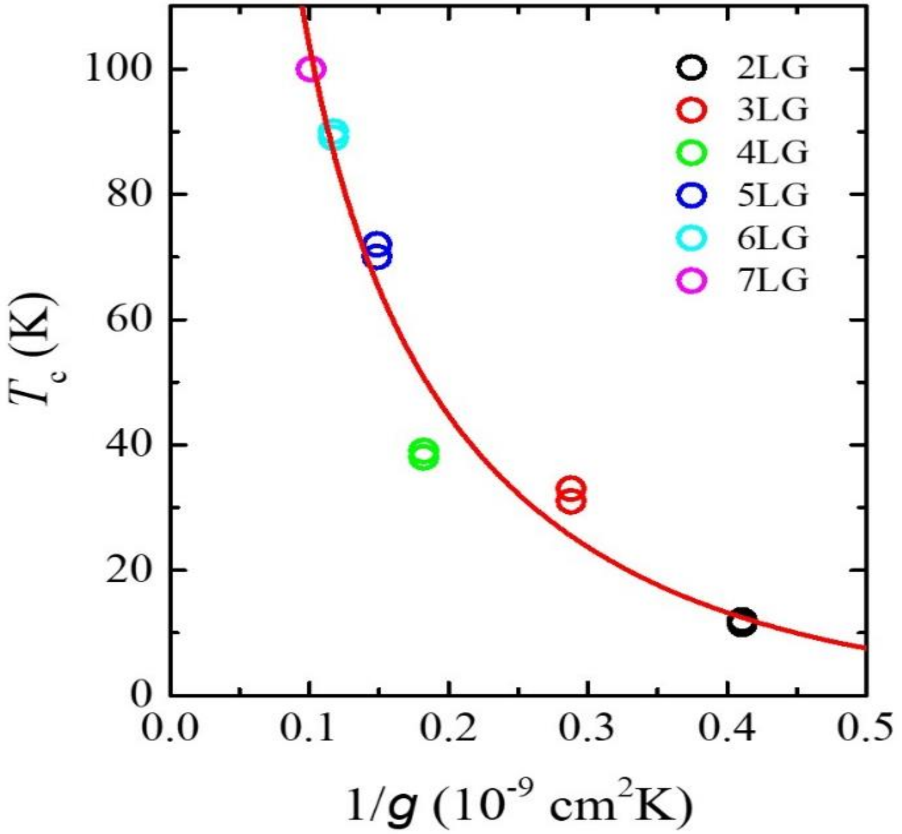


Even-odd



Activation energy is NOT the gap (edge transport?)

Can we understand why T_c increases with increasing thickness?

