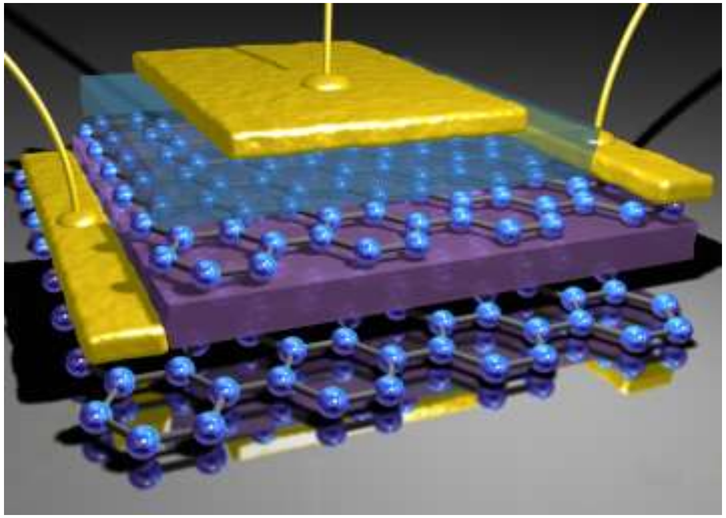
Graphene heterostructures:
electronic properties and potential applications



Leonid Ponomarenko

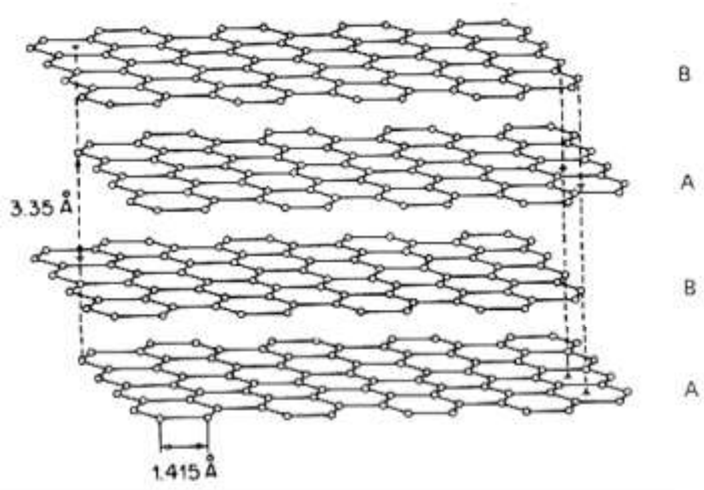
Lancaster University

THE ROYAL SOCIETY

School on Anomalous Transport, Superconductivity and Magnetism in Nanosystems

BITP, 15 - 20 June 2015, Kiev, Ukraine

Graphene



Graphite:

strong in-plane bonding

weak inter-plane interaction

Graphene was isolated and measured for the first time in 2004

Geim

Novoselov

The Nobel Prize in Physics 2010 was awarded jointly to Andre Geim and Konstantin Novoselov "*for groundbreaking experiments regarding the two-dimensional material graphene*"

Graphene Superlatives

thinnest imaginable material

strongest material ever measured

stiffest known material (stiffer than diamond)

most stretchable crystal (up to 20% elastically)

record thermal conductivity (outperforming diamond)

highest current density at room T (million times of those in copper)

highest intrinsic mobility (100 times more than in Si)

lightest charge carriers (zero rest mass)

longest mean free path at room T (micron range)

most impermeable (even He atoms cannot squeeze through)

...

National Graphene Institute

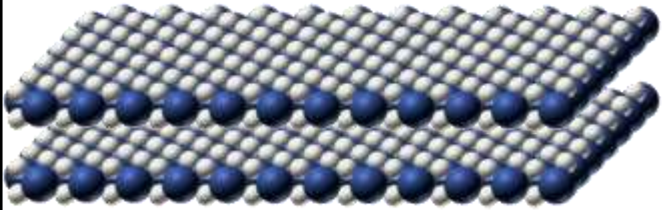
- £61M building funded by UK Government and European Union
- Officially opened in March 2015

Prof V. Fal'ko,

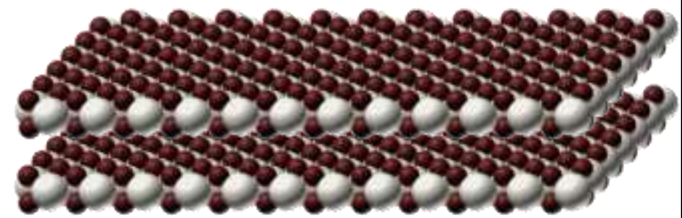
director of NGI

(from 8/06/15)

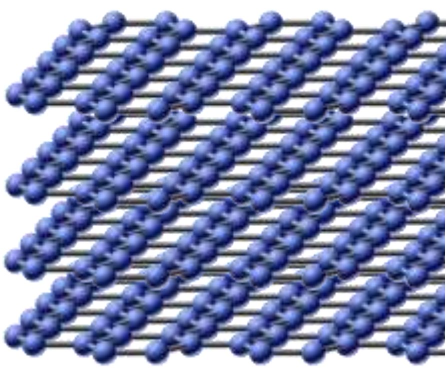
Graphene is not alone



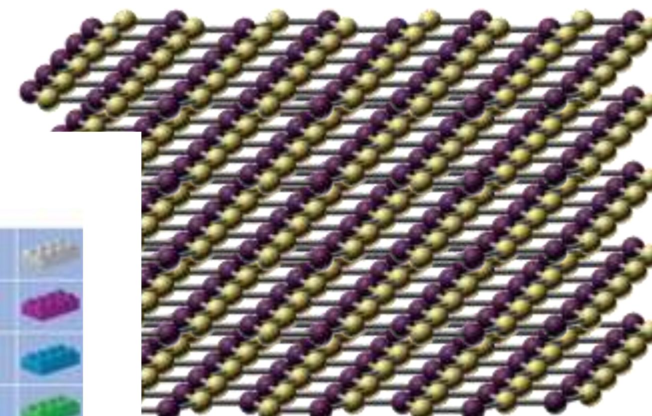
MoS_2



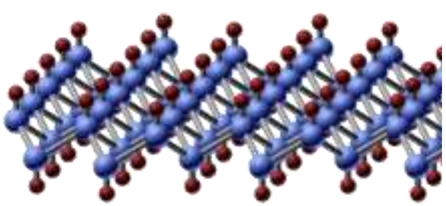
$NbSe_2$



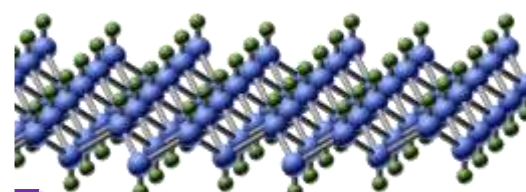
Grap



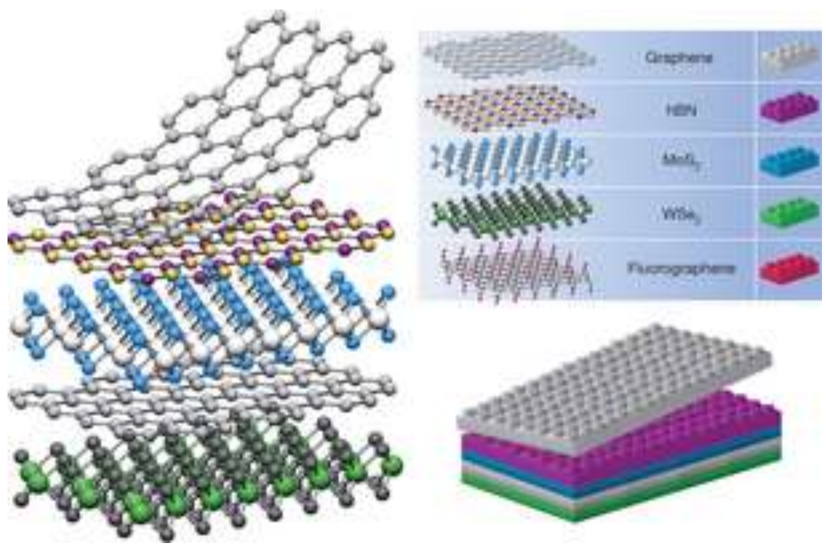
on-Nitride



Gr



Fane



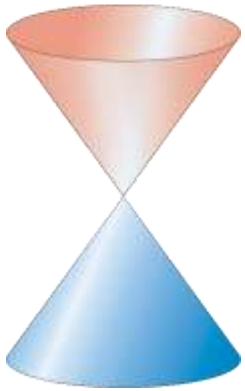
Van der Waals heterostructures. Geim & Grigorieva, NATURE (2013)

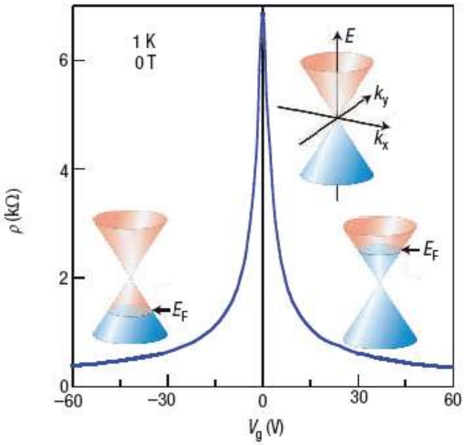
Outline

Properties of graphene monolayer

band structure

field effect



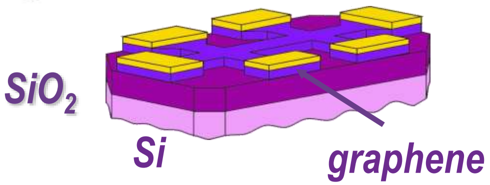


Graphene – BN heterostructures

fabrication

superlattices

Hofstadter butterfly



Graphene multilayer structures

metal-insulator transition

Coulomb drag

tunnelling transistors

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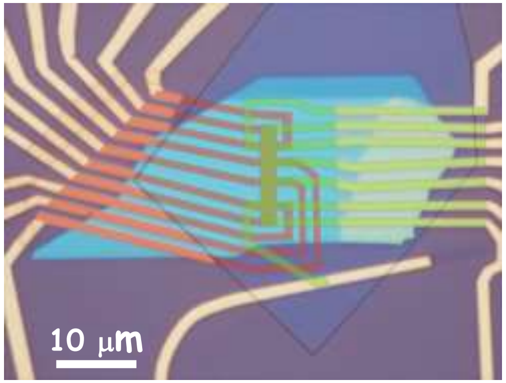

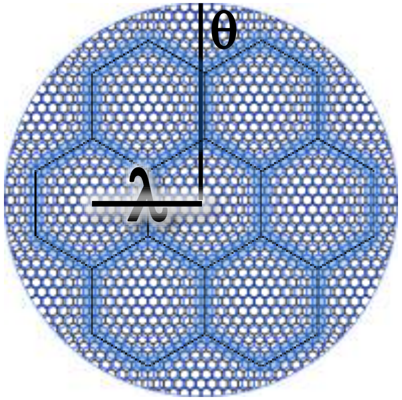

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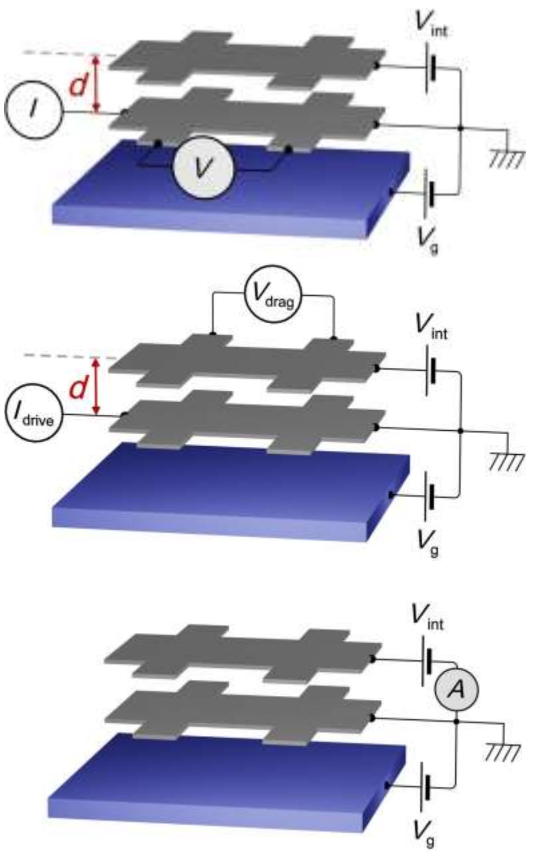
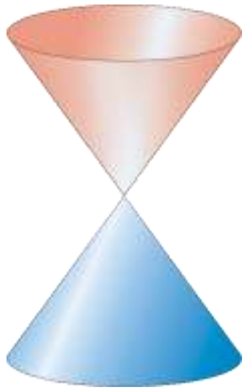
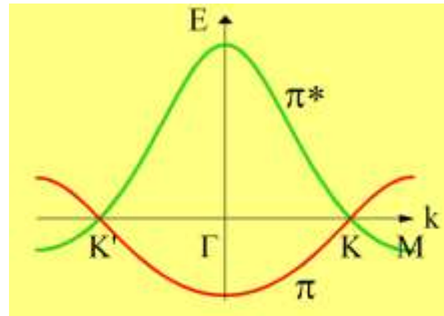
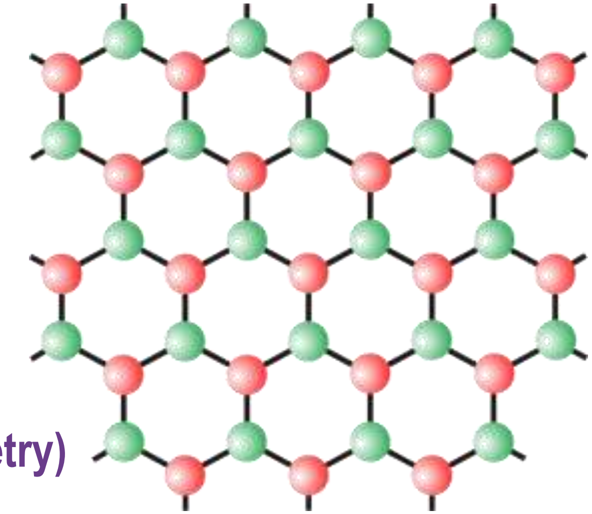


Band Structure and Field Effect

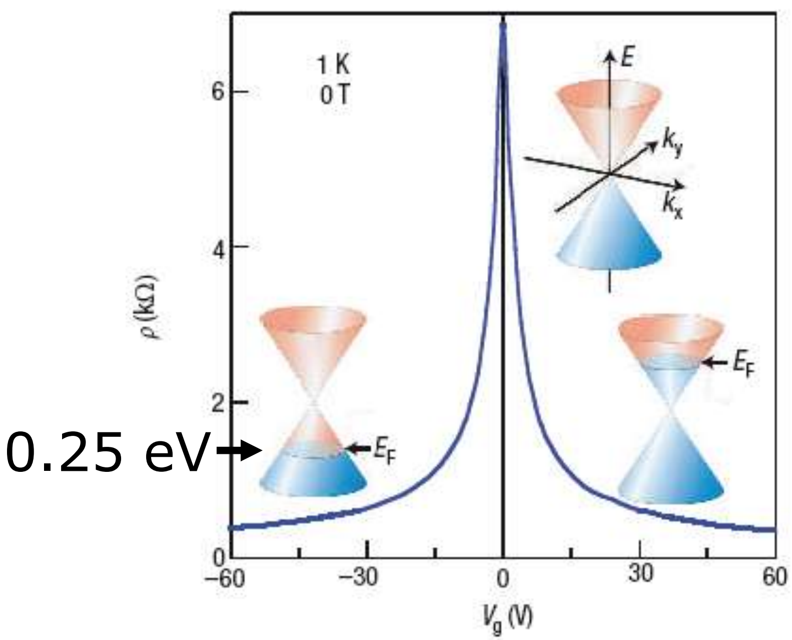
tight binding calculations, Phil Wallace, Phys. Rev. 71, 622 (1947)

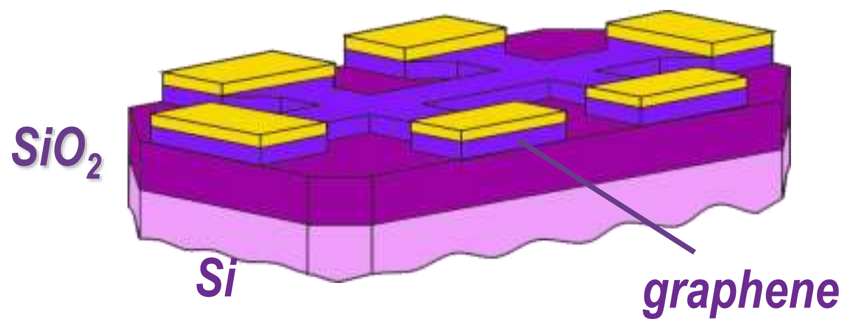






Hexagonal lattice, A-B sublattice symmetry (= inversion symmetry)

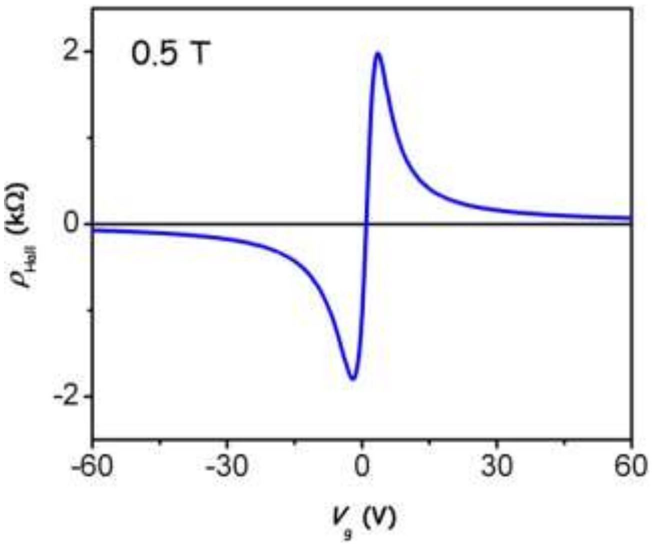




Geim & Novoselov, Science 2004

Mechanical exfoliation ("Sticky tape" method)

Band Structure and Field Effect



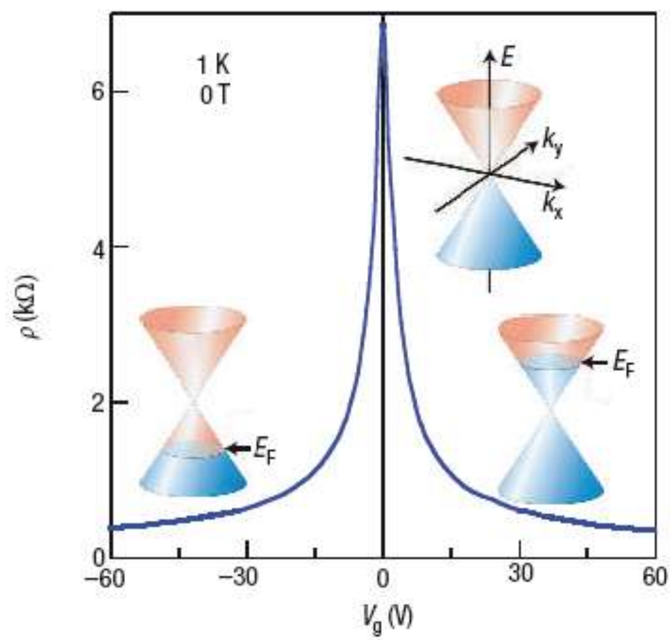
$$\rho_{Hall} = \frac{B}{en}$$

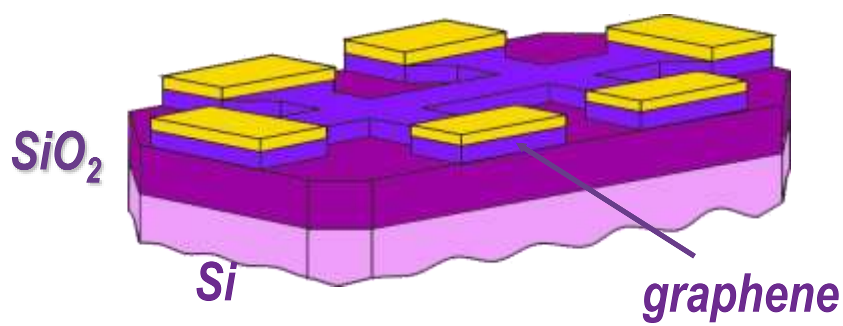
B – magnetic field
n – carrier density

$$\sigma = \frac{1}{\rho} = \mu en$$

μ - mobility

10 000 cm²/Vs (on SiO2)
1 000 000 cm²/Vs (suspended, 4 K)





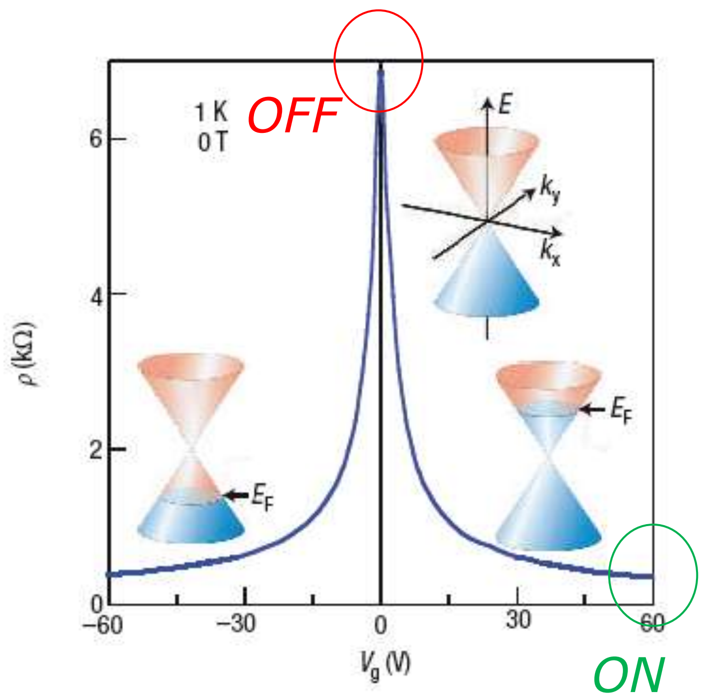
Geim & Novoselov, Science 2004

Mechanical exfoliation ("Sticky tape" method)

End of Introduction Part

2 things to remember:

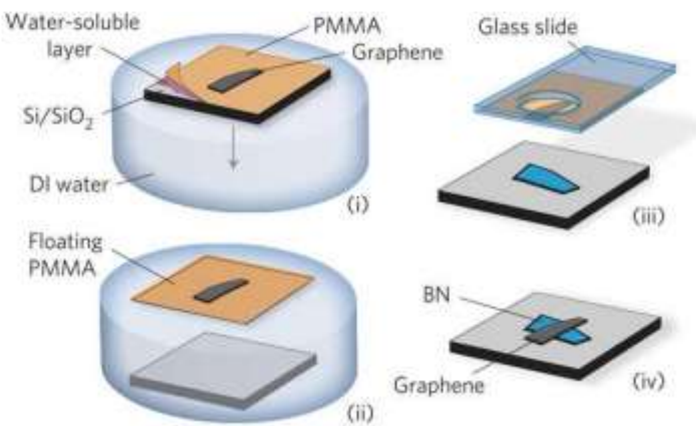
Graphene is always conducting;
FET doesn't work.

