van der Waals Heterostructures: Fabrication and Materials Issues

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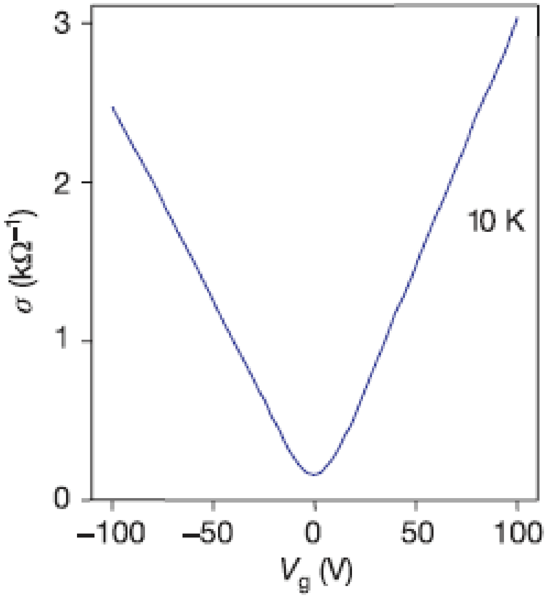
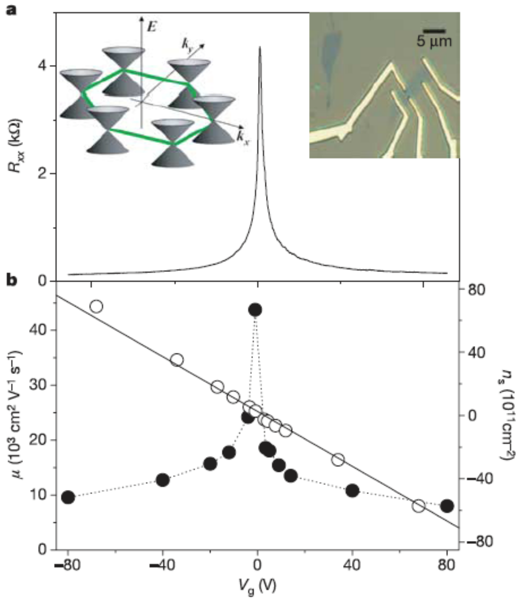
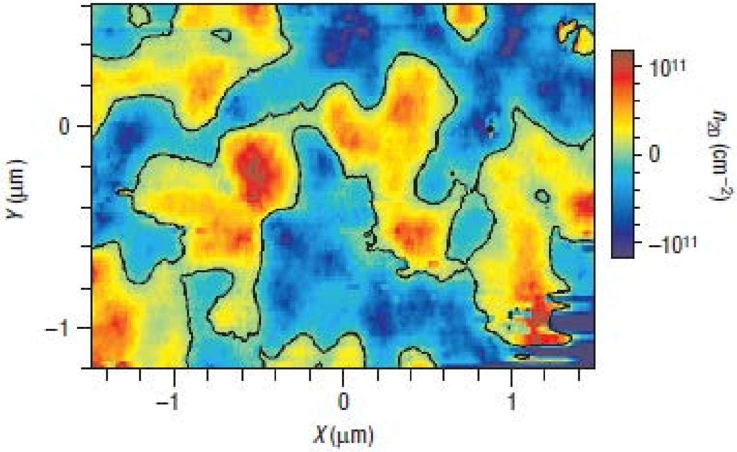
Graphene on SiO_2



Field effect mobility



Novoselov, Geim, Science (2005), Nature (2005); Zhang, Kim, Nat. (2005)

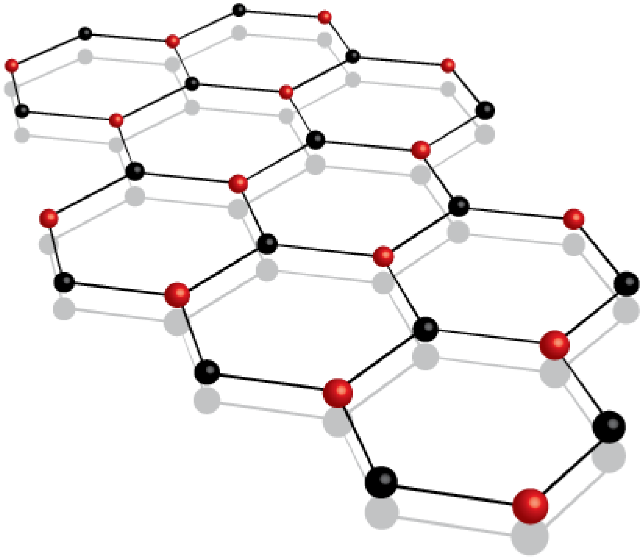


Martin, et al, Nature Phys. (2008)

Graphene on SiO_2:

- mobility ~20,000 cm^2/Vs
- surface roughness
- scattering from charged impurities
- disorder from charged impurities

Boron Nitride – graphene’s insulating ‘cousin’



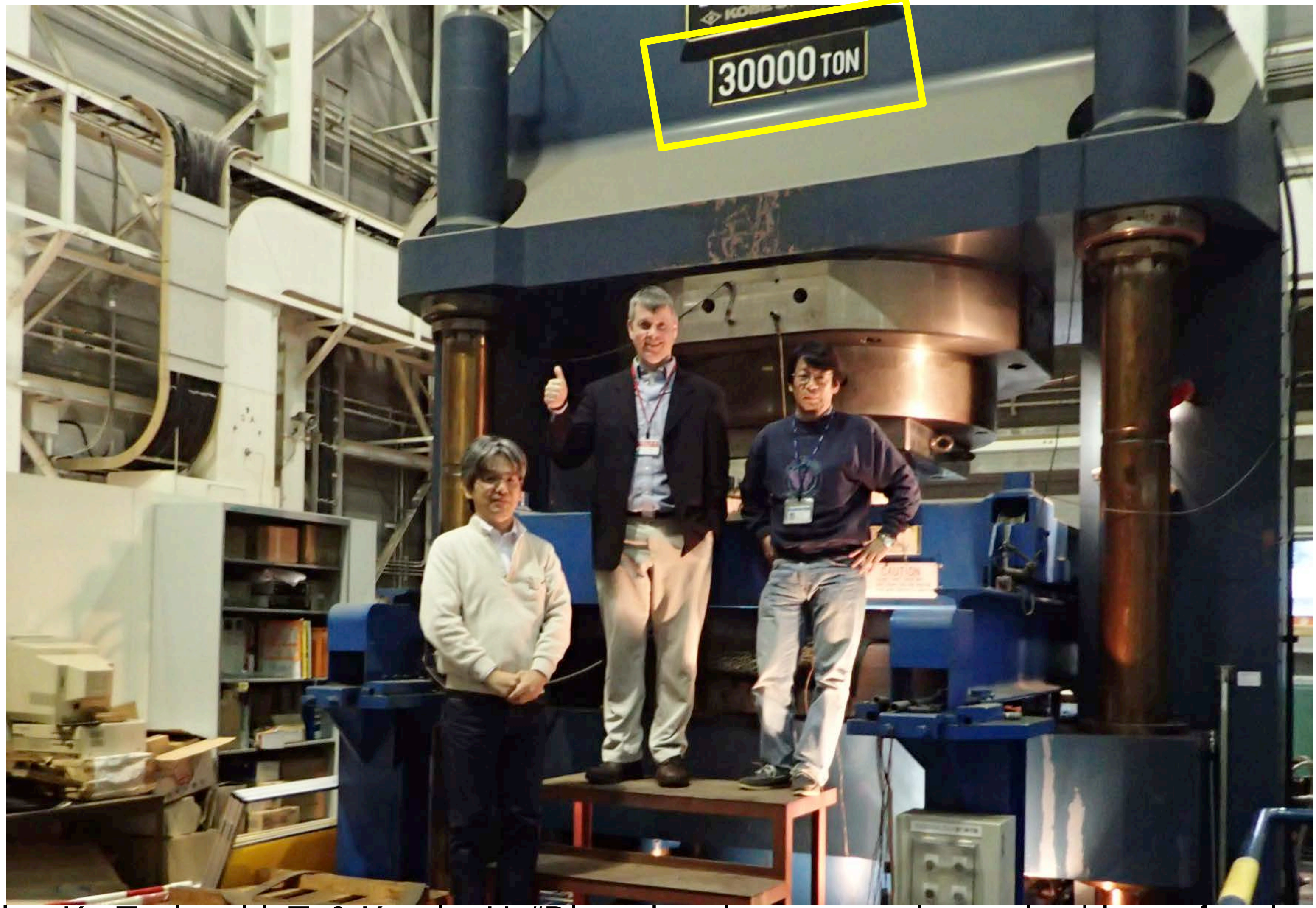
Boron Nitride

- < 2% lattice mismatch to graphene
- atomically flat
- chemically inert, stable to high temp.
- no dangling bonds
- good dielectric properties

Optical properties of h-BN

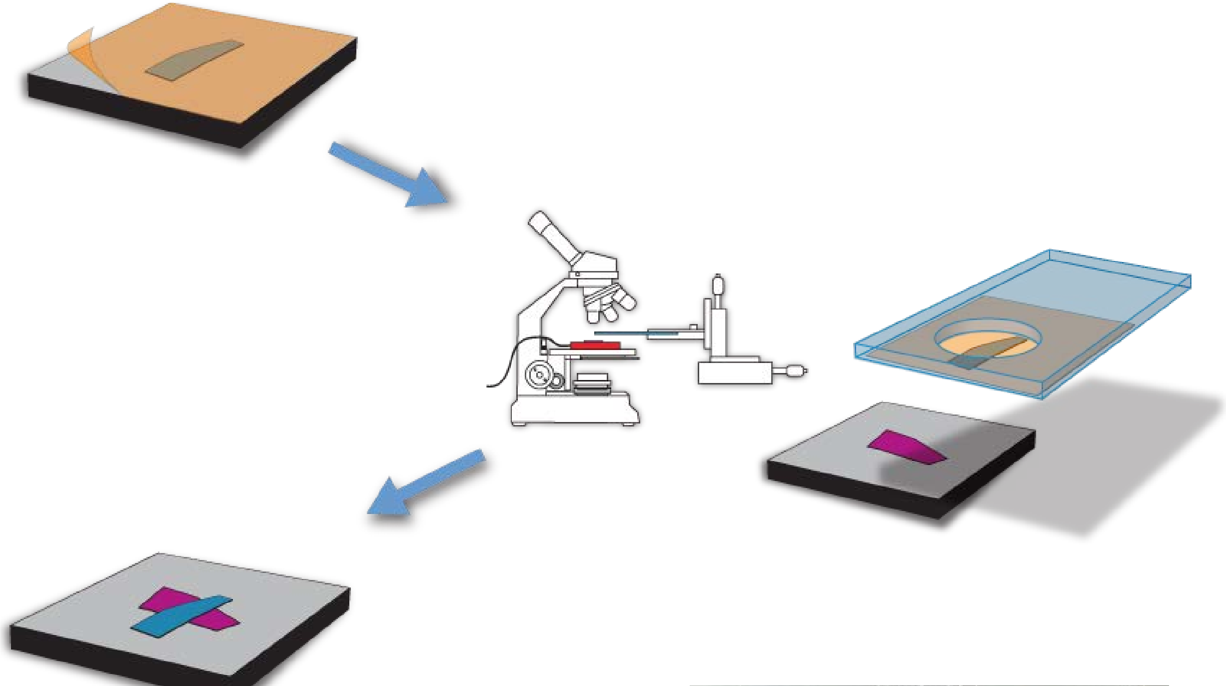
	Band Gap	Dielectric Constant	Optical Phonon Energy
BN	5.8 eV	~3.5	>100 meV
SiO2	8.9 eV	3.9	59 meV

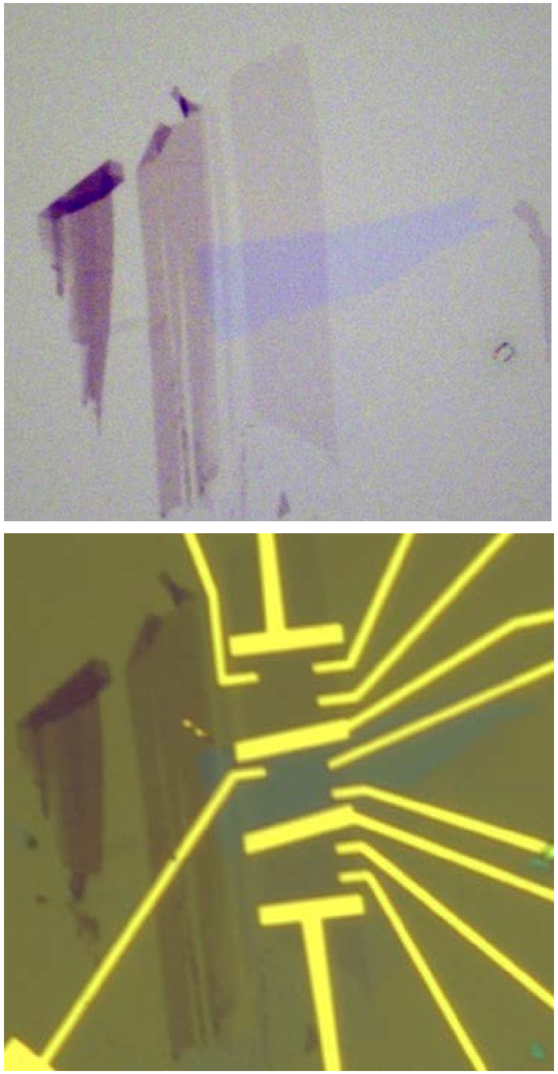
The Source…



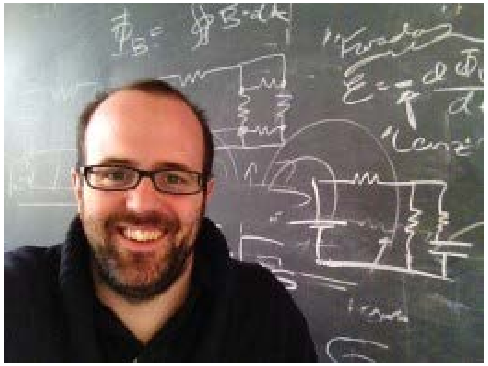
Watanabe, K., Taniguchi, T. & Kanda, H. “Direct-bandgap properties and evidence for ultraviolet lasing of hexagonal boron nitride single crystal.” *Nature Mater*. **3**, 404–409 (2004).
Taniguchi, T. & Watanabe, K. “Synthesis of high-purity boron nitride single crystals under high pressure by using Ba–Bn solvent.” *J. Cryst. Growth* **303**, 525–529 (2007).

‘Polymer transfer’ of graphene onto BN



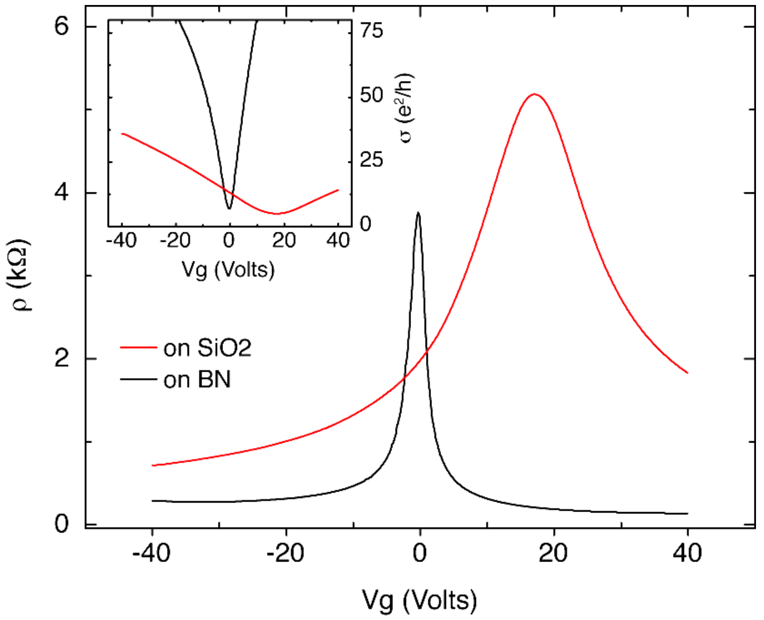


C.R. Dean, A.F. Young, I. Meric, C. Lee, L. Wang, S.Sorgenfrei, K. Watanabe, T. Taniguchi, P. Kim, K.L. Shepard, J. Hone, Nature Nano (2010)

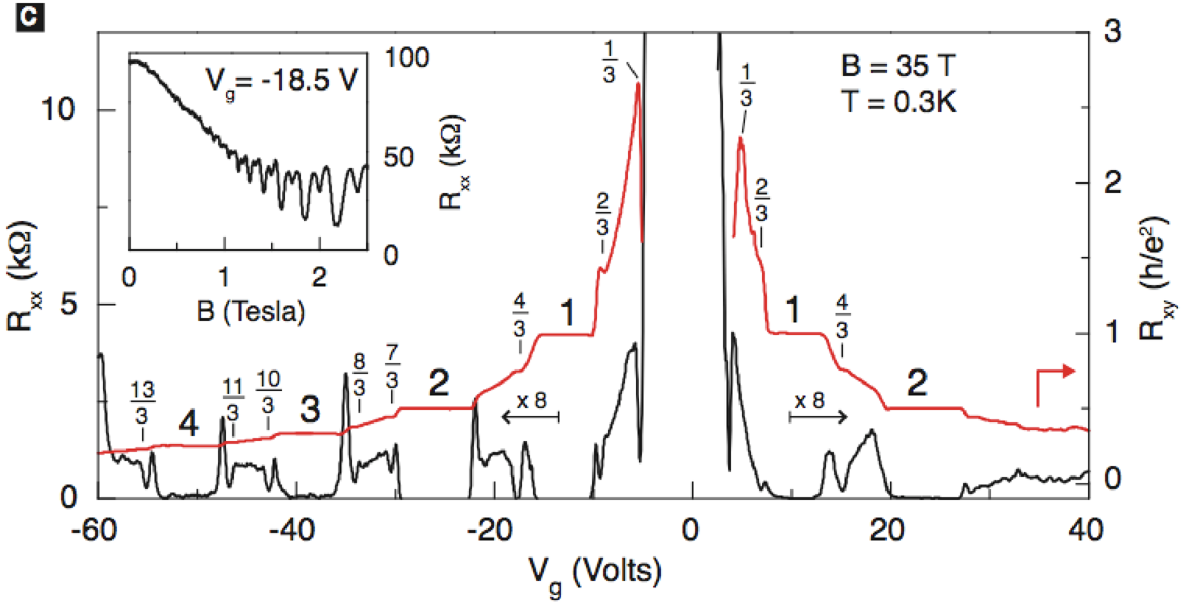


Improved Performance

- Enhanced mobility
- lower disorder

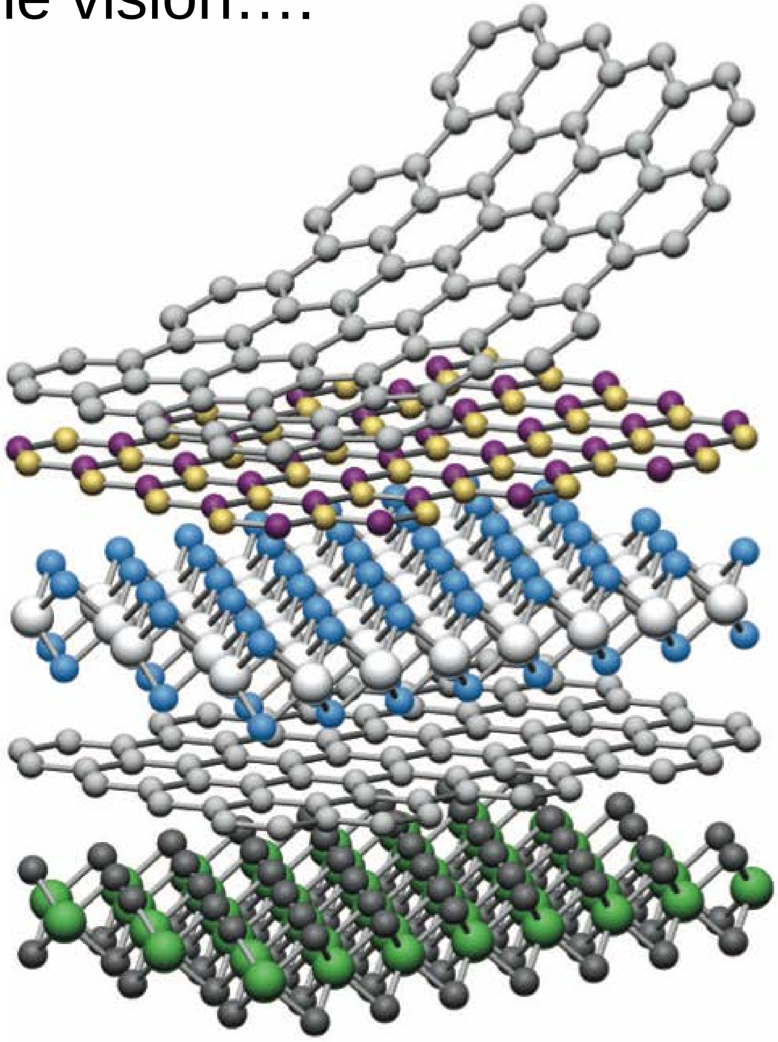


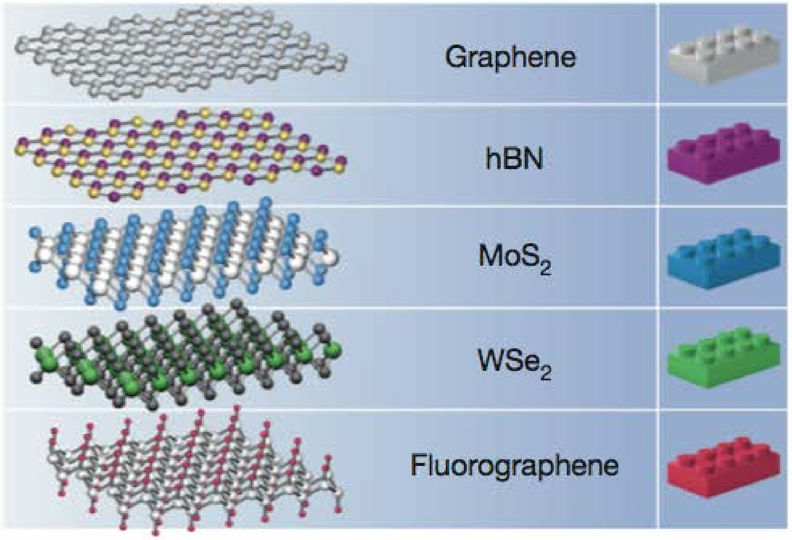
- Many FQHE states

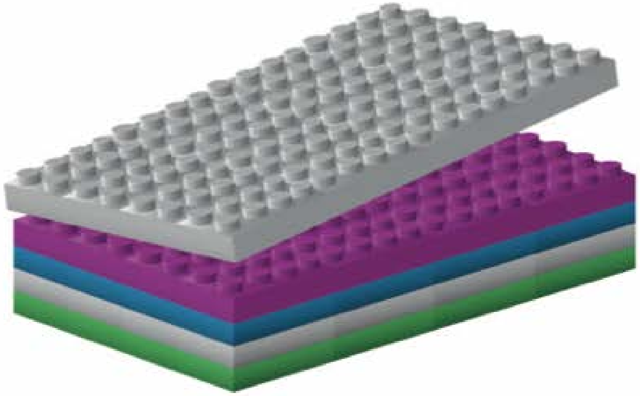


Making Layered Structures

The vision….