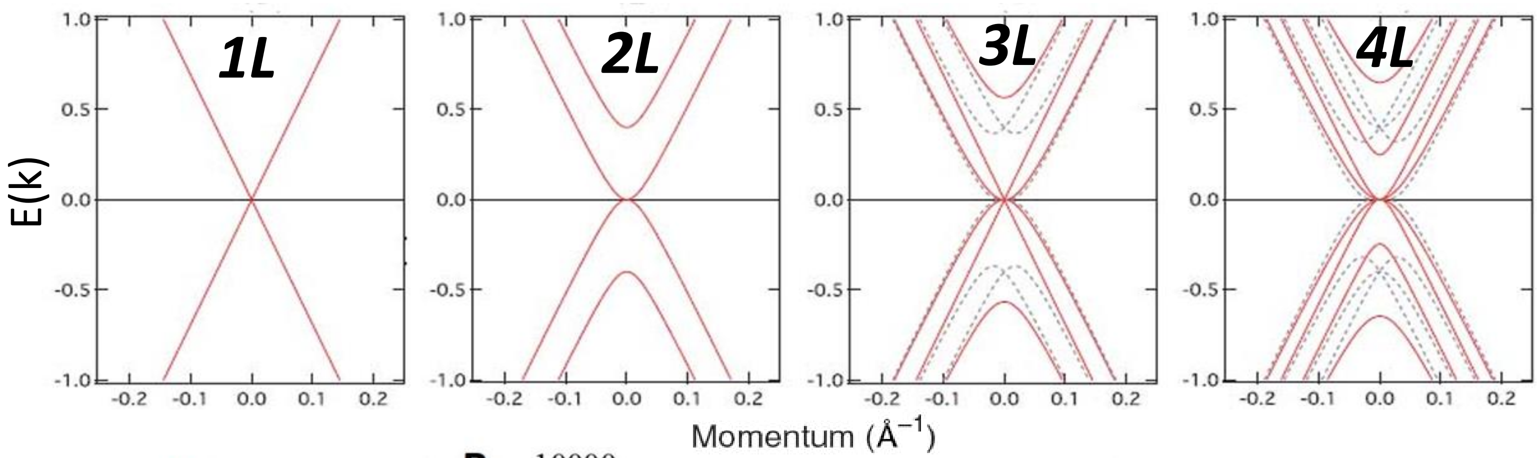


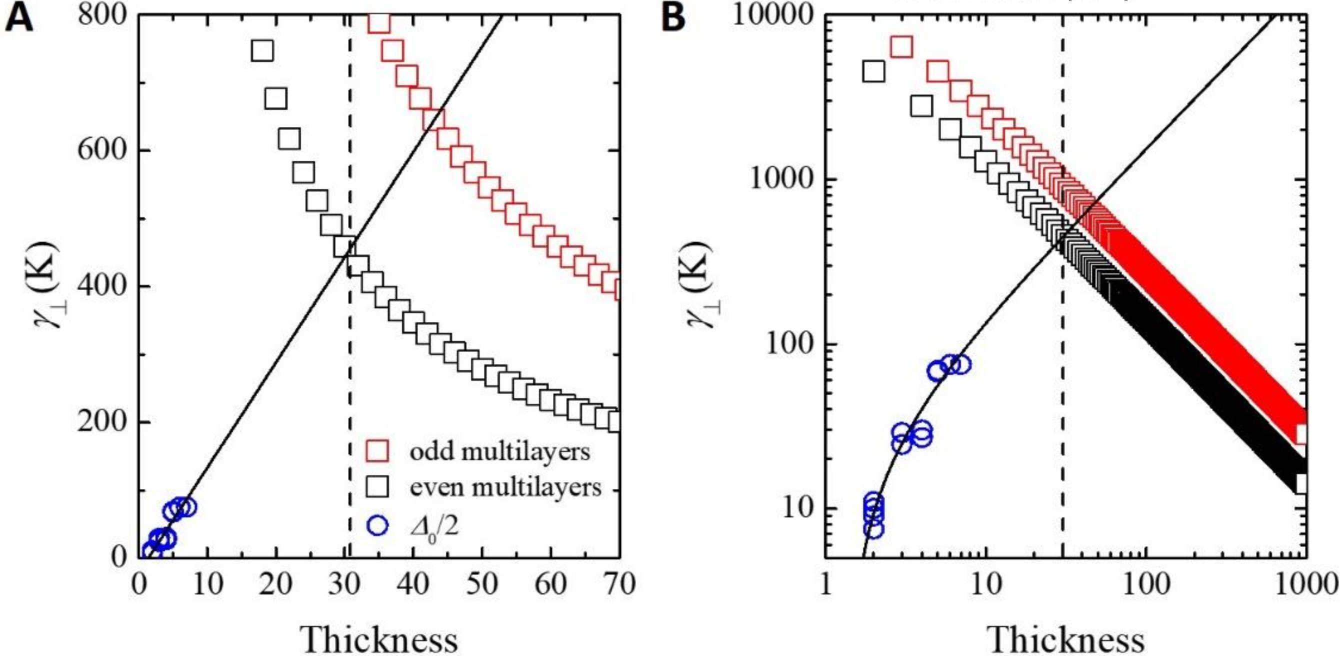
$$k_B T_c = \frac{E_{cut-off}}{2\sinh[1/(gV)]}$$

Maybe, but can it be justified…?

When will it stop? Don't know, but…

If Δ gets too large, can't use low-energy Hamiltonian anymore





At around 20 LG Δ will hit the "high energy bands"

New scenario certainly needed past that thickness

Ballistic transport limited by e-h collisions in charge neutral suspended bilayer graphene

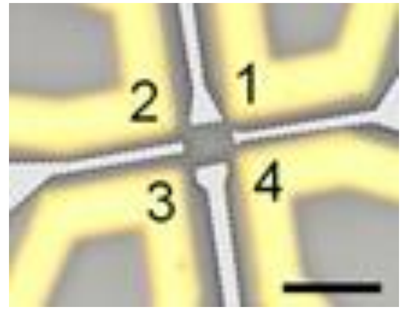
What limits ballistic transport in suspended bilayers?