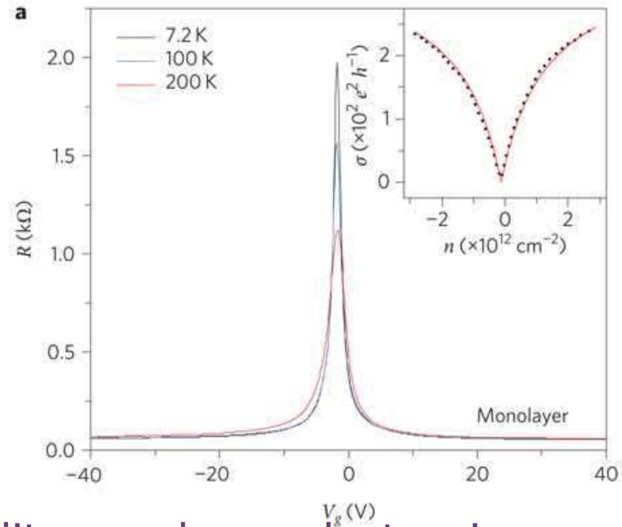


In graphene ON/OFF ratio
is ~ 20 at room T
(needed ~ 10^3 for FET)

there is no proper OFF state
because of zero band gap

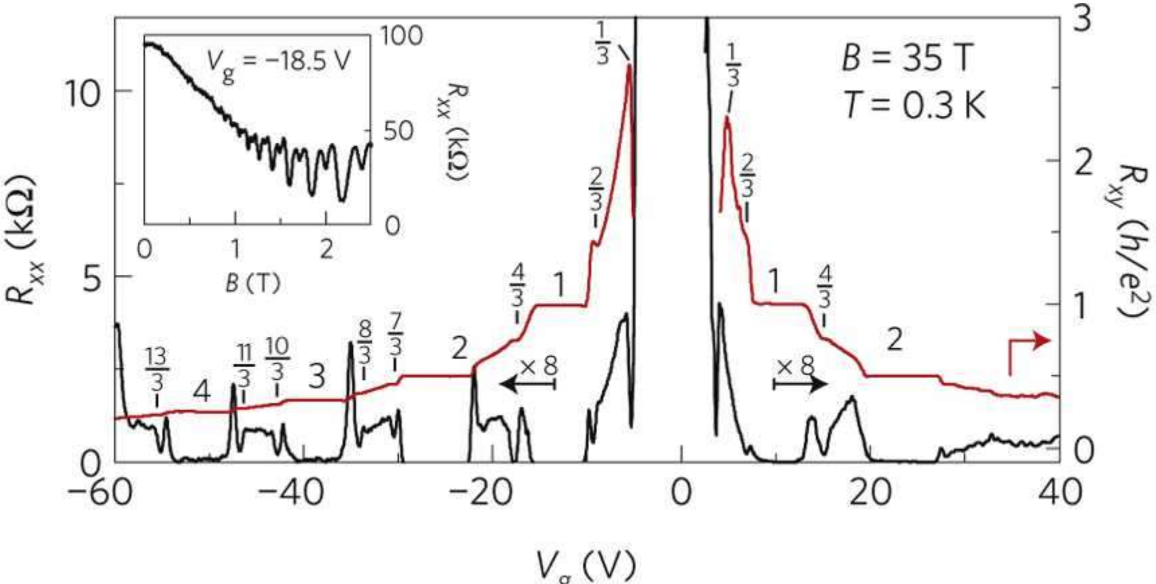
Graphene Heterostructures





Boron nitride substrates for high-quality graphene electronics
C. R. Dean et al. *Nature Nano* 5, 722 (2010)

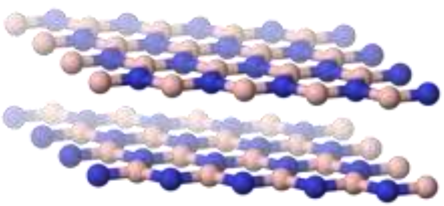
One order improvement in mobility.



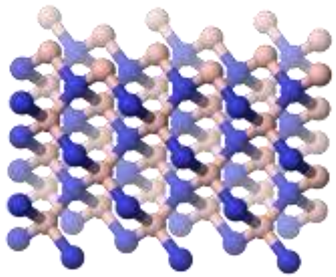
Fractional Quantum Hall Effect

Dean et al., *Nature Phys*. *(2011)*

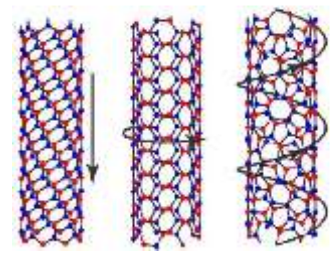
Different forms of Boron Nitride



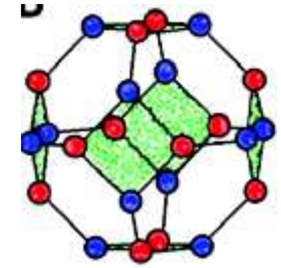
Hexagonal – "white graphite"



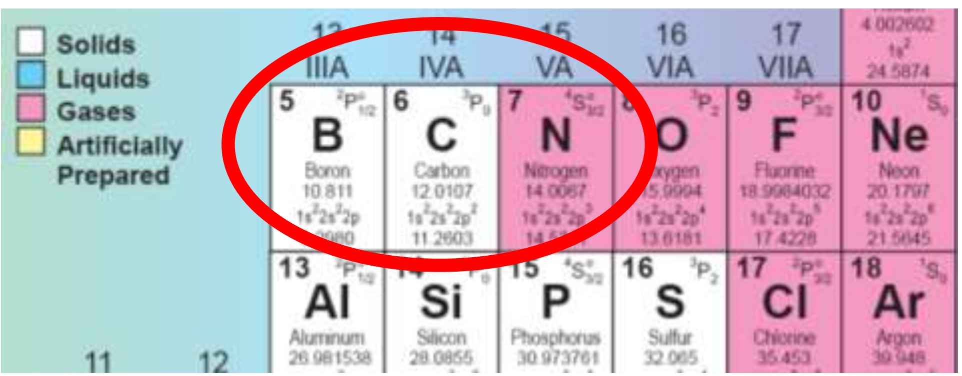
Cubic – almost as hard as diamond



BN nanotubes



Fullerene-like molecule $B_{12}N_{12}$

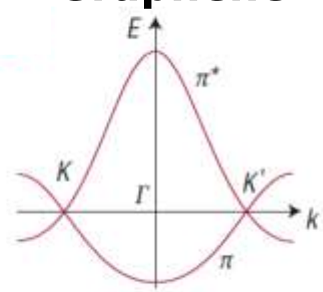


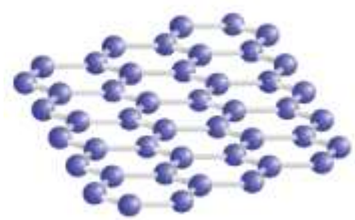
BN: insulator with gap 5.8 eV

K. Watanabe et al, Nature Materials 2004.

Inversion symmetry is broken!

