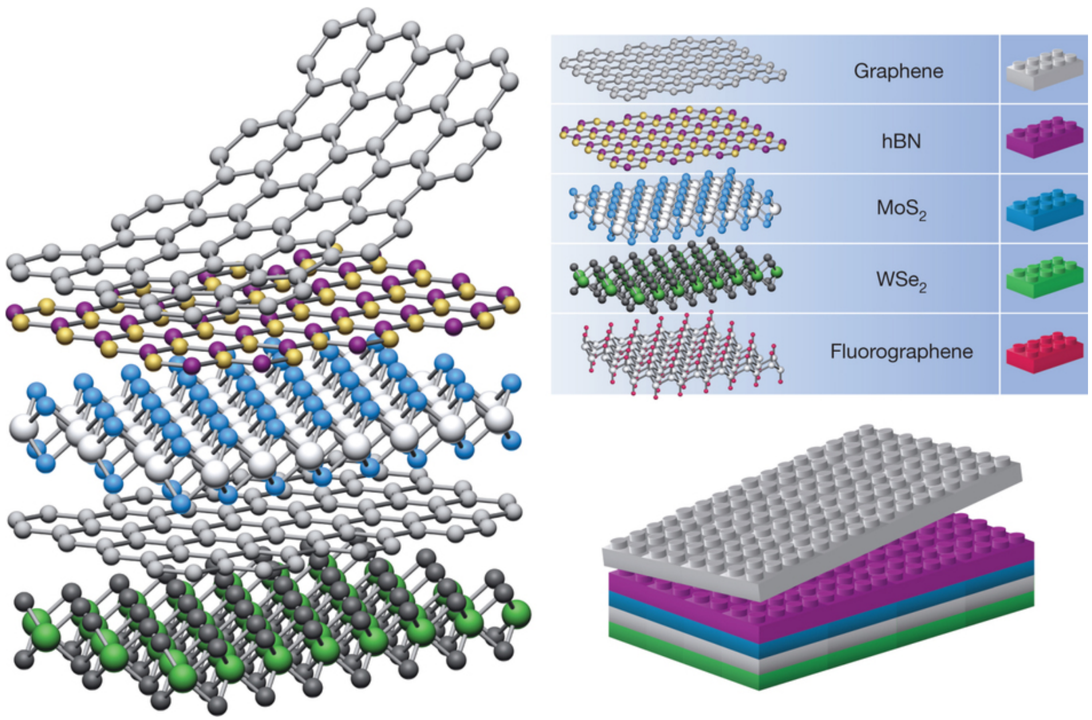
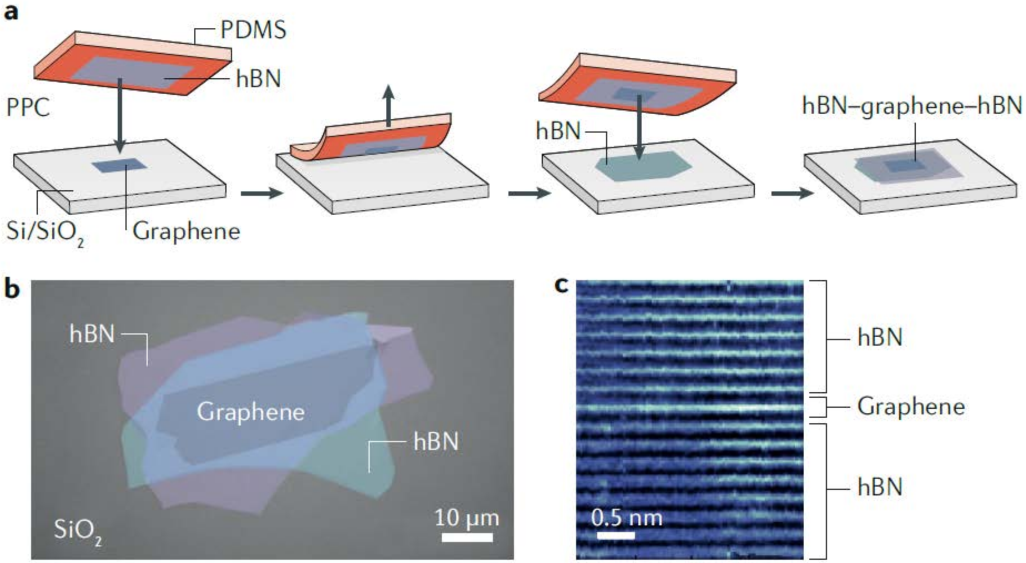
Fundamentals of nanophysics

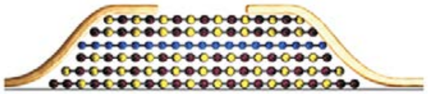
Budapest University of Technology and Economics 2024 spring