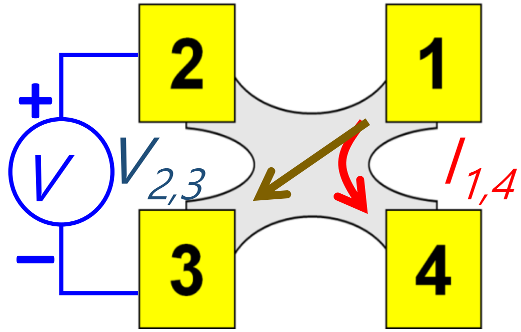
Suspended bilayer devices of very high quality

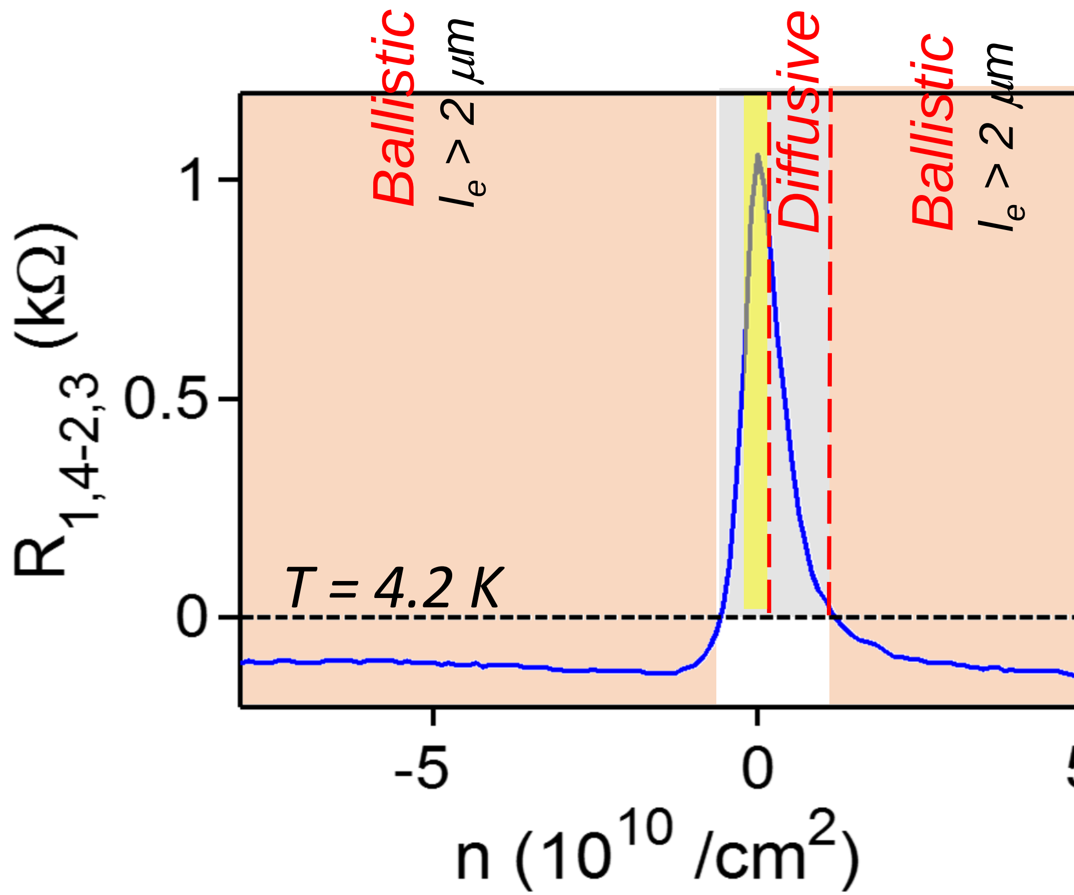
- *Charge fluctuations* *$< 10^9$ cm^{-2}*