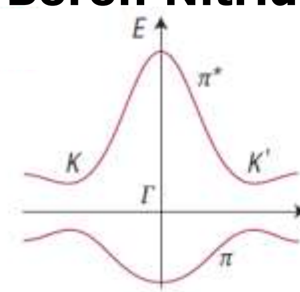
Graphene

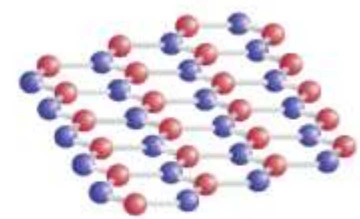




a = 2.462 A, c = 6.708 A

Boron Nitride





a = 2.504 A, c = 6.661 A

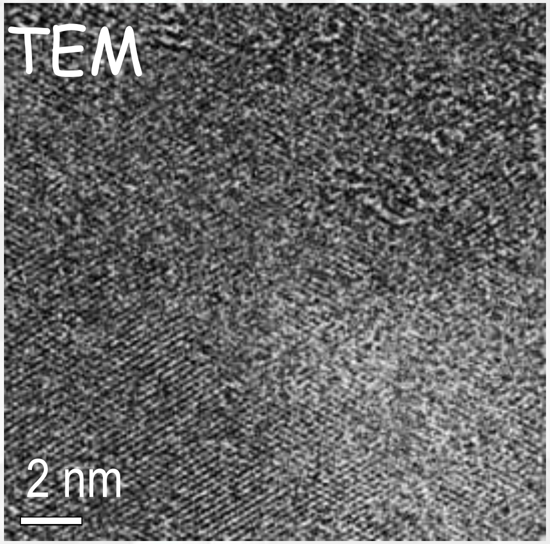
Dry-peel transfer

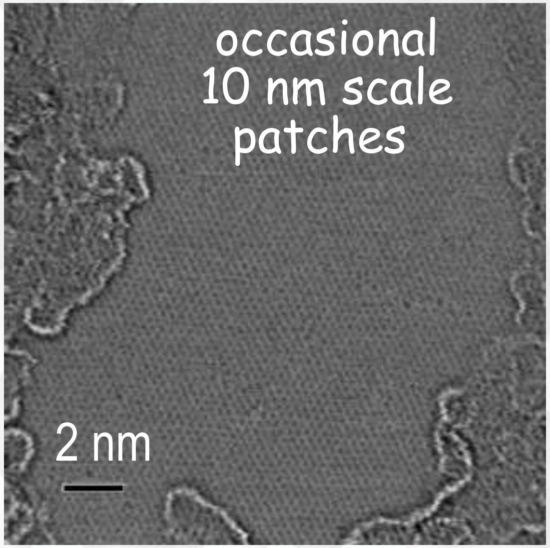
de-lamination

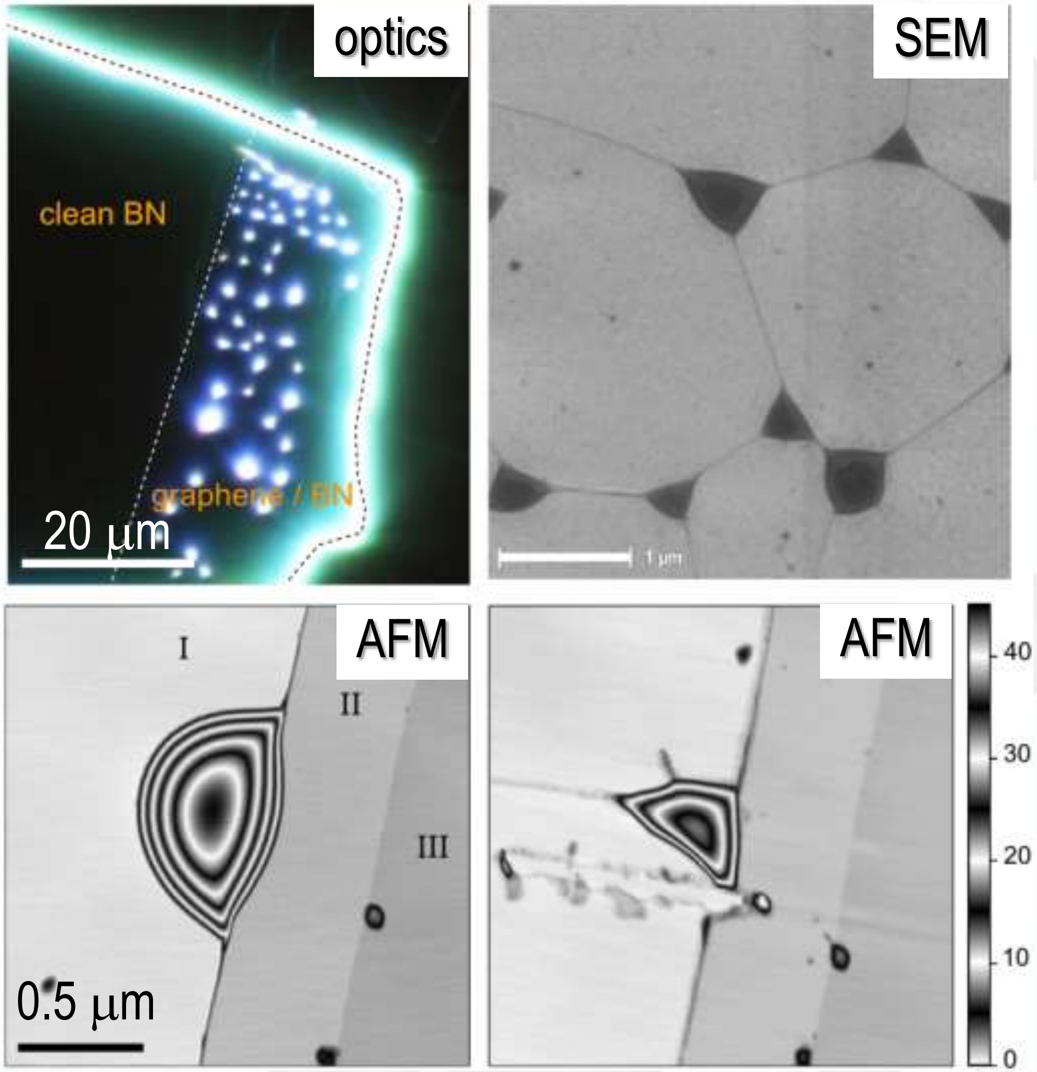
release

Bubbles

everything covered with contamination

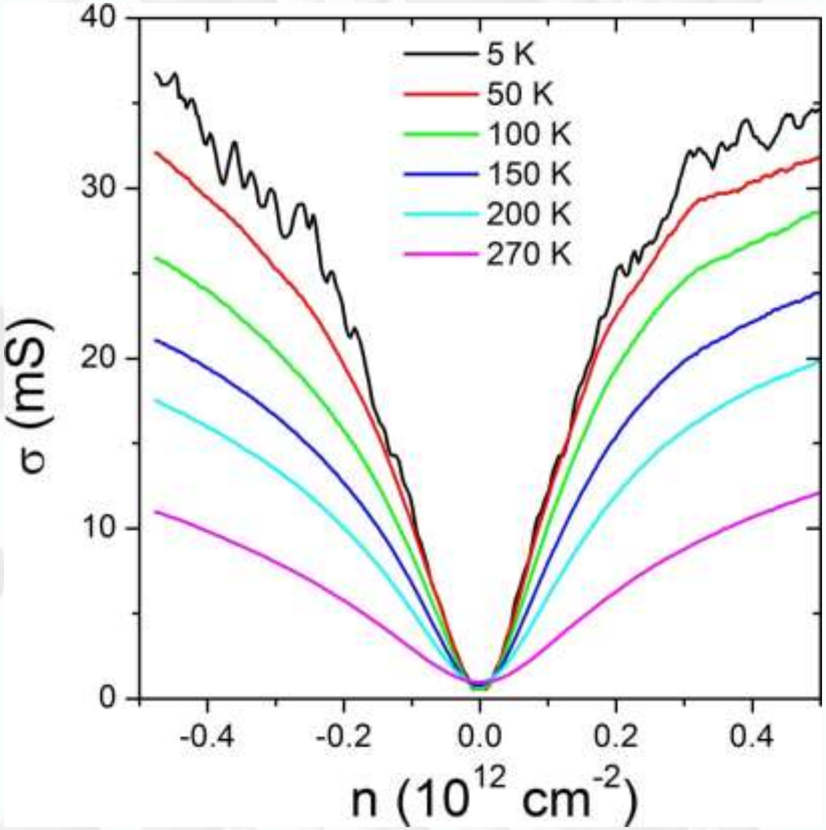


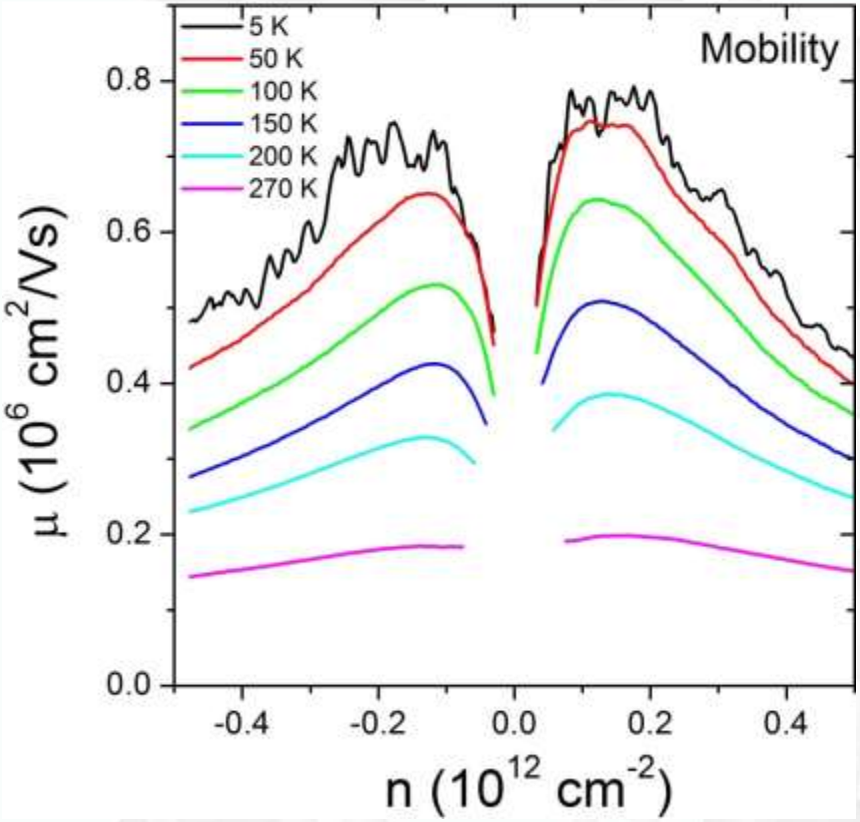




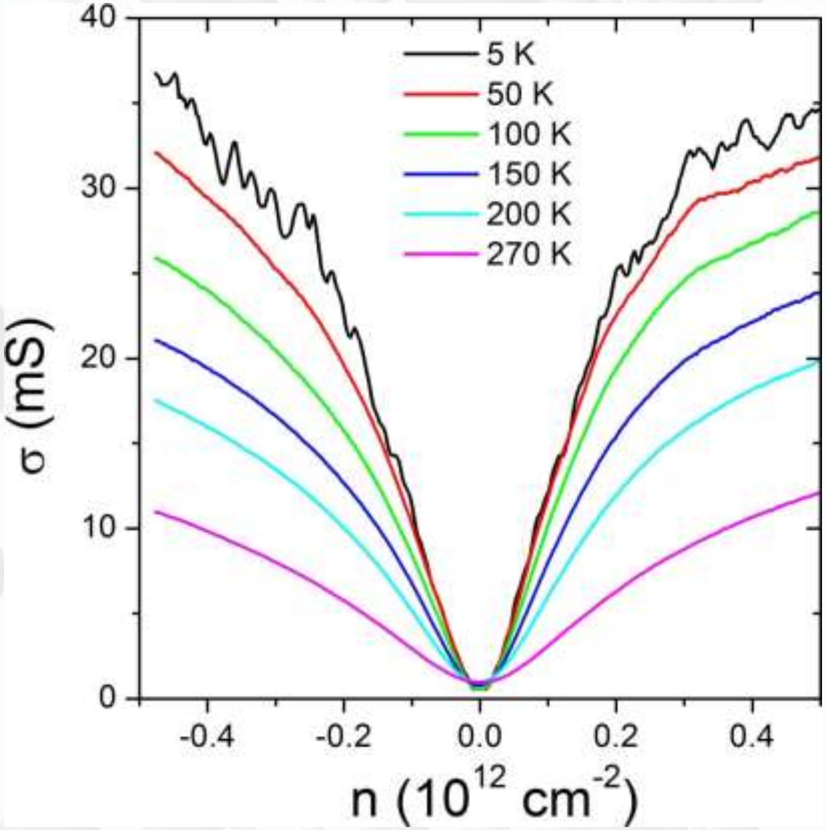
so much contamination → BUBBLES

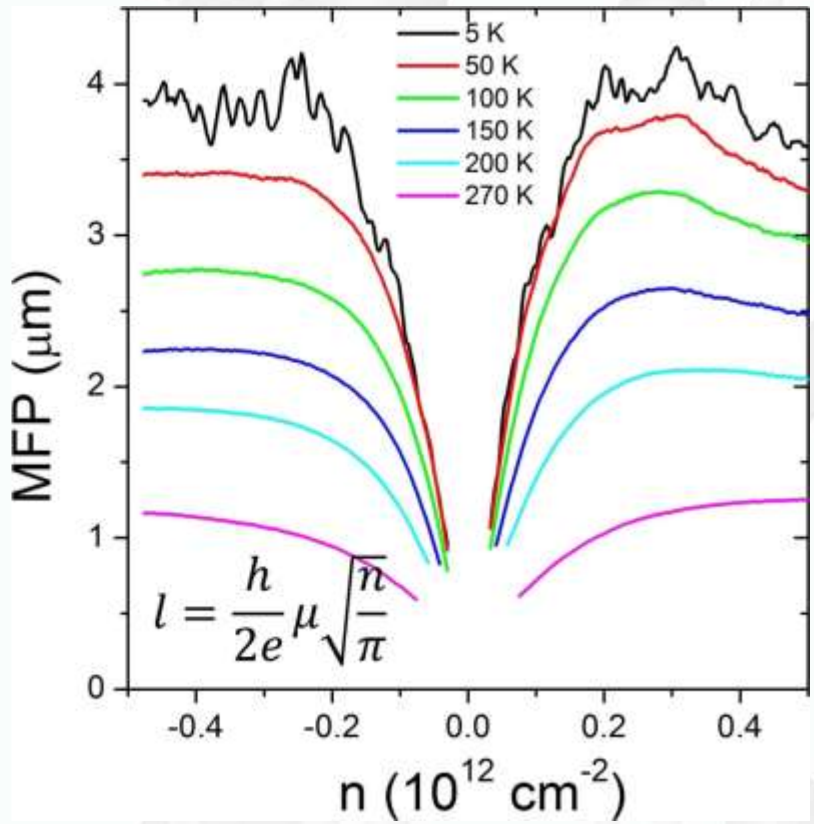
Mobility and Mean Free Path





Mobility and Mean Free Path

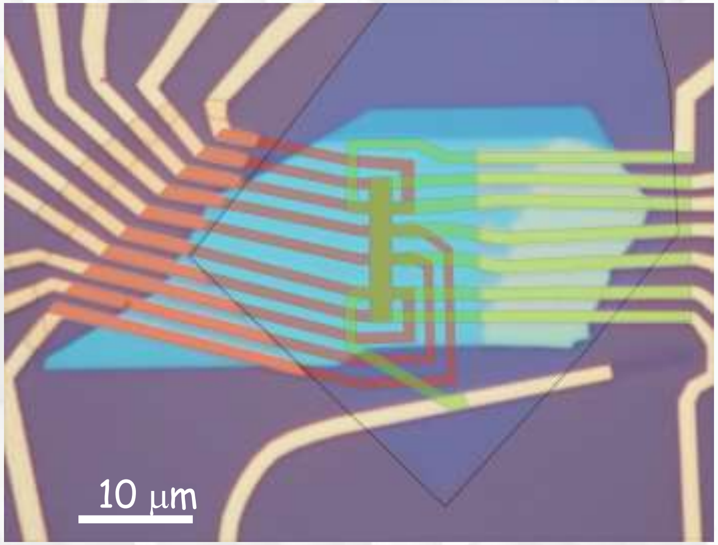


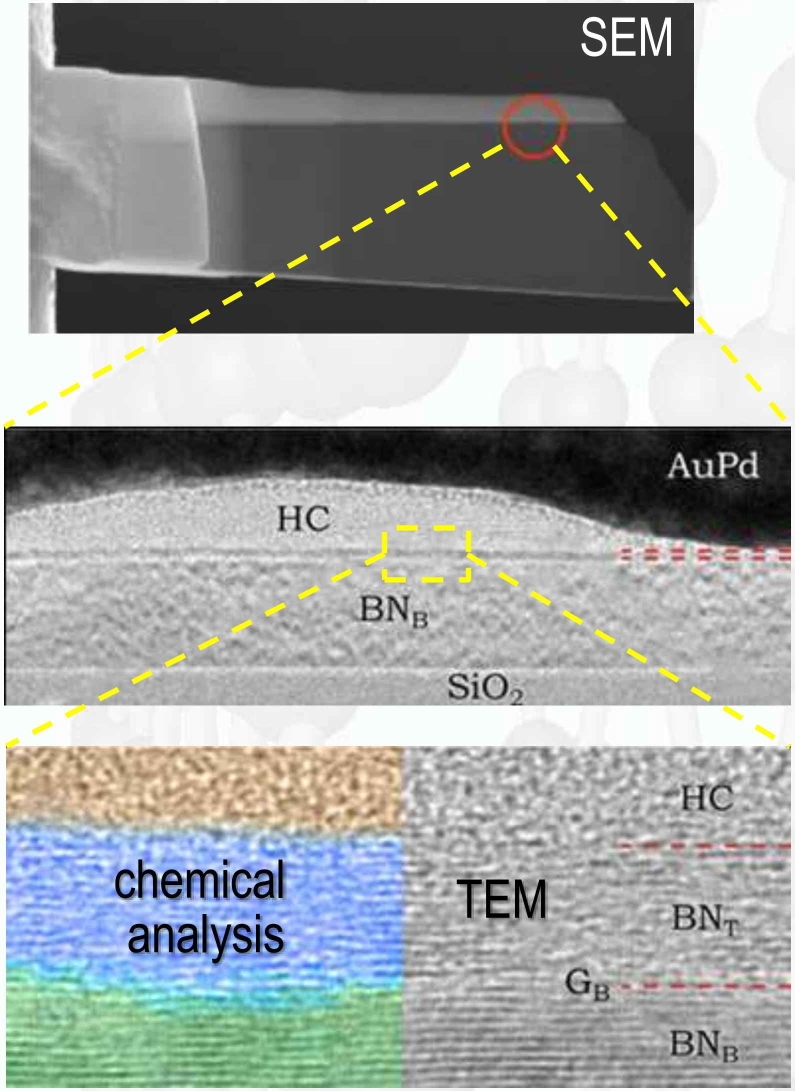


The width of the Hall bar is 2 μm

At low temperatures the MFP is limited by the size of the device

graphene-hBN interface in TEM



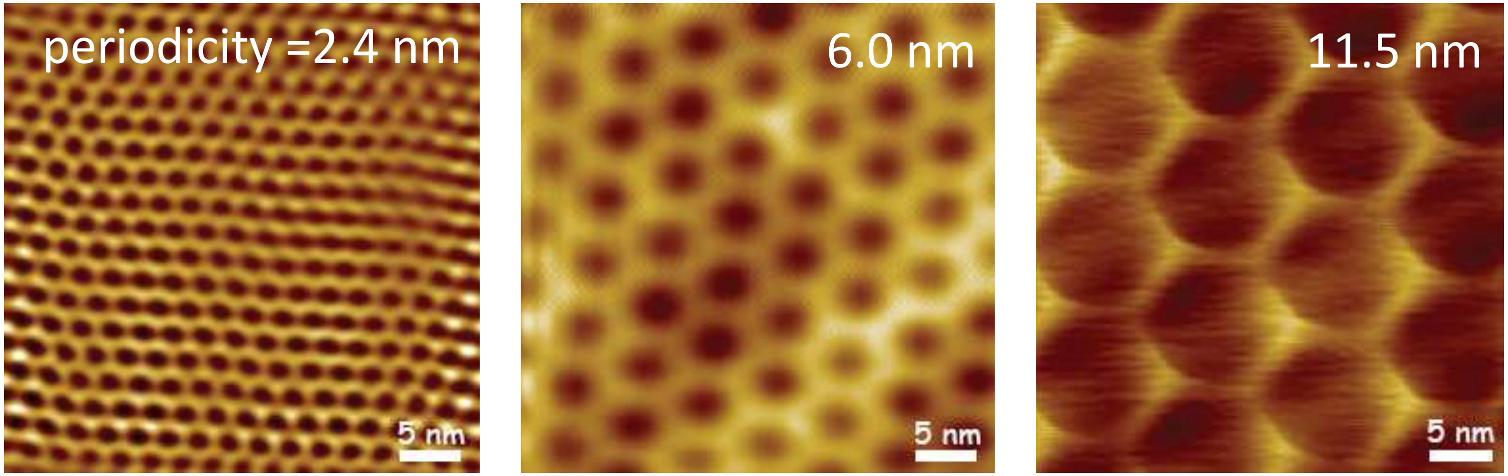


NO CONTAMINATION LAYER
at the interface
between graphene & hBN

Focused ion beam (FIB) milling + STEM
Slices 20-70 nm thick

Manchester, *Nature Materials* 2012

moiré patterns: graphene on hBN

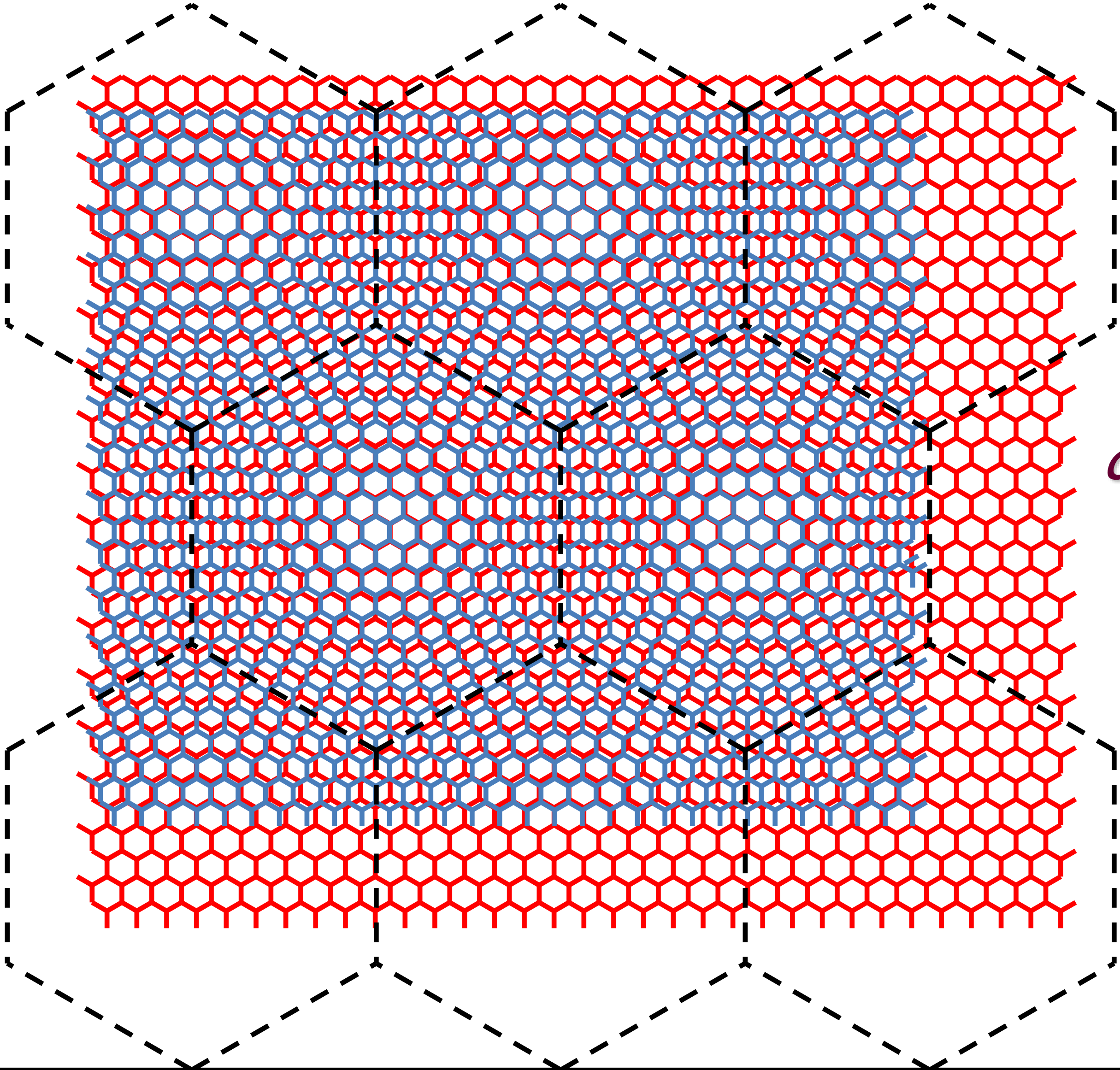


Yankowitz et al, *Nature Phys* 2012
Xue et al, *Nature Mat* 2011; Decker et al, *Nanolett* 2011

Scanning tunnelling microscopy (STM):

- the period is much larger than graphene lattice constant
- conductance electrons “feel” periodic potential

Graphene on Substrate with Similar Lattice Constant

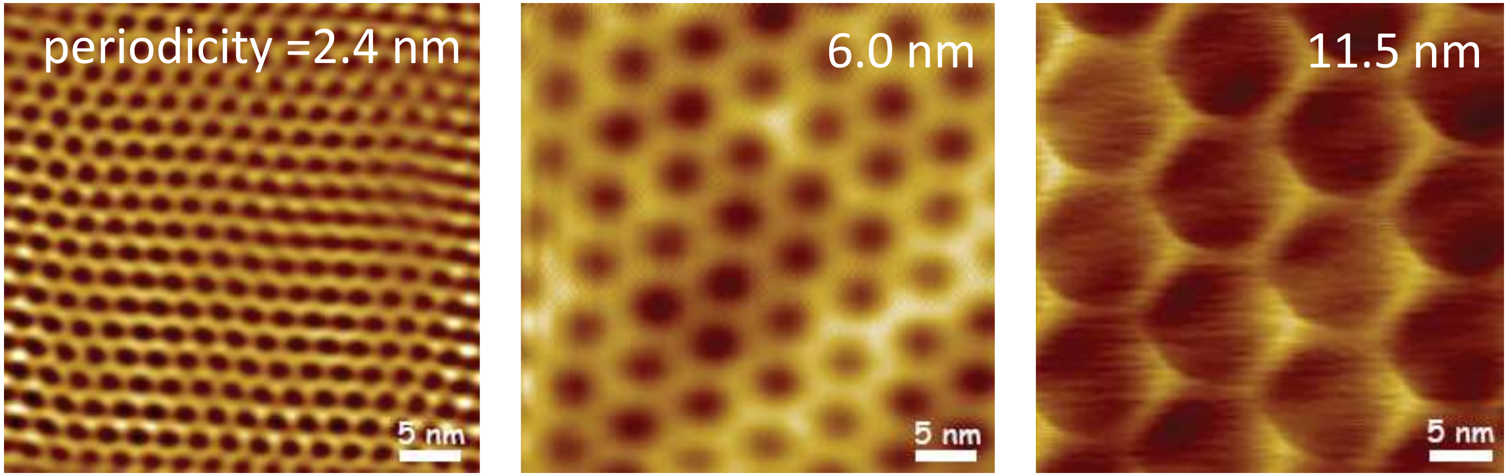


Moiré pattern: well defined long range order

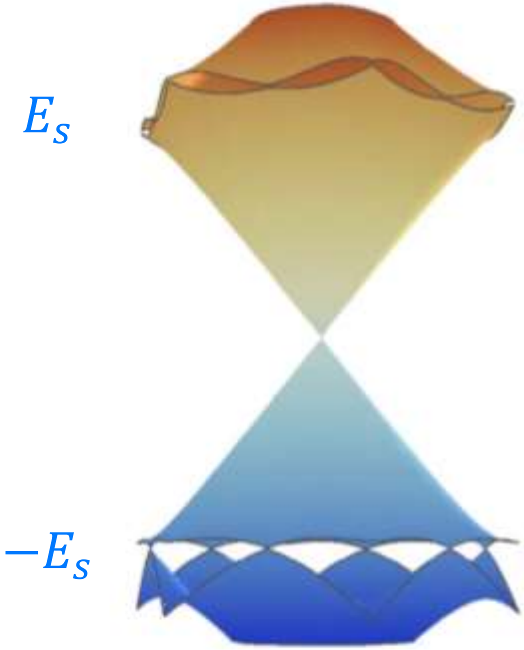
Graphene is just one atom thick. Electrons feel atoms of the substrate (if the interface is clean)

What happens in strong magnetic field?

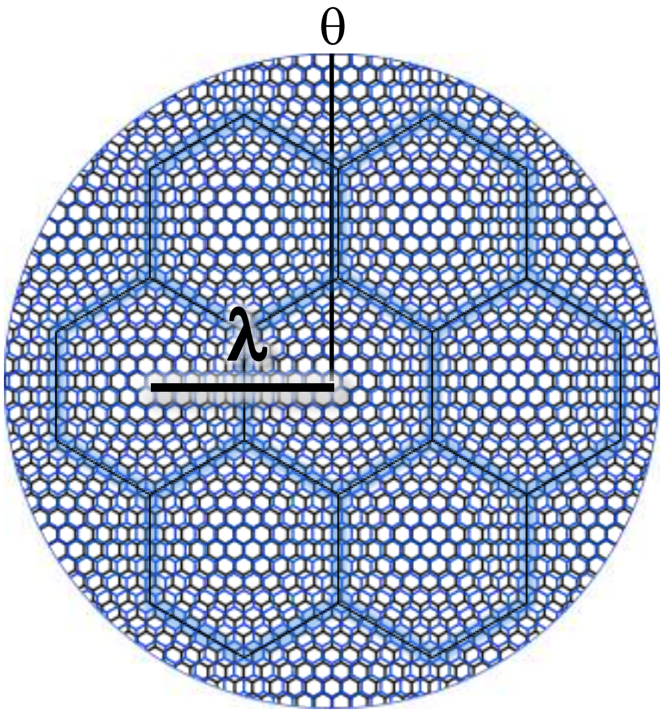
moiré patterns: graphene on hBN



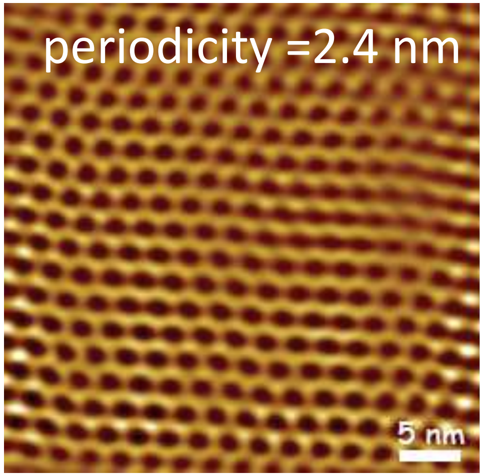
Yankowitz et al, *Nature Phys* 2012
Xue et al, *Nature Mat* 2011; Decker et al, *Nanolett* 2011

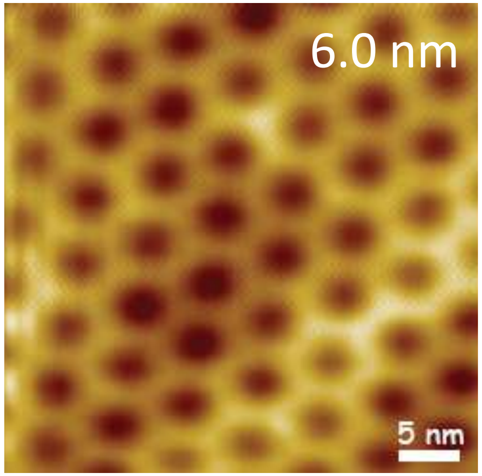


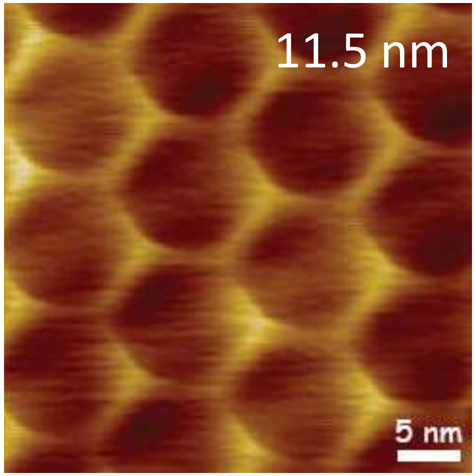
THEORY:
Steve Louie's group
Nature Phys 2008, *PRL* 2008
Francois Peeters' group
PRB 2010-2012
Burset et al, *PRB* 2011
Ortix et al, *PRB* 2012
Kindermann et al, *PRB* 2012
Fal'ko et al, *PRB* 2013
and SOME MORE