Transport in 2D van der Waals heterostructures



Van der Waals engineering

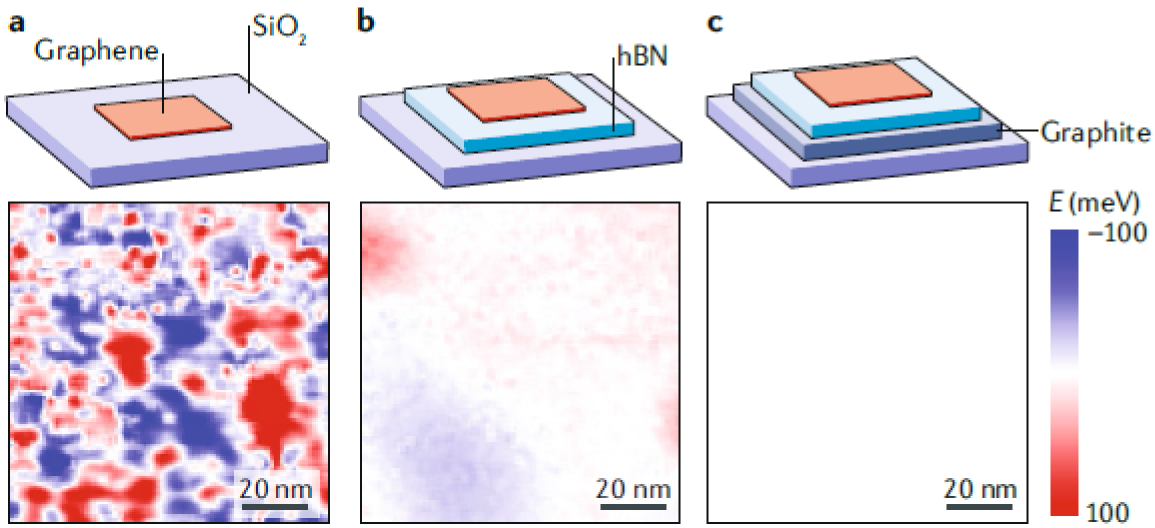


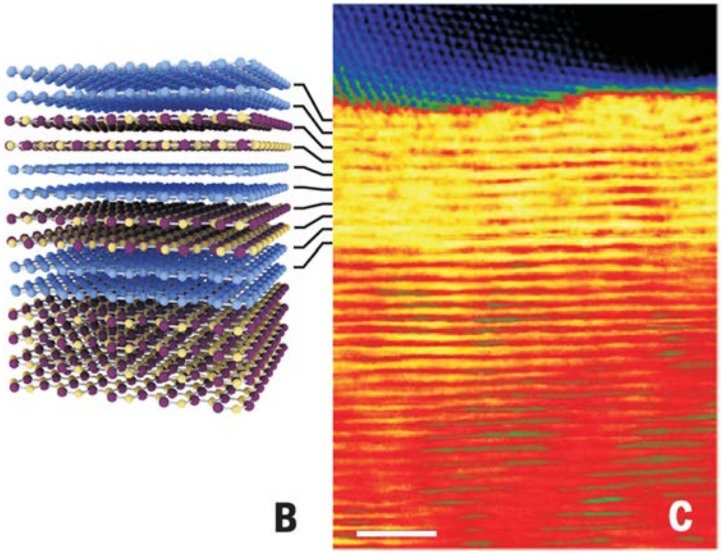


hBN as an ideal substrate
Step forward: fully encapsulate graphene in hBN – high quality but hard to access – side contacts are needed
Clean interfaces (see next slide)

Higher quality since:
Reduced charge fluctuation
Measurement: STM spectroscopy measurement
Plotted is the spatial fluctuation of the CNP (measured by spectroscopy mode)
Graphite gate: flat surface and screens additional fluctuations

M. Yankowitz et al., Nat. Rev. Phys. 1, 112 (2019)

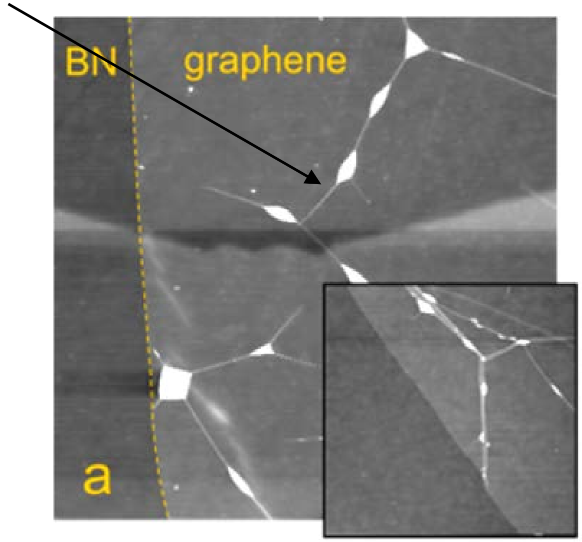




Cross sectional TEM imaging shows that there are no adsorbates stuck between the layers – here blue is graphene inbetween hBN layers can be seen

During the transfer process the adsorbates are pushed out (affinity of 2D layers to each other is larger)

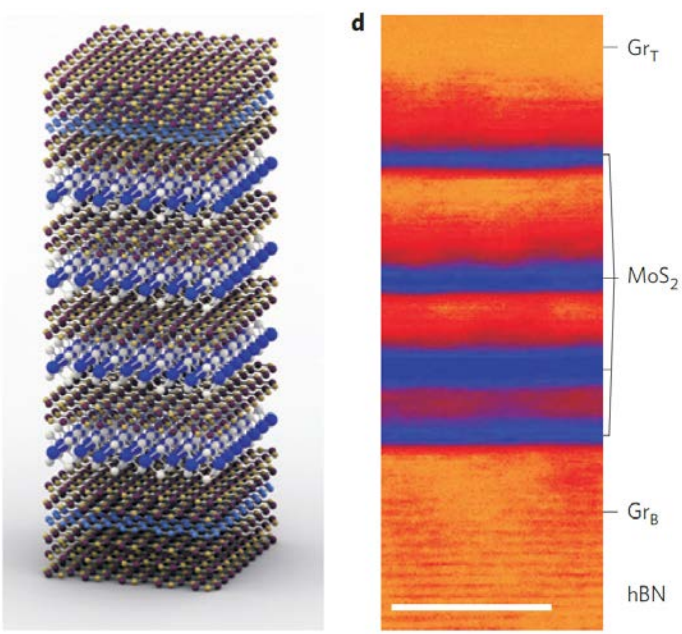
Sometimes they merge in to *bubbles* – usually containing hydrocarbons