- *Quantum Hall plateaus* *starting from 300 Gauss*

- *Observed* *even-denominator FQHE*

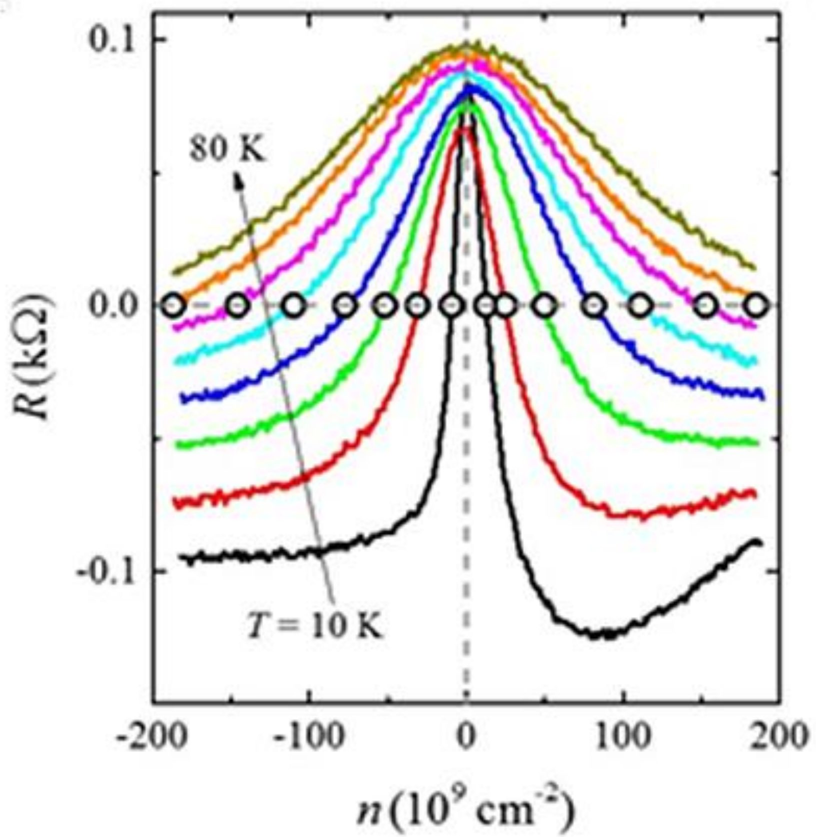


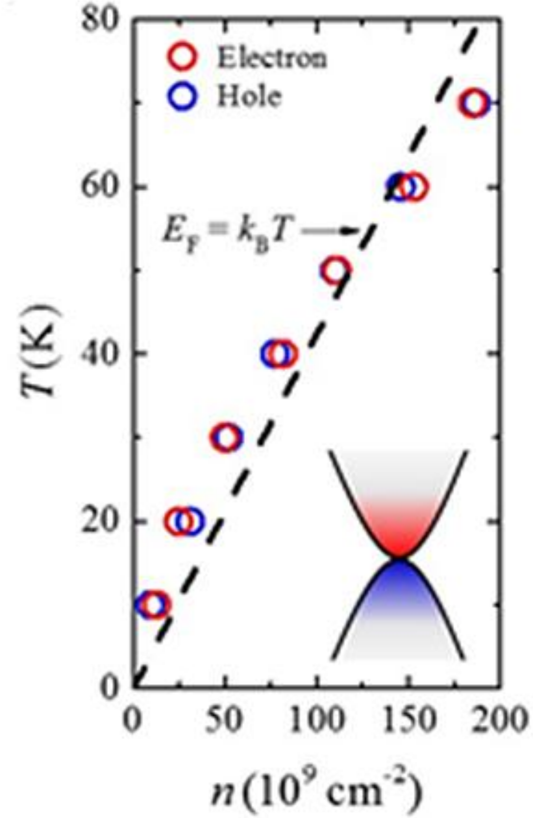




Strongly T & n dependent scattering mechanism

Onset of negative resistance (=ballistic transport)
depends strongly on temperature and carrier density