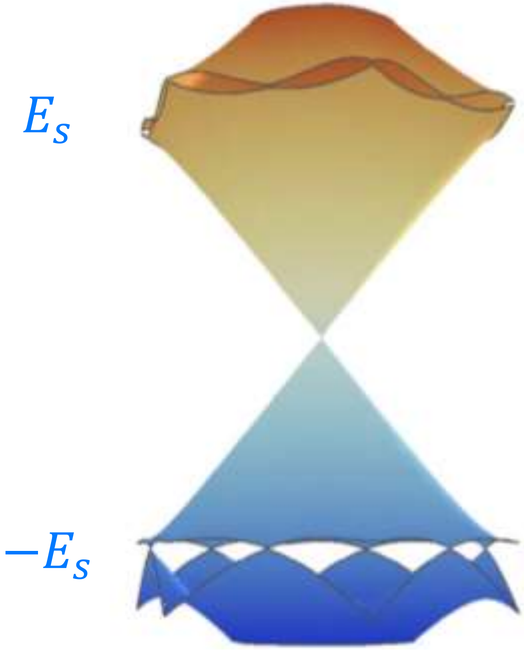
moiré patterns: graphene on hBN



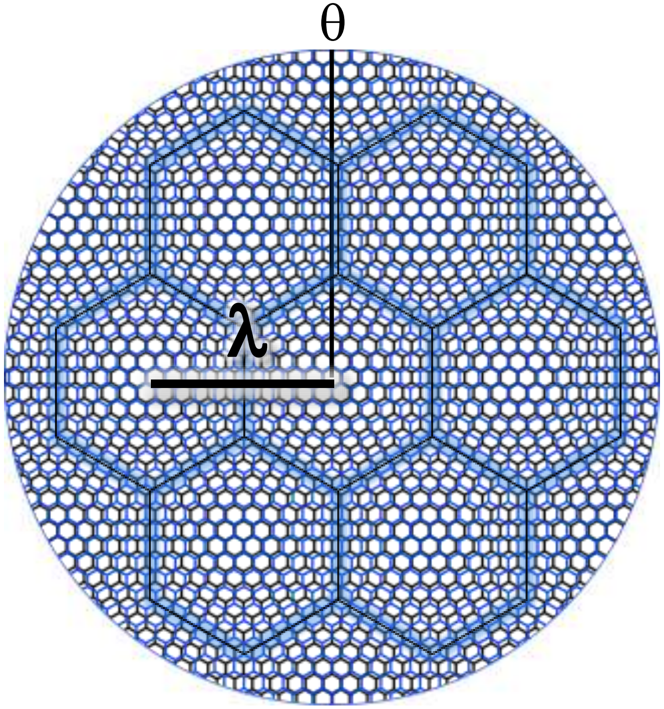




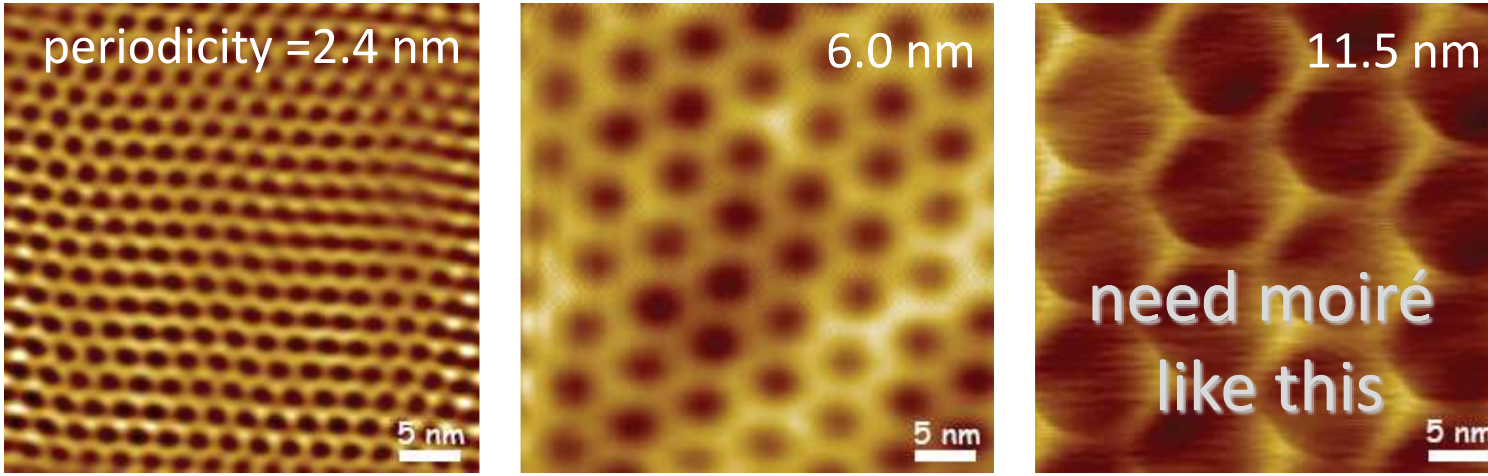
Yankowitz et al, *Nature Phys* 2012



New Dirac points generated at the edges of the superlattice Brillouin zone



can we probe in transport?



Yankowitz et al, *Nature Phys* 2012

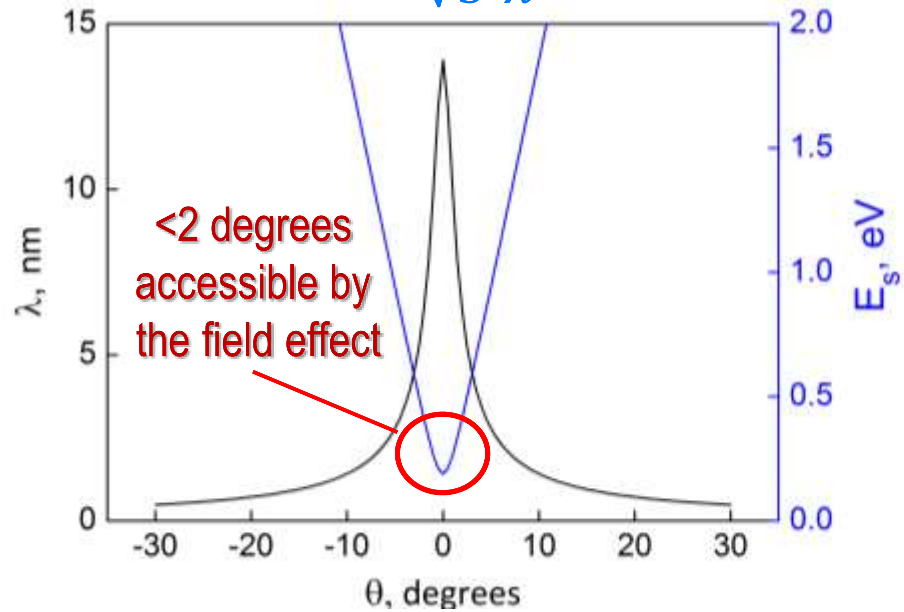
E_s

$-E_s$

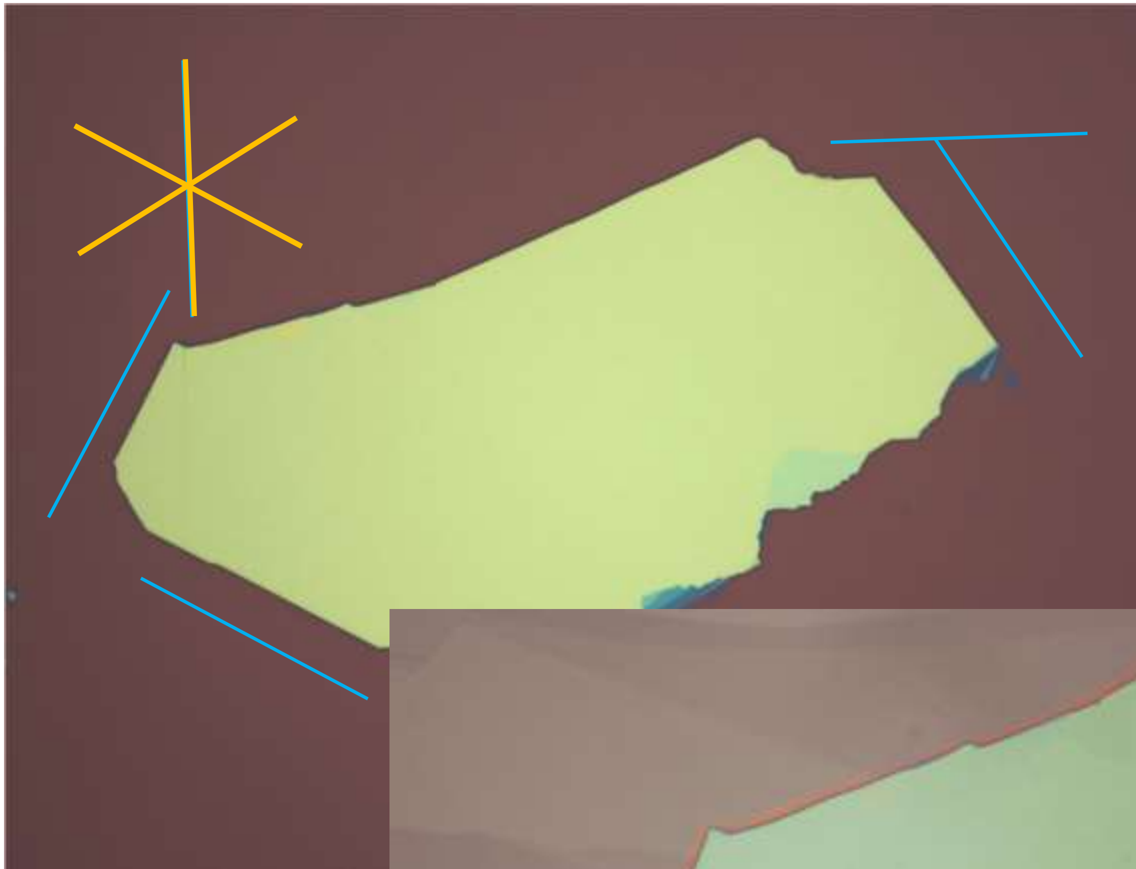
for short periods,
gaps lie at
too high energies

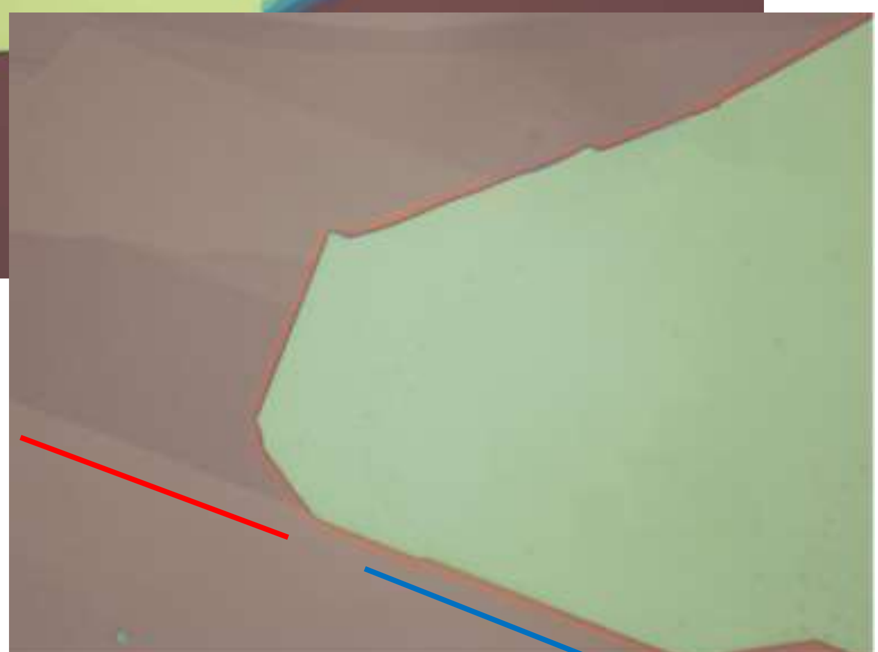
“invisible”
for transport
measurements

$$E_s = \frac{2\pi\hbar V_F}{\sqrt{3}\ \lambda}$$

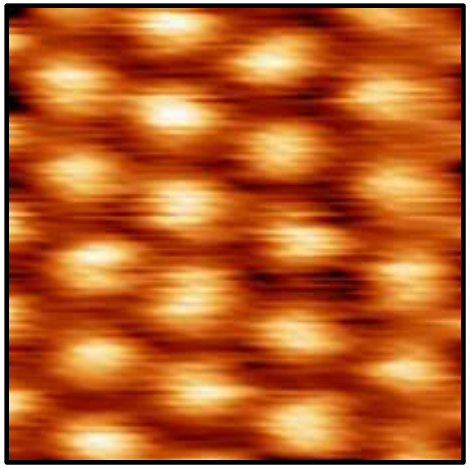


specially aligned graphene devices

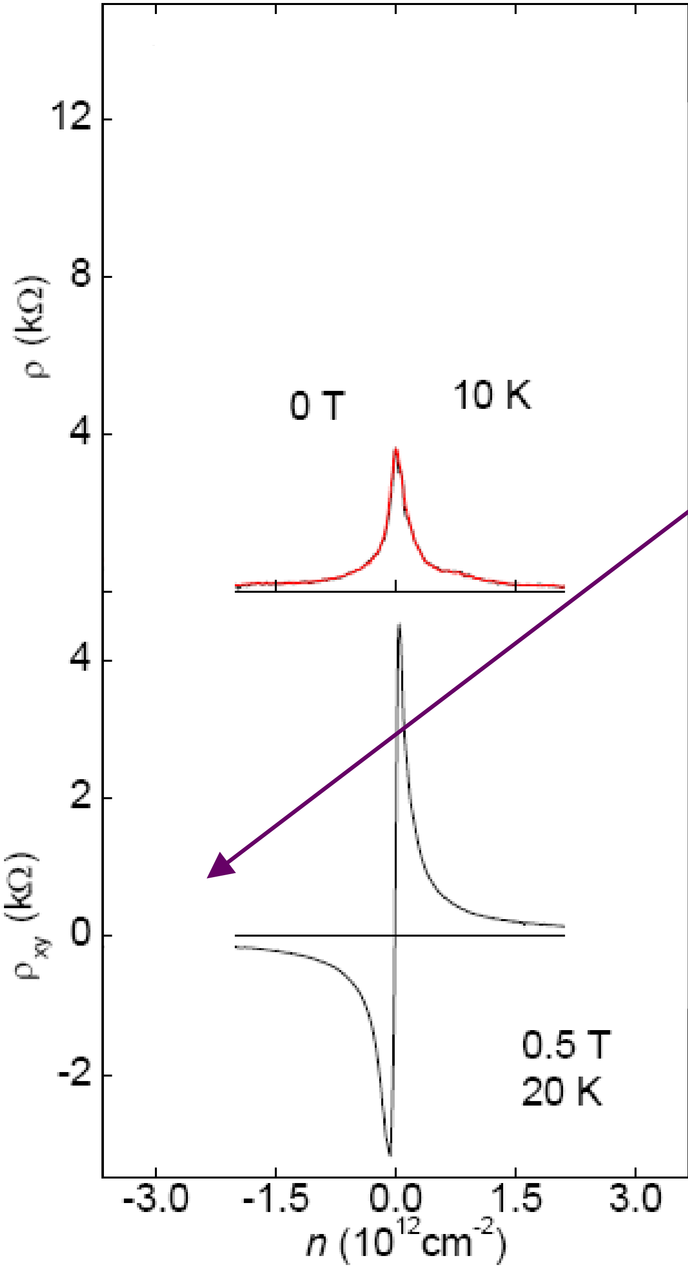




ambient CAFM:
moiré in graphene on BN
with ~12 nm period



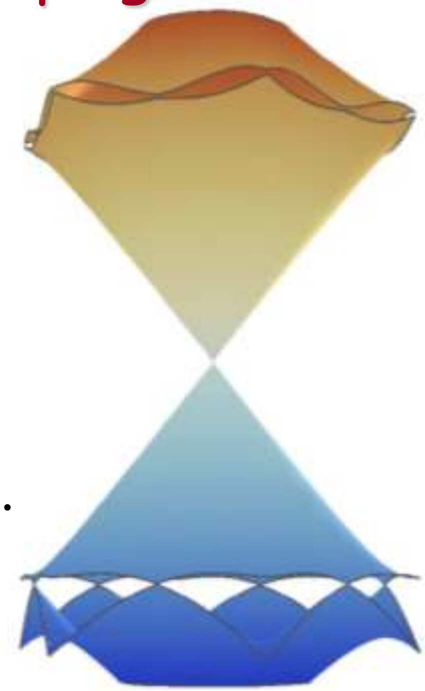
specially aligned graphene devices
second Dirac points



new neutrality points @0.2-0.35 eV

electron-like orbits for strong hole doping and vice versa

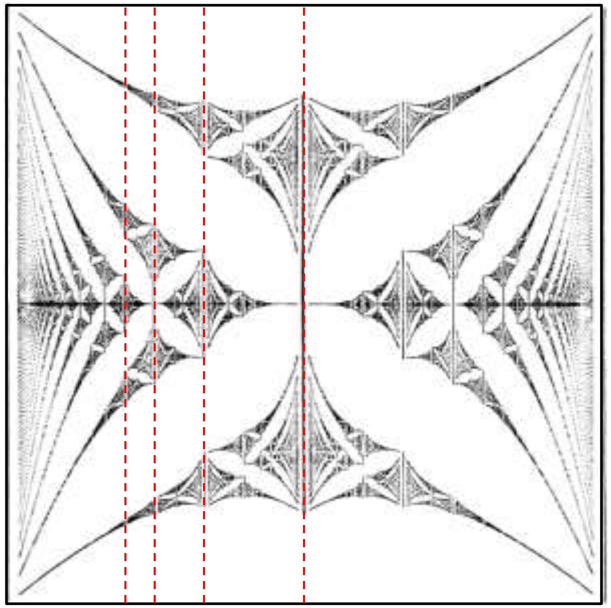
$n \sim 3\times10^{12}$ cm^{-2},
λ = 13 nm,
(A = 150 nm^2)
4 electrons/u.c.



second generation Dirac fermions

Ponomarenko et al. Nature (2013)

what happen in magnetic field?



Two competing lengthscales:

a : lattice periodicity

l_B : magnetic length

Duglas F. Hofstadter, Phys. Rev. B 14, 2239 (1976)

$$\frac{\phi}{\phi_0} = \frac{a^2 B}{\phi_0}$$ flux quanta per unit cell

Energy levels develop fractal structure when magnetic length is of the order of the lattice period

graphene:

$$l_B = \left(\frac{\hbar c}{eB}\right)^{1/2} = \frac{25\ \text{nm}}{\sqrt{B[T]}}$$

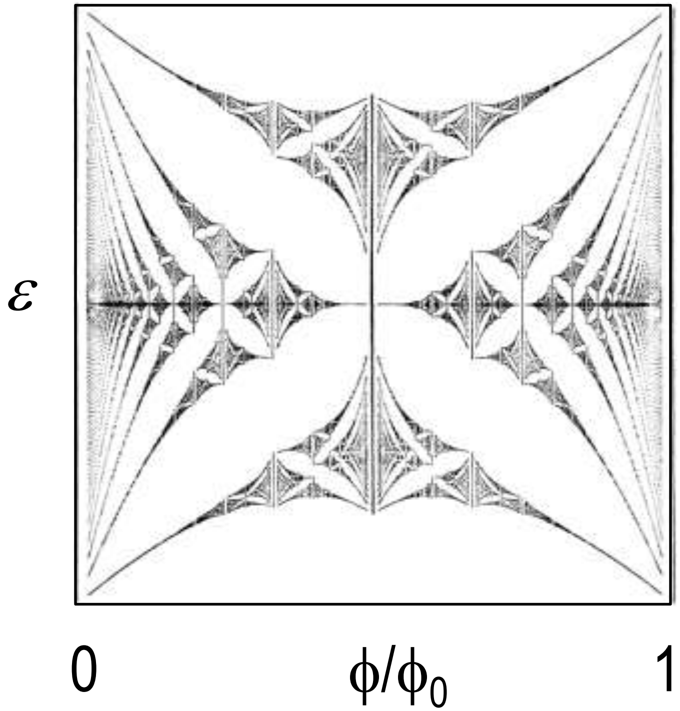
$a \approx 0.25$ nm $\Rightarrow$ B ≈ 10^4 T

graphene/BN superlattice:

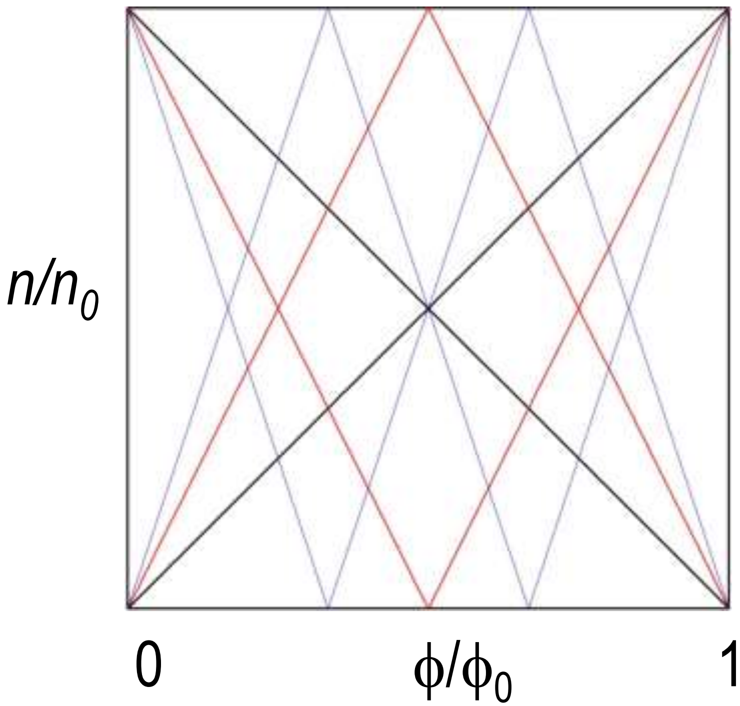
$\lambda \approx 14$ nm A ≈ 170 nm^2

B ≈ 24 T ($\phi/\phi_0 = 1$)