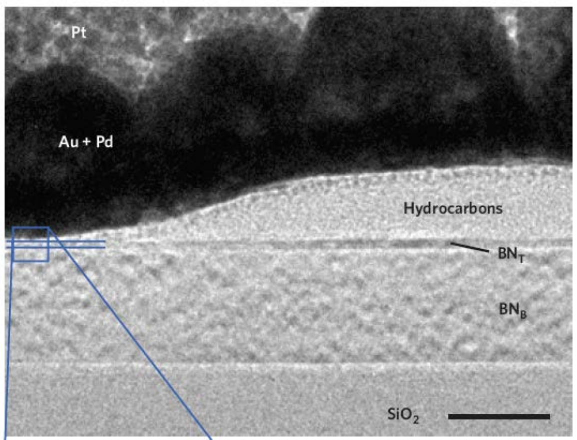


Bubbles can be seen in AFM and avoided during design stage

Cross sectional TEM shows such a bubble – scalebar 25nm

Quite complex structures can be engineered

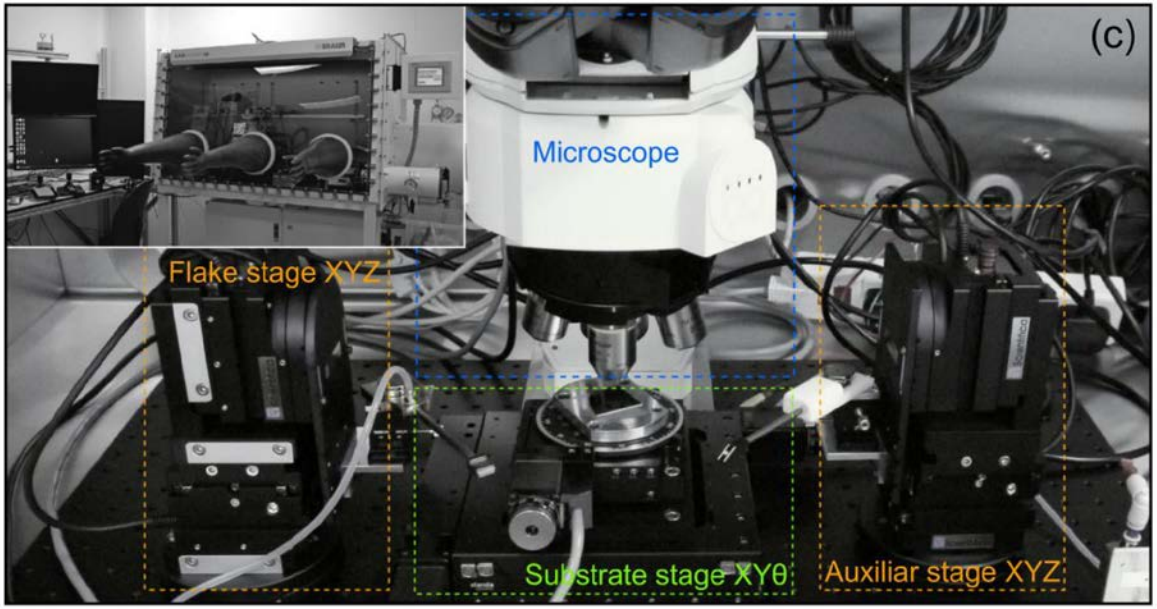


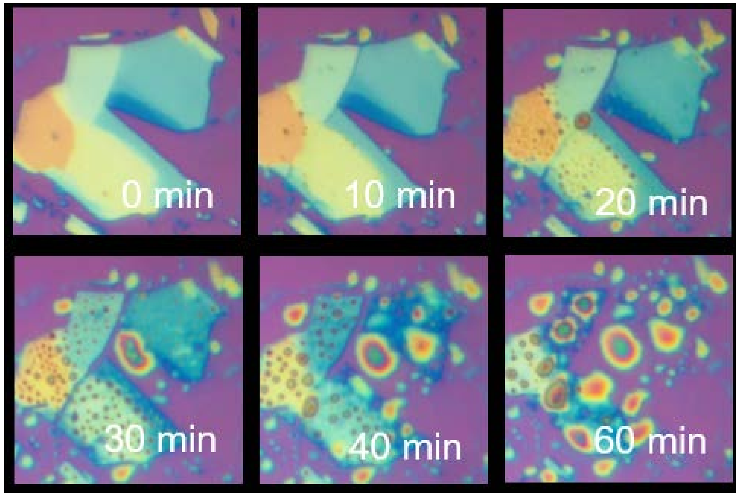


S. J. Haigh et al., Nature Materials 11, 764 (2012)

A. V. Kretinin et al., Nano Lett., 14, 3270 (2014)

Air-sensitive materials





Some flakes are air/humidity sensitive

Different strategies

- Fabricate fast and run
- Fabricated in glove-box and encapsulate in hBN. Motorized stages for stacking
- First trials towards full UHV fabrication

R. Frisenda et al., Chem. Soc. Rev., 47, 53 (2018)

D. Shcherbakov et al., Nano Lett., 18, 4214 (2018)

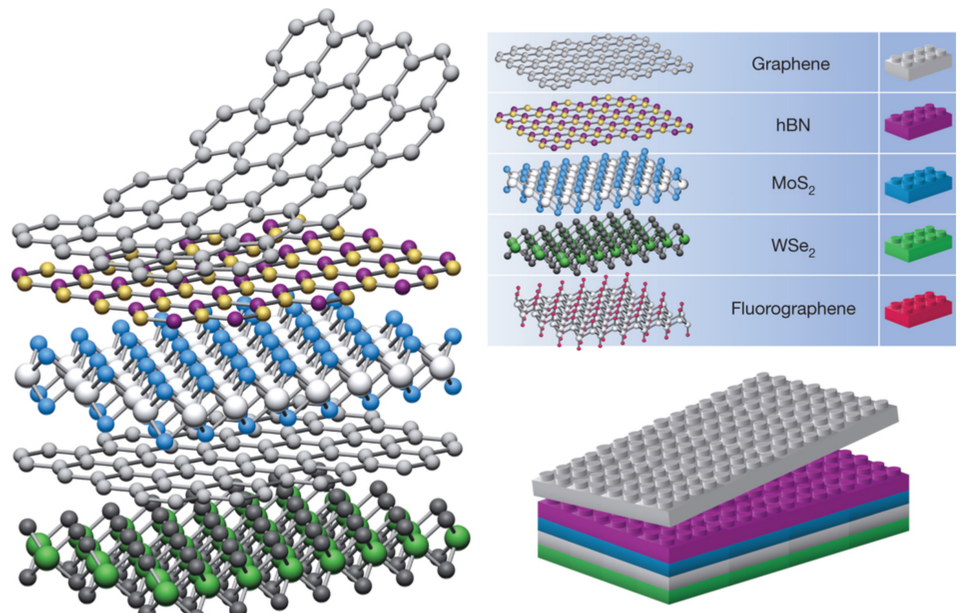
Zoo of materials – beyond graphene

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	MoO_3, WO_3	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	
	Layered Cu oxides	TiO_2, MnO_2, V_2O_5, TaO_3, RuO_2 and so on		Others	

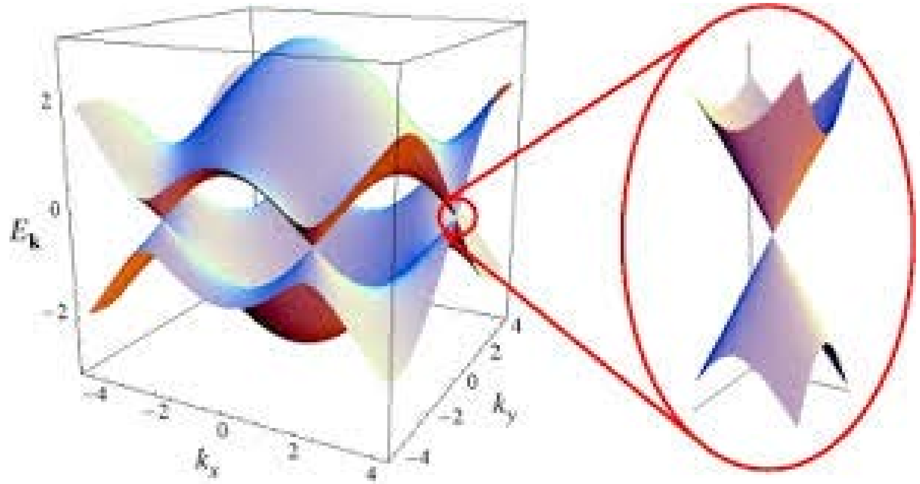
Other materials exist – quite many…

- Insulators
- Semiconductors
- Magnetic materials
- Superconductors
- ….