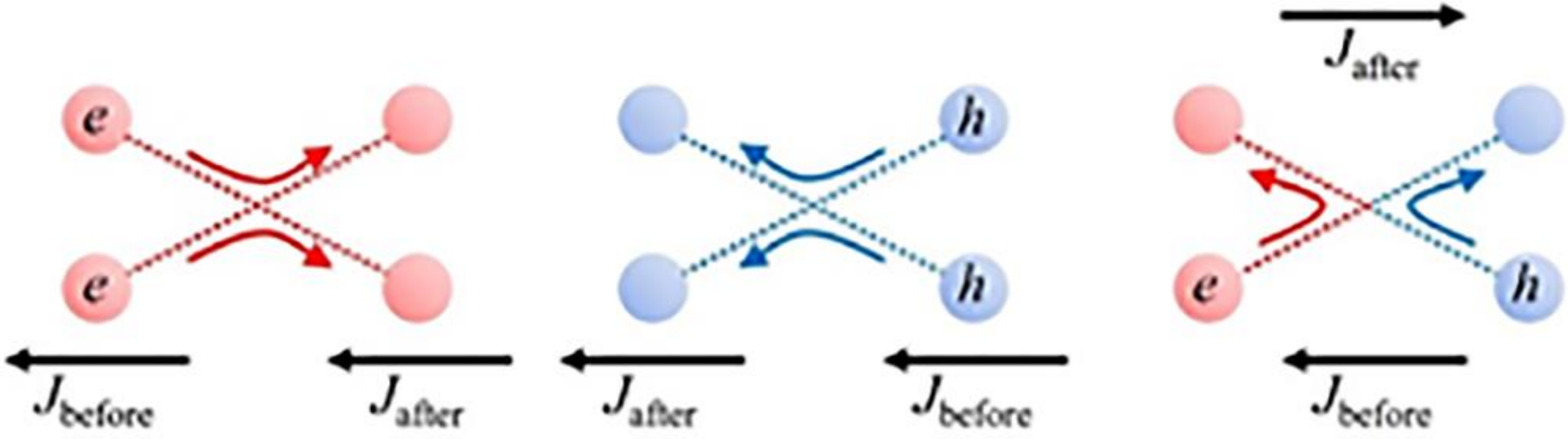


- *Onset of negative resistance transport at $E_F \sim kT$*
- *Ballistic transport for $E_F > kT$*

Hint for the role of e-h collisions

How does it work?

Coulomb scattering always conserves momentum (we do not consider umklapp processes)



But in the presence of multiple bands momentum conservation does not imply velocity conservation

Electron-hole scattering “dissipates” current

Can e-h collisions explain transport in the diffusive regime?

At charge neutrality (= $kT > E_F$) we have electron and holes

(1) $\sigma = n_e e\mu_e + n_h e\mu_h \qquad \mu_{e/h} = \dfrac{e\tau_{e/h}}{m}$

Assume: e-h collision determine velocity relaxation

(2) $\dfrac{1}{\tau_e} = \Gamma \dfrac{n_h}{n_e+n_h} \qquad \dfrac{1}{\tau_h} = \Gamma \dfrac{n_e}{n_e+n_h} \qquad \Gamma = C\dfrac{kT}{\hbar} \qquad C \sim 1$

We obtain:

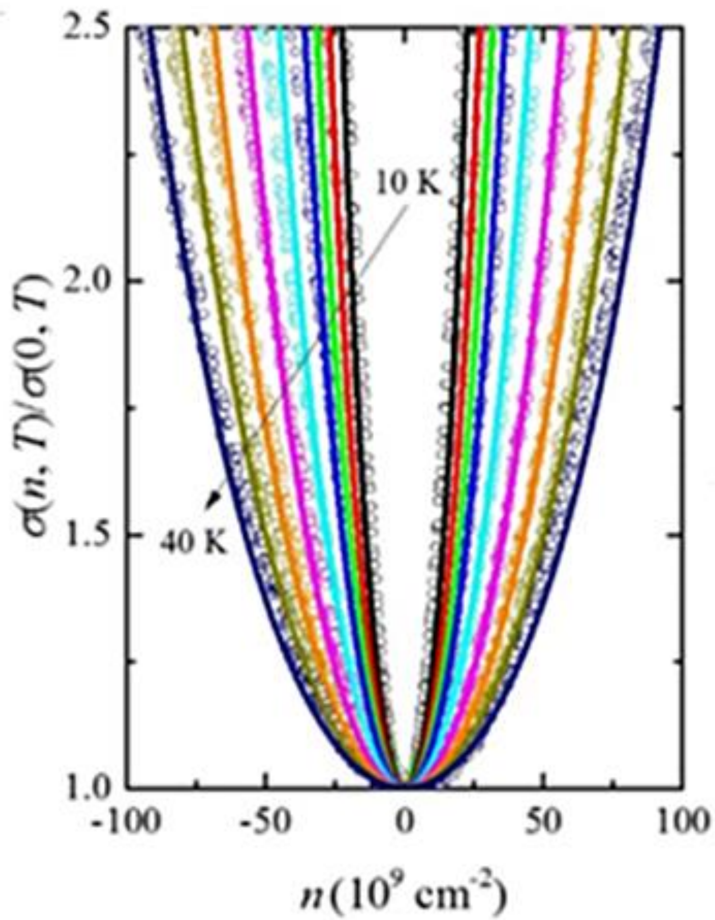
(1) + (2) $$\sigma(n,T) = C^{-1}\frac{\hbar}{kT}\frac{e^2}{m^*}(n_e+n_h)\frac{{n_e}^2+{n_h}^2}{n_e n_h}$$