Tracing gaps in B and n



Hofstadter's energy spectrum



Wannier diagram

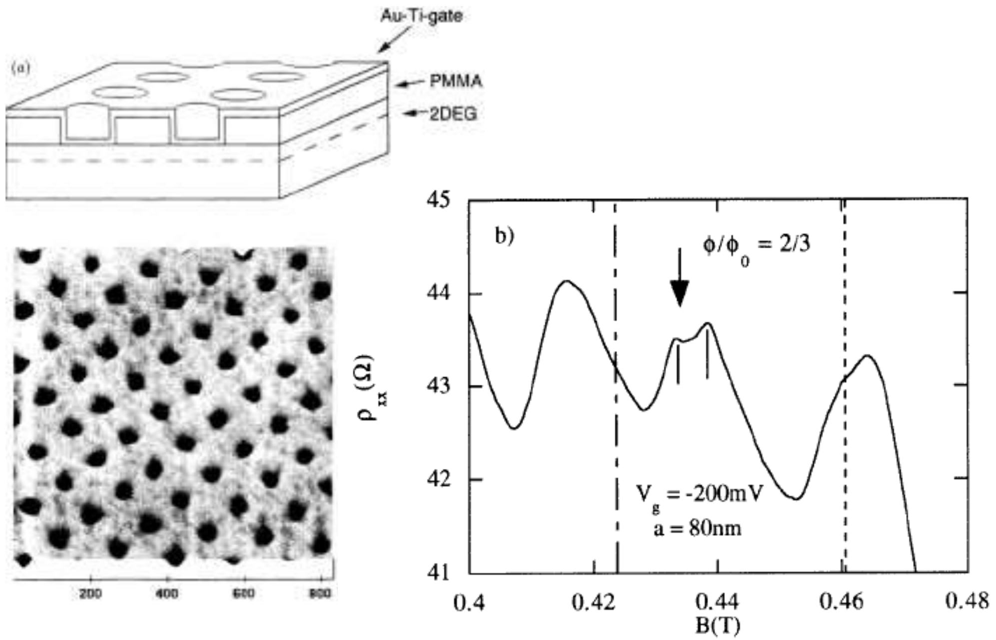
Phys. Status Solidi **88**, 757 (1978)

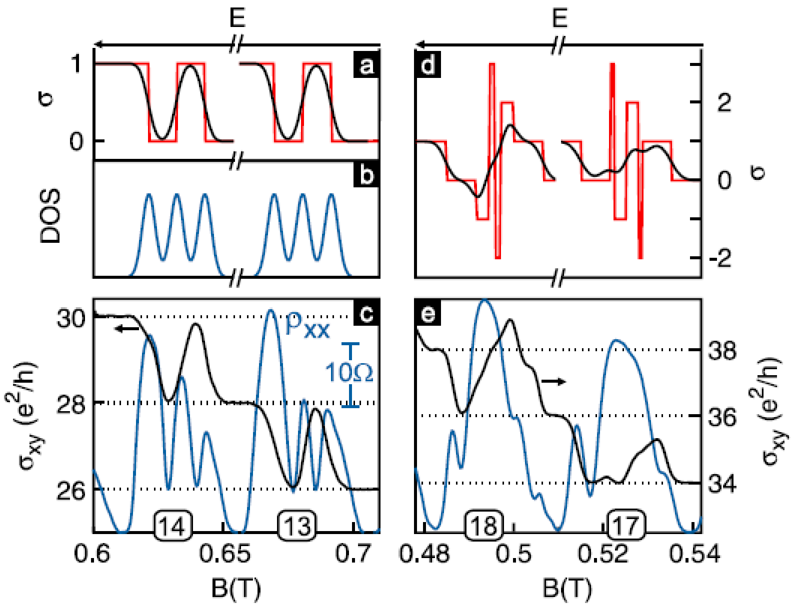
gaps are constrained to linear trajectories in the B-n diagram:

n/n_0 – normalised density (n_0 = 4/A, where A is the area of supercell)

Glimpse of Hofstadter's butterfly

Earlier attempts in GaAs based structures





T. Schlosser et al, Semicond. Sci. Technol. (1996)

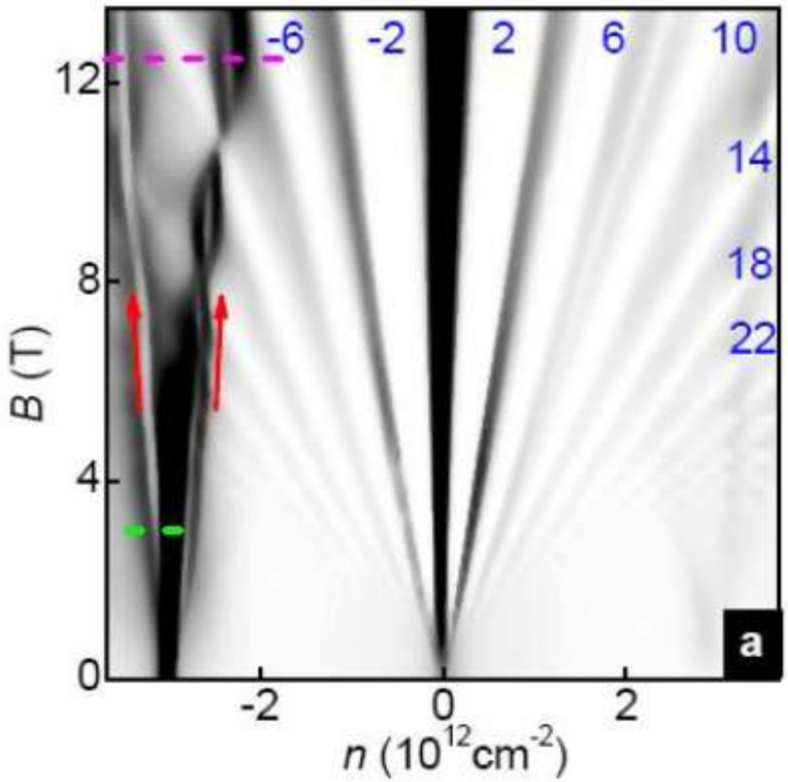
T. Schlosser et al, Europhys. Lett.(1996)

C. Albrecht et al, PRL (2001)

M. C. Geisler et al, PRL (2004)

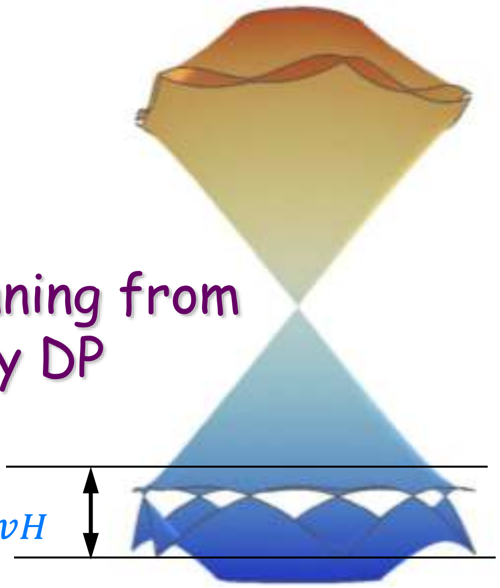
- Large unit cell (100 nm or larger)
- Limited range of densities
- Significant disorder

Magnetotransport measurements



Some of the features of Hofstadter's energy spectrum are seen in magnetotransport

"Landau levels" fanning from the secondary DP



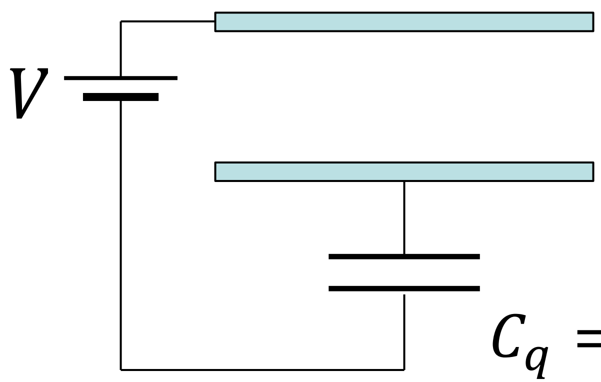
$E_{LL}(12\ T) > \Delta_{vH}$

L. A. Ponomarenko et al. Nature 497, 594–597 (2013)
C. R. Dean et al. Nature 497, 598–602 (2013)
B. Hunt et al., Science 340, 1427-1430 (2013)

Capacitance Measurements

Simple and reliable technique for studying details of the band structure

"geometrical" capacitance



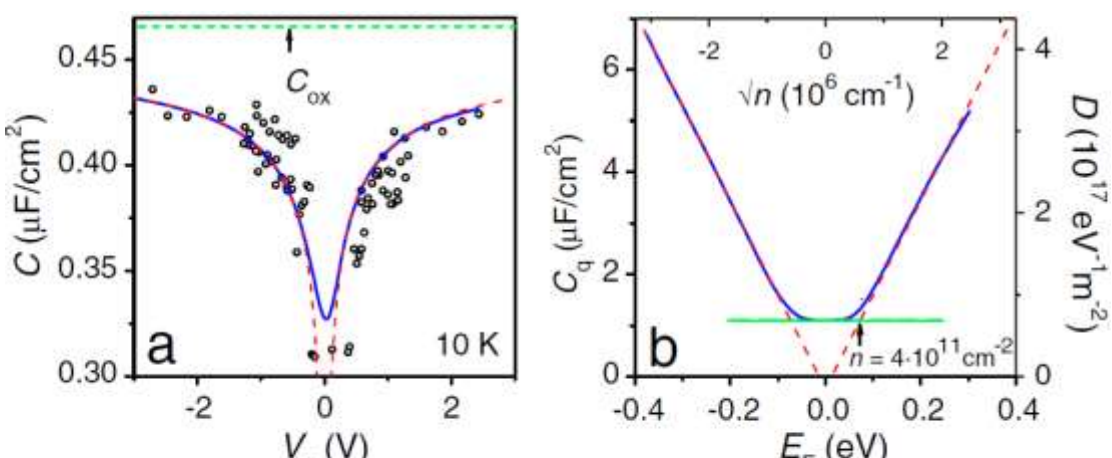
$$V = Ed + \frac{1}{e}\mu(n)$$

~~$V \propto n$~~

$$C_q = e^2 \frac{dn}{d\mu}$$

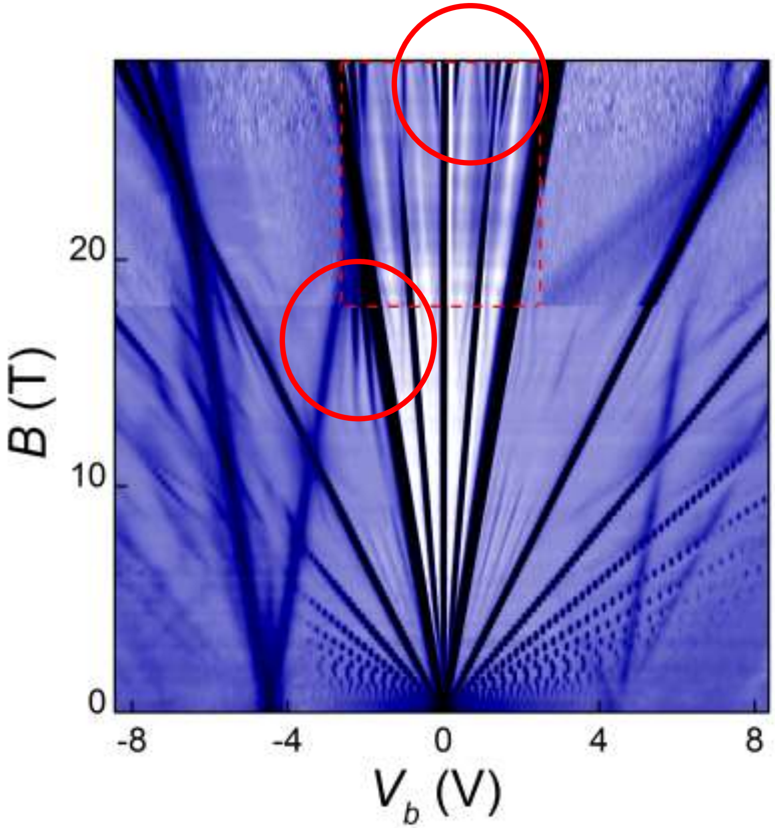
density of states

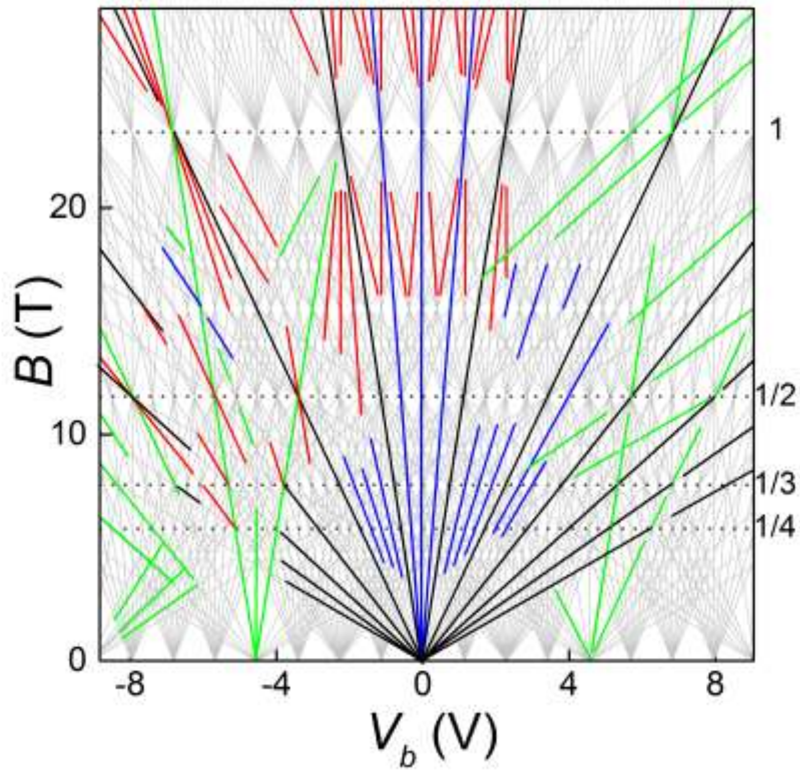
$$\frac{1}{C} = \frac{1}{C_g} + \frac{1}{C_q}$$



L.A.Ponomarenko et al, PRL 105 136801 (2010)

Signature of Hofstadter's butterfly





Black – Landau fan (4x degenerate)

Blue – Landau fan (degeneracy lifted, QHFM)

Green – "Landau levels" from secondary DPs

Red – gaps in Hofstadter spectrum

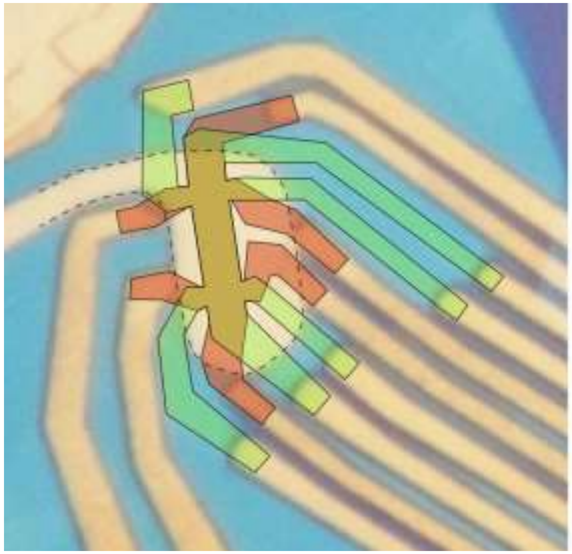
G.L.Yu et al. Nature Physics 10, 525 (2014)

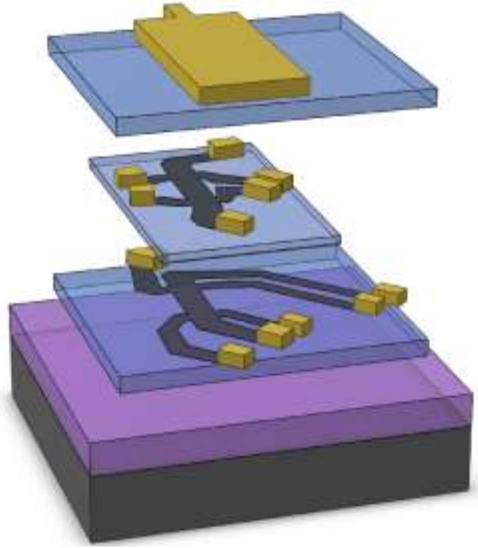
Conclusions for Part 2

Graphene superlattice is an excellent example of "band structure engineering" and creating artificial structures with on demand properties (simply by controlling the orientation of graphene with respect to the properly chosen substrate)

Hofstadter butterfly has been finally observed (almost 40 years after its prediction)

Graphene Double Layer Structures





Layer-by-layer material engineering

BN-Gr-**BN**-Gr-BN

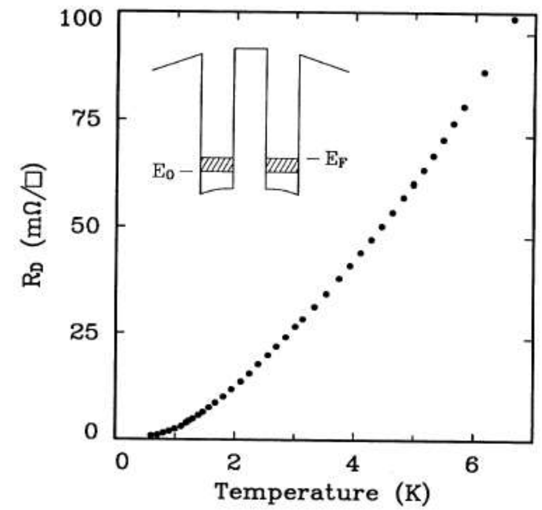
$\mu_B \approx$50,000 to 120,000 cm^2/Vs
$\mu_T \approx$30,000 to 60,000 cm^2/Vs

BN thickness:
anything down to monolayer
(drag measurements - 3 layers)

High quality, perfect interface, versatile system

Double Layer Structures

GaAs/AlGaAs double-quantum-well structures have been studied for more than 20 years (in particular J. Eisenstein group, Caltech)



also Cavendish and Sandia Labs on e-h bilayers

T.J. Gramila et al, Phys. Rev. Lett. (1991)

Weakly coupled layers ($d \ll n^{-1/2}$): e-e scattering (Coulomb drag)

Strongly coupled layers: support coherent state (excitonic condensation)

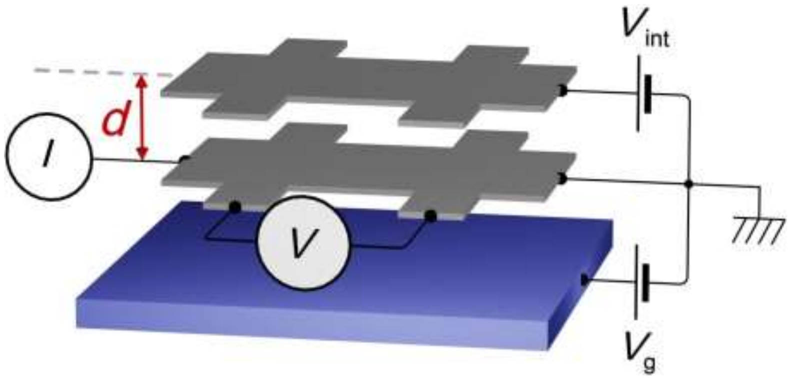
Tunnelling spectroscopy: details of bend structure

Graphene Double Layers vs Double Quantum Well

	GaAs/AlGaAs	Gr/BN/Gr
Separation	> 15 nm	>1 nm
Temperature range	< 5 K	up to room T
Contacts	Split gates needed	No need of split gate
Carriers	Either electrons or holes in each layer	Ambipolar (both e and h)
Size	a few mm	a few μm (UCF at low T)
Mobility (cm^2/Vs)	> 1 000 000	> 100 000

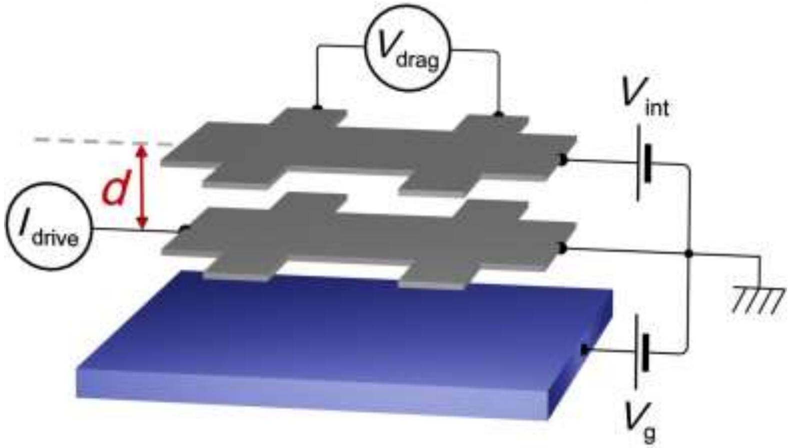
Graphene double layers - strongly interacting regime ($k_F d \sim 0.1$-1)

Graphene Double Layer Structures



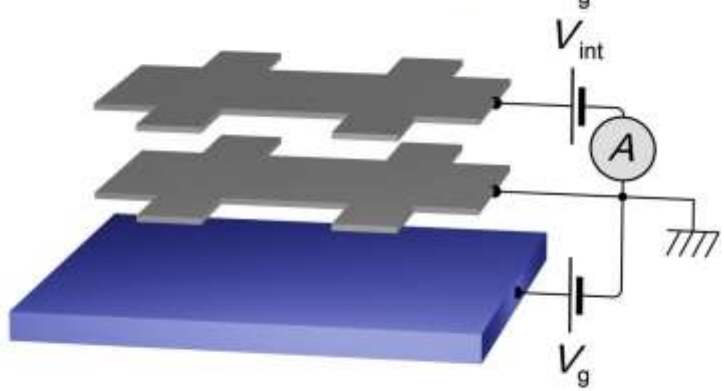
Graphene as a tunable metal plate:
Screening of charged impurities
Metal-Insulator Transition

Ponomarenko et al., Nature Physics (2011)