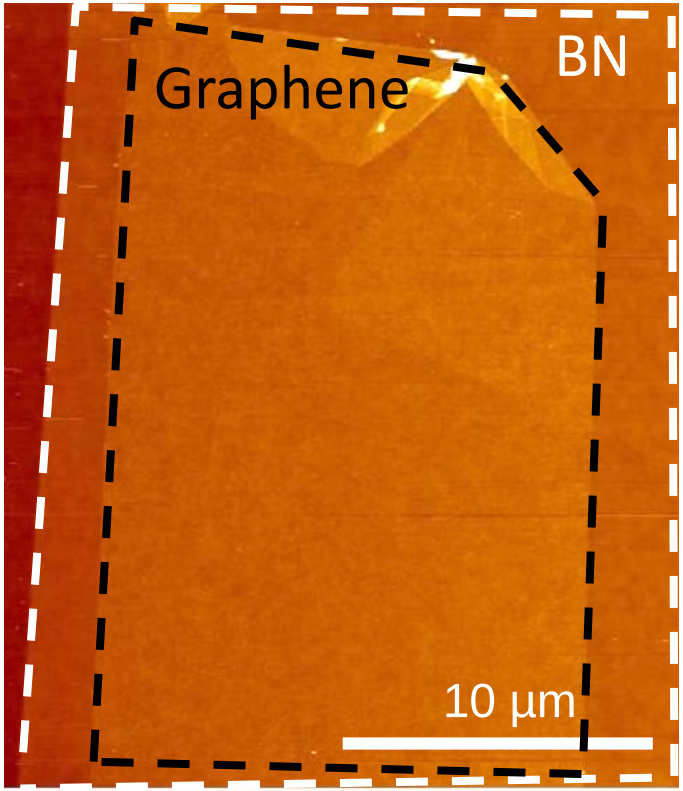




Geim, Nature 2013

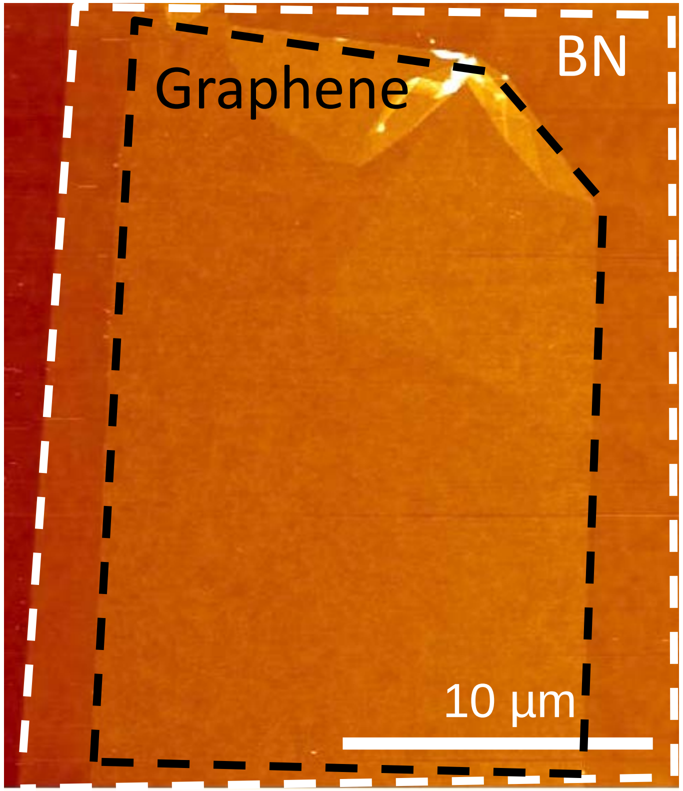
Making Layered Structures

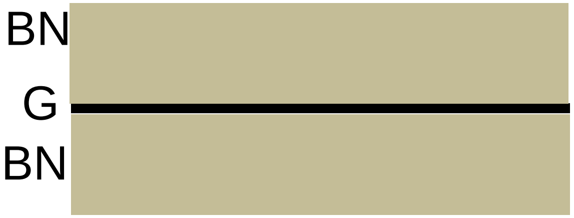
The reality…



Making Layered Structures

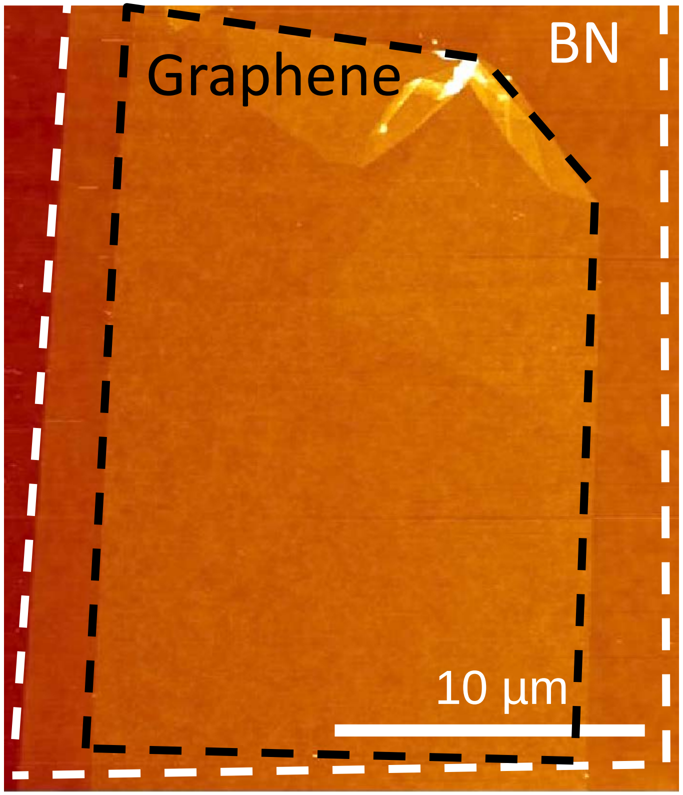
The reality…

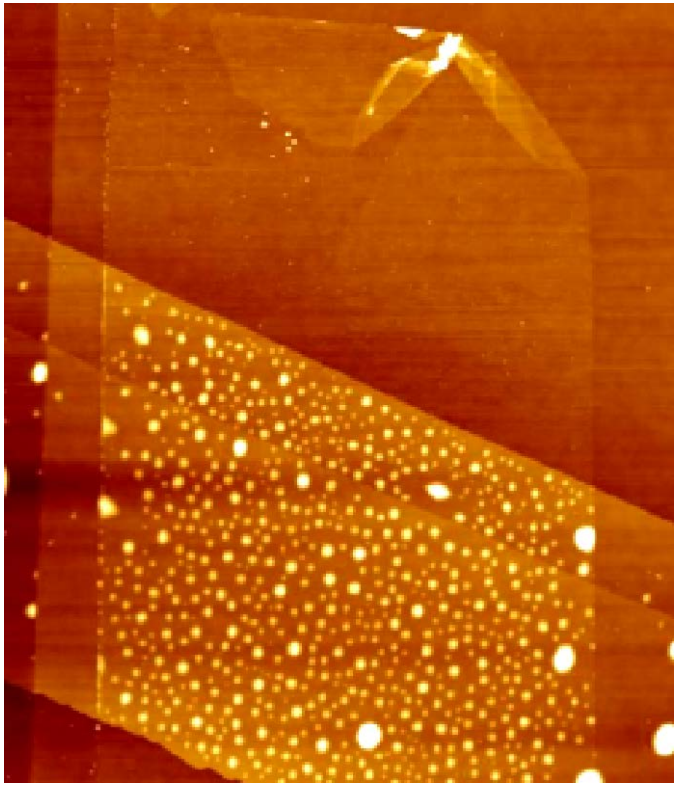


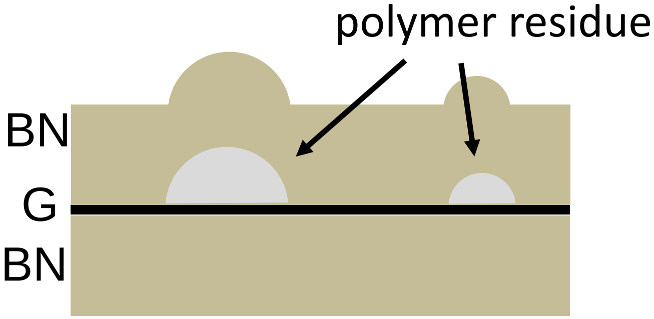


Making Layered Structures

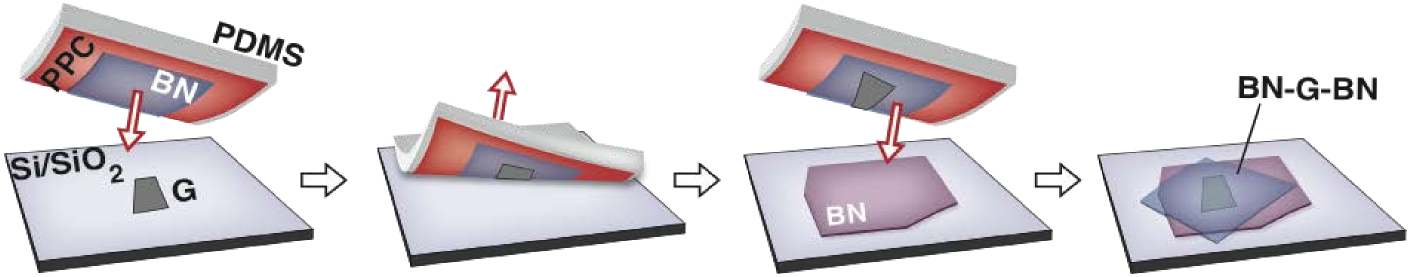
The reality…

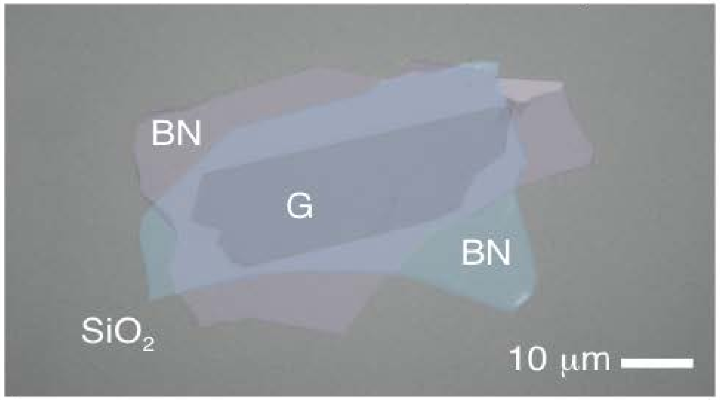


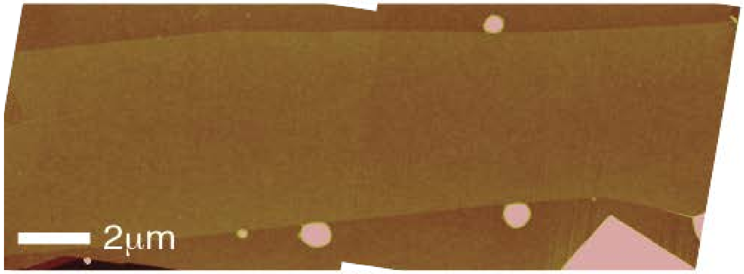


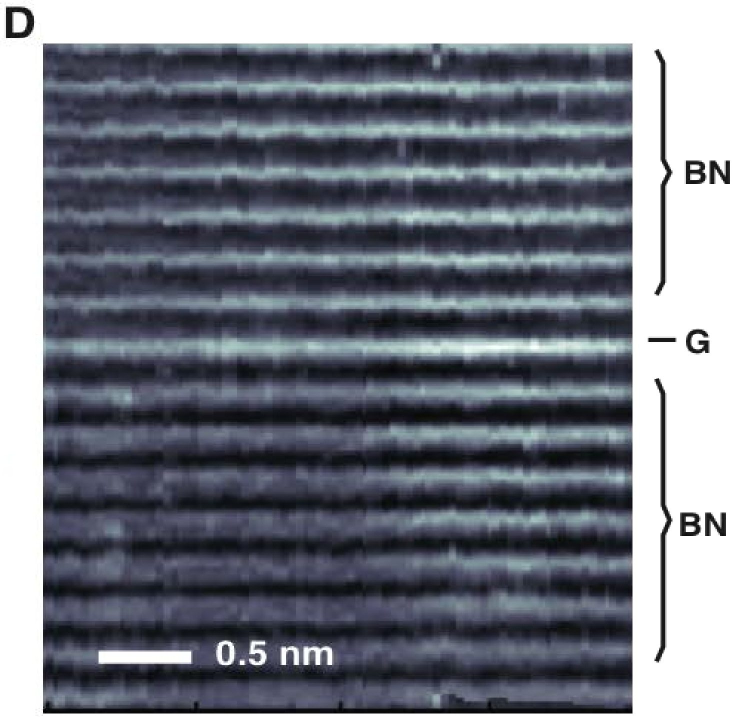


Van der Waals Assembly





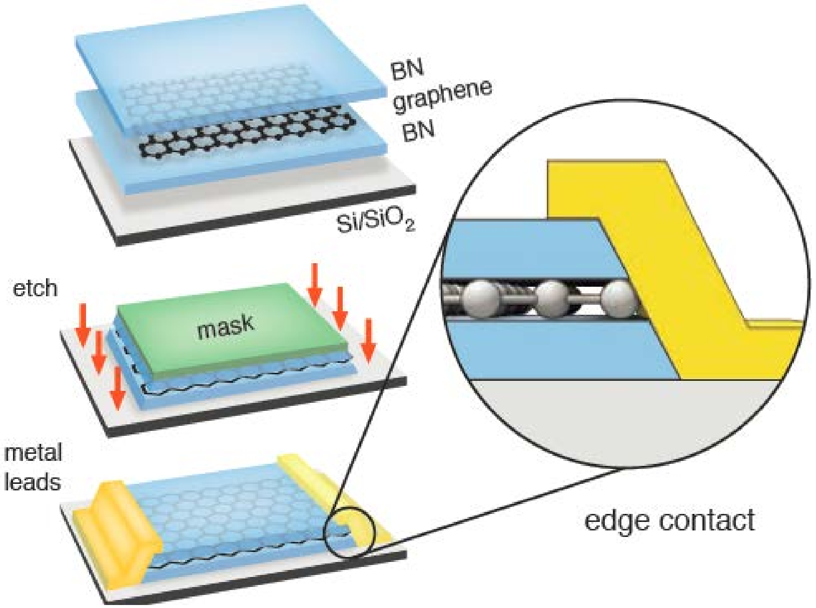


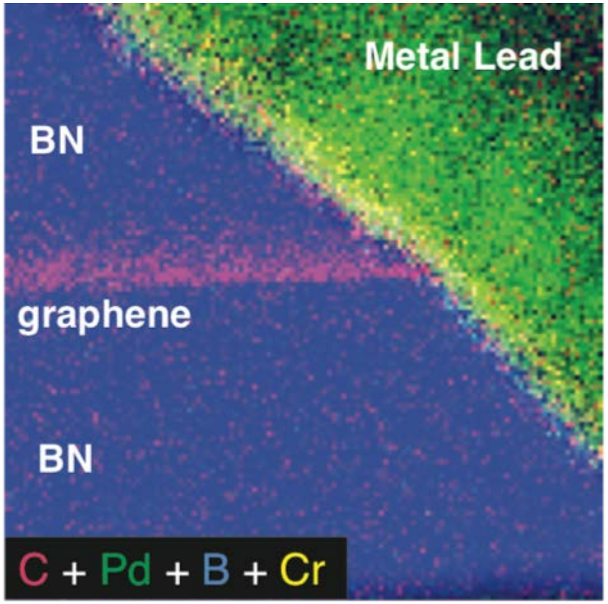


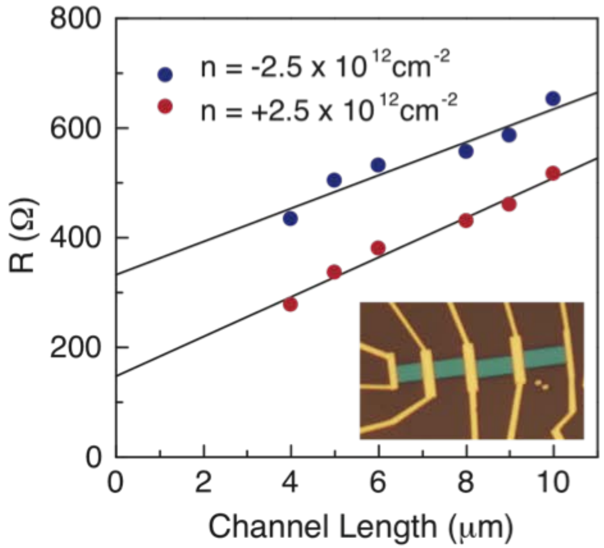
L. Wang et al, Science (2013)

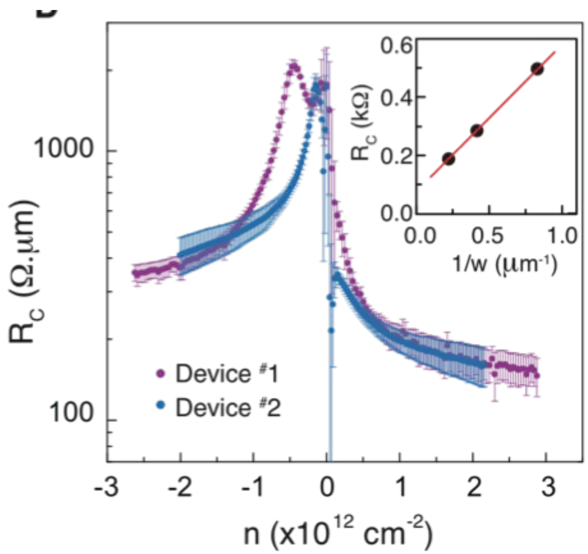
Ultraclean technique – graphene never exposed to polymer

Side Contacts

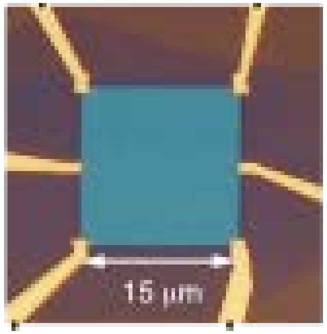


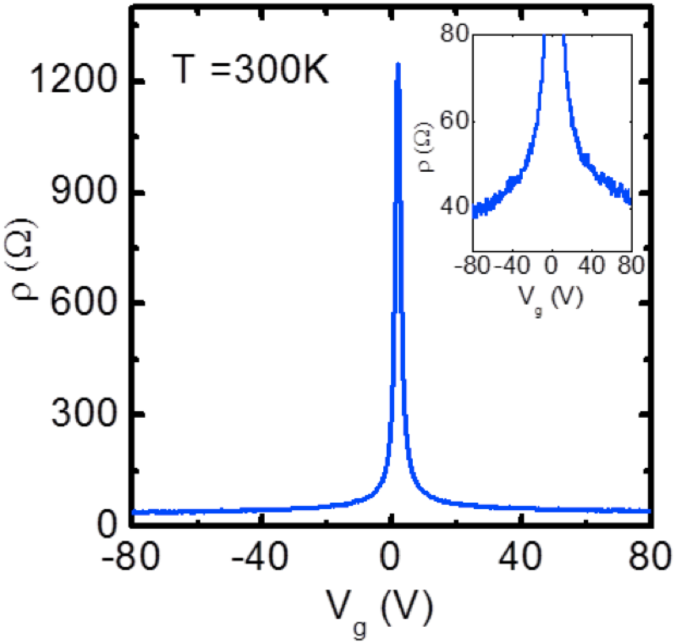


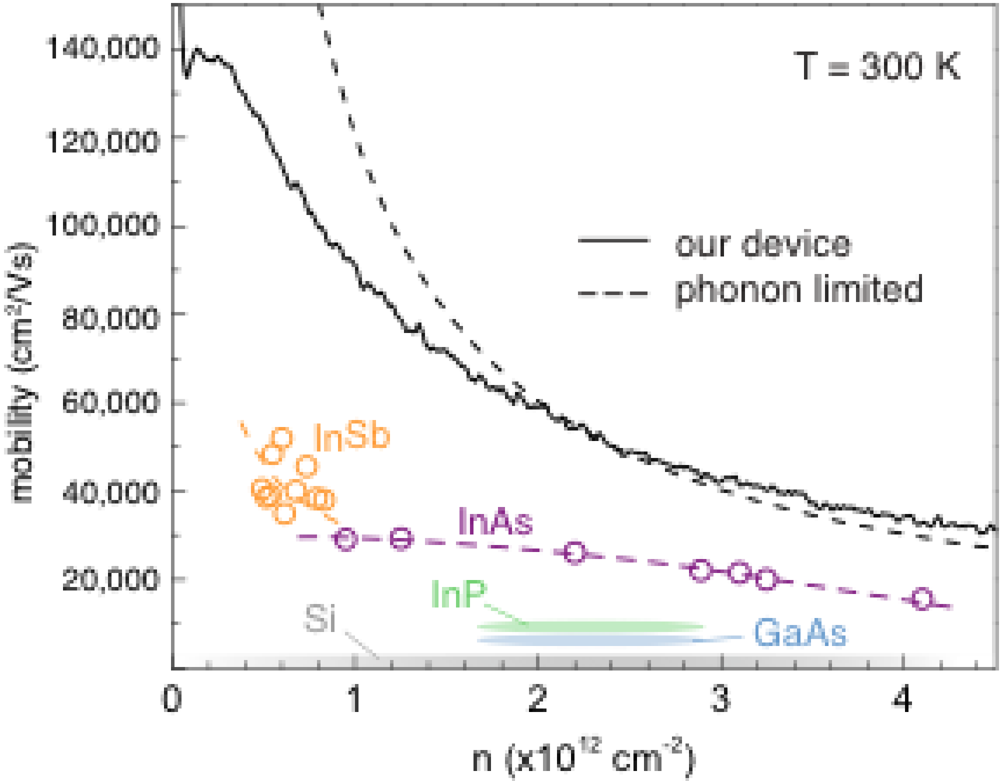




Achieving ideal electrical performance



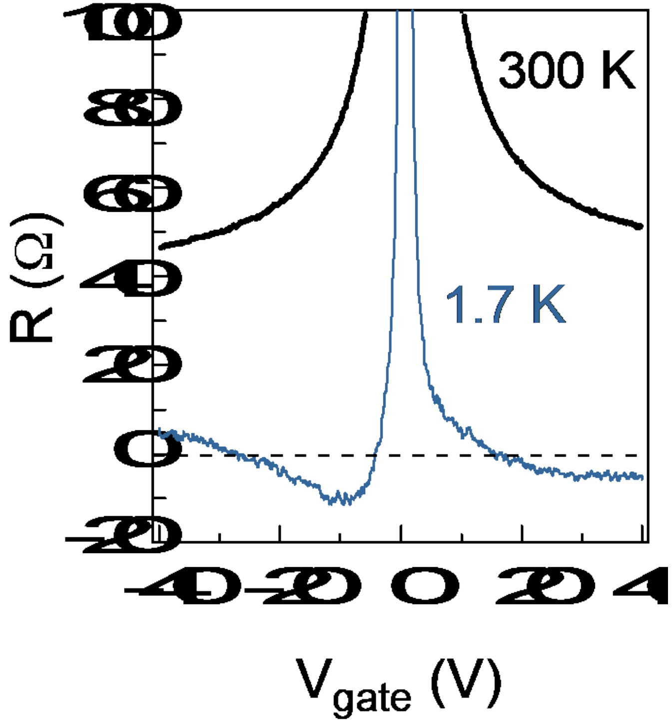


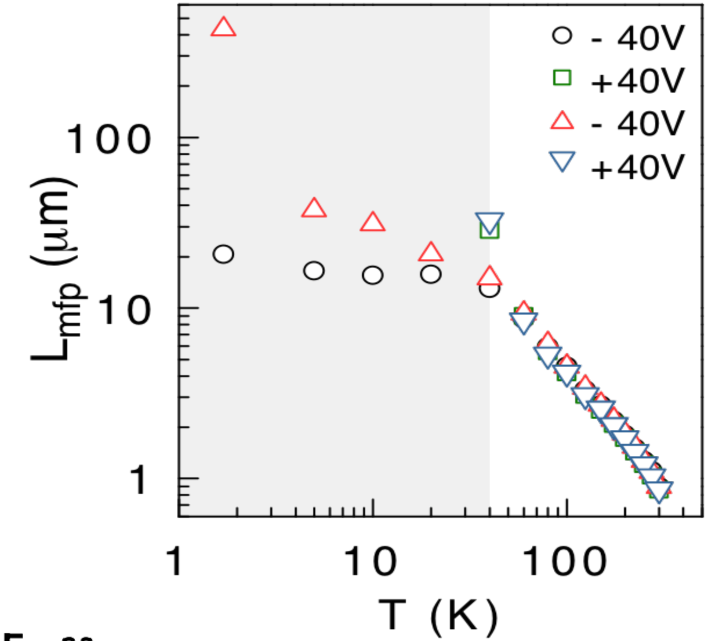


Sheet resistance below 40 Ω – more conductive than silver or copper!

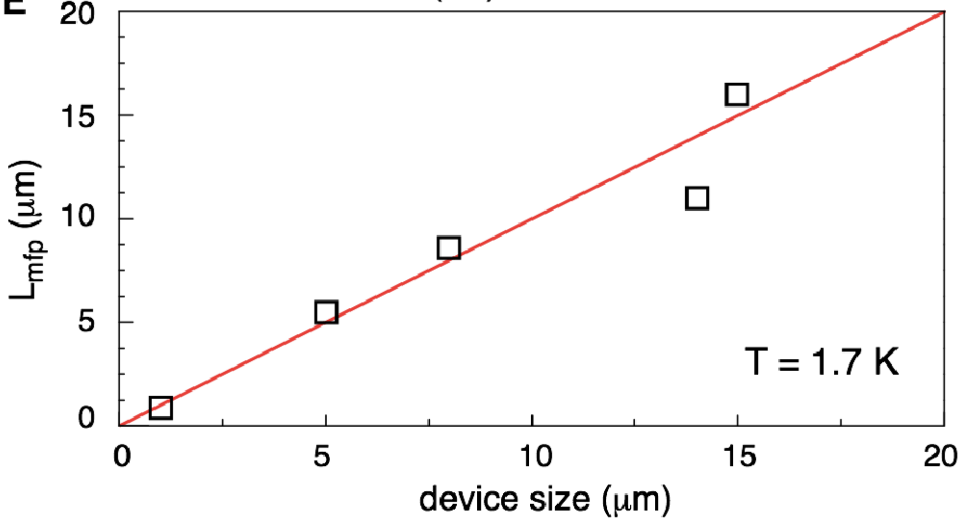
At limit of acoustic phonon scattering. Highest R.T. mobility of any material!