They can be combined to engineer new materials assembled on the nanoscale



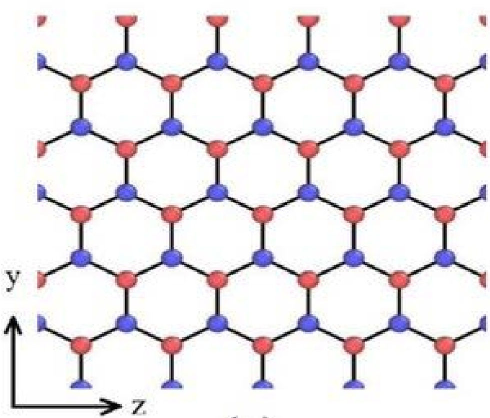
K. Geim et al., Nature 499, 419-425 (2013)

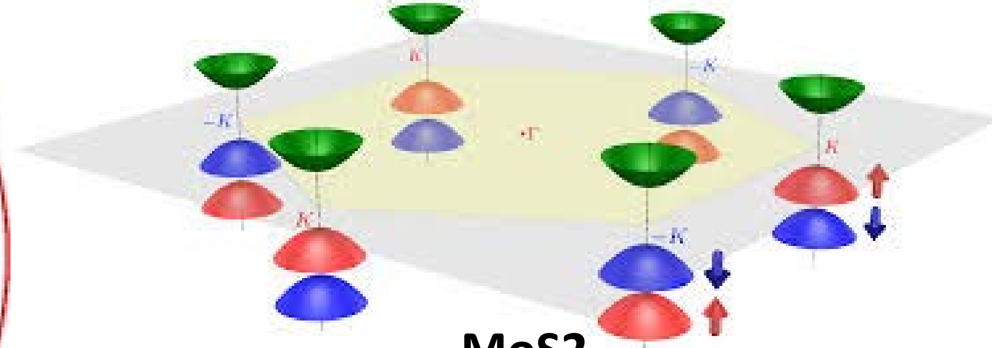


Graphene

$$H_0 = \hbar v_F \left(\kappa \sigma_x k_x + \sigma_y k_y \right) s_0$$

σ: sublattice
κ: valley
s: spin





MoS2

$$H_{VZ} = \lambda_{VZ} \kappa \sigma_0 s_z$$

Spin orbit term in case of broken inversion symmetry

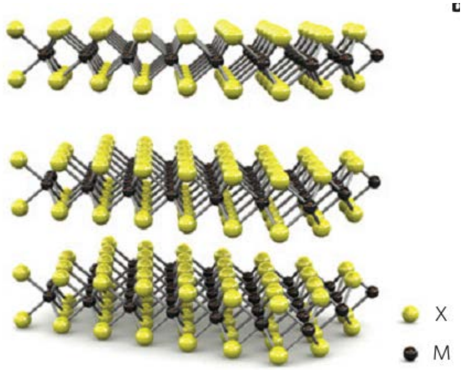
hBN

$$H_\Delta = \Delta \sigma_z$$

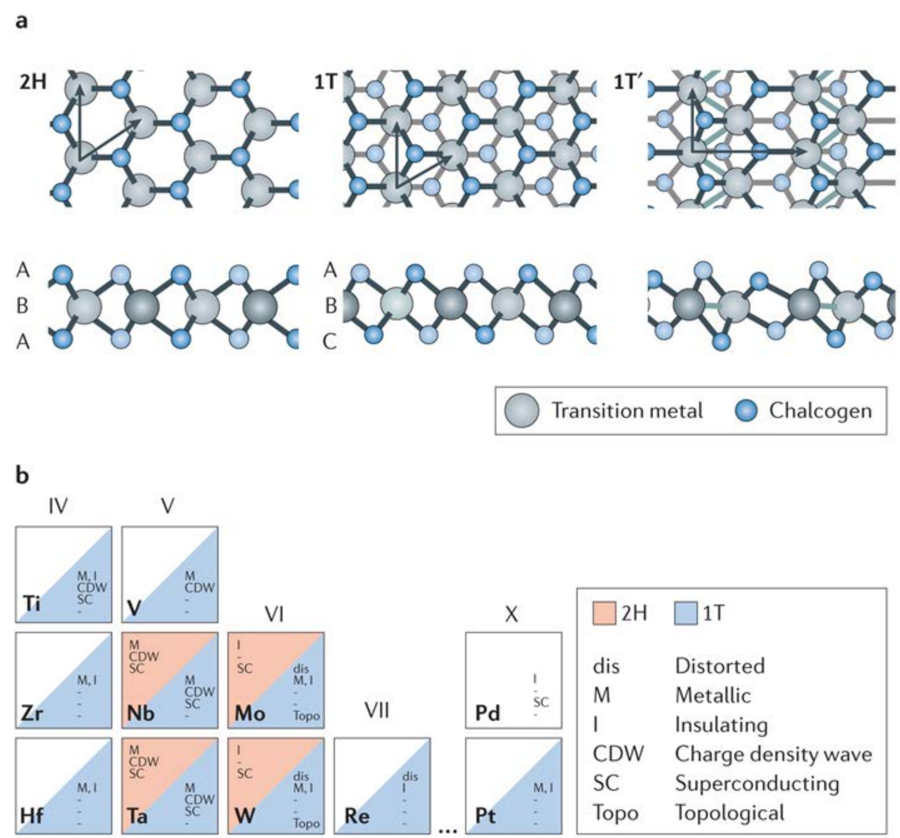
Gap term appears due to different on-site energy on A and B sublattice (broken inversion)

TMDCs

Layered structures with MX_2 structure
Weak interlayer coupling

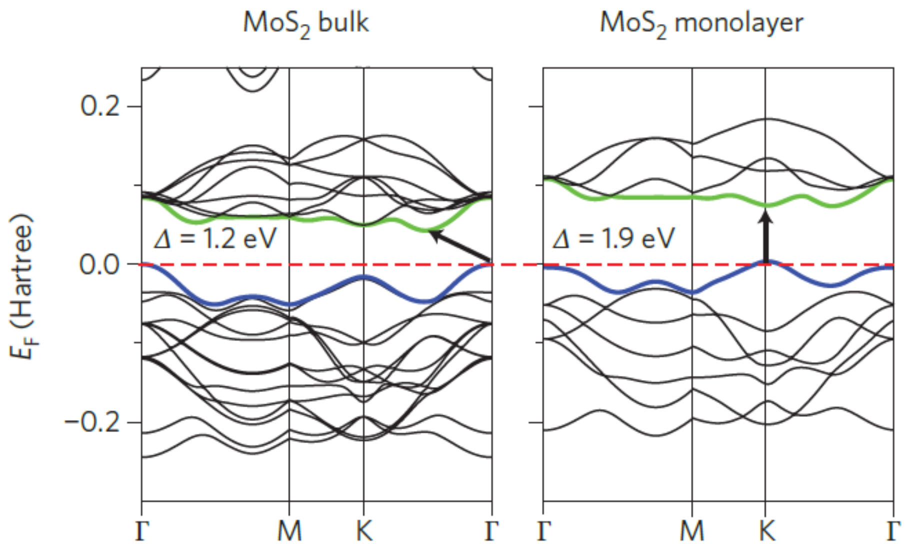


TDMCs have different properties
- Metallic
- Semiconducting
- Superconducting
- Topological
- ...