Coulomb drag

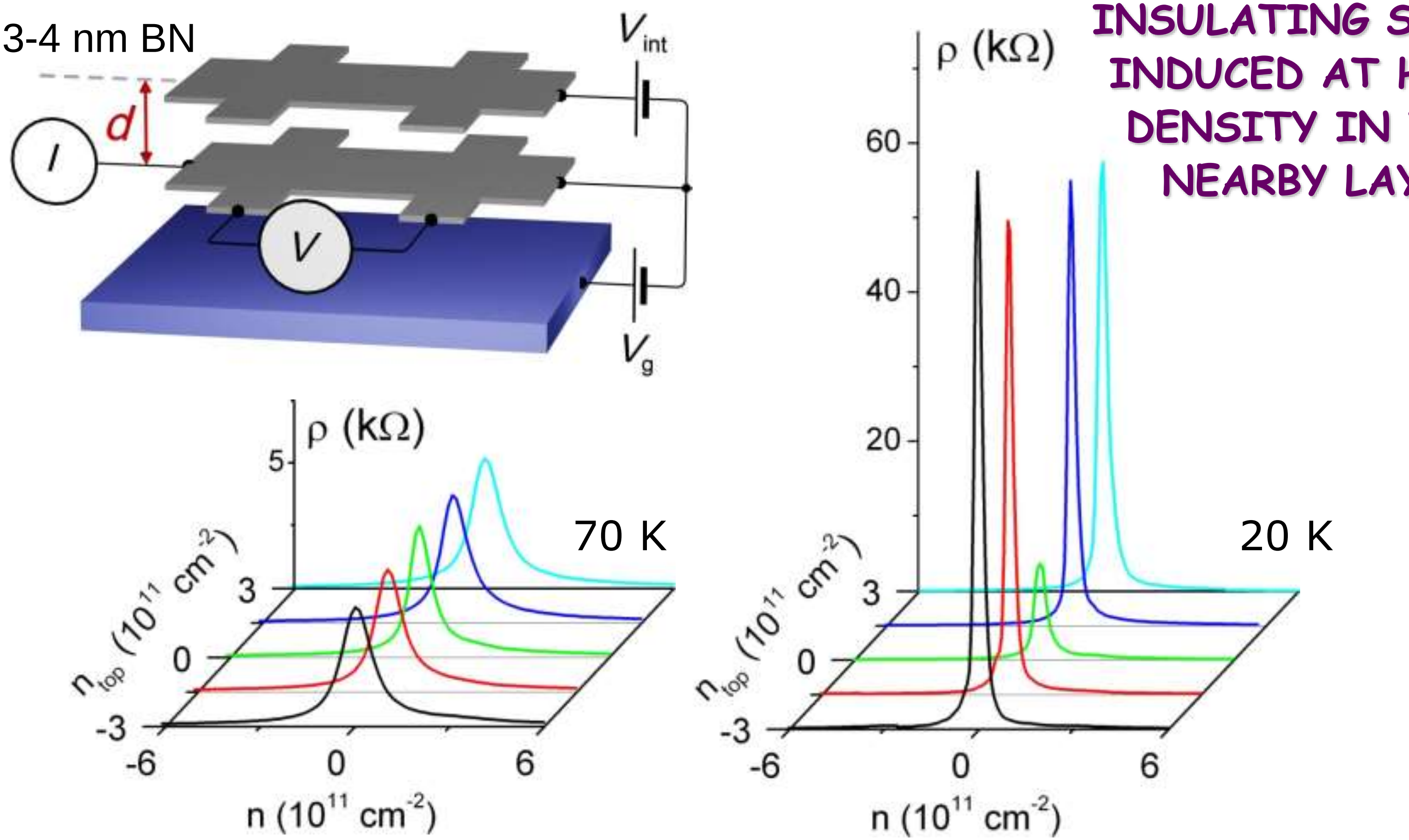
Gorbachev et al. Nature Physics (2012)
Titov et al. Rhys. Rev. Lett (2013)



Tunneling Transistors

Britnell et al., Science (2012)
Georgiou et al. Nature Nano. (2013)

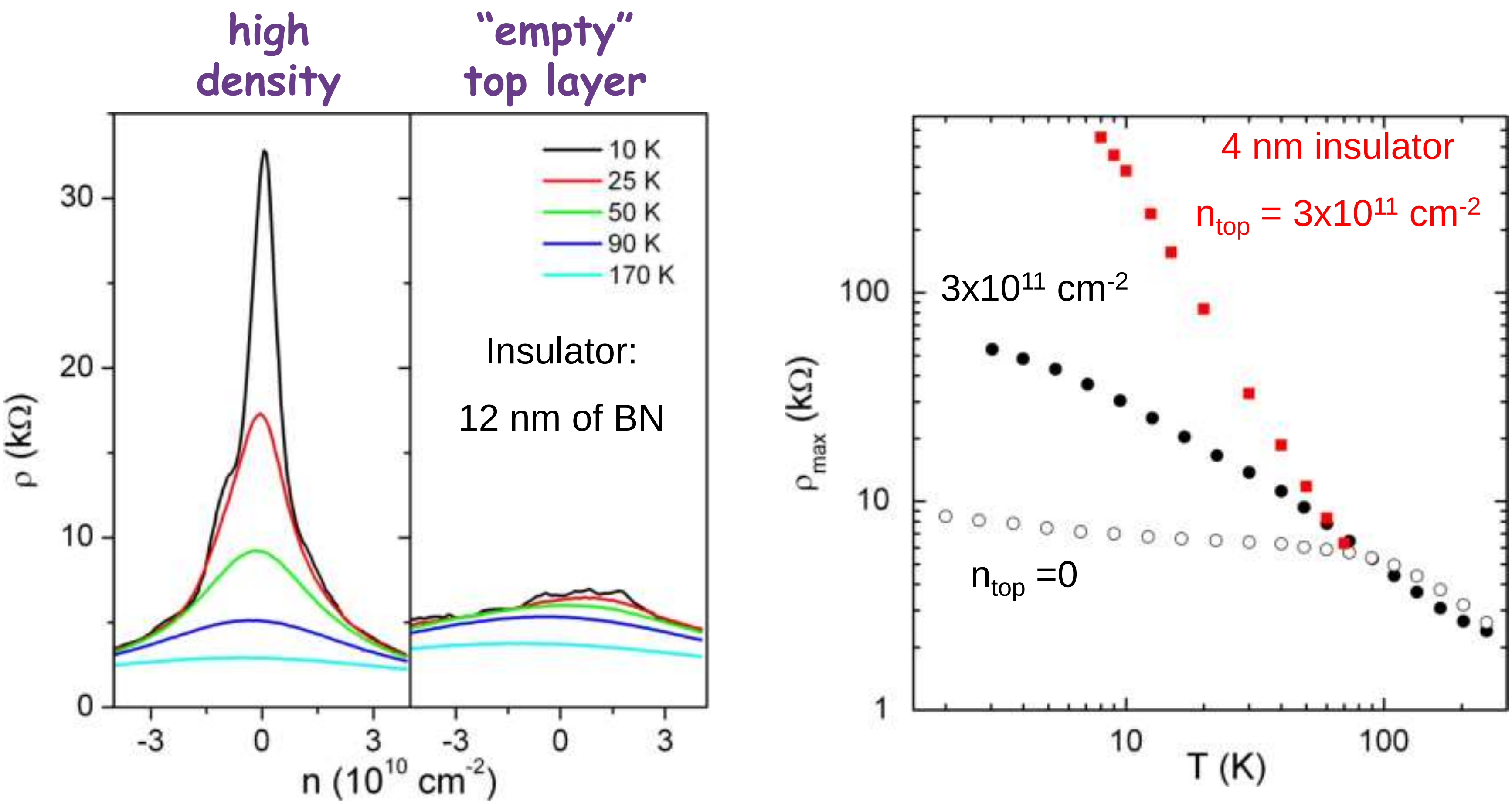
Tunable Metal-Insulator Transition



INSULATING STATE INDUCED AT HIGH DENSITY IN THE NEARBY LAYER

Top layer as a metallic plate with tunable density

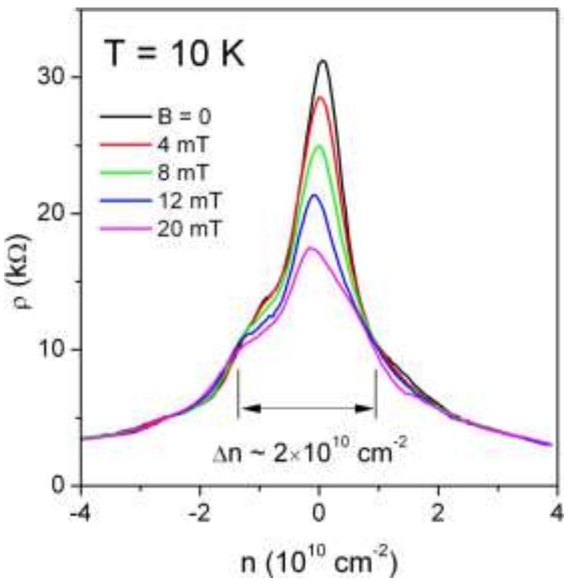
Temperature dependence

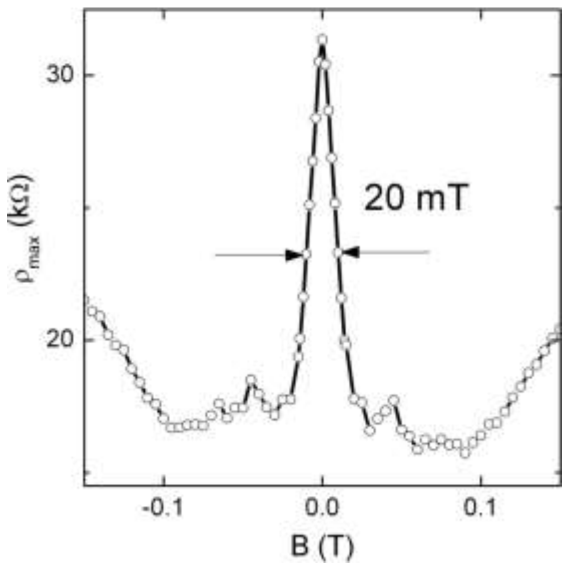


Power-law rather than activation behaviour ⇒ no gap

$$\rho_{max} \sim T^{-\alpha},\ \ 1 < \alpha < 2$$

Field dependence





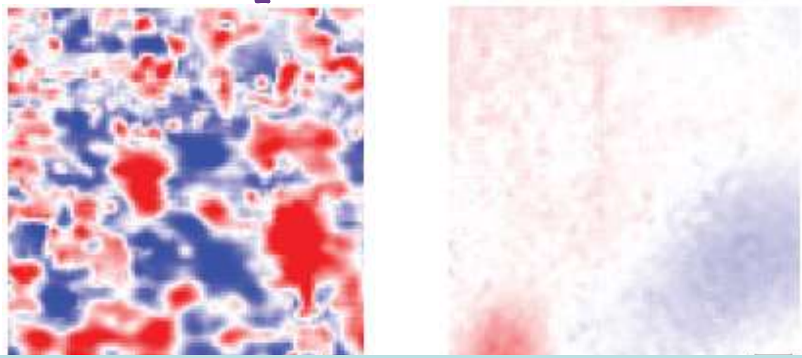
Insulating state suppressed by weak magnetic field ⇒

MI transition is interference phenomenon (localization)

Screened-Out Puddles

Disorder results in e-h puddles

on SiO_2 | on BN



Yacoby, Nature Phys 2007
LeRoy, Nature Mat 2011
Crommie, Nano Lett 2011

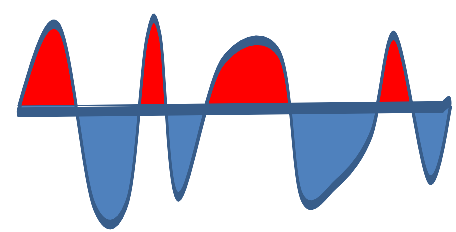
Message:

By screening puddles we can make graphene insulating

typical puc … $10^{10}cm^{-2}$)

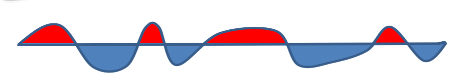
$\rho \approx h/4e^2$ due to percolation of e-h puddles

Falko et al PRL 2007; Das Sarma et al PNAS 2007; Fogler PRL 2009



second graphene acts as a metallic plate and screens out e-h puddles

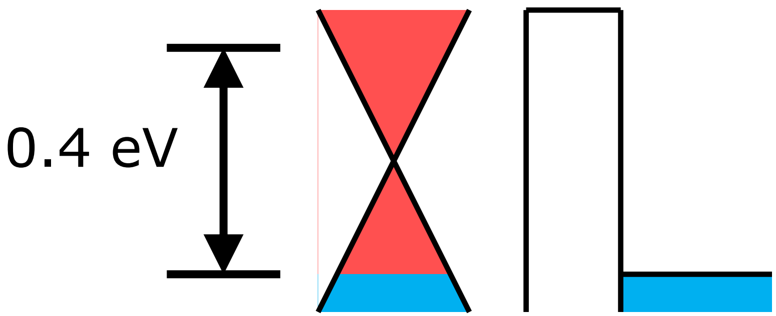
MAKING PUDDLES SHALLOWER THAN CRITICAL CAUSES THE LOCALIZATION TRANSITION



Tunnelling Transistor

Graphene doesn't have a band gap, but…

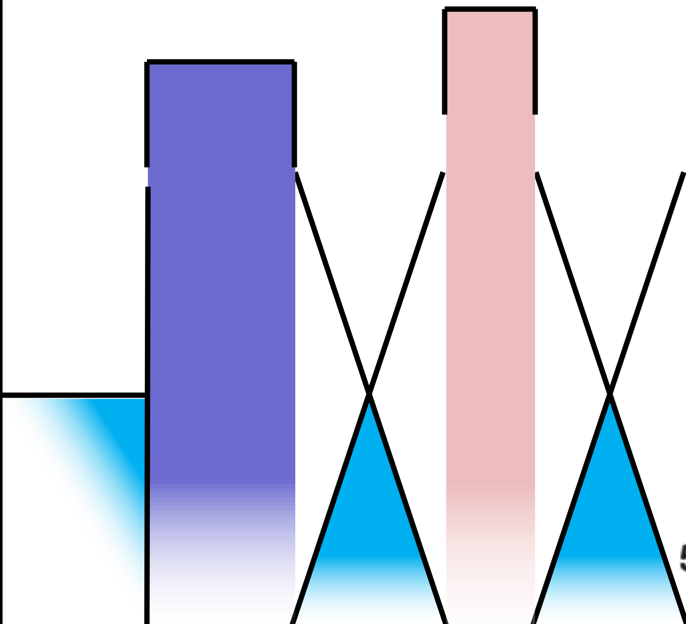
Fermi level can be moved by ±0.2 eV easily



The barrier has to be insulating 2D-crystal

The second electrode can be any conductor, but graphene (or graphite) works the best.

Tunnelling Transistor

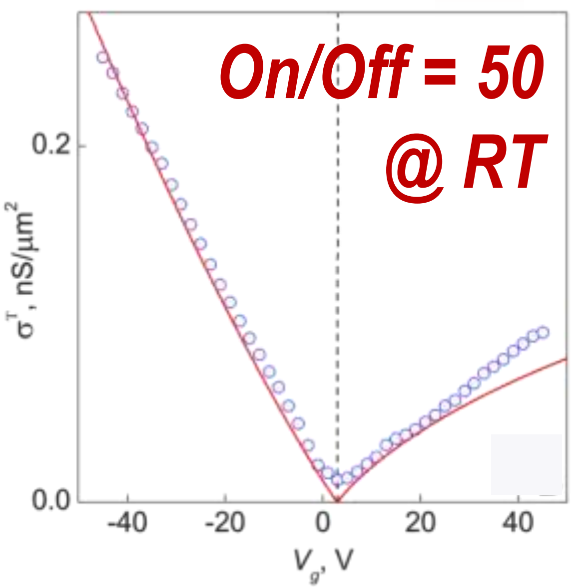


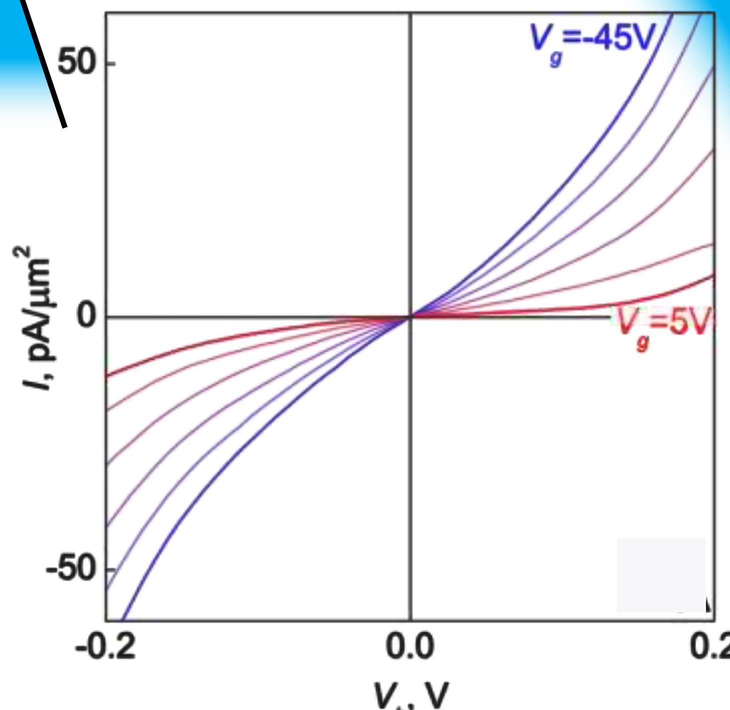
$$I \propto DoS_b \, DoS_t \, T/W'$$

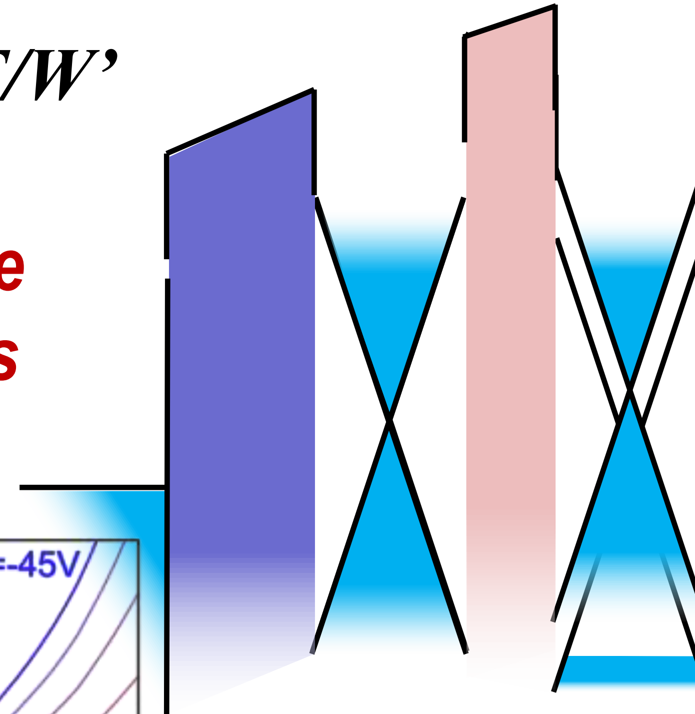
for BN barrier:

dominated by the Density of States

On/Off = 50 @ RT



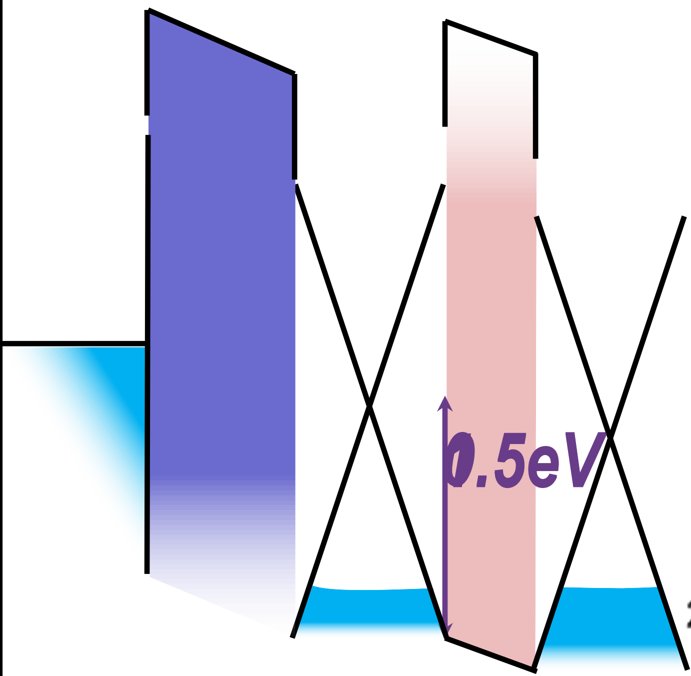




Graphene-based vertical tunnelling transistor

L. Britnell et al Science '12

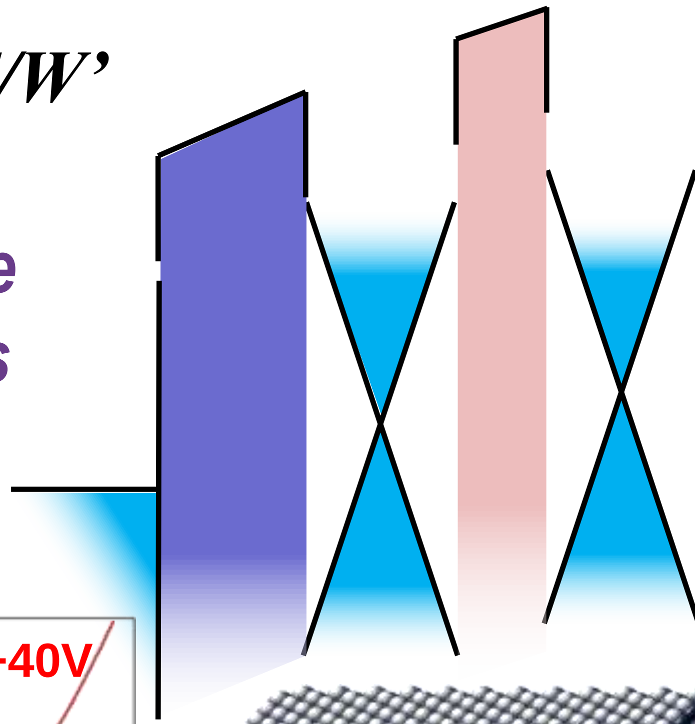
Increasing On/Off

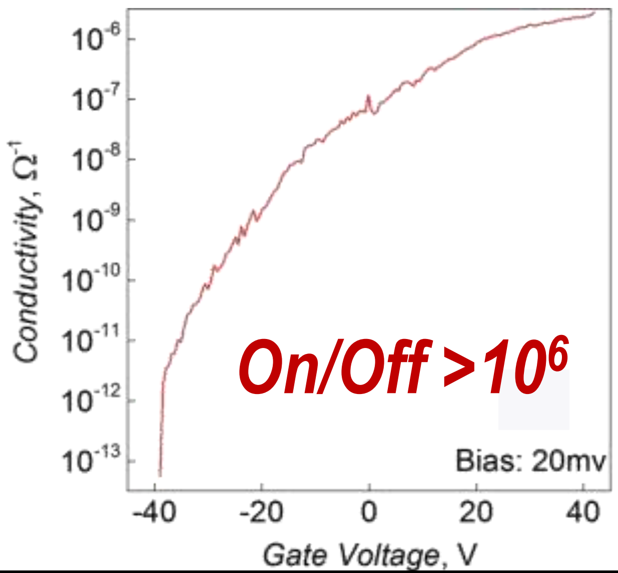


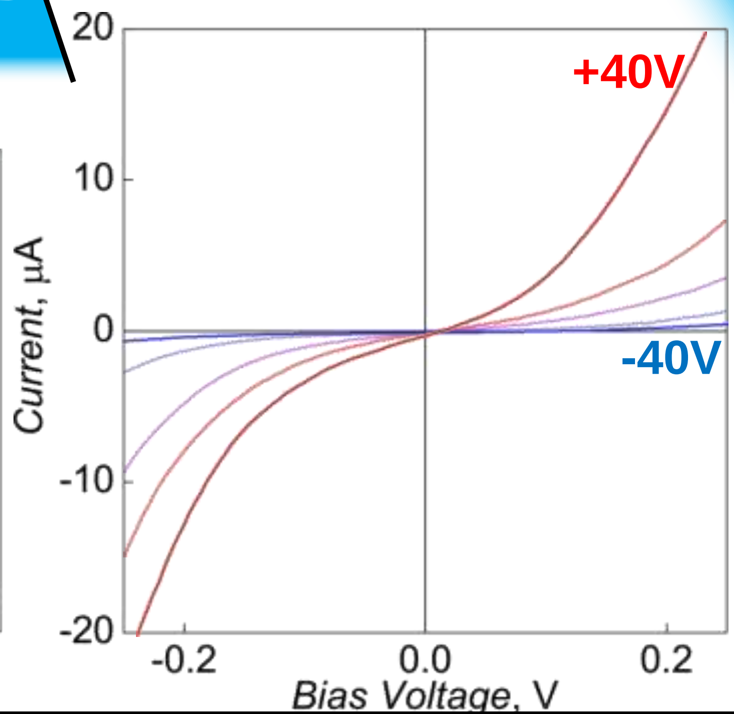
$I \propto DoS_b \, DoS_t \, T/W'$

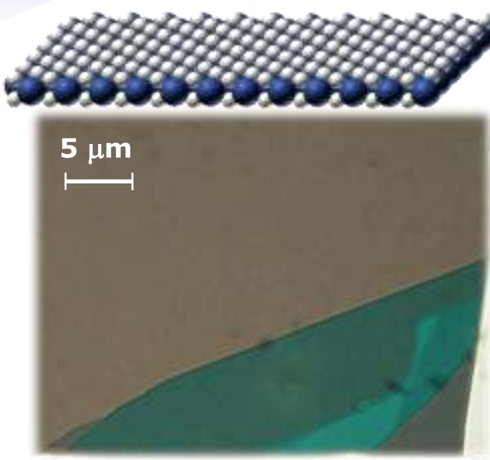
for BN barrier: dominated by the Density of States

Dominated by $T \propto \exp(-d \cdot \Delta^{1/2})$









2D MoS_2 in optics

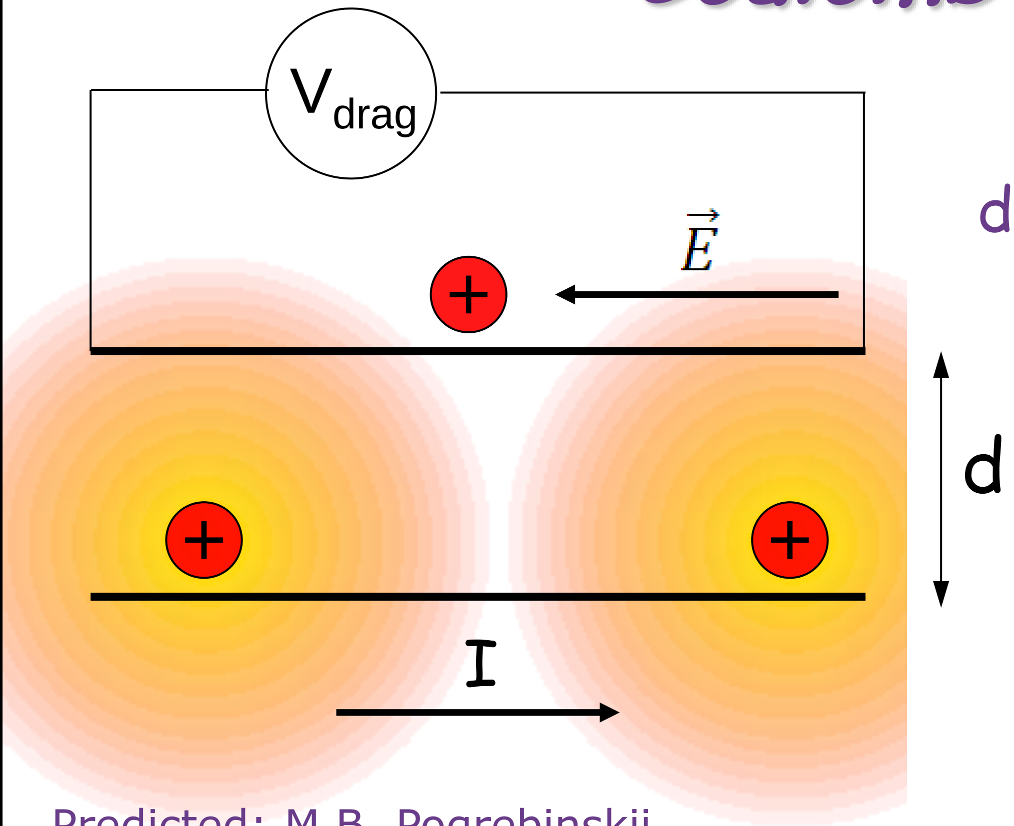
L. Britnell et al Science '12

MESSAGE TO TAKE AWAY

VERTICAL TUNNELING DEVICES
OFFER ALTERNATIVE ROUTE
TO GRAPHENE-BASED ELECTRONICS

remains to be evaluated by engineers

Coulomb Drag



Momentum transfer

d is of the order of the distance between charge carriers

d ~ 1 nm of hBN (20 times smaller than in traditional GaAs systems)

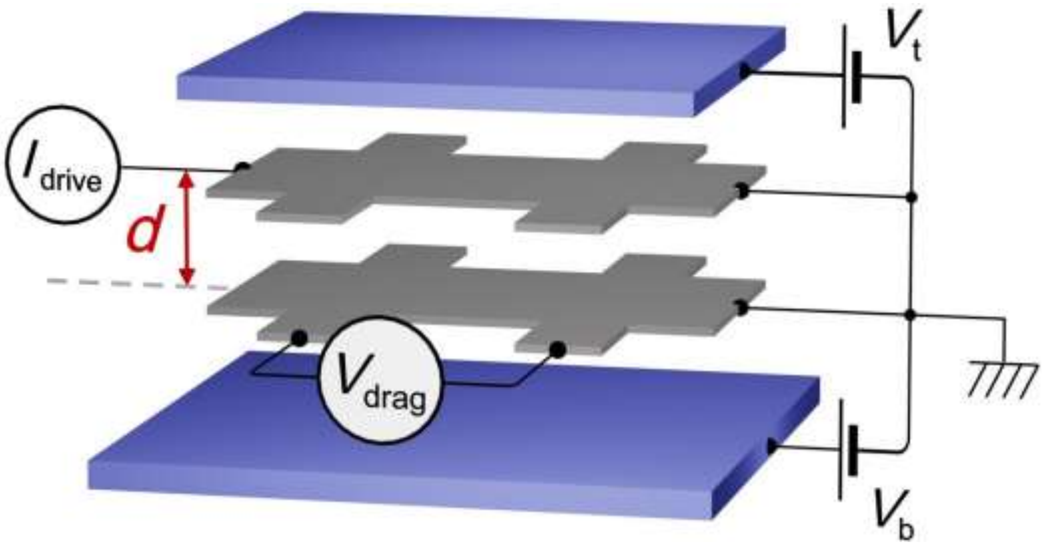
Predicted: M.B. Pogrebinskii, Fiz. Tekh. Poluprovod. 11, 637 (1977)

Nandi *et al.* Nature 488, 481 (2012)

Direct measurements of e-e scattering rate in simple transport experiment

d as small as 1 nm of BN $\Rightarrow$ $k_F d < 1$ (strong interaction)

Drag in Double Layer Structures



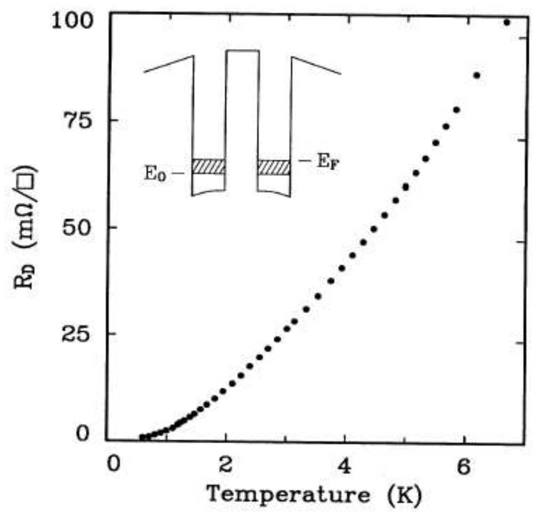
$$R_{drag} = \frac{V_{drag}}{I_{drive}}$$

Two gates to control densities in both layers

Note: no tunnelling between graphene layers

Weakly coupled layers ($d \gg n^{-1/2}$): e-e scattering (Coulomb drag)

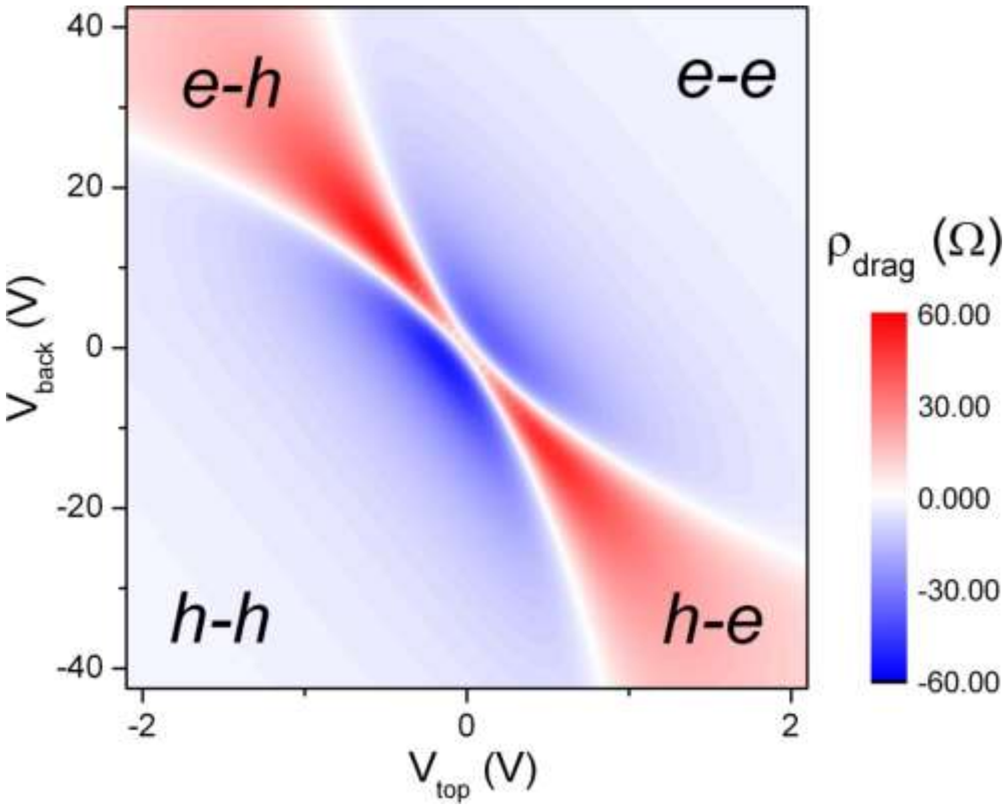
Strongly coupled layers: support coherent state (excitonic condensation)

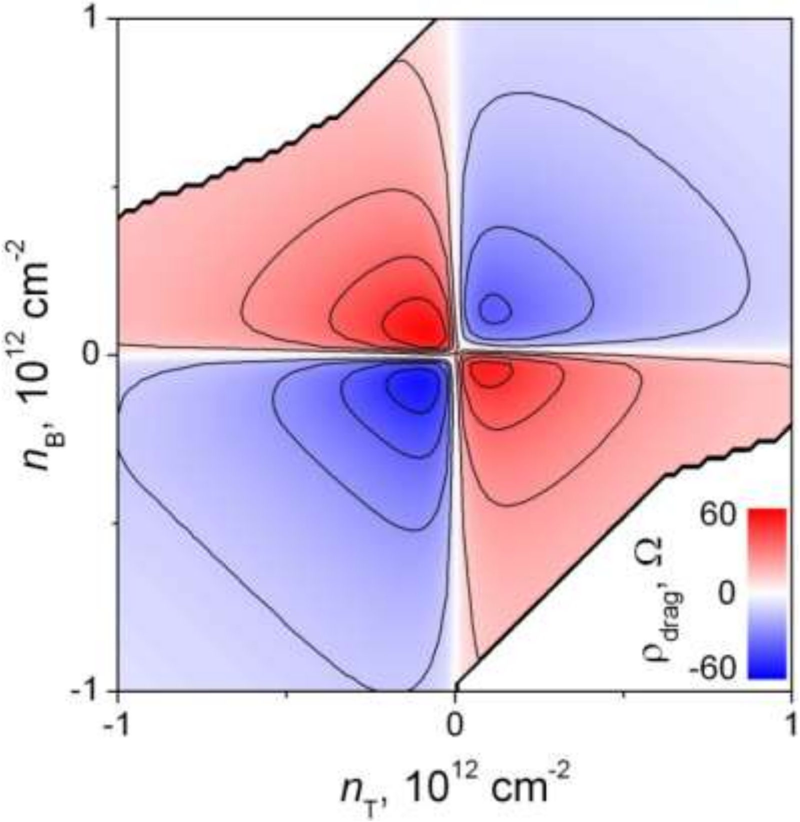


T.J. Gramila et al, Phys. Rev. Lett. (1991)

Drag measurements

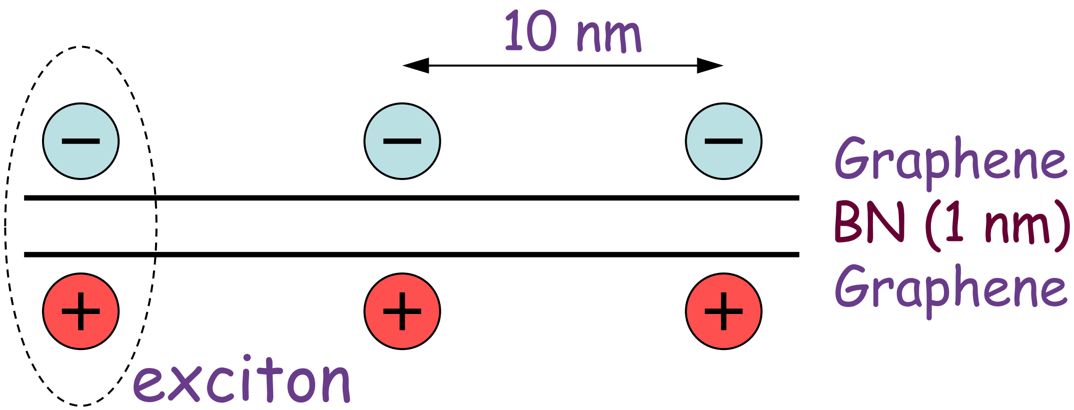
240 K, 3 layers of BN (~ 1 nm)

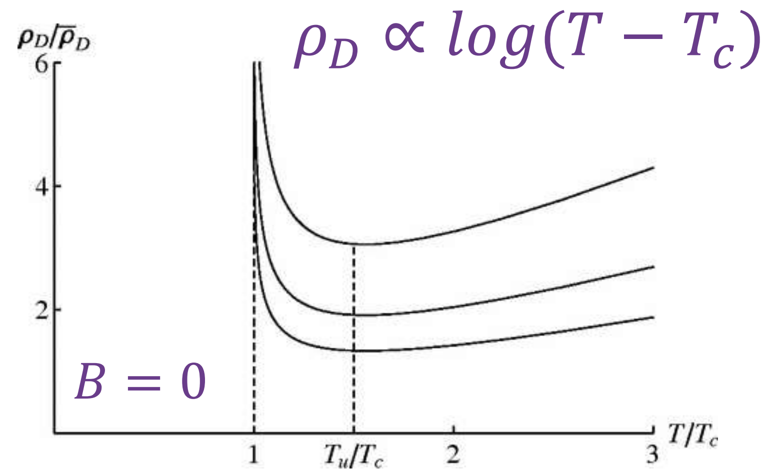




Drag resistance has a right sign.

Interlayer Excitons





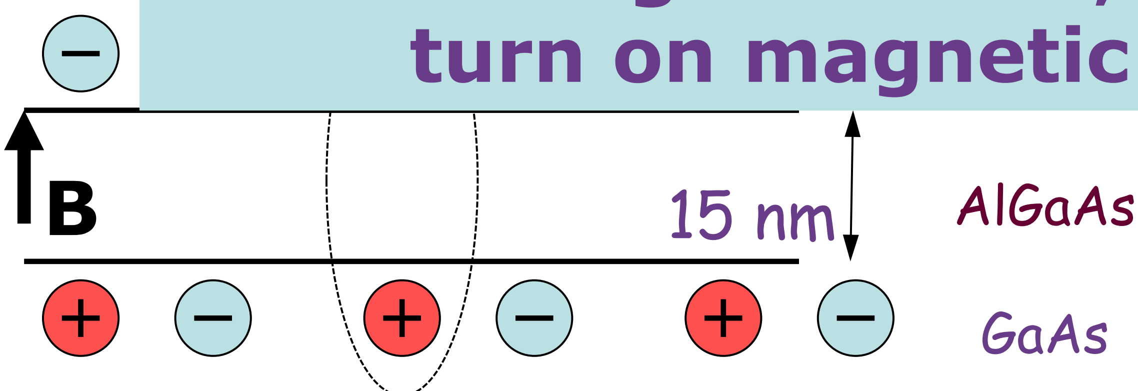
Excitons are bosons ⇒ condensation?