Low-T Ballistic Transport





$$l_{mfp} = \frac{\sigma h}{2e^2 k_f}, \qquad k_f = \sqrt{\pi n}$$



Transport is ballistic at low T up to at least ~15 µm

Ultra-clean graphene: magneto-transport

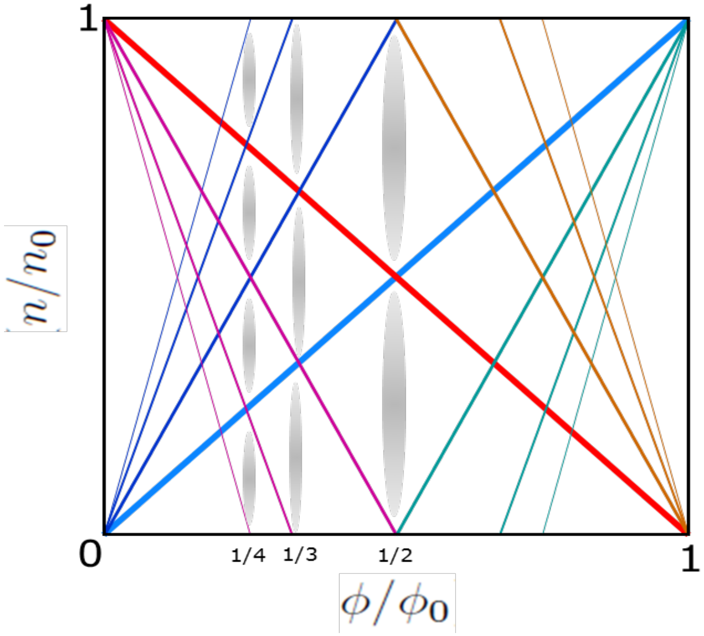
1. ’Hofstader Butterfly’
2. Negative Refraction
3. Exciton Condensation

Hofstadter's Butterfly

Hofstadter, PRB 1976:

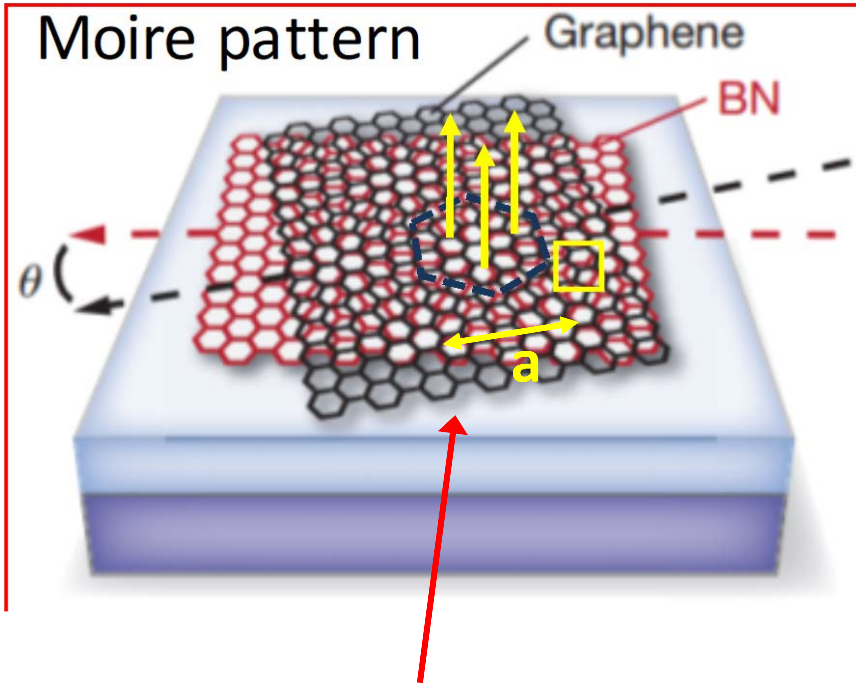
- Basic problem: What are Bloch states in 2D with lattice + magnetic field? (2 length scales)
- Energy bands develop fractal structure when magnetic length is of order the periodic unit cell

Wannier (1977): Fractal gaps trace out straight lines in n vs. flux, with two quantum numbers (s,t)



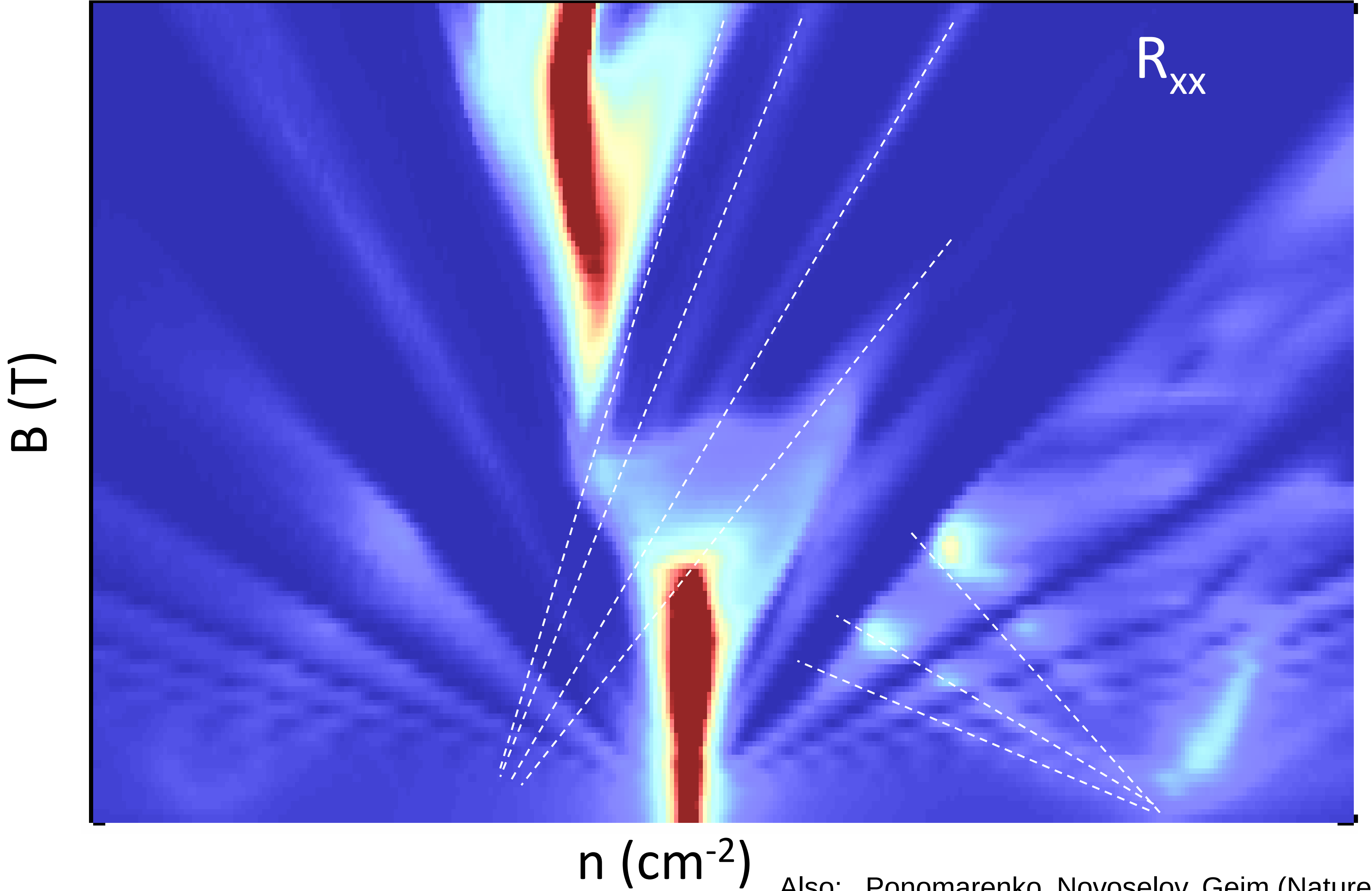
$$\frac{n}{n_o} = t\frac{\phi}{\phi_o} + s, \quad s,t \in I$$

graphene – BN moire' pattern



For well-aligned structure, a ~ 15 nm

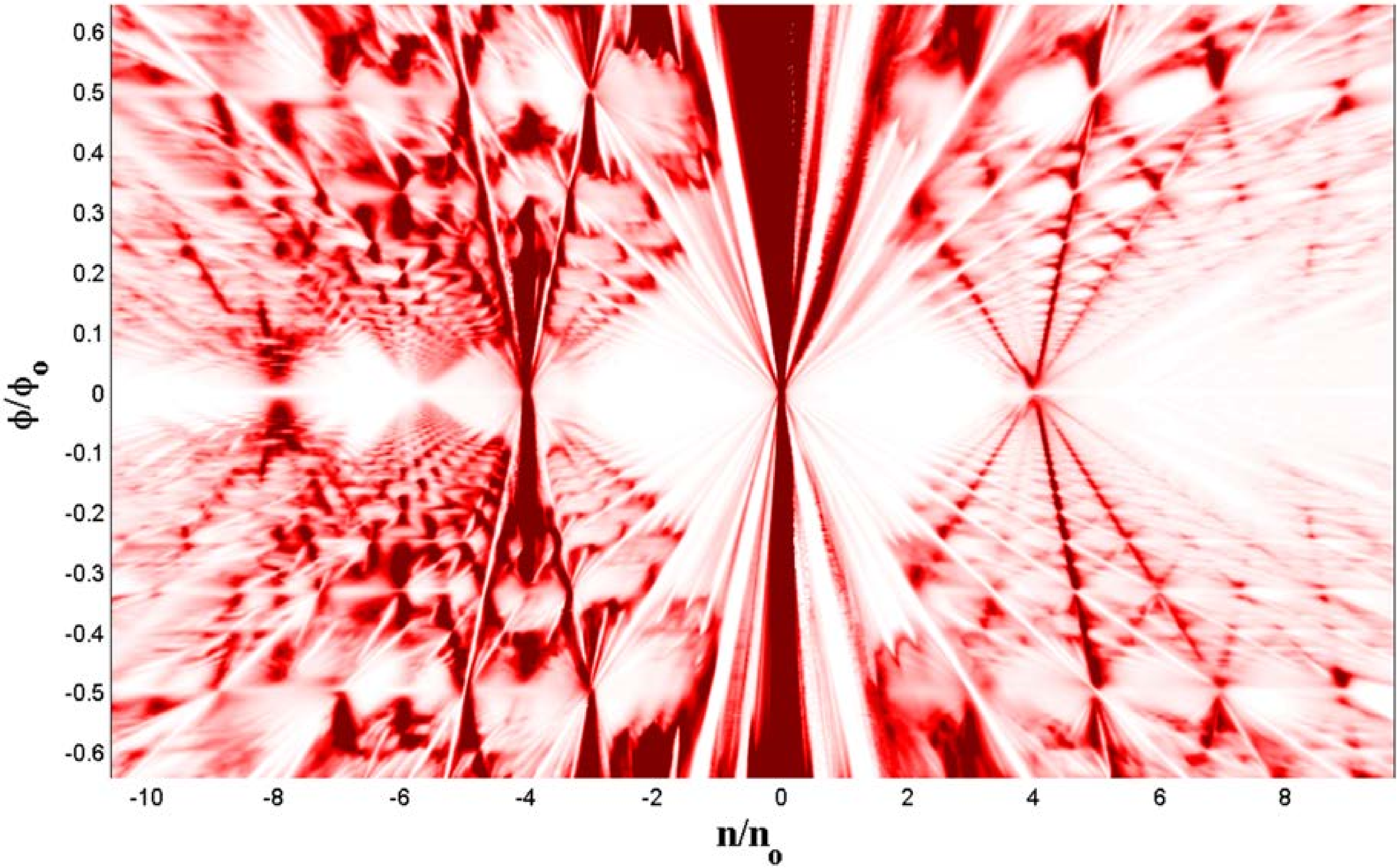
1^{st} generation: polymer transfer



C. R. Dean, L. Wang, …, JH, P. Kim, Nature (2013)

Also: Ponomarenko, Novoselov, Geim (Nature);
Hunt, Jarillo-Herrero (Science)

2nd generation – van der Waals assembly

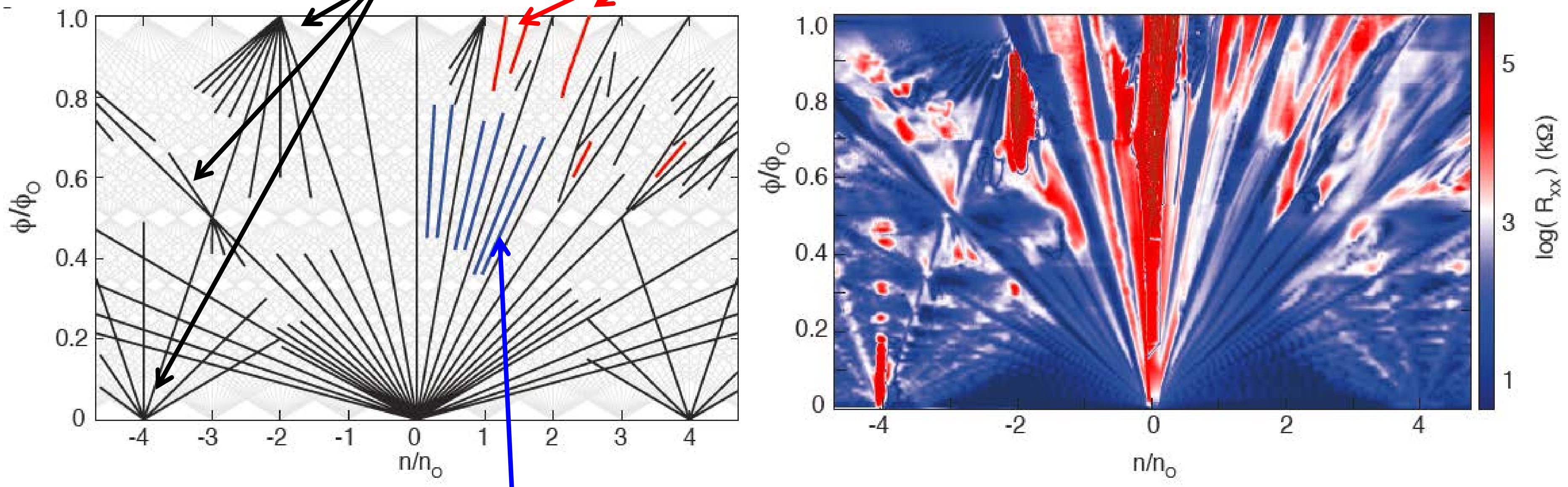


L. Wang, JH, C. Dean, in preparation

States within Hofstader spectrum

New states

Fractal states



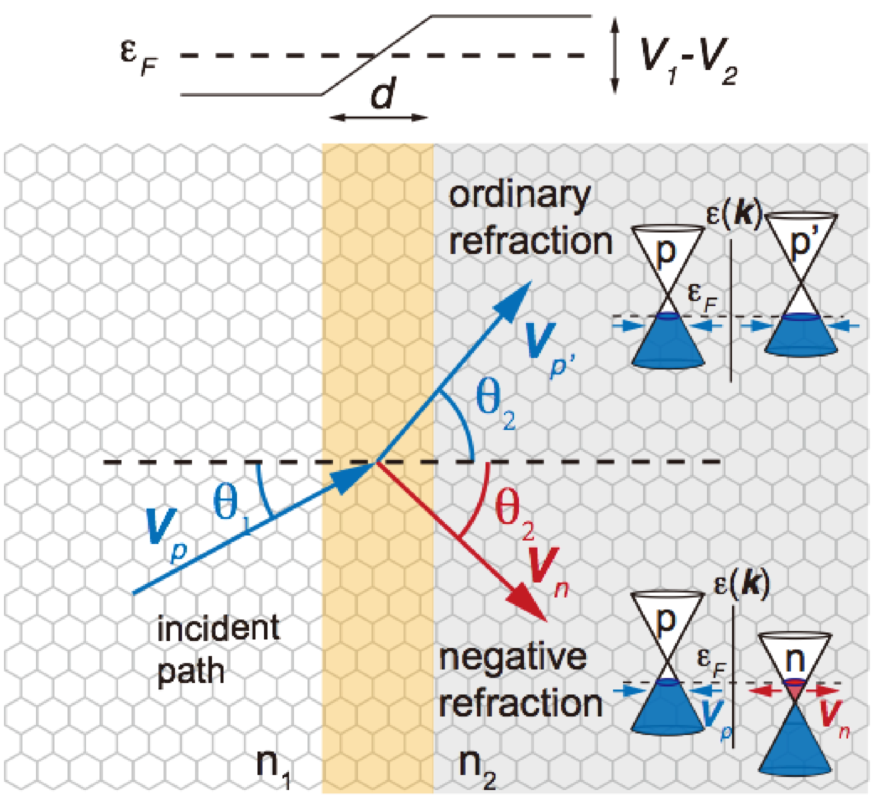
fractional quantum Hall states

$$\frac{n}{n_o} = t\frac{\phi}{\phi_o} + s$$

Fractal states:	integer *t, s*
FQHE:	fractional *t*
New states:	fractional *s*

Wang, L., Gao, Y., Wen, B., Han, Z., Taniguchi, T., Watanabe, K., Koshino, M., Hone, J. & Dean, C. R. *Science* **350,** 1231-1234, (2015).

Using Magnetic Focusing to Study Refraction



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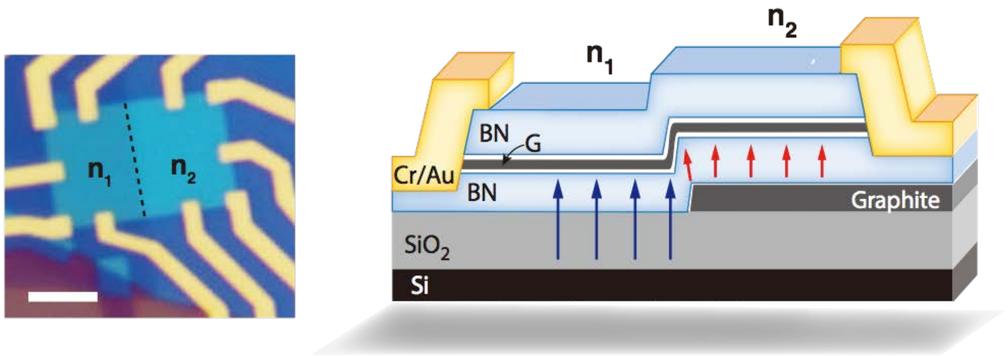
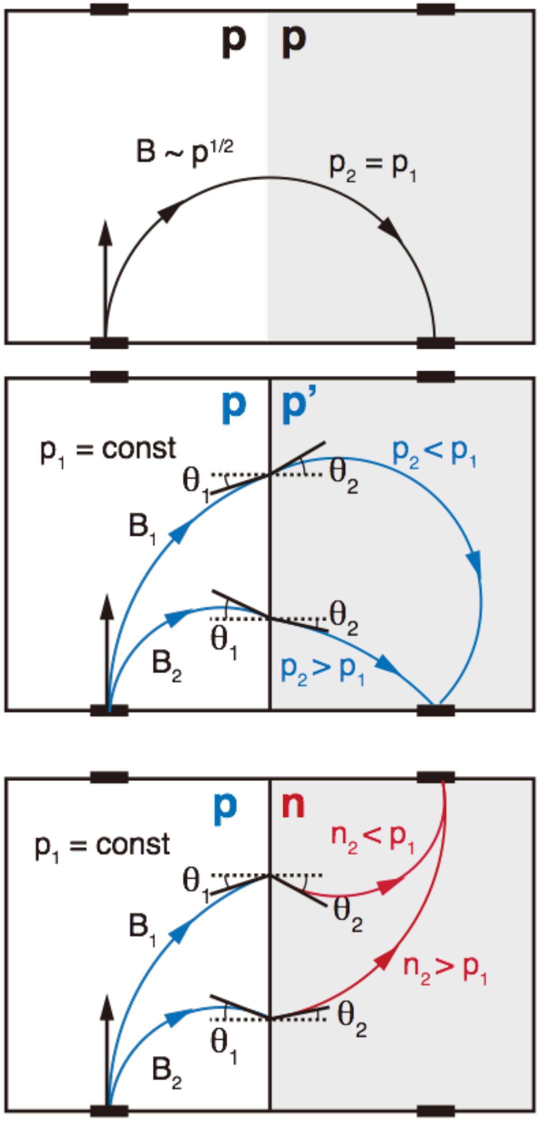
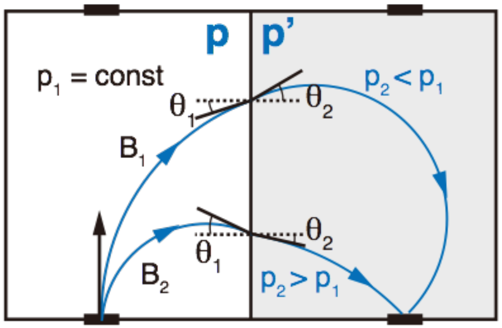
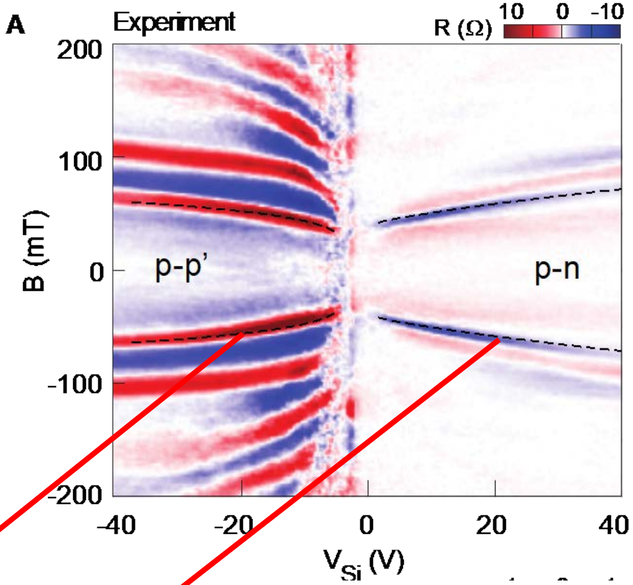


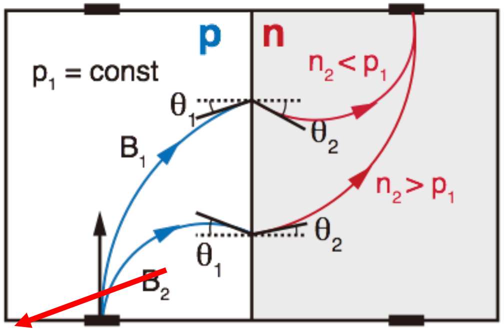


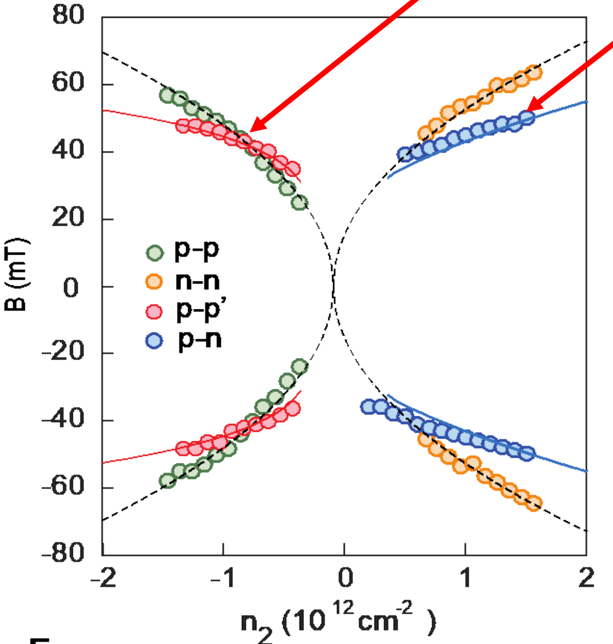
S. Chen… JH, A. Ghosh, C. Dean, *Science* (2017)