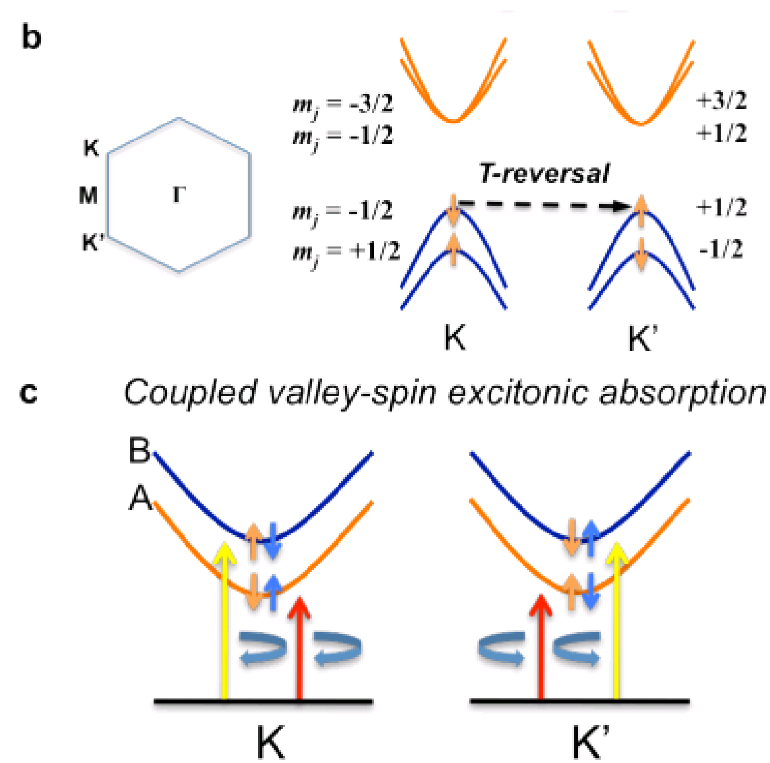
S. Manzeli, S. *et al. Nat. Rev. Mater. 2, 17033 (*2017)
D. Zappa et al., Materials 10, 1418 (2017)

Example: MoS2



MoS_2: indirect band gap semiconductor, small gap: 1.3 eV
Single layer MoS_2: direct band gap 1.8 eV

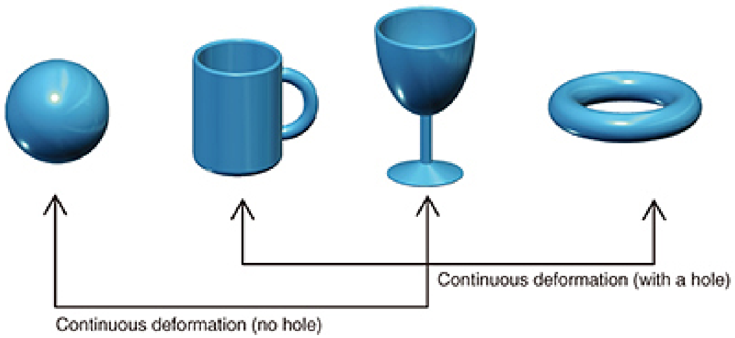
Cond band: localized d orbitals on Mo atom, unaffected by interlayer coupling (K point)
Valance band at (Γ) point p_z orbitals on S, strongly affected by interlayer coupling



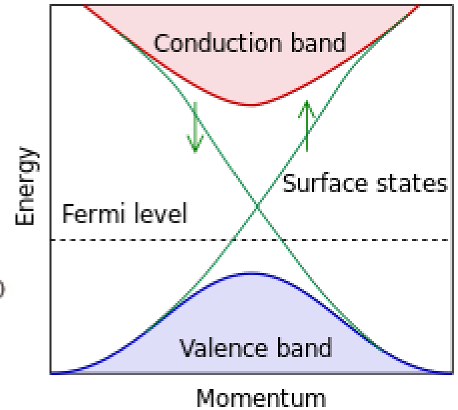
-Inversion symmetry is broken, since different atoms occupy A and B site
-Split valance band due to SO interactions in the lack of inversion (160 meV)
-$E_{\uparrow}(k) \neq E_{\uparrow}(-k)$ but $E_{\uparrow}(k)=E_{\downarrow}(-k)$ (Time Reversal S.)
→Coupling of spin and valley and the spin arrangement is different in different valleys
E crystal field (in plane), $B_{SO} \sim k \times E$ out of plane
- For bilayer, inversion is recovered and $E_{\uparrow}(k) = E_{\uparrow}(-k)$
- Valley dependent selection rules