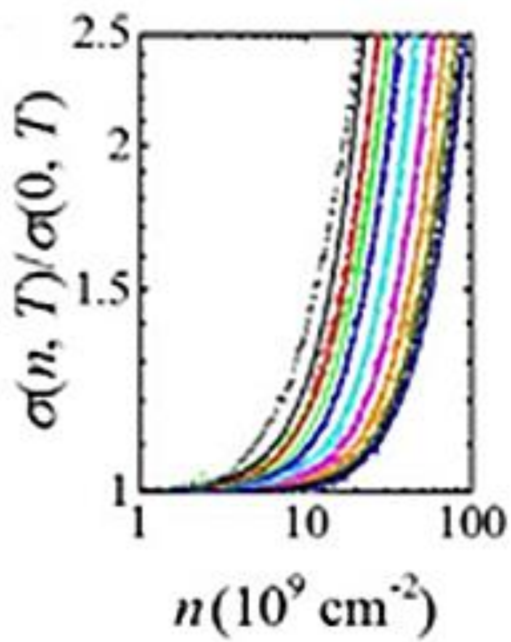
$$\frac{\sigma\,(n,T)}{\sigma\,(0,T)} = \frac{\pi\hbar^2}{8kTm^*\ln(2)}\frac{(n_e+n_h)({n_e}^2+{n_h}^2)}{n_e n_h}$$

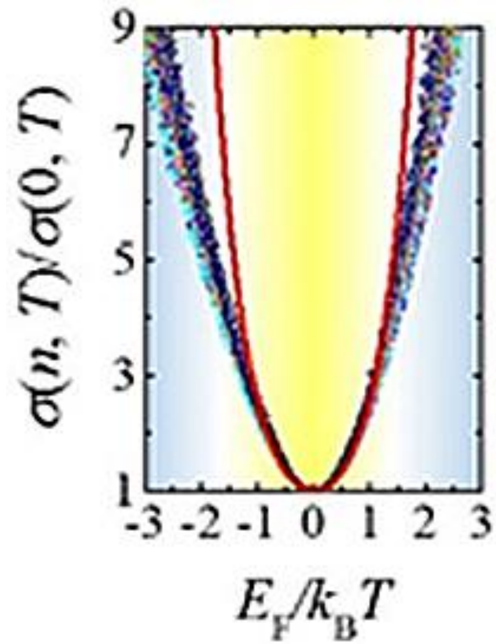
$m^* = 0.033\ m_o$

No free parameters: either it reproduces the data or it does not

Perfect agreement with no free parameters when $kT > E_F$







Excellent agreement:
over 2 orders of magnitude in n

Normalized conductivity:
Near CNP it only depends on E_F/kT

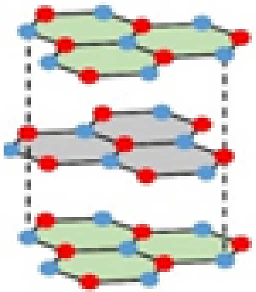
Observed in 4 out of 4 samples investigated in detail