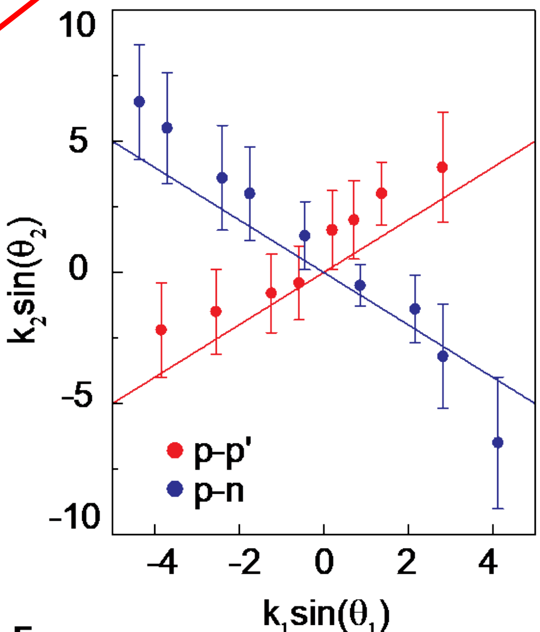
Negative refraction in p-n junctions





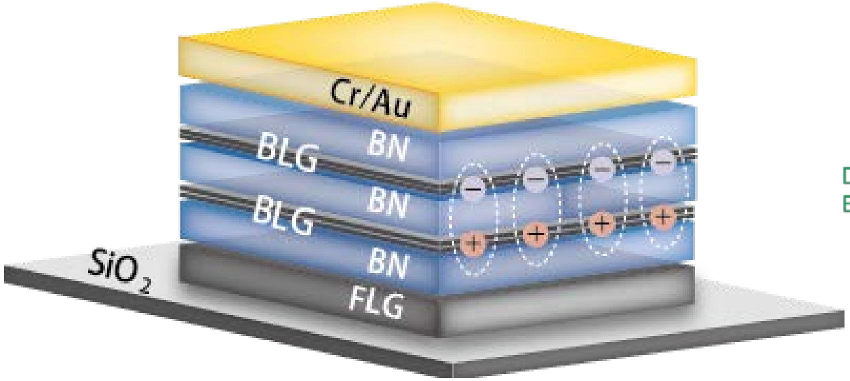


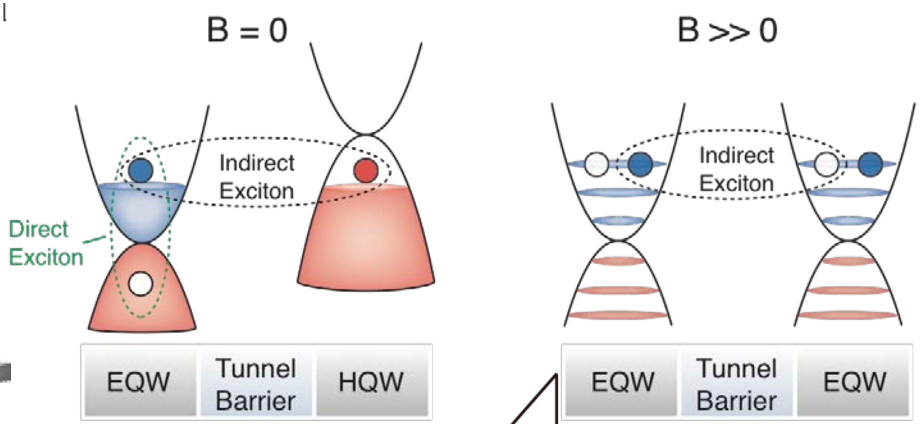


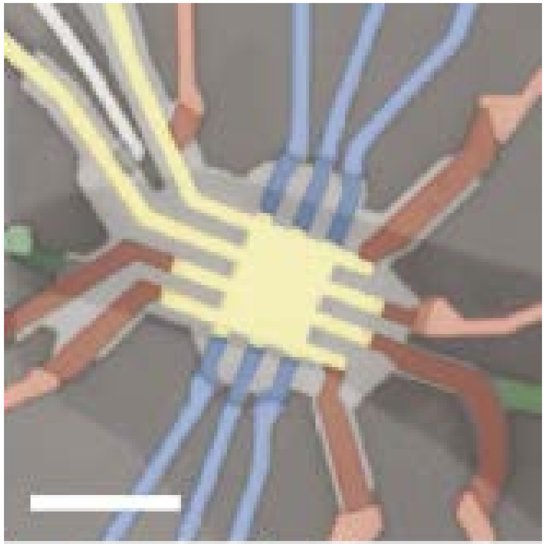


- Simple geometric model fits shape of focusing peaks
- Positive and negative Snell's law confirmed

Exciton Condensation in Bilayer Graphene Superlattices



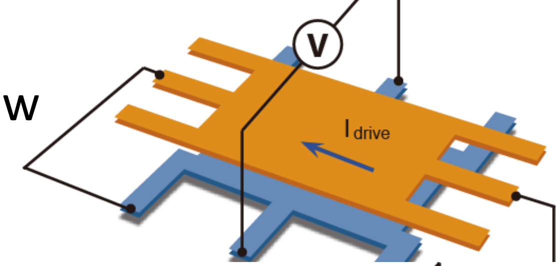




🟥 Top BLG 🟨 Top Gate

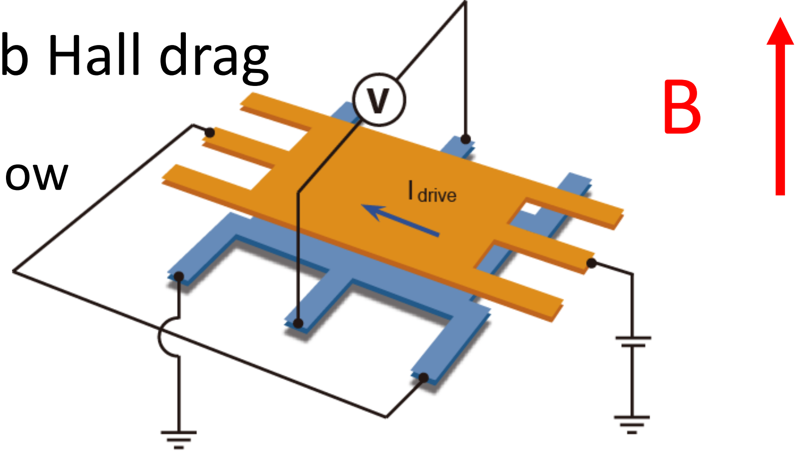
🟦 Bot. BLG 🟩 Bot. Gate

Counterflow



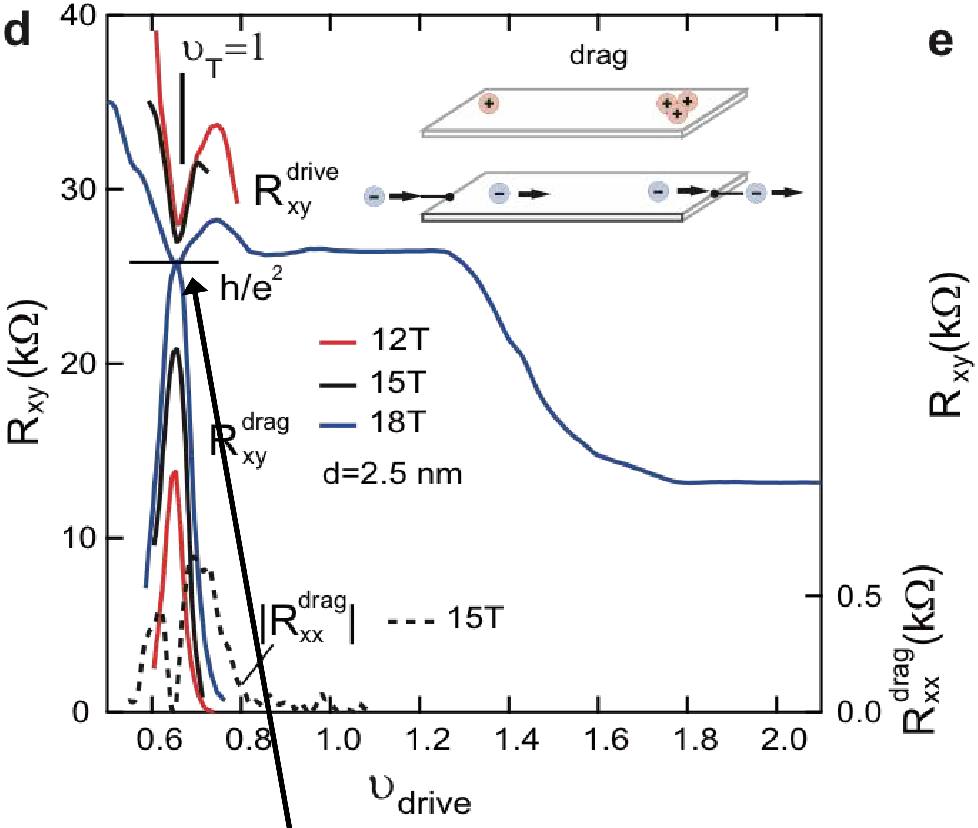
Coulomb Hall drag

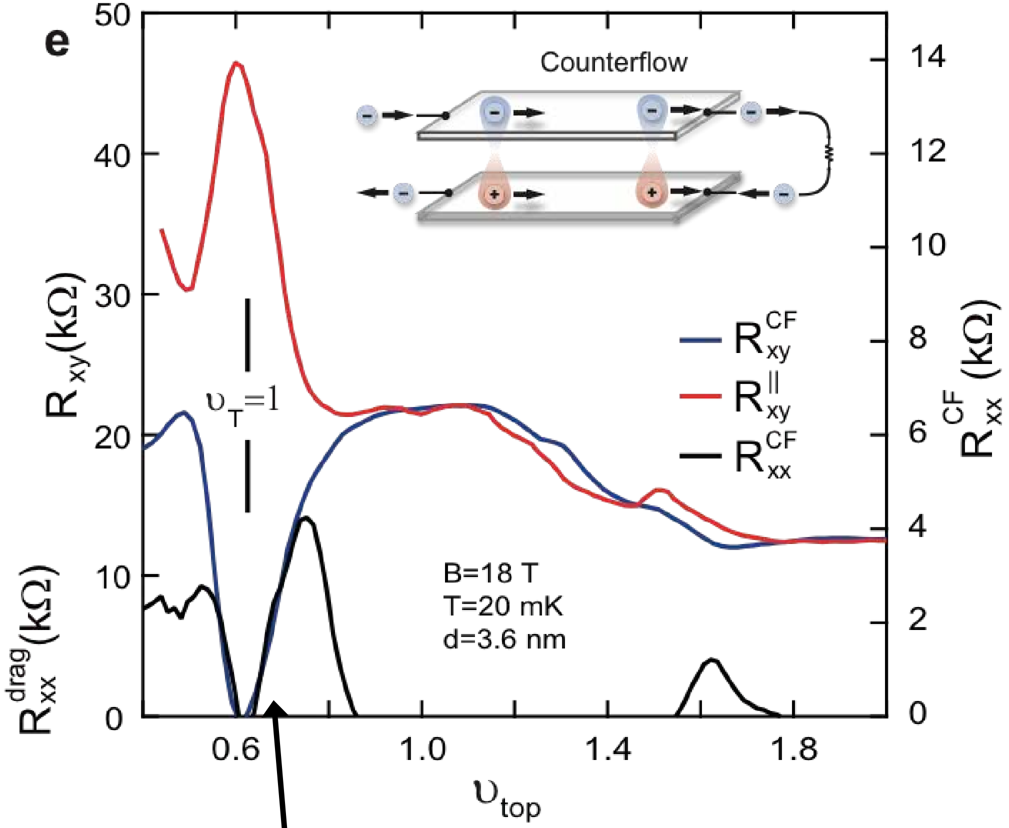
Parallel Flow



J. Li, JH, C. Dean, under review (Nature). Also P. Kim group

Evidence for Condensate phase at $\nu_{tot} = 1$





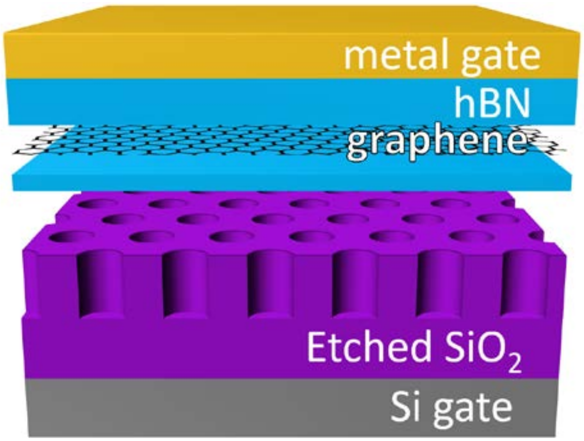
At ν_T = 1, drive and drag layer both show quantized Hall resistance

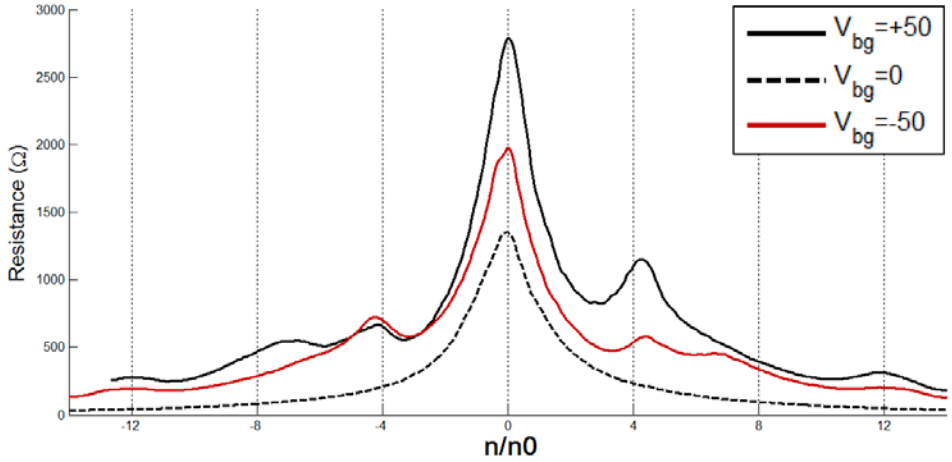
In counterflow geometry, Hall drag = 0 at ν_T = 1

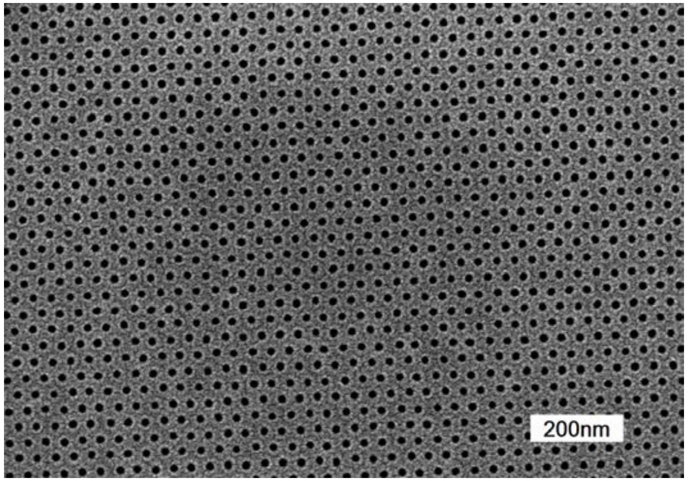
New Control of Graphene Devices

1. Artificial Superlattices
2. Gate-defined structures
3. Interlayer Rotation

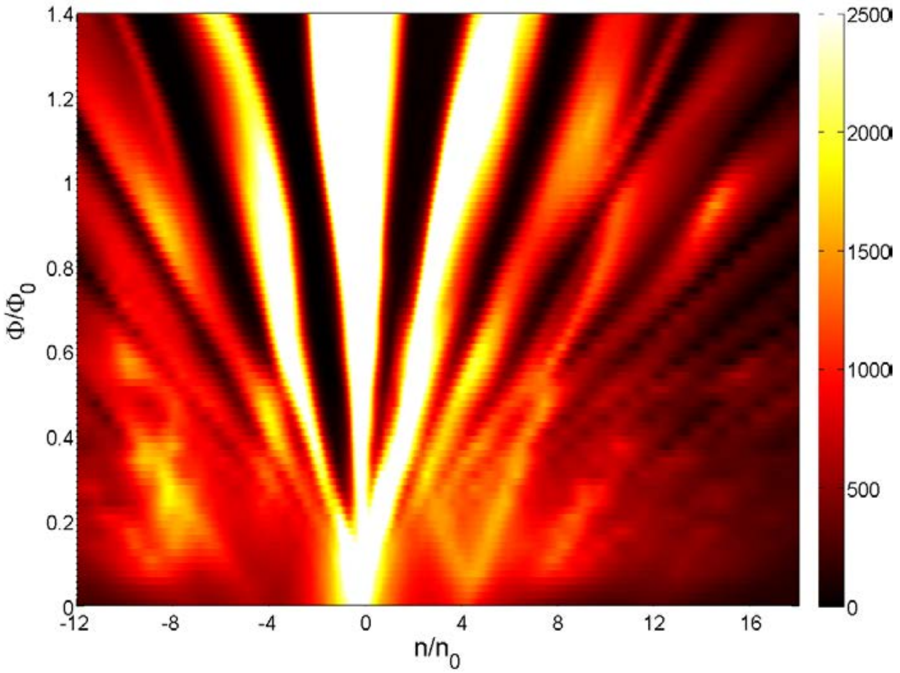
Artificial Superlattices



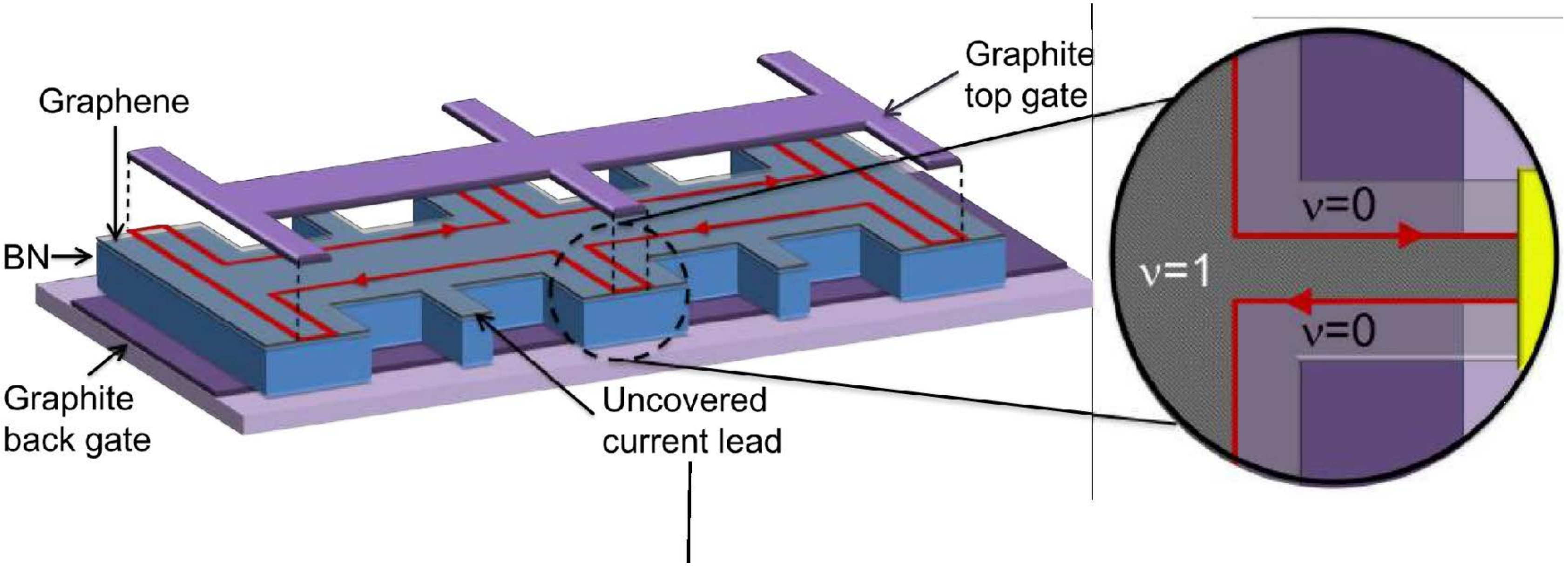




Triangular lattice of holes in SiO$_2$, 40nm periodicity



How can we make graphene even better?

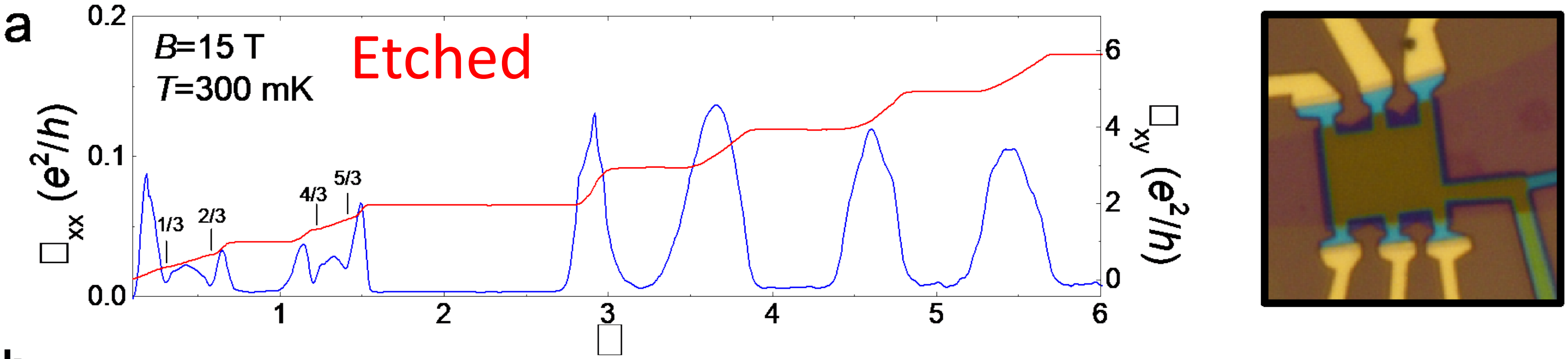


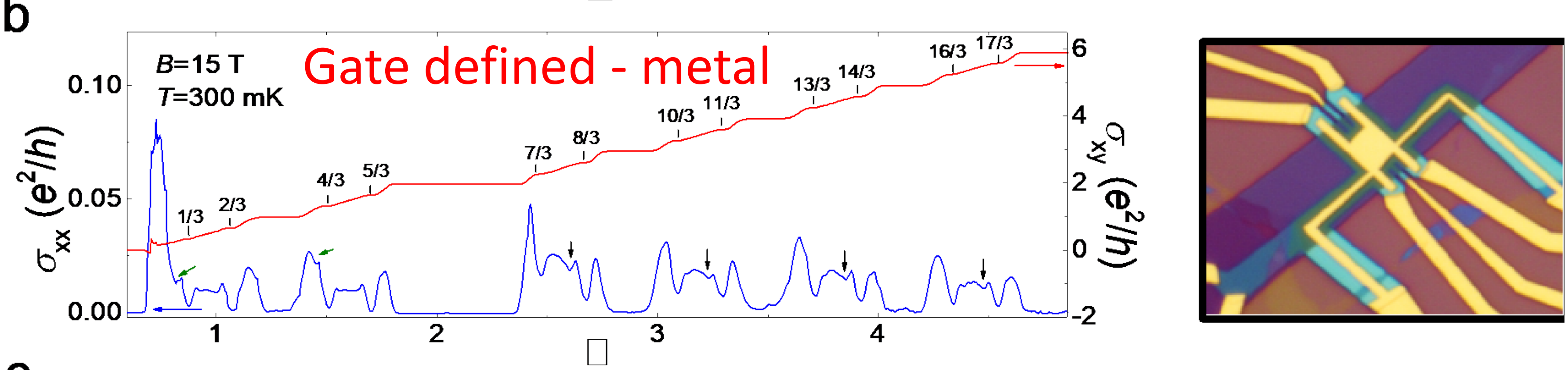
Use graphite as top / bottom gates: better electrostatic uniformity