A. Kuc et al., Phys. Rev. B. 83, 245213 (2011)
K. F. Mak et al., Nature Nanotech. 7, 494 (2012)

Quantum Spin Hall effect – WTe_2

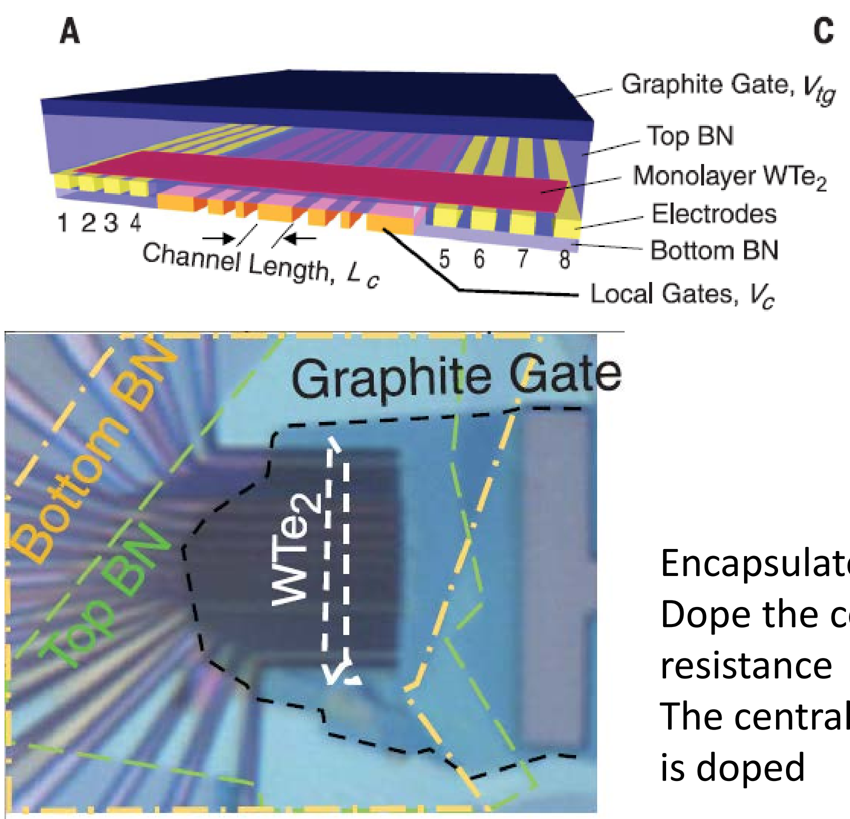


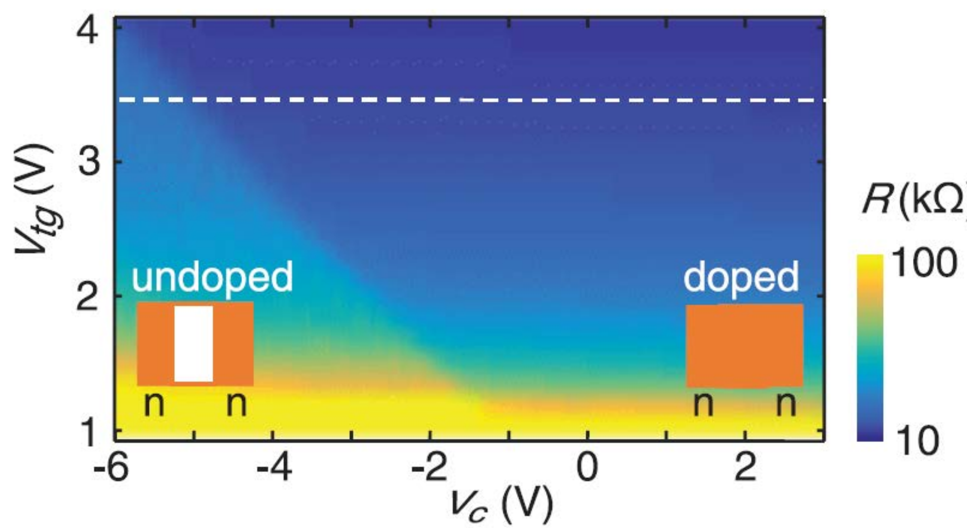
H. Yamaguchi (NTT Review)



Quantum Spin Hall Effect (QSHE)

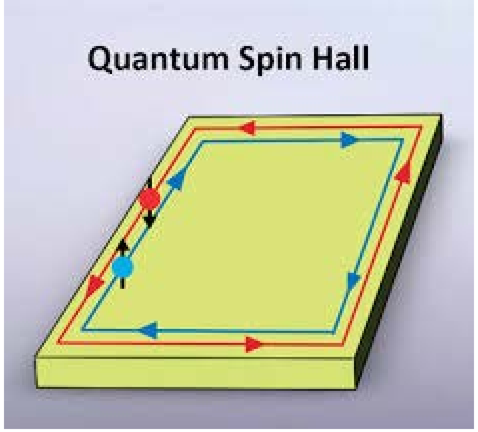
- Edge states with opposite spin circulating opposite direction
- Bulk is gapped
- $2e^2/h$ conductance in 2-terminal
- Suppresssion of quantization with magnetic field: breaks time reversal symmetry
- Topological origin



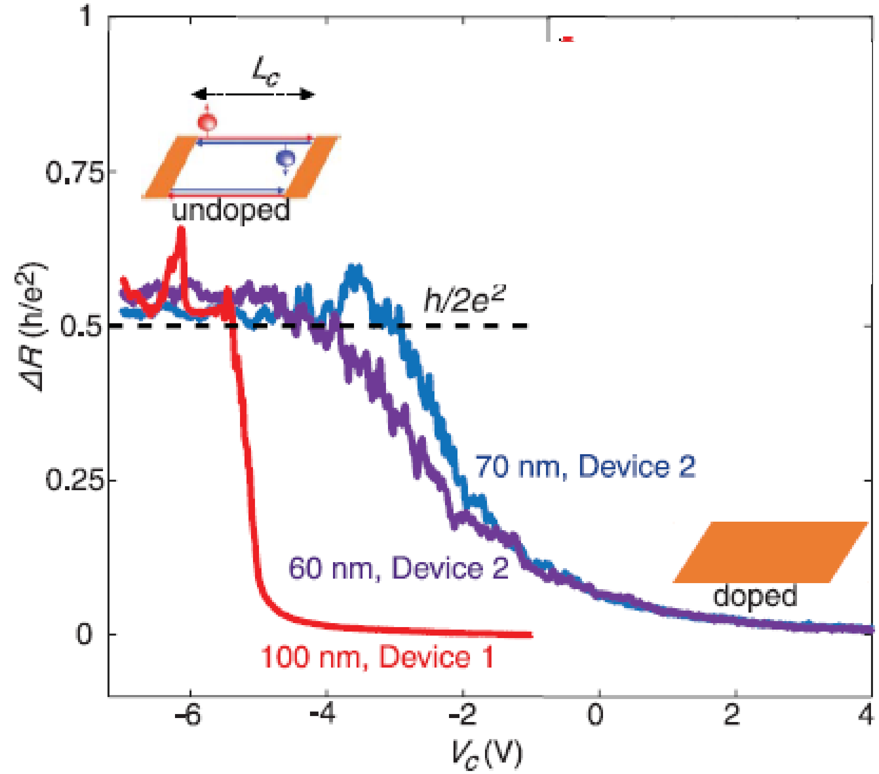


Encapsulated in hBN with local gates and local electrodes
Dope the contact regions (using topgate) to achieve small series resistance
The central region is tuned by Vc: for small voltages gapped, for large it is doped