- between 10 and 100 K,

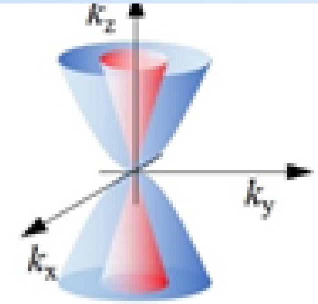
- with $m^* = 0.031\text{-}0.034\ m_o$

It also works on Bernal-stacked trilayer graphene

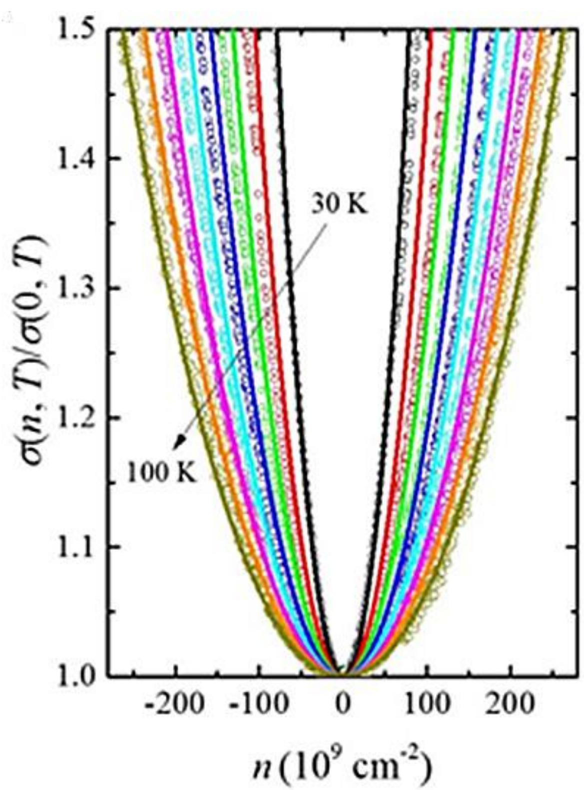
Bernal stacked trilayer

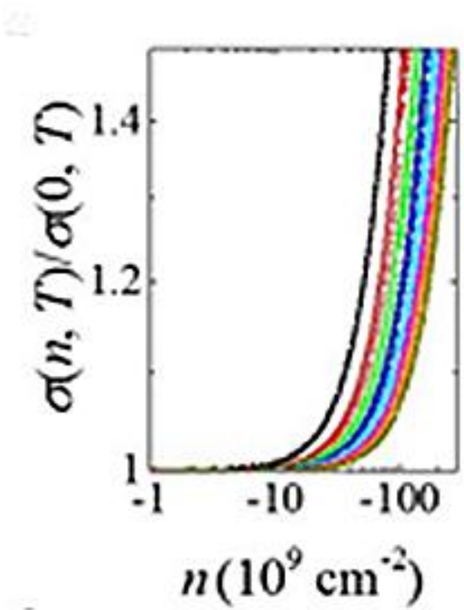


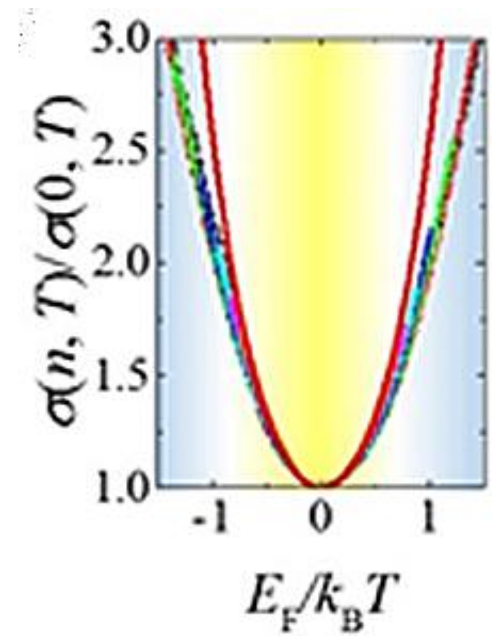
Bands:
1 x linear
1 x quadratic



- *For E ~ kT > E_F quadratic band DOS > 100 x linear band DOS*
- *Quadratic band dominates transport*







Perfect agreement with $m^* = 0.06\ m_o$

(expected $m^* = 0.05\ m_o$)

Also quantitative agreement with no free parameters

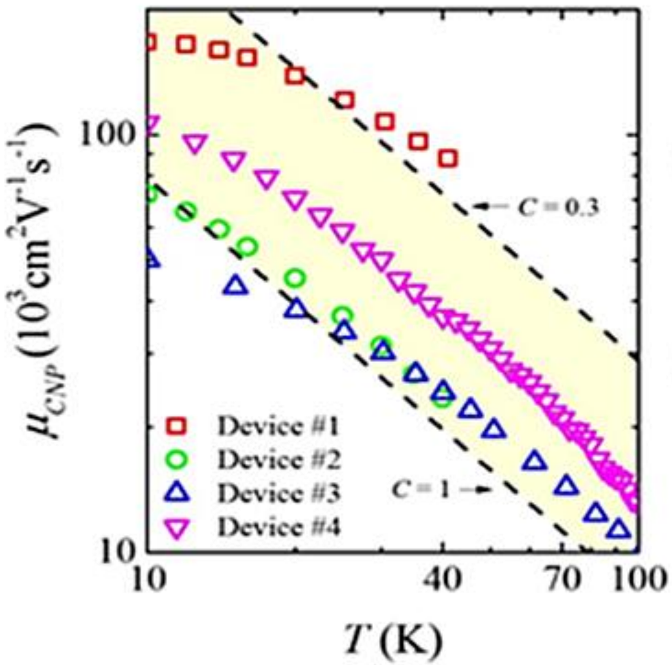
$$\sigma(n,T) = C^{-1}\frac{\hbar}{kT}\frac{e^2}{m^*}(n_e+n_h)\frac{{n_e}^2+{n_h}^2}{n_e n_h}$$