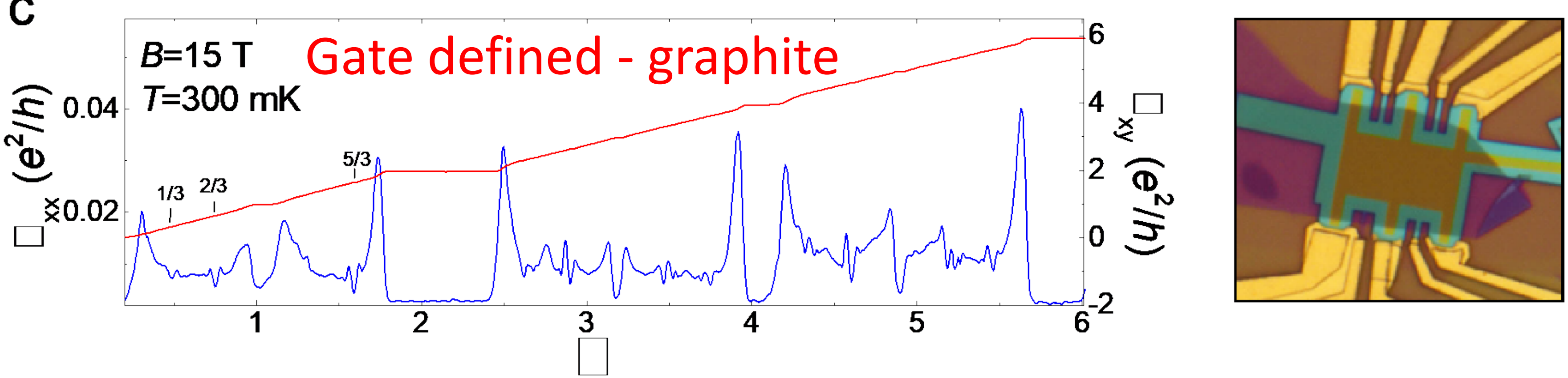
Use top gate to define channel region

    Adjust back gate / Magnetic field to maintain insulating ($\nu=0$) state

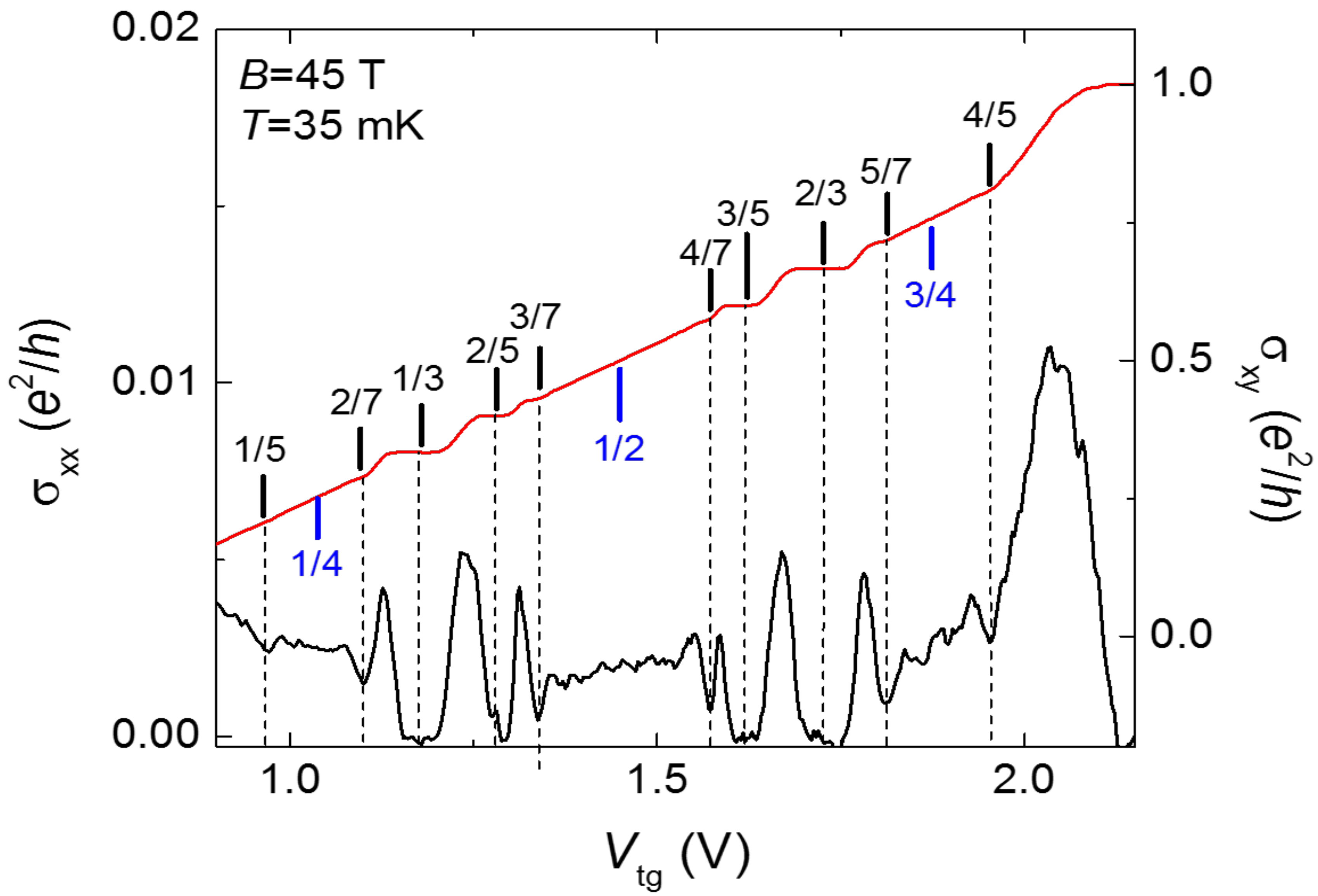
Qualitative Improvement in QHE



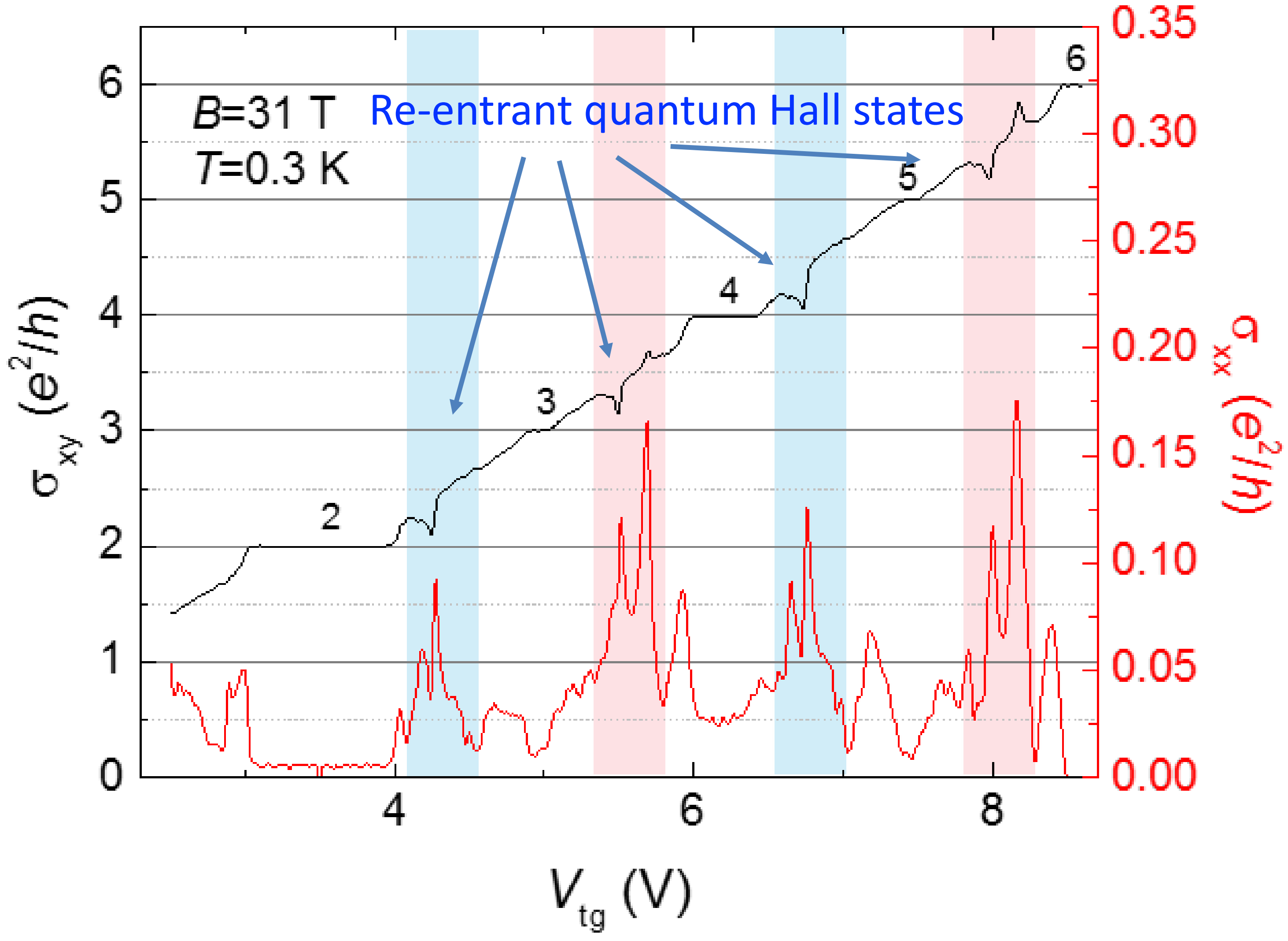




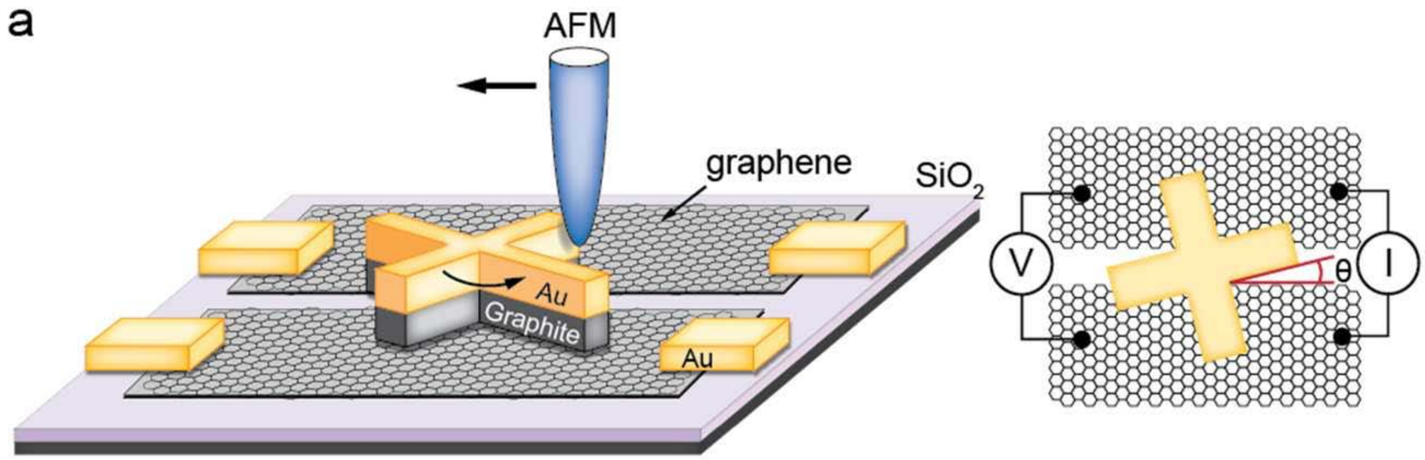
New physics reachable thanks to the gate defined devices

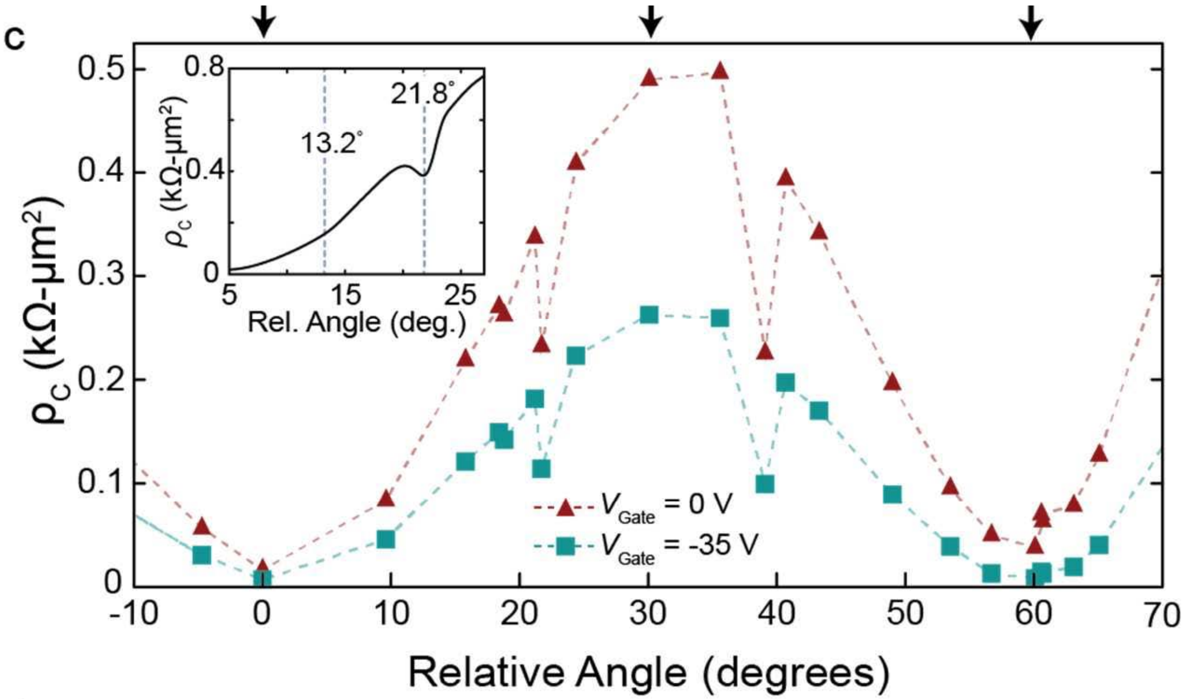


New physics reachable thanks to the gate defined devices



Controlling Interlayer Rotation



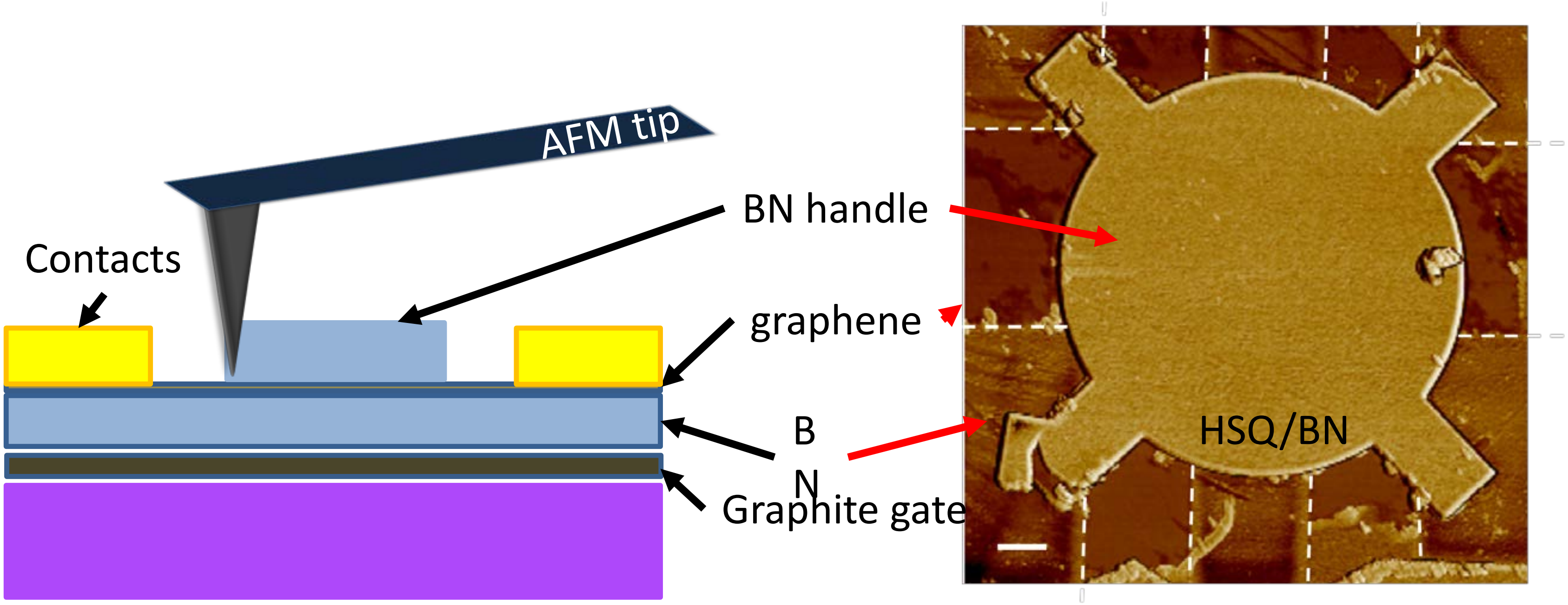


Angle-dependent contact resistance between graphene and graphite

Chari, Ribero, Dean, Shepard, Nano Lett (2016)

Controlling Interlayer Rotation: Graphene / BN

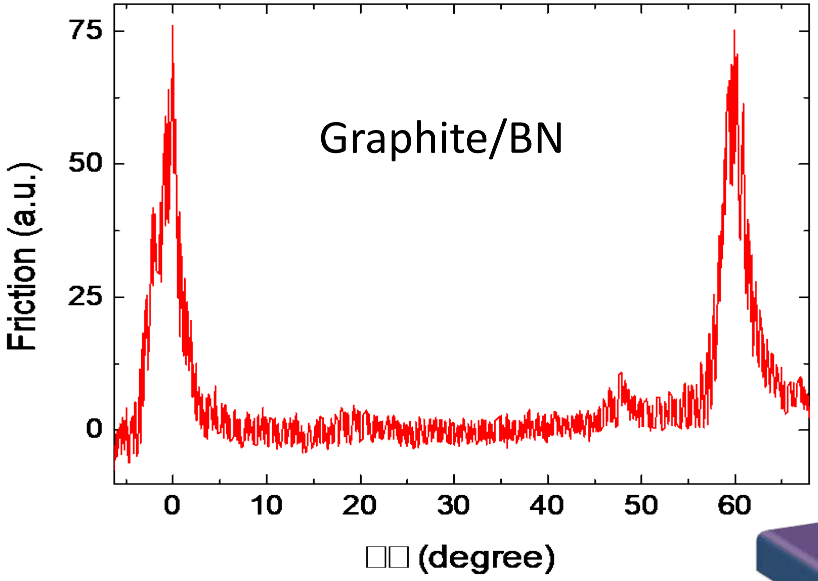
BN on graphene (band structure manipulation)

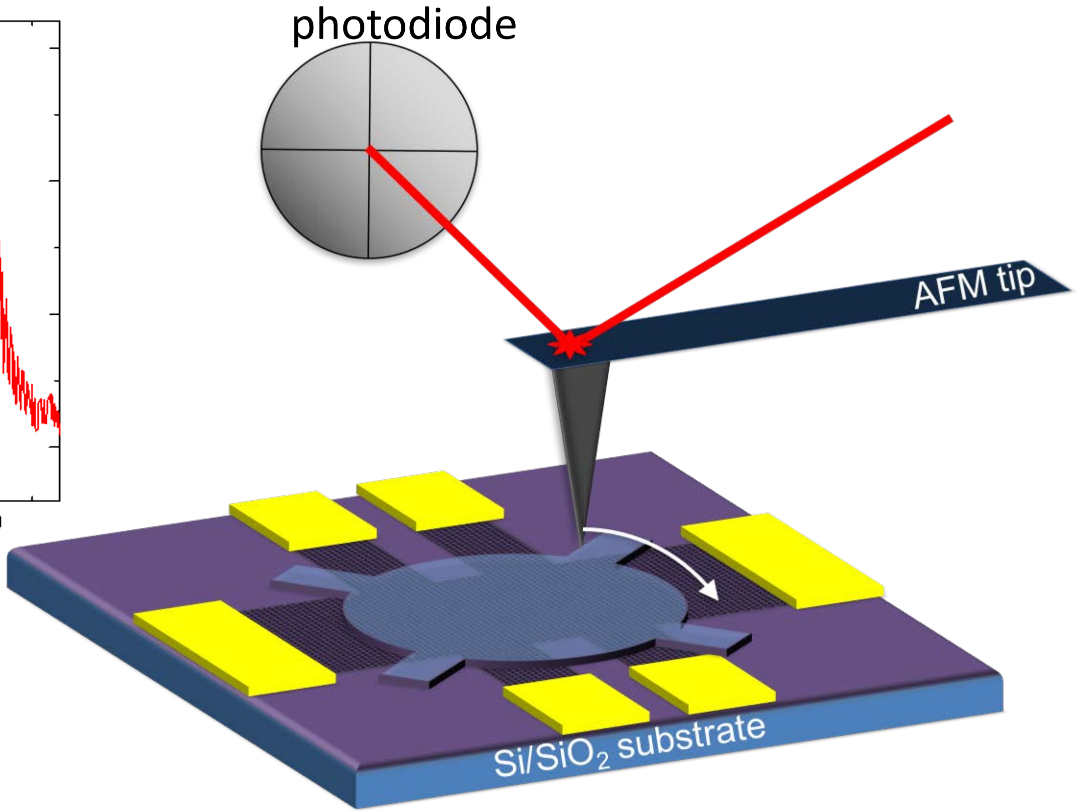


- Shape ‘handle’ structure
- Rotate with AFM
- Measure transport

Interlayer friction

BN on graphene (friction)

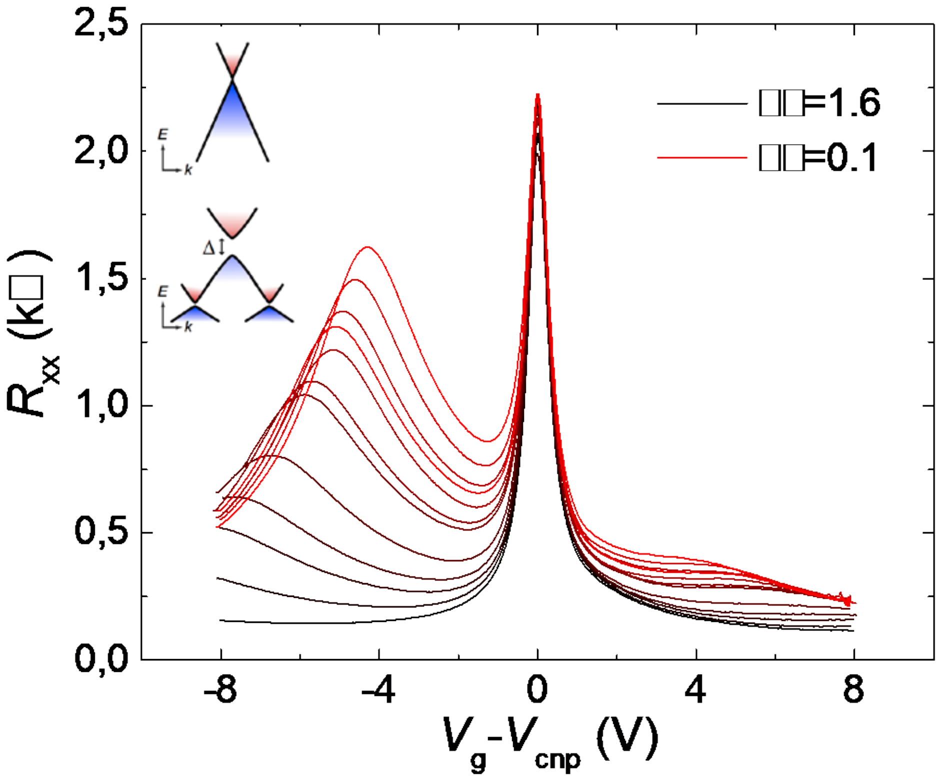


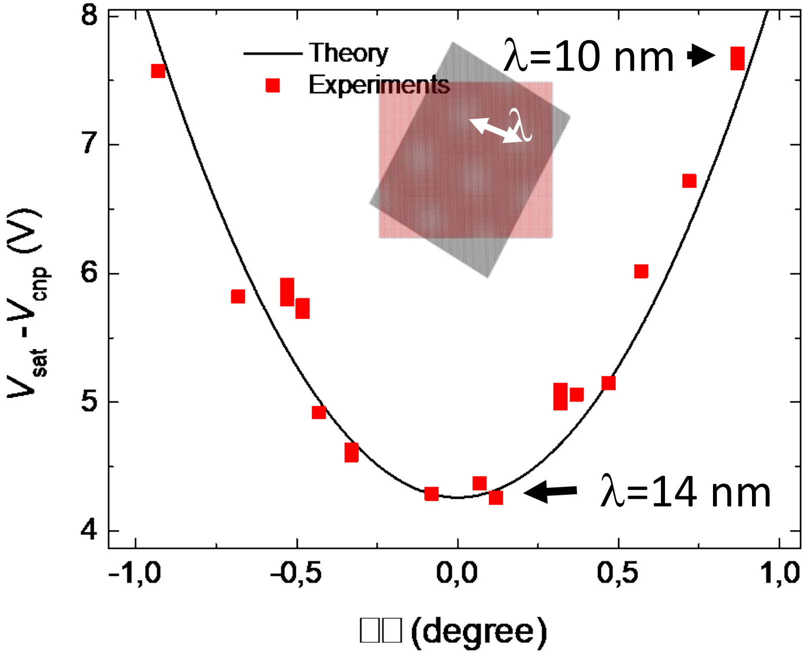


Layers ‘lock’ together at commensurate angles

Controlling Moire’ Wavelength

BN on graphene (band structure manipulation)