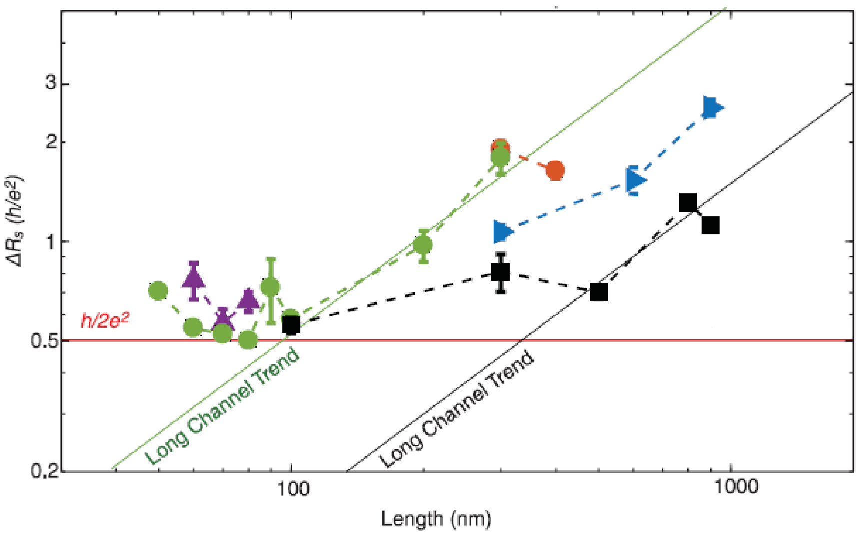
S. Wu et al., Science 359, 76 (2018)



A plateau at $2e^2/h$ apppears
The bulk is gapped (from other measurements)

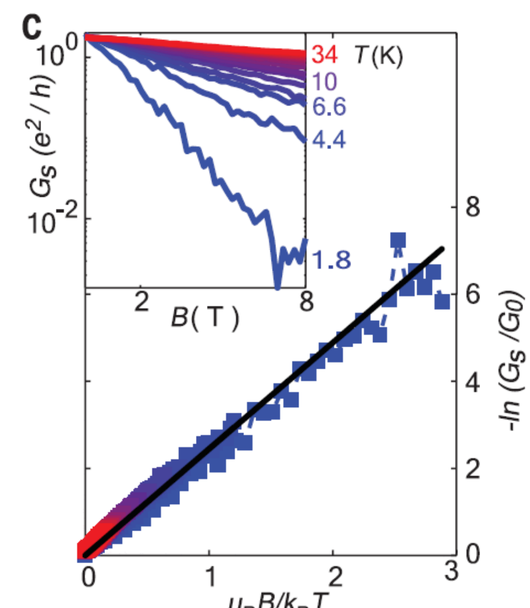


C. Cao (Quantum Spin Hall materials)



For longer channels the condcuctance becomes smaller:
Spin flip backscattering between the edge channels

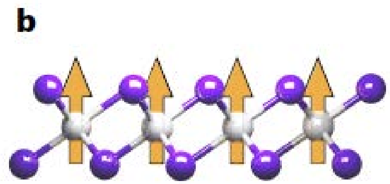
It also disappears (becomes gapped) in magnetic fields. Gap can be seen in activation plot
It persists up to temperatures of 100K



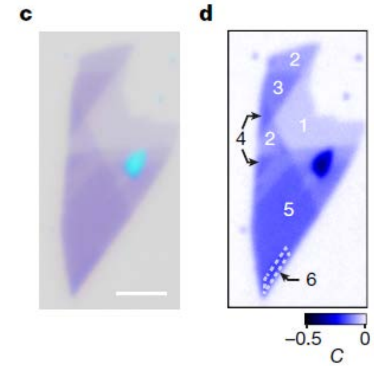
S. Wu et al., Science 359, 76 (2018)