M.Yu. Kharitonov and K.B. Efetov PRB (2008)
H. Min et al, PRB (2008)
Yu. E. Lo
D. K. Efin
A. Perali
Abergel,
D. Neilso

B.Y.-K. Hu.PRL 85, 820 (2000)
M.P. Mink et al. PRL 108, 186402 (2012)

Strategy: check drag at room T,
stay close to Dirac point,
go to low T,
turn on magnetic field

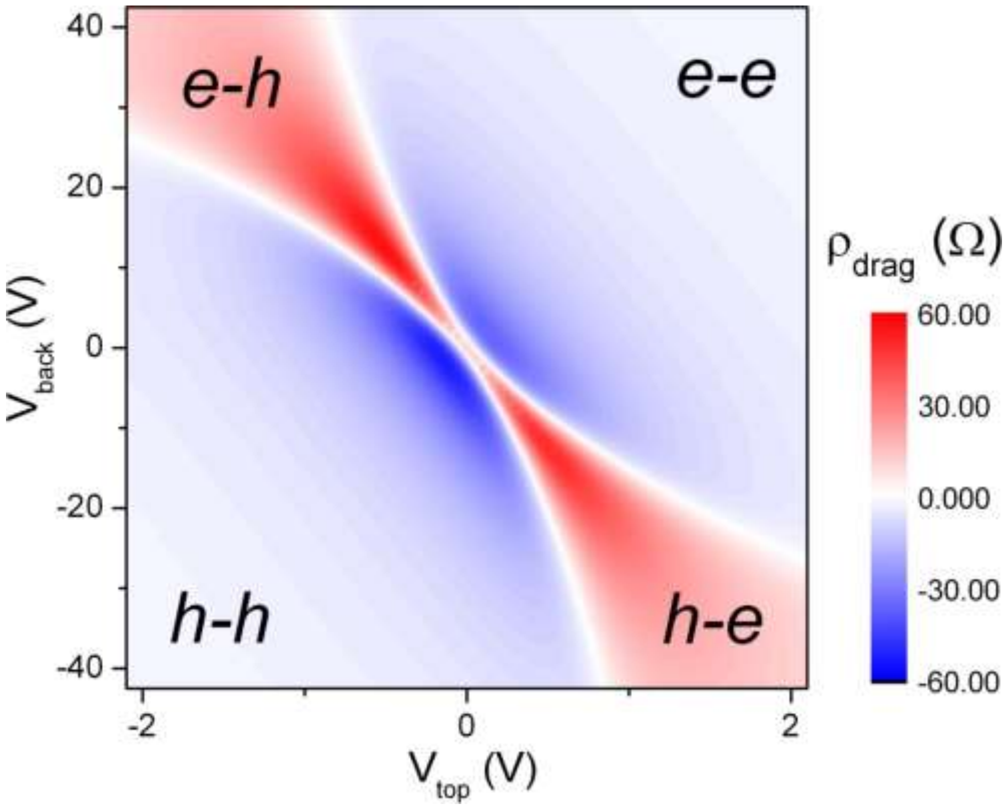
au level
n, 90's

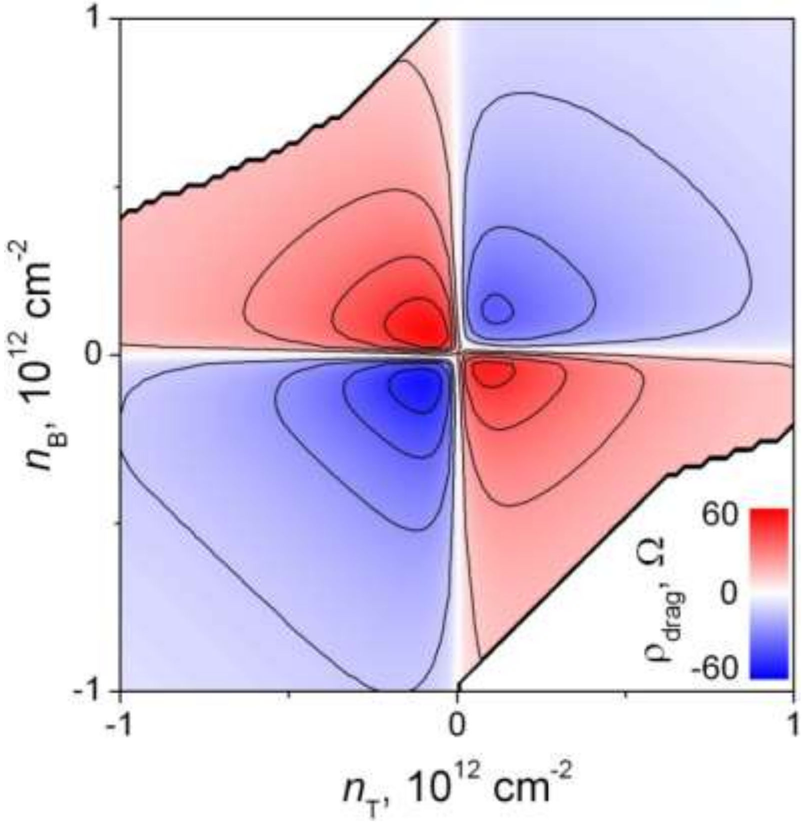


Graphene: half filled
Landau level is
at neutrality point

Drag measurements

240 K, 3 layers of BN (~ 1 nm)

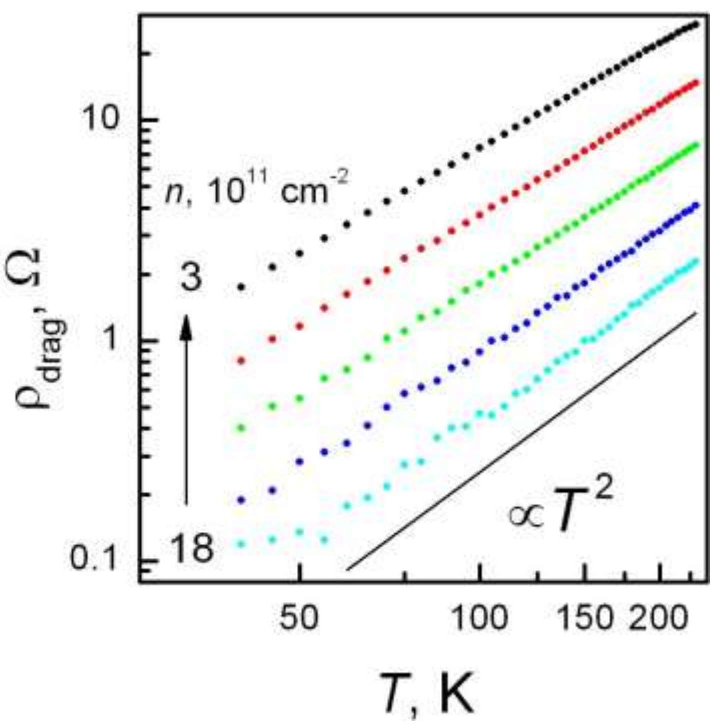


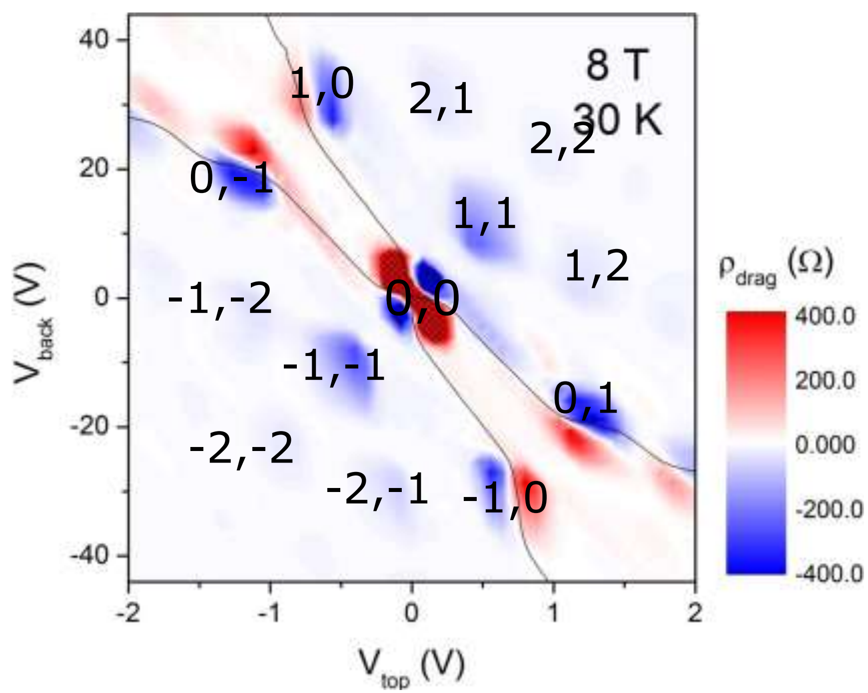


No significant difference between $n_t = n_b$ and $n_t = -n_b$

(no excitons at room T)

Temperature and Field Dependence





Between LLs drag vanishes

Temperature dependence is quadratic down to 40 K (no sign of exciton condensation)

Large drag at the double neutrality point (0,0)

Conclusions

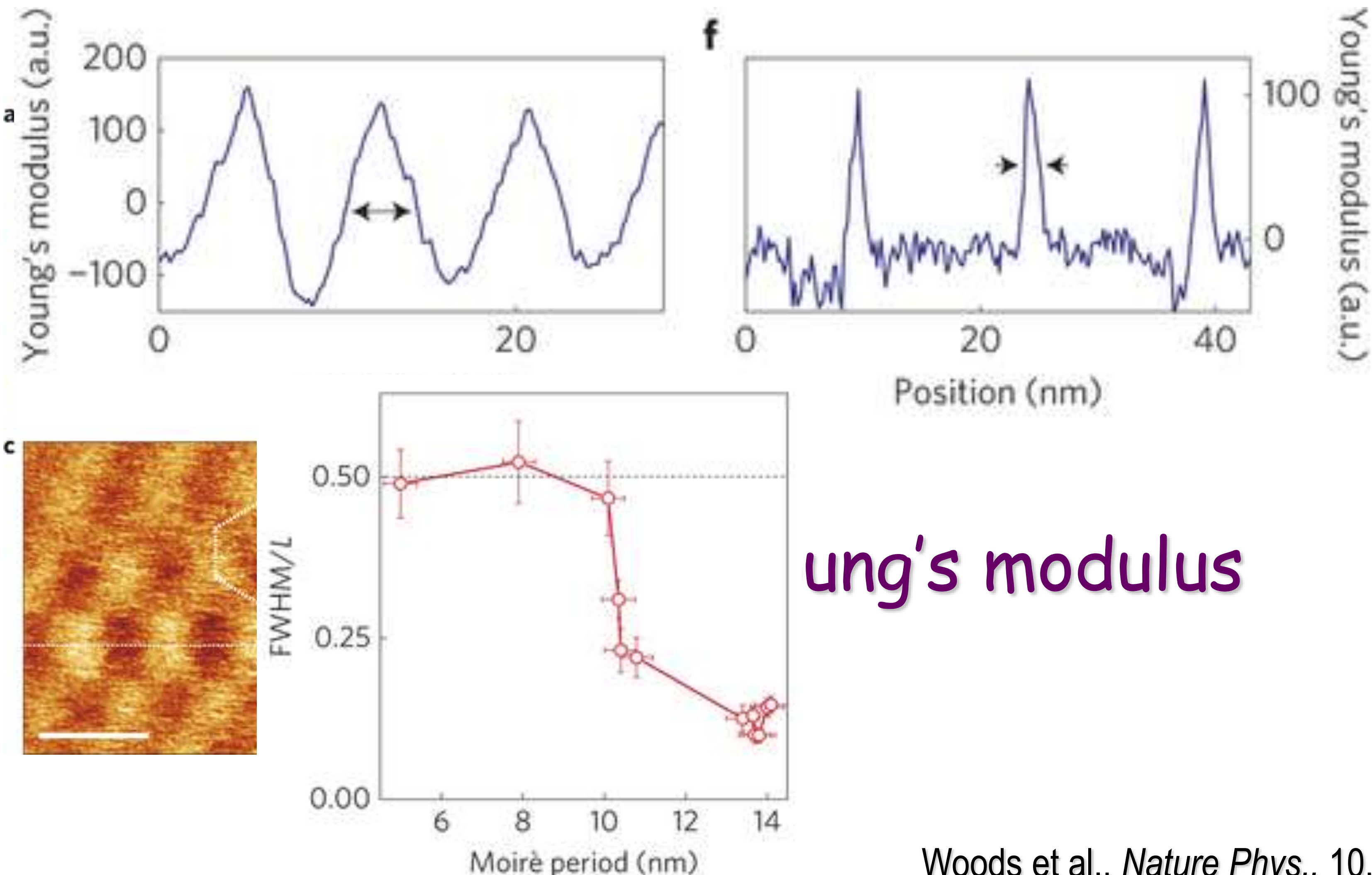
Graphene multi-layer structures:
there is a lot of new and interesting physics beyond graphene

Vertical tunnelling transistors offer ON/OFF ratio suitable for digital applications.

By screening the charged impurities one can make graphene insulating

Coulomb drag in double-layer structures: towards excitonic Bose-Einstein condensation

Commensurate-incommensurate transition in graphene on hBN



ung's modulus

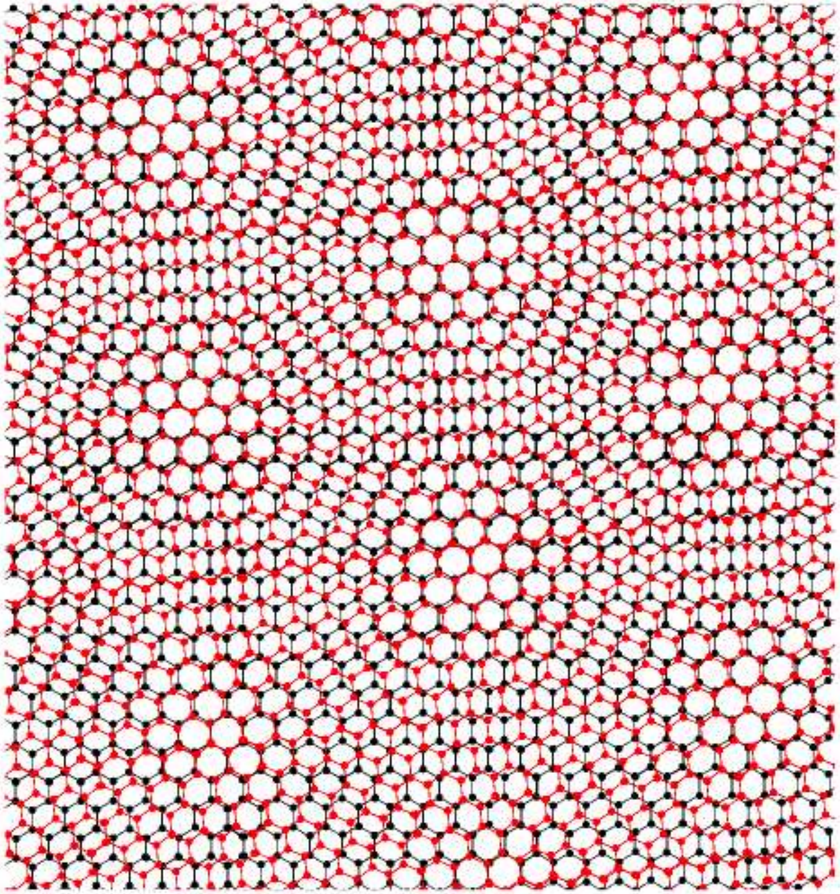
Woods et al., *Nature Phys.*, 10, 2014, 451.

Two types of G/hBN superlattices

Incommensurate:

Not aligned

No global gap

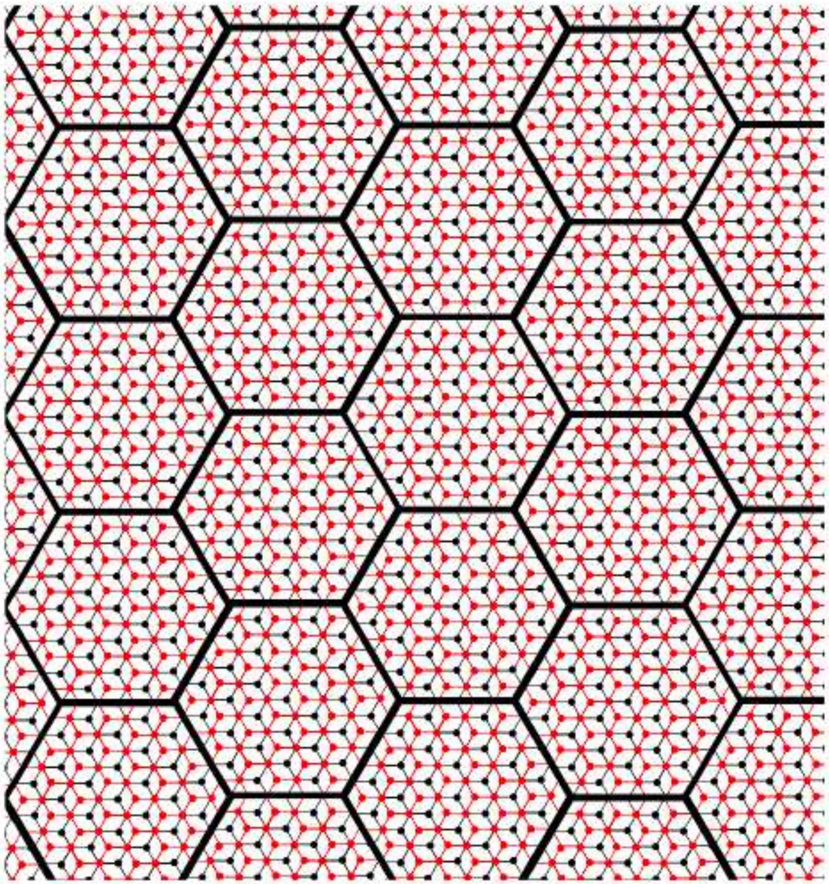


Ponomarenko et al., *Nature,* 497, 2013, 594.

Commensurate:

Global A/B asymmetry

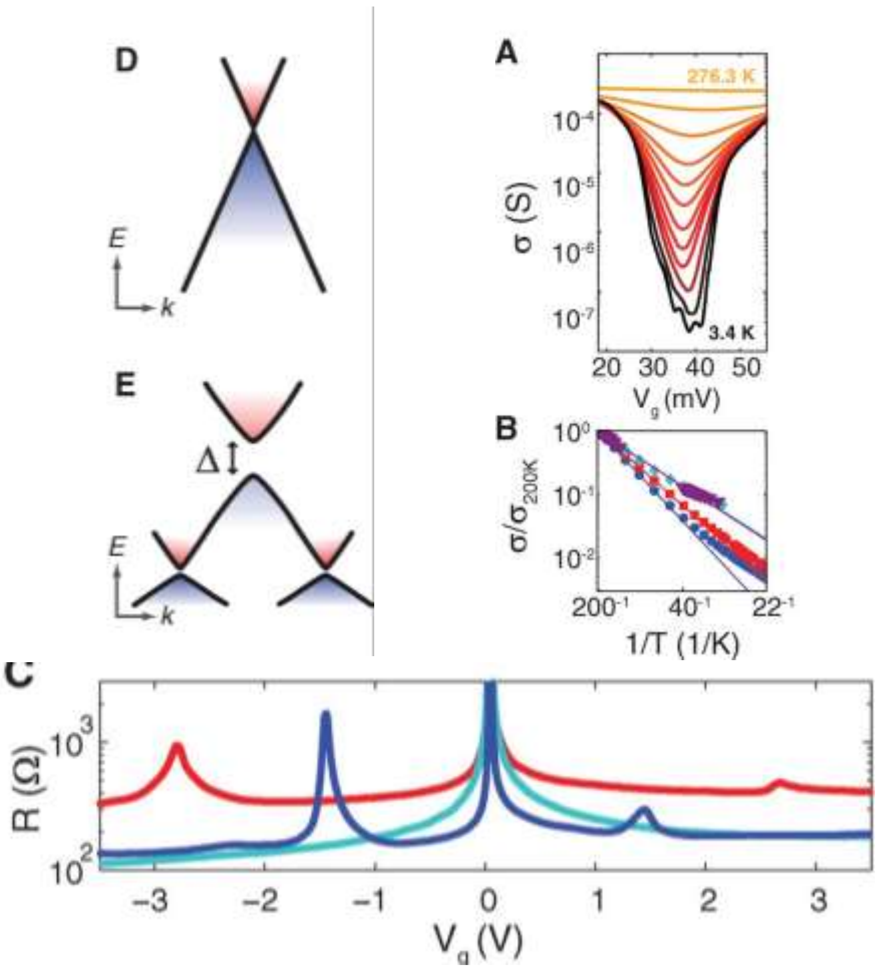
Global gap



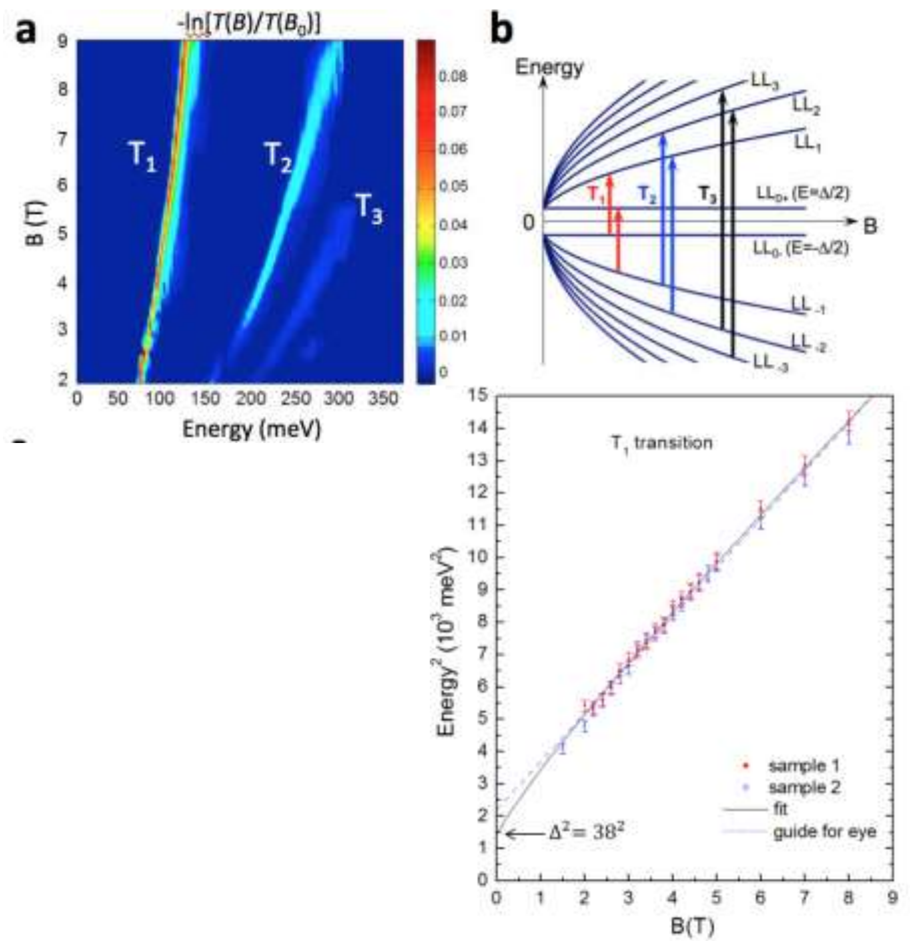
Woods et al., *Nature Phys.,* 10, 2014, 451.

Massive Dirac Fermions in van der Waals Heterostructure

A gap opens at the Dirac point in aligned structures, $\Delta \sim 38\ \mathrm{meV}$



Hunt et al., *Science.*, **340**, 1427 (2013)



Chen et al., Nature Comm. **5**, 4461 (2014)