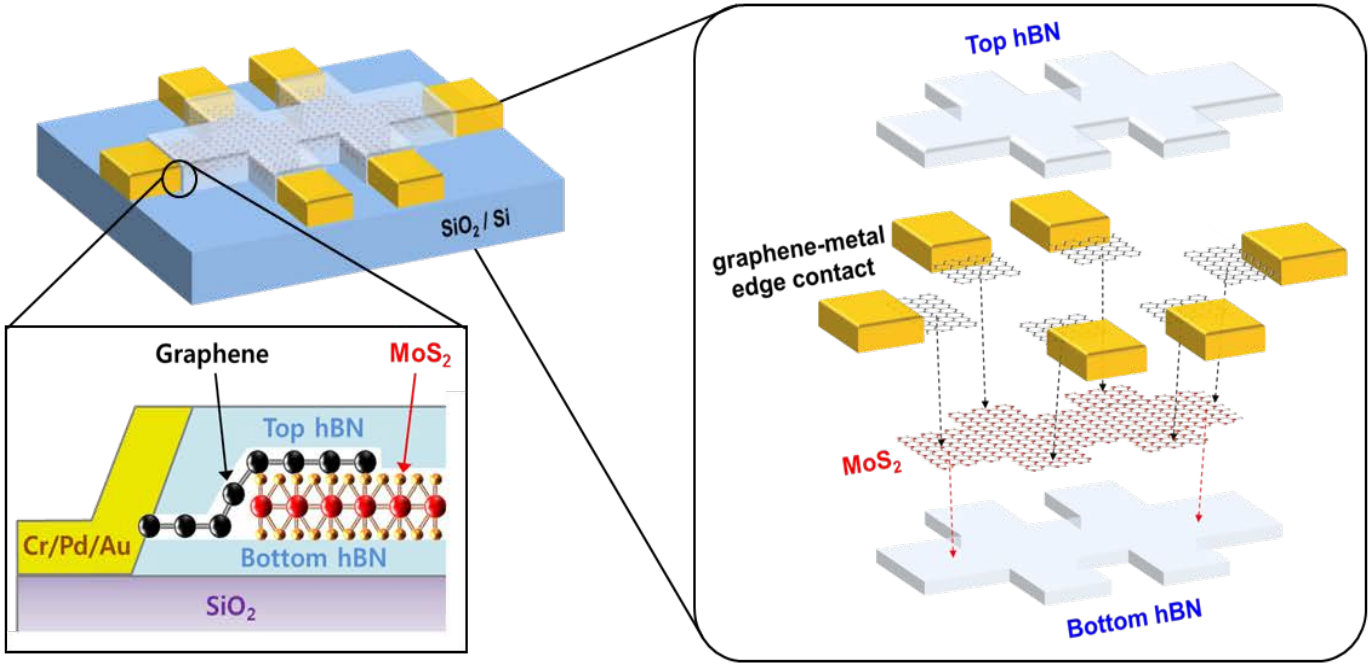


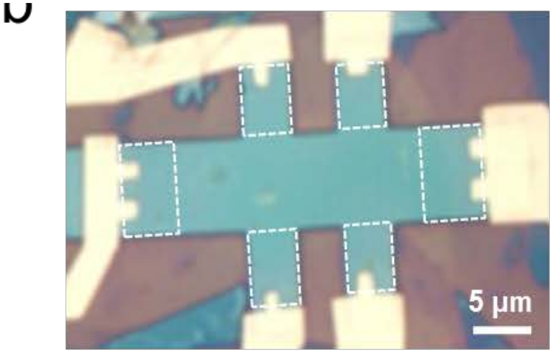
Graphene Light Sources

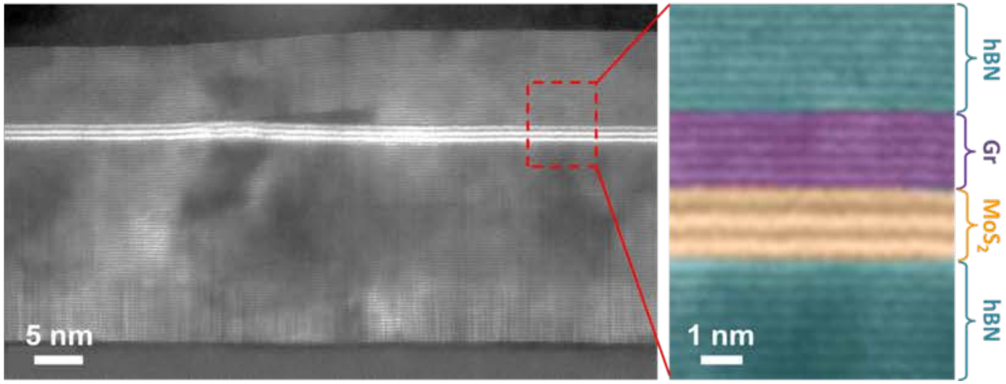
1. Suspended Graphene Emitters
2. Ultrafast BN-encapsulated Emitters

2D Semiconductors

Encapsulated MoS_2 with graphene contacts

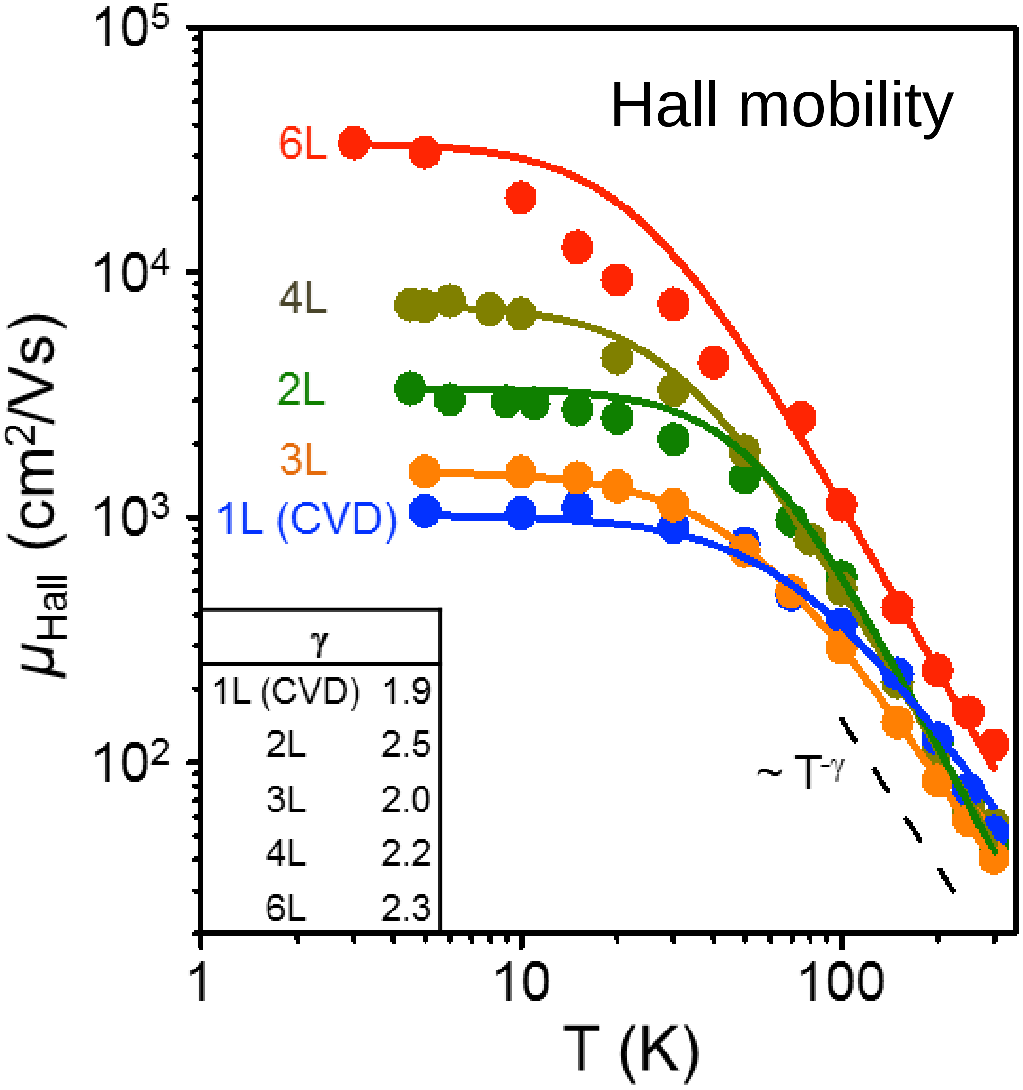


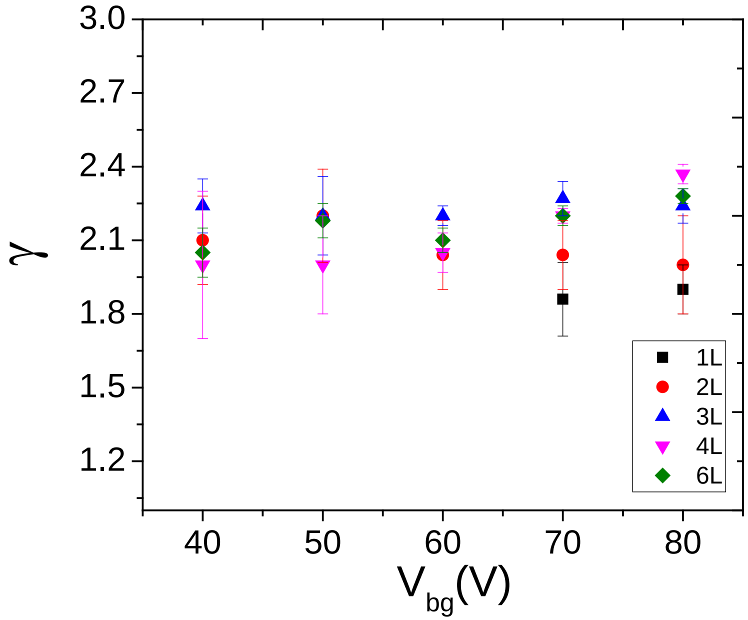




X. Cui, G. H. Lee et al.,Nature Nano (2015)

Multi-terminal transport



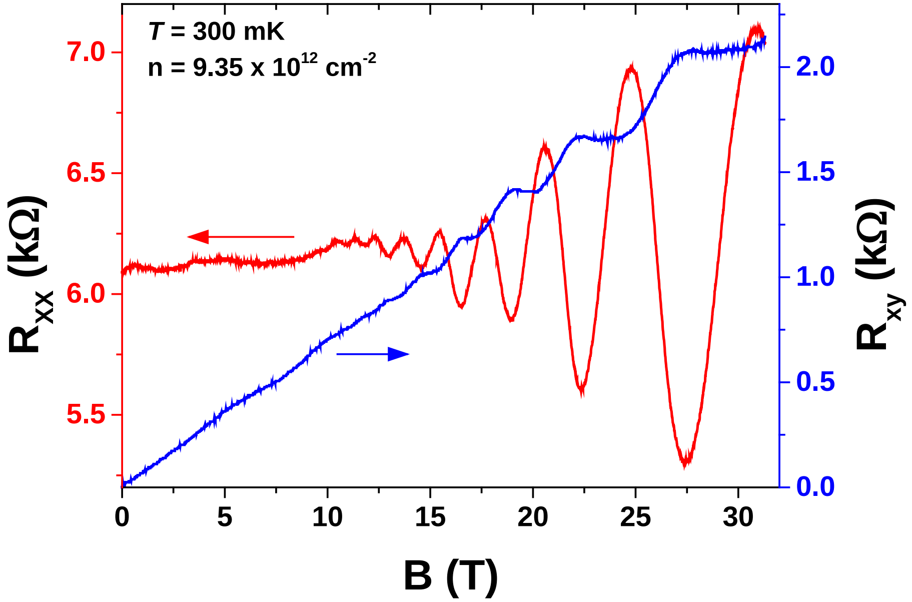


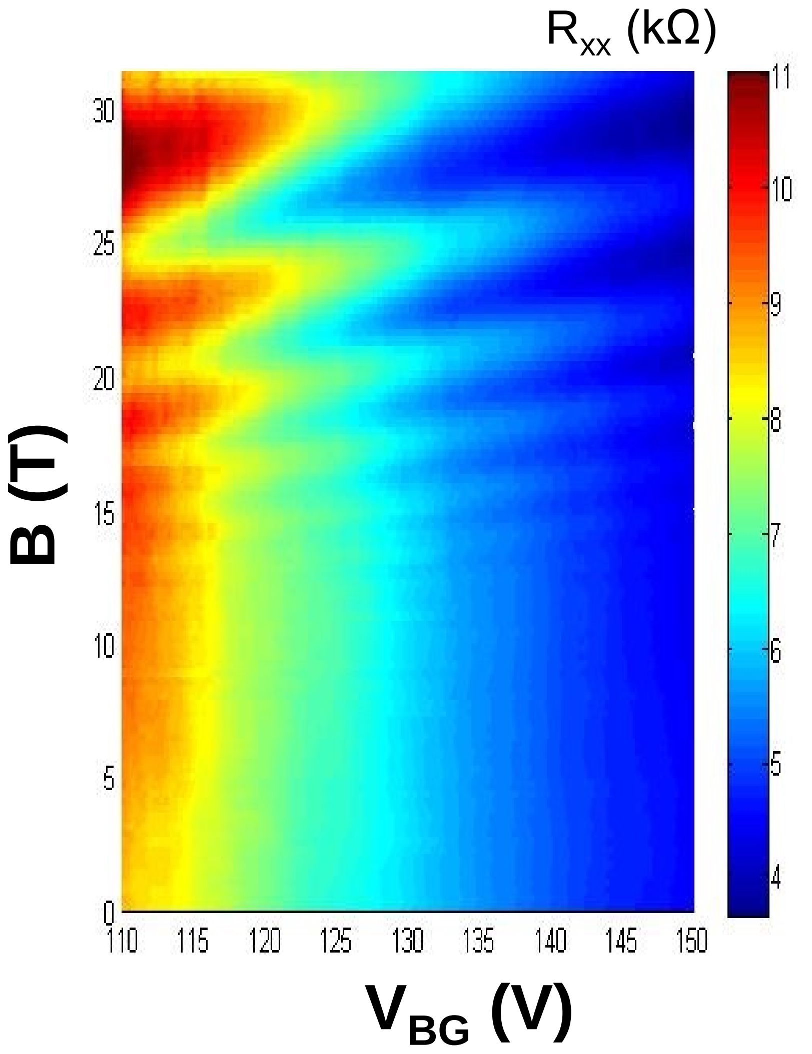
Electron-phonon mobility:

μ~T^{-2} for all samples

- contrast to on SiO_2
- Independent of gate voltage
- 50-150 at R.T.

Quantum Oscillations





Tunable SdH Oscillations in monolayer MoS_2

2D Semiconductors – current challenges

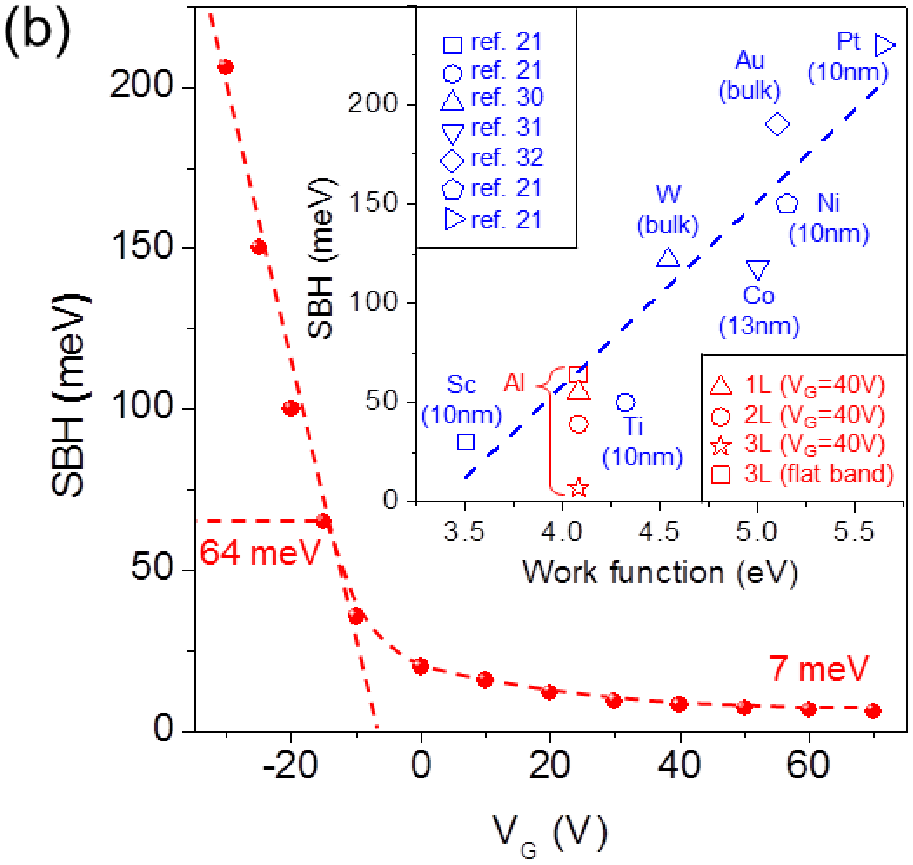
Contacts

Achieiving intrinsic optical properties

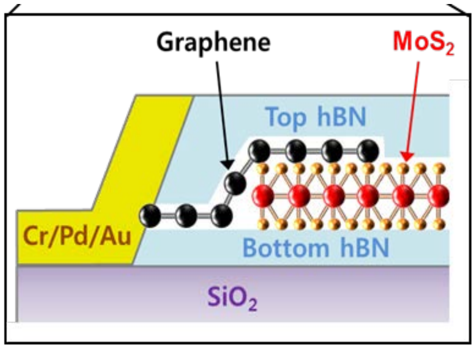
Materials Quality

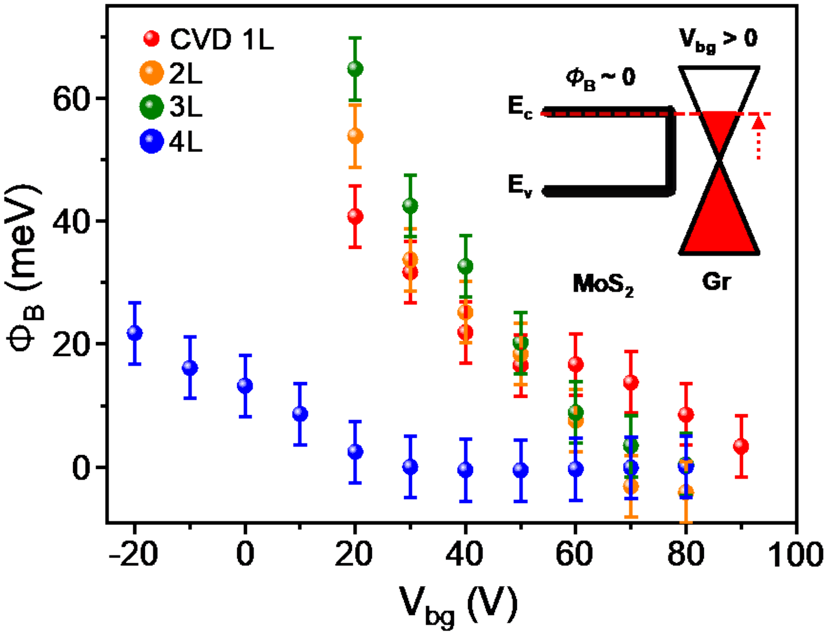
Contacts to TMDCs

A. Allain. Nature Mat. (2015)



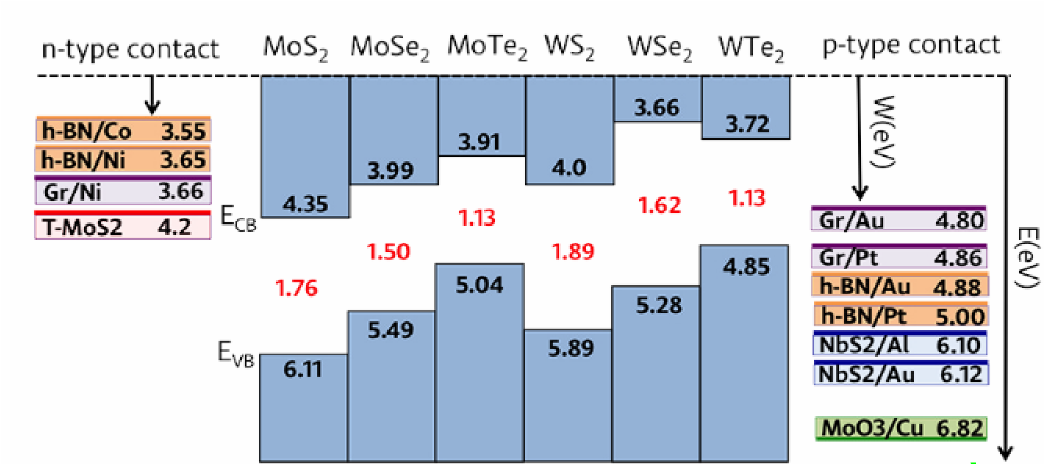
Metal contacts: Fermi Level Pinning maintains Schottky barrier even with large change in metal work function



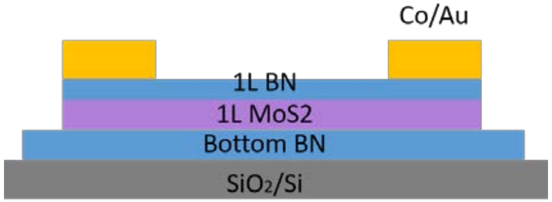


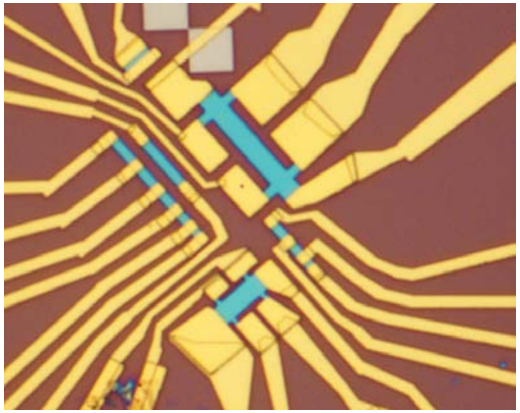
Can make Ohmic contacts with graphene – but only at very high carrier density for monlayers

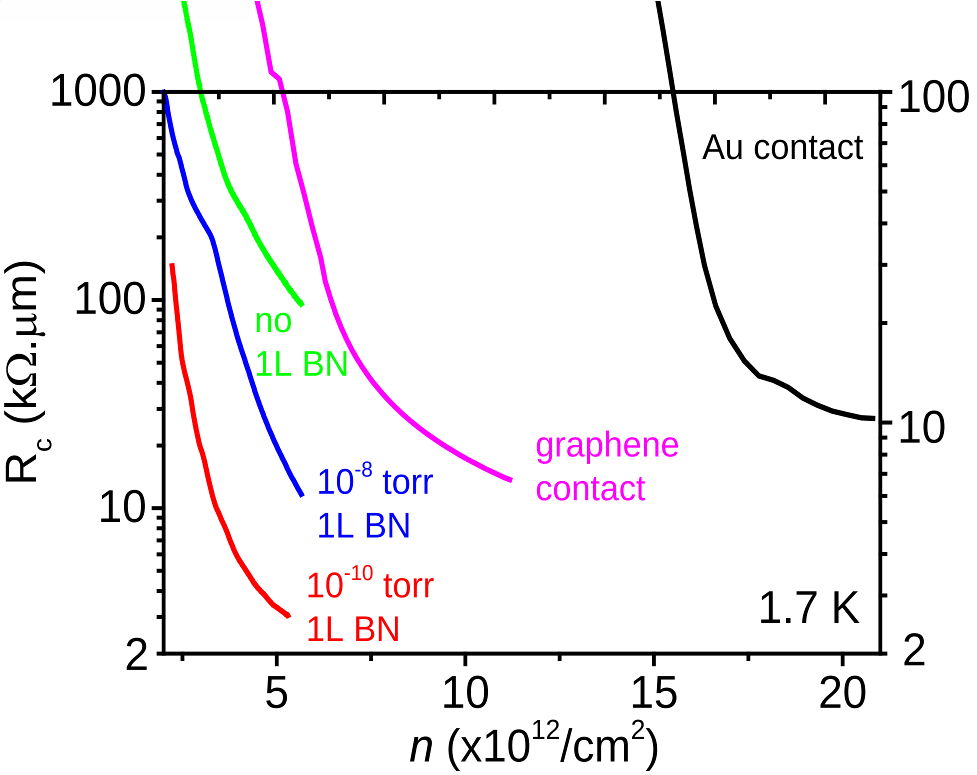
Hybrid Metal / hBN Contacts



Theory: strong hybridization between transition metals and hBN/graphene strongly shifts work function
(M. Farmanbar et al. PRB. (2016))





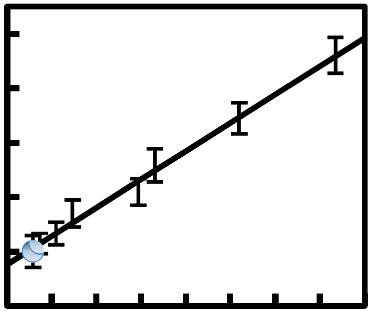


Approaching the Intrinsic Limit for TMD PL

2DFTS measurements:

Extrinsic
broadening: > 10 meV

Intrinsic
broadening: ~ 2 meV

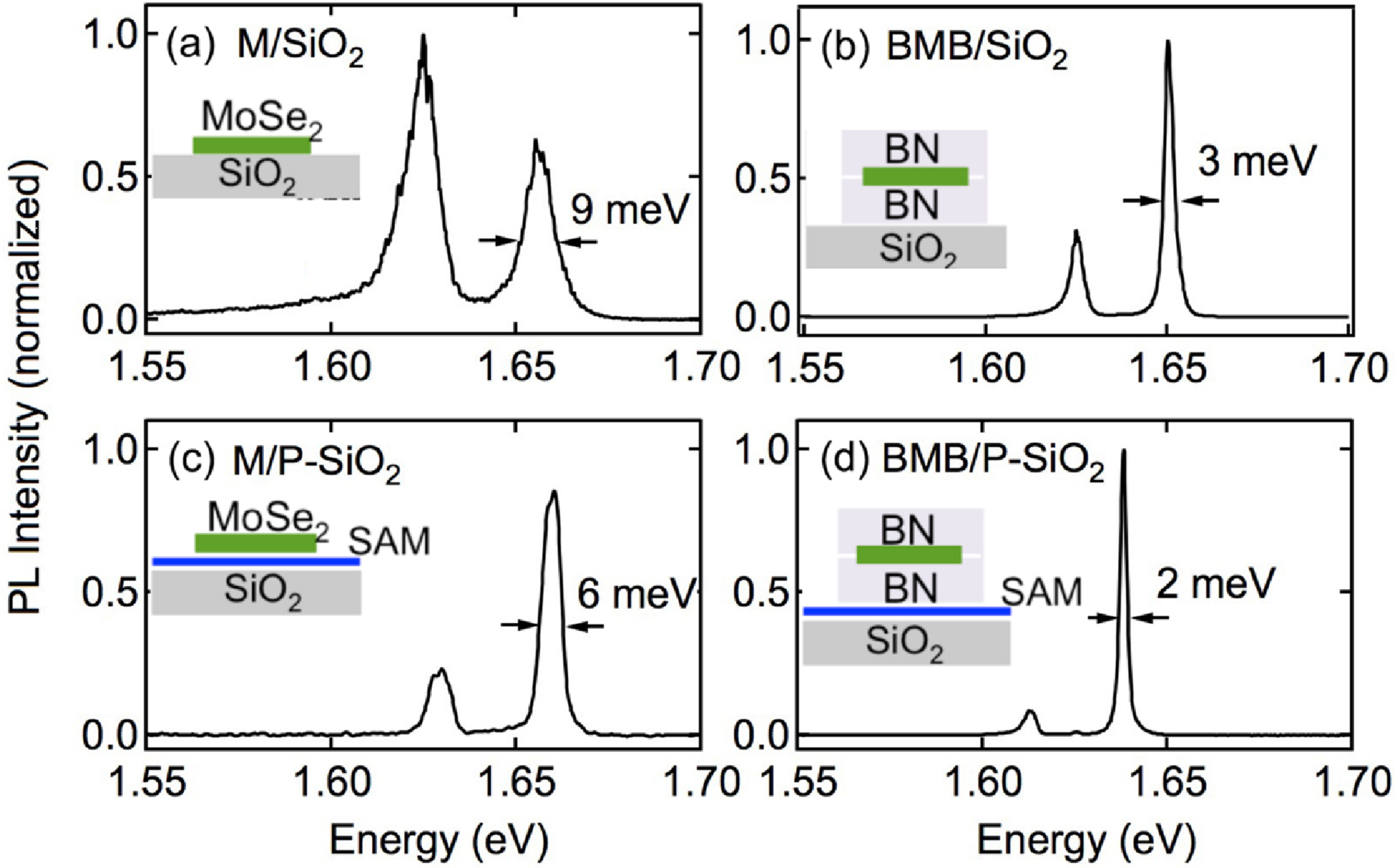


Can we minimize or eliminate extrinsic effects?