C ~ 1

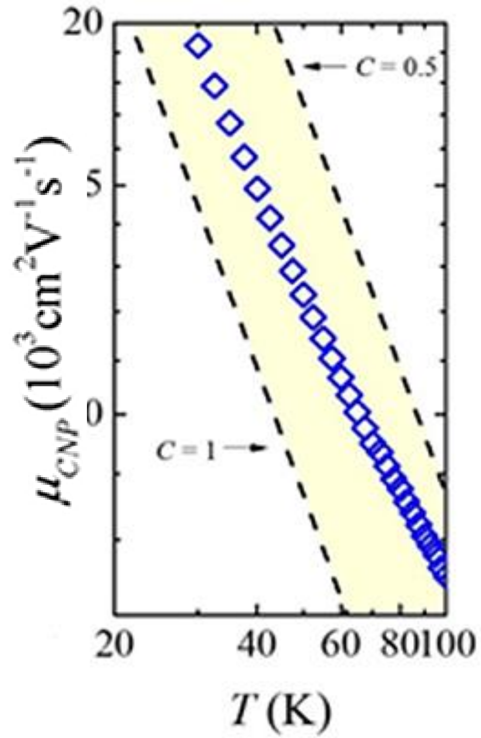
T-dependent mobility at CNP

$$\mu(T) = \frac{\sigma(n=0,T)}{n_e(n=0,T)+n_h(n=0,T)}$$

Bilayer

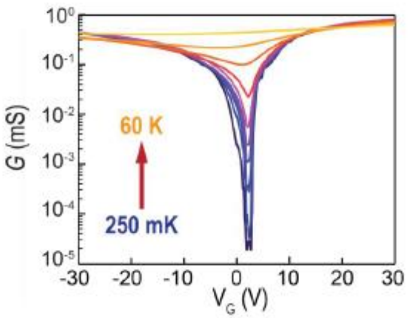


Trilayer

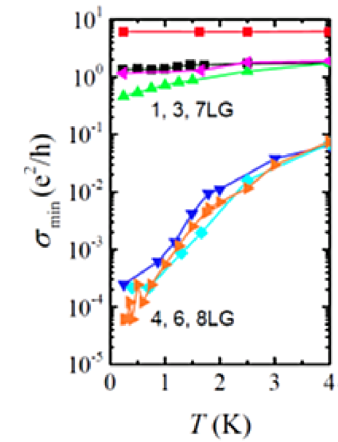


- *Quantiative agrement within a factor of 2-3 or better*
- *Expected due to indtermination on C and precise geometry*

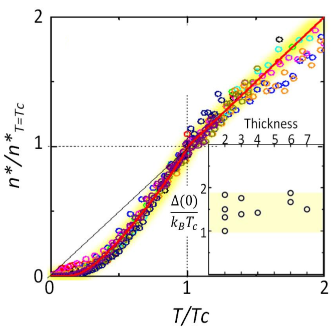
Conclusions



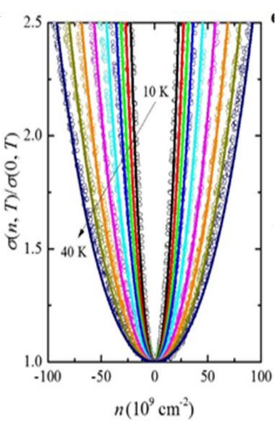
Unexpected insulating state @ CNP in all even Bernal multilayers



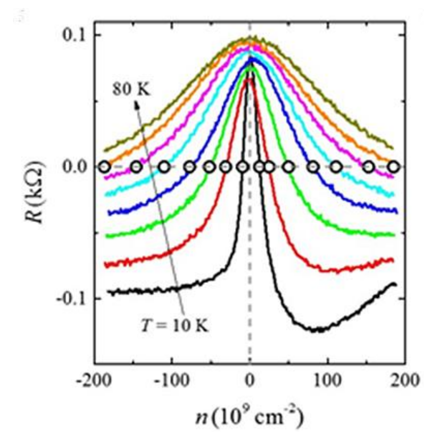
Even-odd effect: at low T all odd multilayers remain conducting with conductivity of 1 Dirac band



2^{nd} order phase transition gapping quadratic bands In all layers, with T_c increasing with thickness



e-h scattering limits ballistic motion at CNP and cause diffusive transport



Inelastic scattering rate @ CNP is $\Gamma = C\,\frac{kT}{\hbar}$