G. Moody, …., A. Knorr, X. Li, Nat. Comm 2015

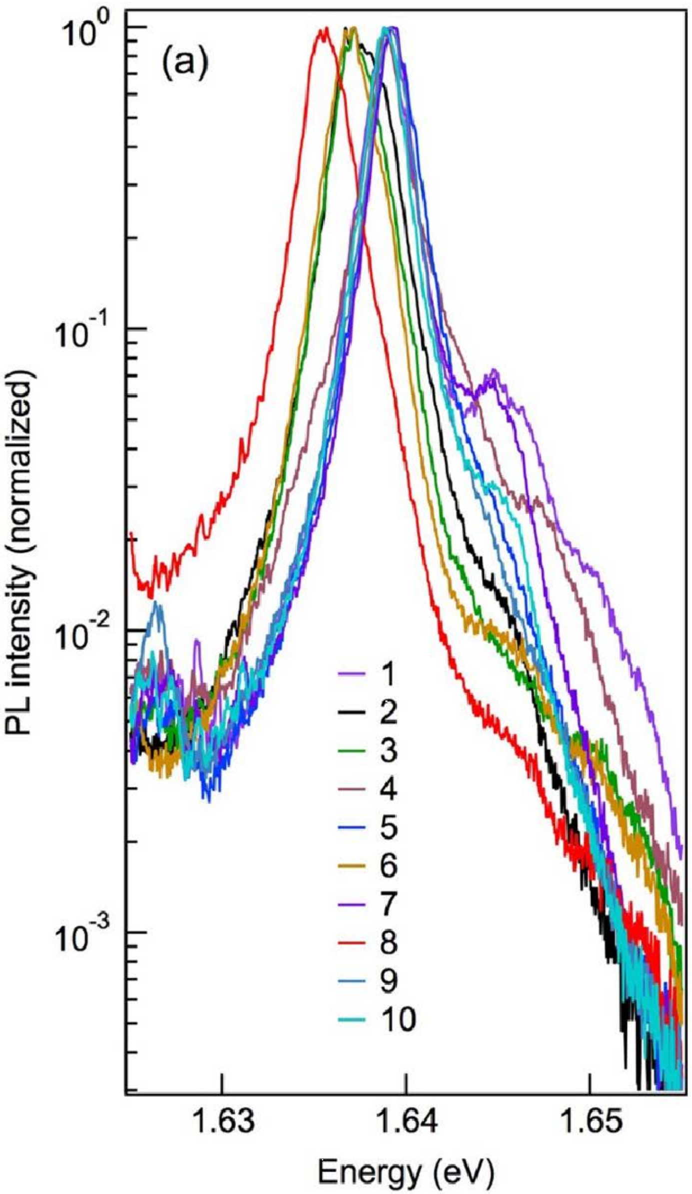
Approaching the Intrinsic Limit for TMD PL

T = 4 K



O. Ajayi, T. Heinz, S. Strauf, XY Zhu, JH, submitted

Approaching the Intrinsic Limit for TMD PL

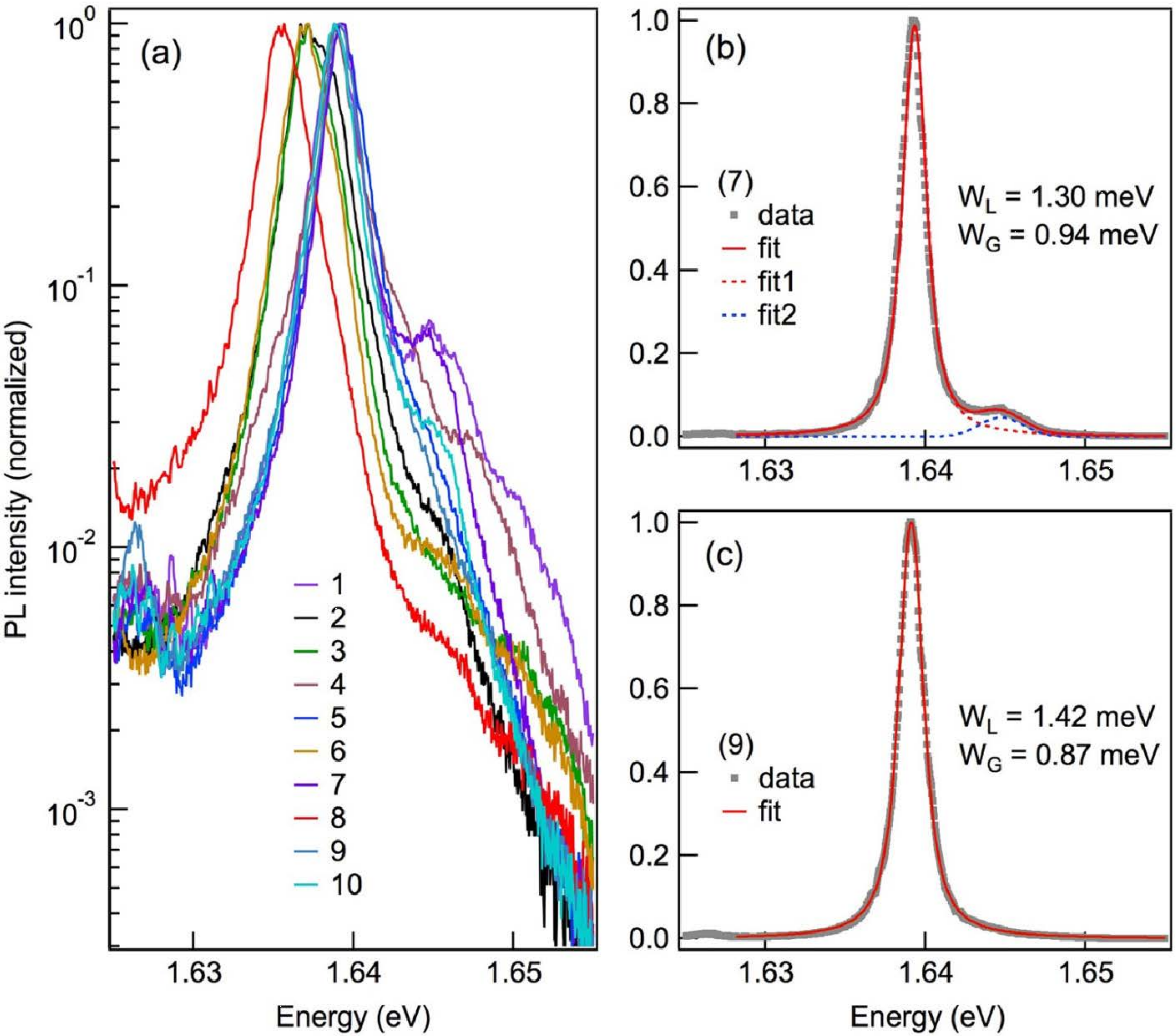


Highly consistent spectra

Broad low-E absorption

High-E ’shoulder’

Approaching the Intrinsic Limit for TMD PL



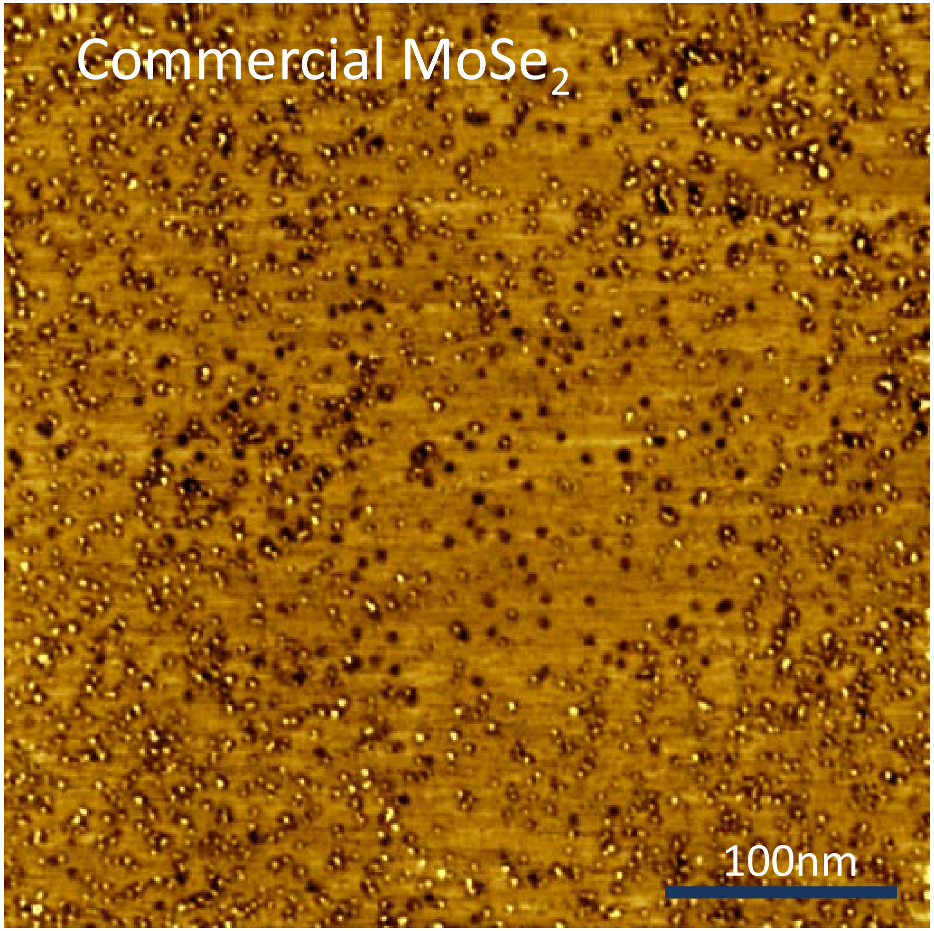
FWHM ~ 2 meV

Voigt Fit (Lorentzian with Gaussian broadening)

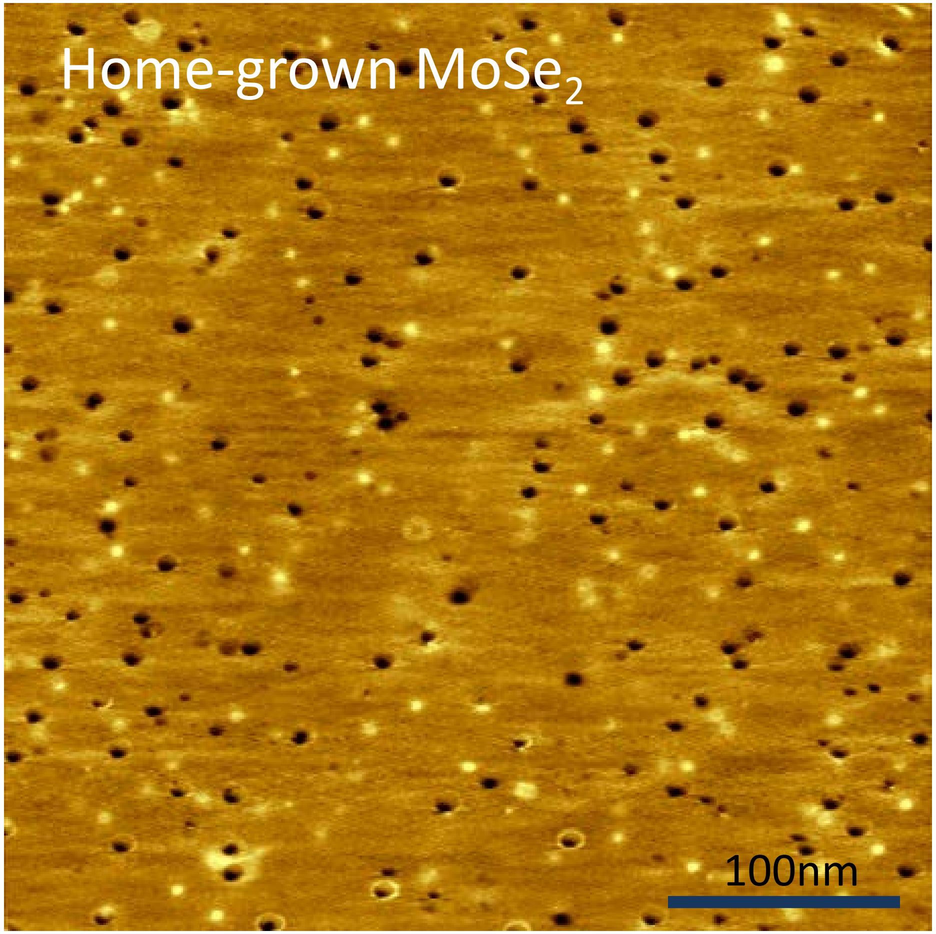
Homogeneous width ~ 1.4 meV

Heterogeneous brodening ~ 0.9 meV

Atomic defects in TMDs



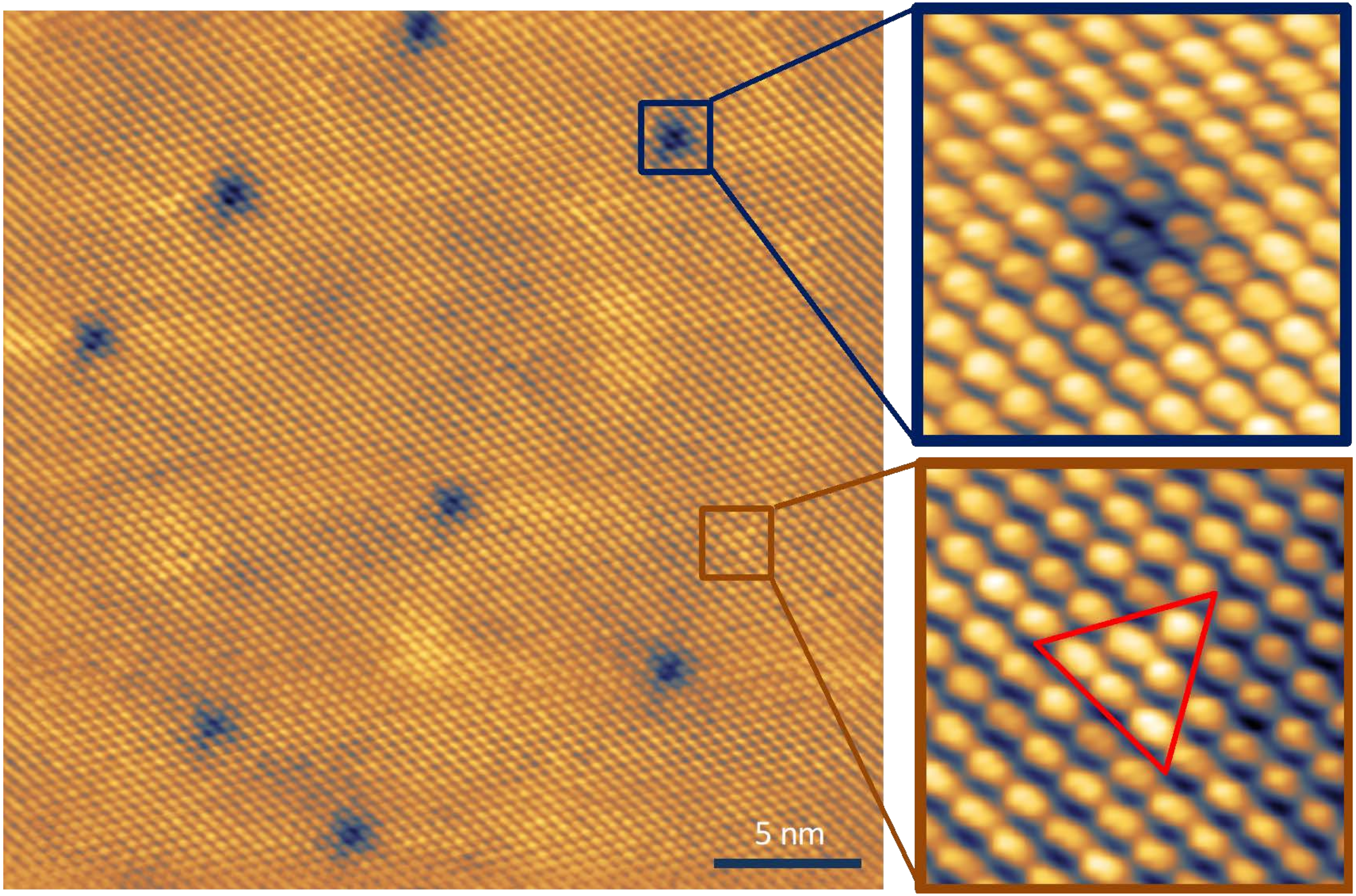
2.6% of Mo are vacancies
1.28 % Interstitial or Antisites per Mo



.33% of Mo are vacancies
.16 % Interstitial or Antisites per Mo

Growth: Hone / Barmak groups   STM imaging: Pasupathy group

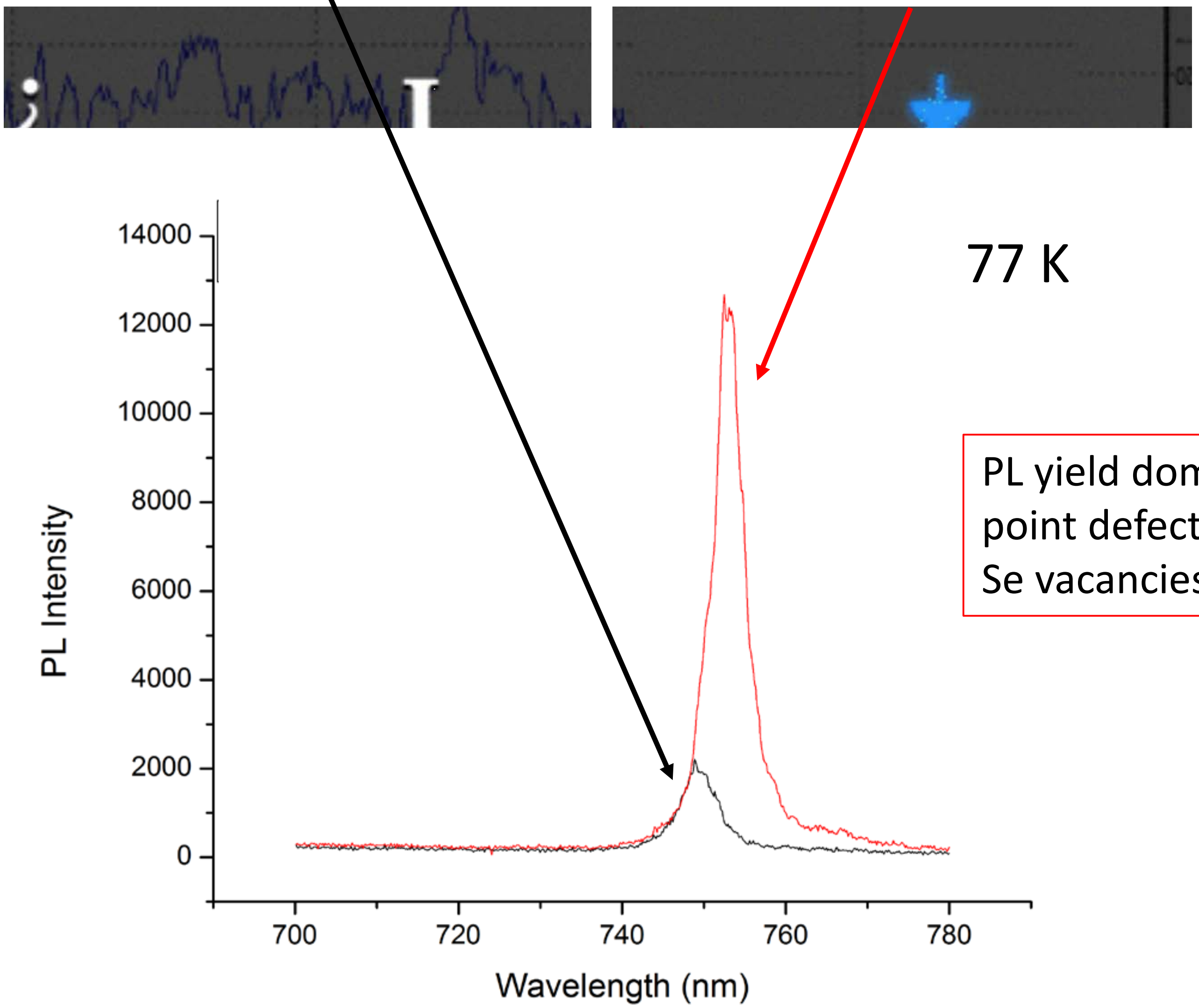
Atomic defects in TMDs



Most common defects are metal vacancies or interstitials

How can we get better?

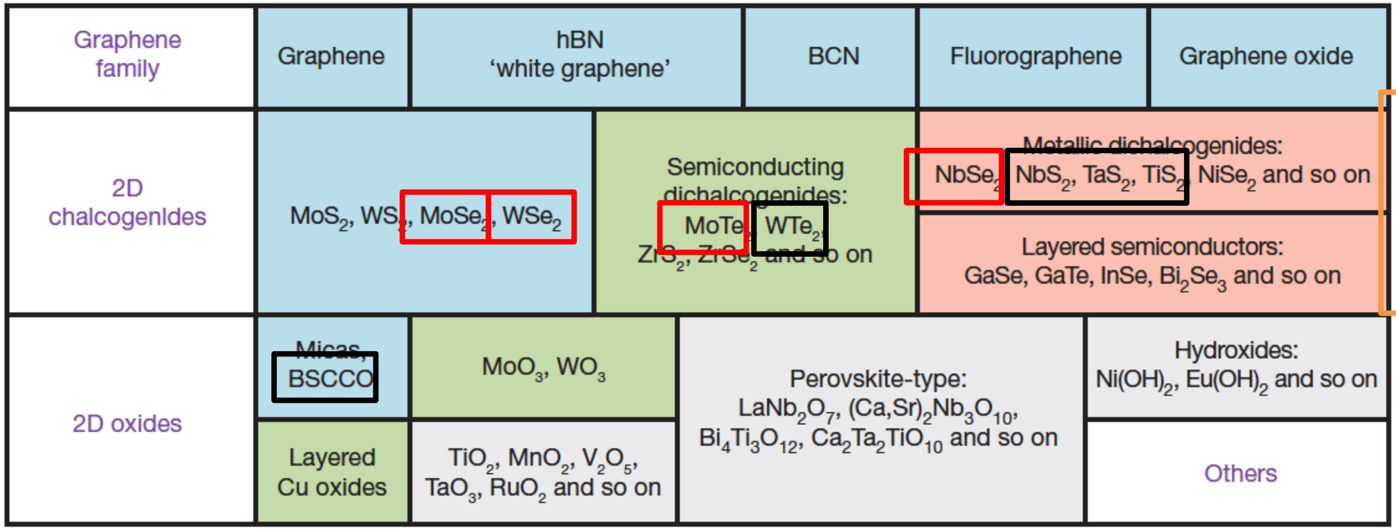
Commercial $MoSe_2$ Home-grown $MoSe_2$



77 K

PL yield dominated by point defects (Mo and Se vacancies)

Air-sensitive 2D Materials



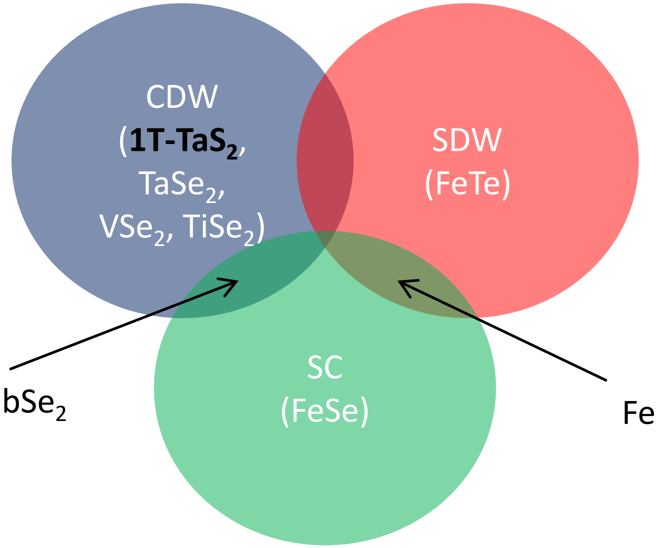
Graphene family	Graphene	hBN 'white graphene'	BCN	Fluorographene	Graphene oxide
2D chalcogenides	MoS_2, WS_2, $MoSe_2$, WSe_2	Semiconducting dichalcogenides: $MoTe_2$, WTe_2, ZrS_2, $ZrSe_2$ and so on	Metallic dichalcogenides: $NbSe_2$, NbS_2, TaS_2, TiS_2, $NiSe_2$ and so on Layered semiconductors: GaSe, GaTe, InSe, Bi_2Se_3 and so on		
2D oxides	Micas, BSCCO Layered Cu oxides	MoO_3, WO_3 TiO_2, MnO_2, V_2O_5, TaO_3, RuO_2 and so on	Perovskite-type: $LaNb_2O_7$, $(Ca,Sr)_2Nb_3O_{10}$, $Bi_4Ti_3O_{12}$, $Ca_2Ta_2TiO_{10}$ and so on	Hydroxides: $Ni(OH)_2$, $Eu(OH)_2$ and so on Others	

FeSe

FeTe

$FeSe_xTe_{1-x}$

Air-sensitive materials



CDW (**1T-TaS_2**, $TaSe_2$, VSe_2, $TiSe_2$)

SDW (FeTe)

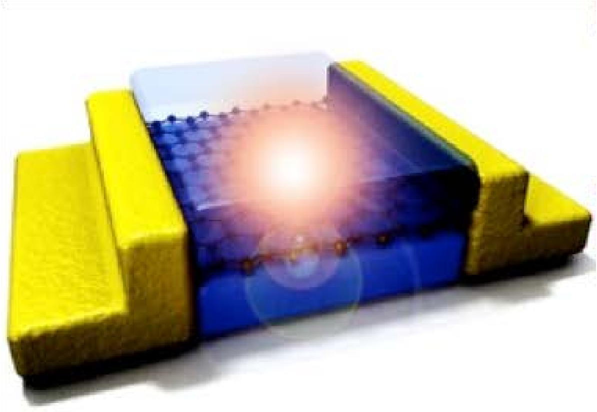
SC (FeSe)

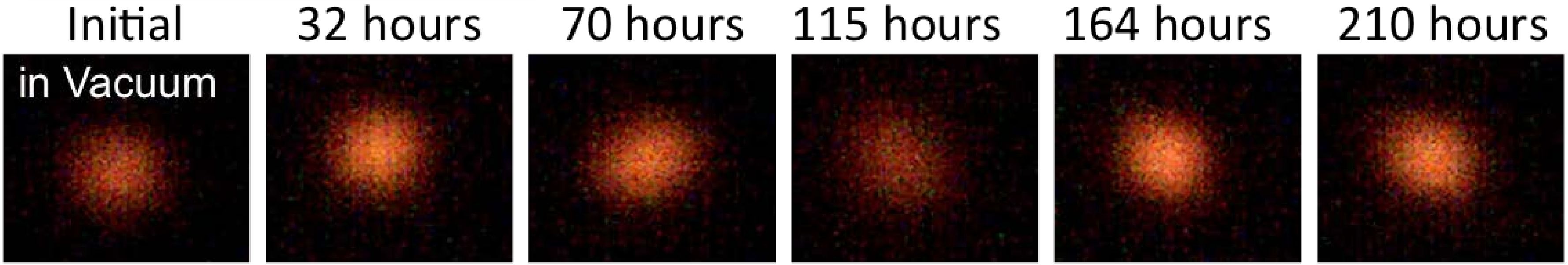
2H-TaS_2, $TaSe_2$, $NbSe_2$

FeTeS, FeTeSe

Novel Phenomena

Incandescent Visible Light Emission

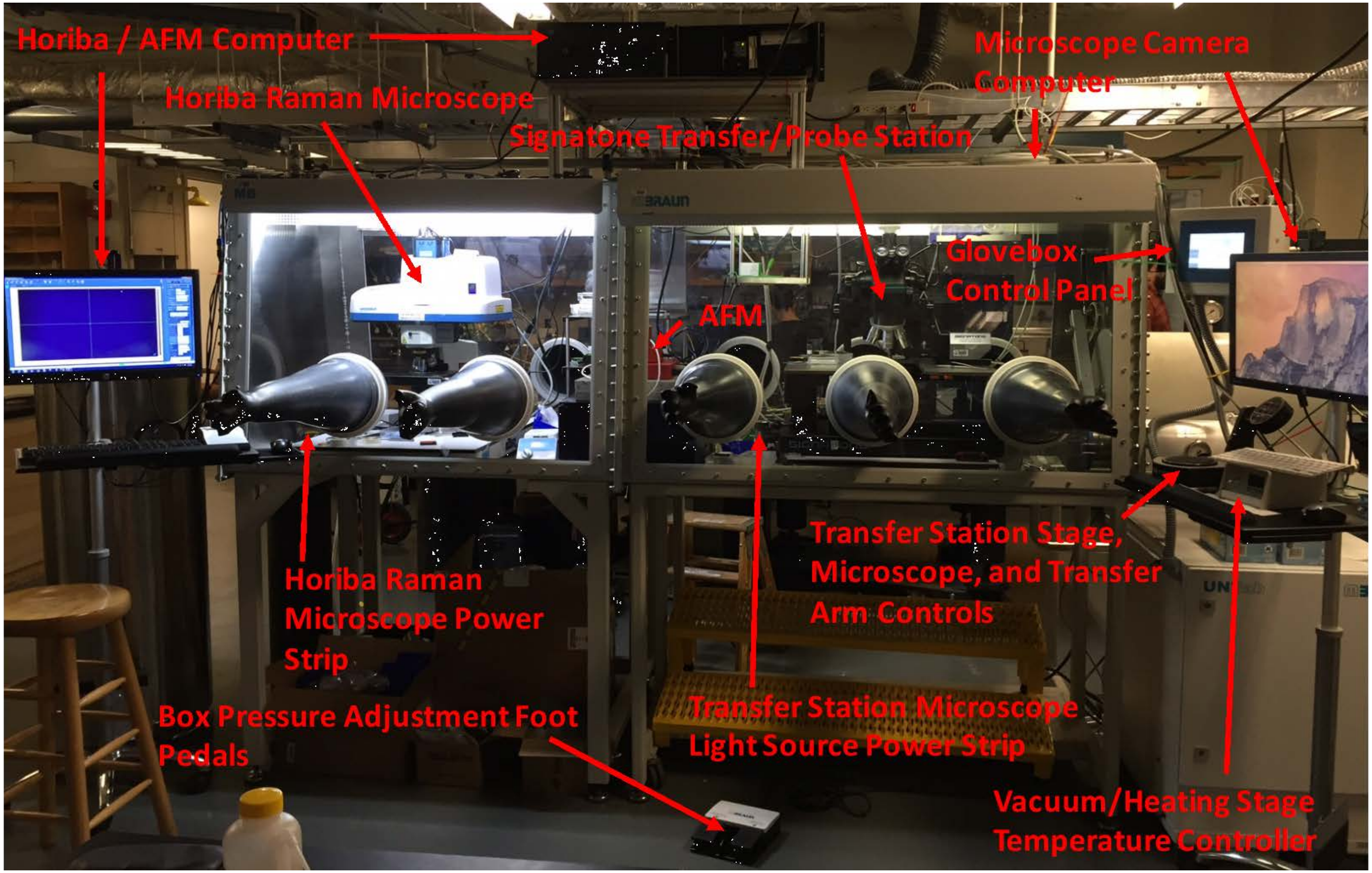



Initial
32 hours
70 hours
115 hours
164 hours
210 hours

- Stable emission for ~ 10 minutes in air
- Stable emission for ~ 10 years in vacuum

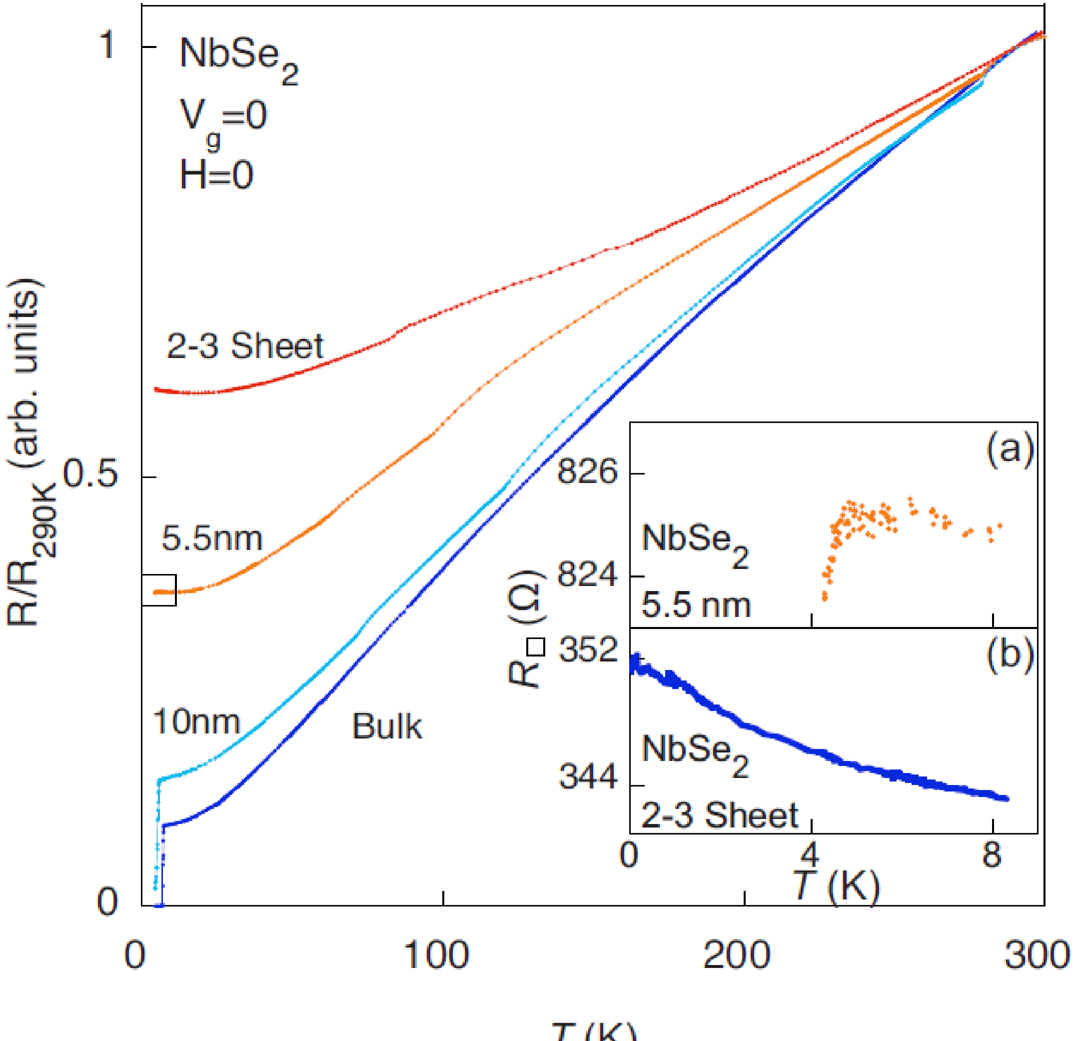
Unpublished

2D ‘Stacking’ system in glovebox



- Encapsulate in glove box using hBN
- Fabricate electrodes etc. afterward

Superconductivity in 2D $NbSe_2$

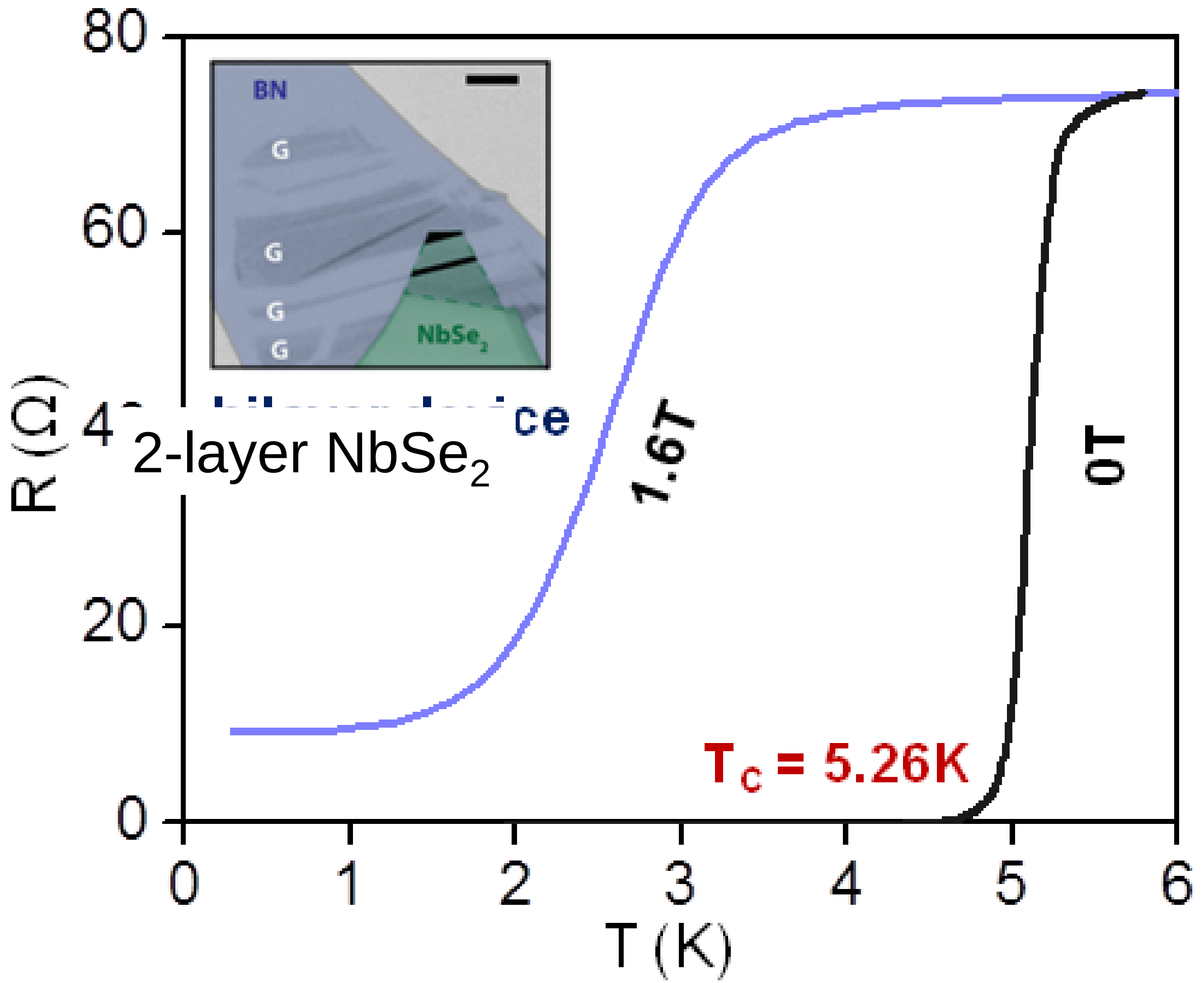


Staley, PRB 2009

Prior work:

- Bulk superconducting with T_c ~ 5 K
- Thin $NbSe_2$ is not superconducting
- High sheet resistance (disordered limit?)

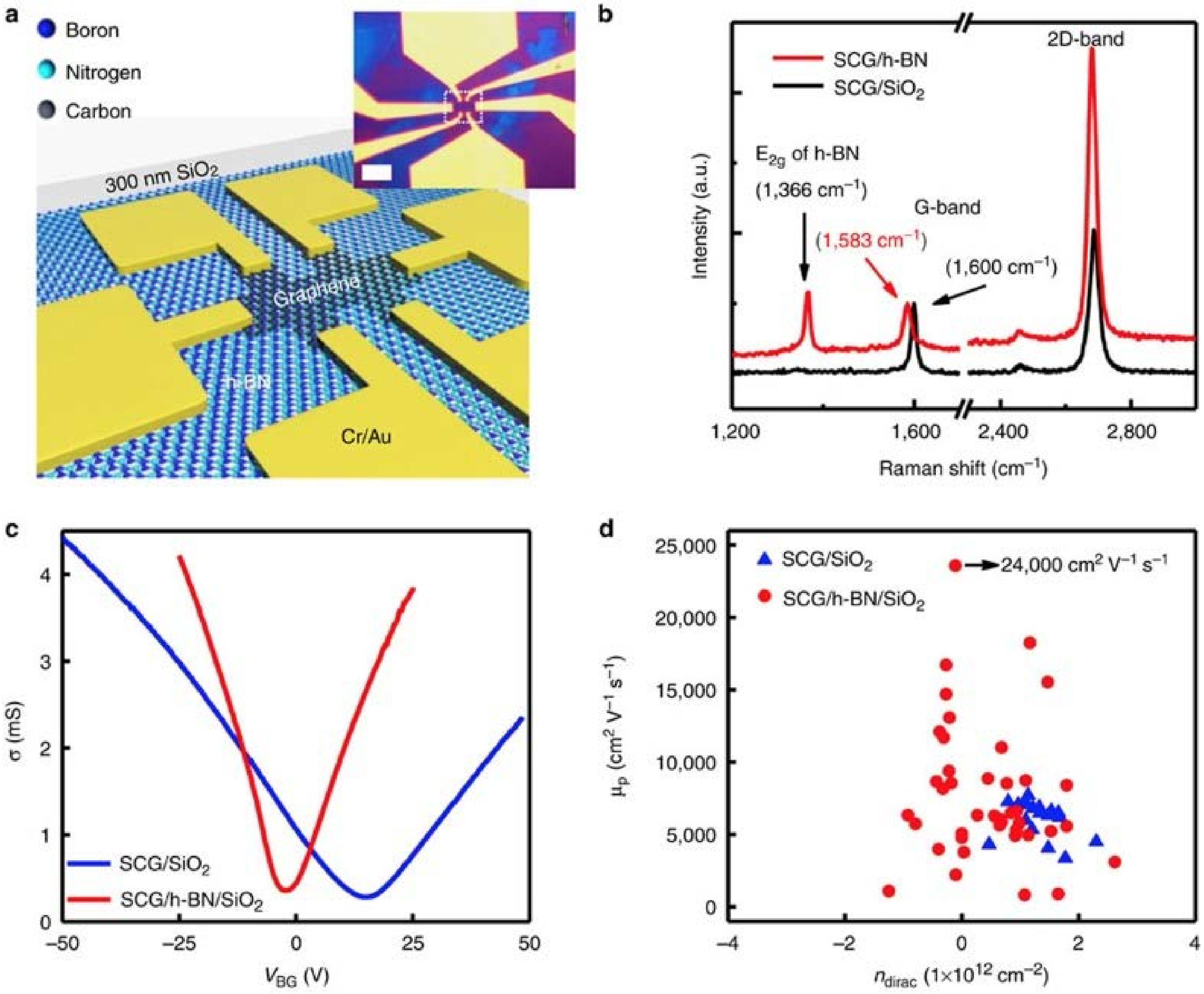
Superconductivity in BN-encapsulated bilayer $NbSe_2$



2-layer $NbSe_2$

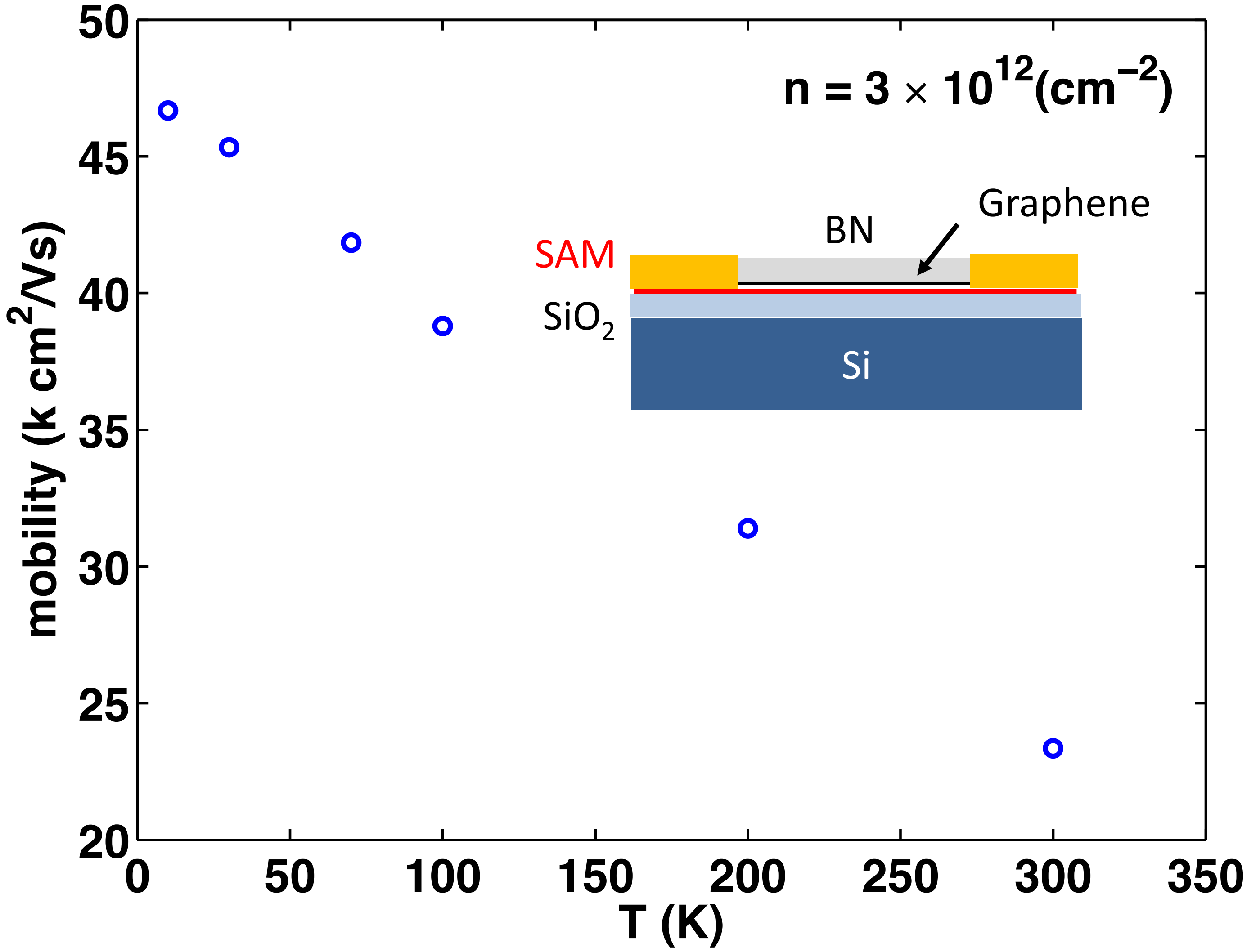
AW Tsen, B Hunt, YD Kim, ZJ Yuan, S Jia, RJ Cava, J Hone, P Kim, CR Dean, AN Pasupathy, Nature Physics (2016)

Scaleup – CVD-grown hBN



S. Kim… YH Lee, J. Kong, Nat. Comm (2015)

Scaleup – passivating SiO_2



C. Zhang, XY Zhu, JH, unpublished

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Funding: NSF MRSEC, NSF DMR, SRC / INDEX, DOE BES, ONR, AFOSR BRI