Magnetic materials

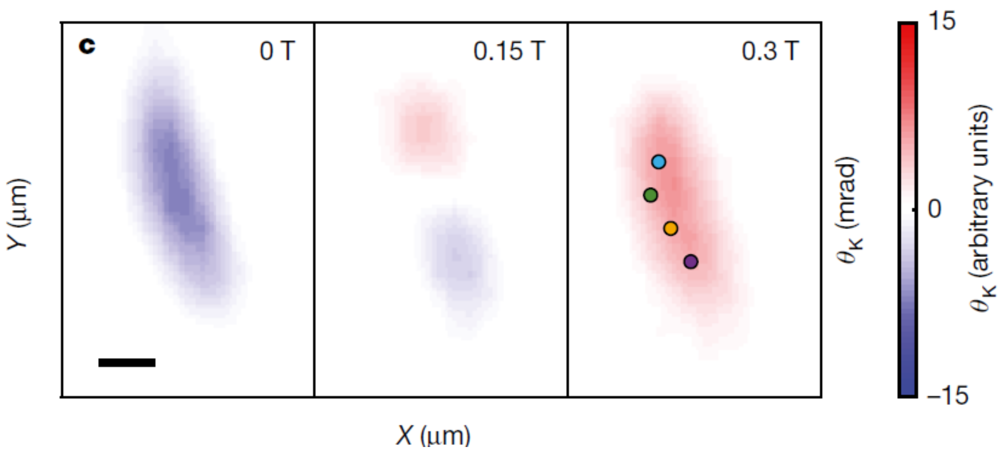
e.g. CrI3

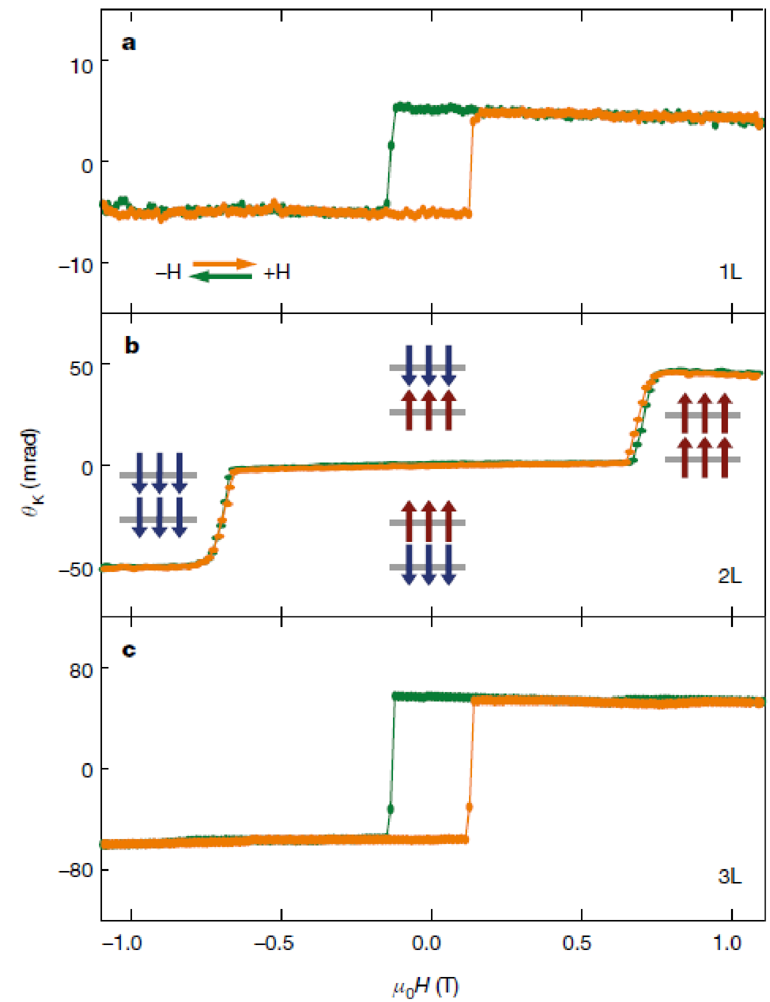


Different thicknesses can be measured on the same flake. Thickness identified from optical contrast and AFM.



Air- sensitive materials. Ising type of magnetism in bulk, with out of plane anisotropy and Tc of 61K. Here FM order is observed down to monolayer – real 2D ferromagnetism. Measure magnetism with Magneto optical Kerr effect (MOKE). See hysteresis loops and Tc=45K. Domains formed as seen from spatial mapping.
Trilayer FM state.
Bilayer AF state – different layer polarization. At large fields becomes polarized fully.

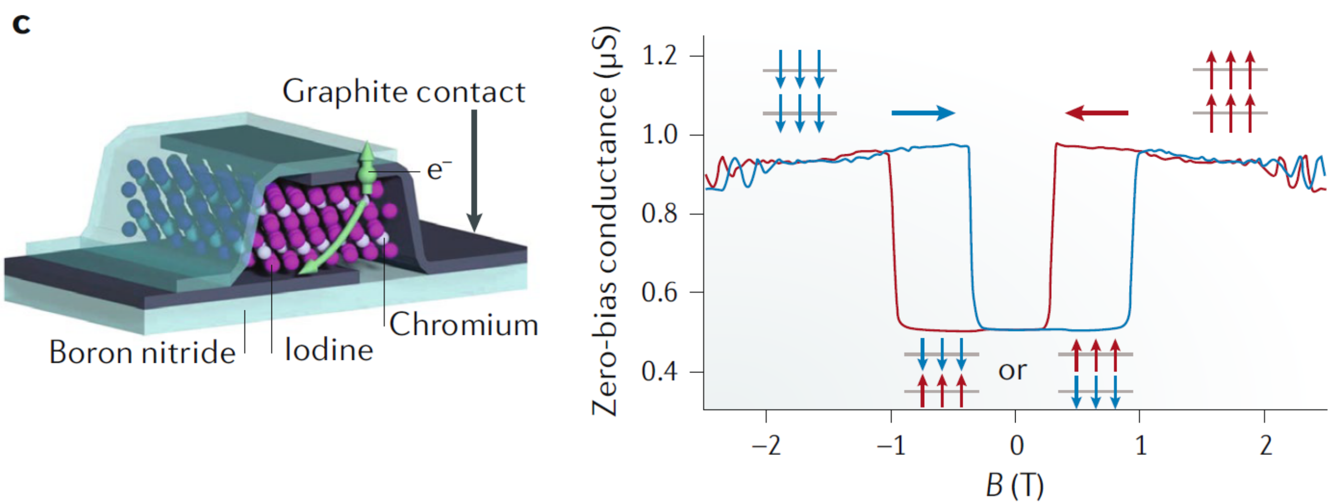




B. Huang et al., Nature 546, 270 (2017)

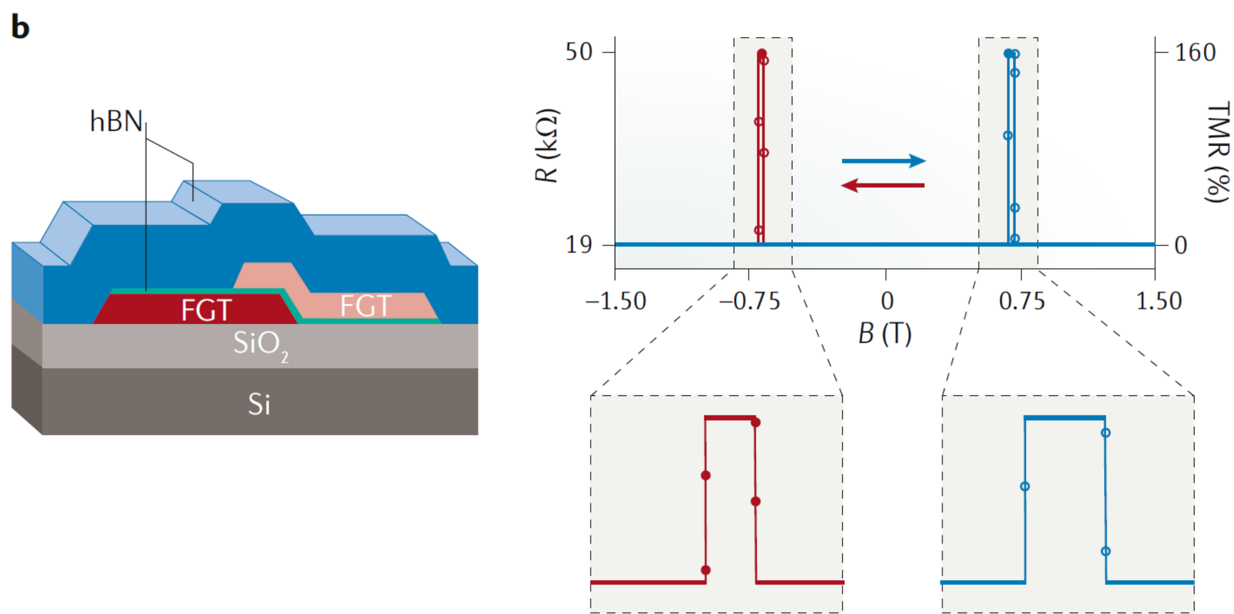
Magnetic materials

Spin filtering through BL CrI3
Tunneling probability depends on the orientation of the magnet (spin-dependent tunneling)
If in AFM phase (small B fields) for both spin direction the tunneling through one layer is really hard. In case of parallel aligment, for both layers it is intermediately hard – lower tunnel resistance.



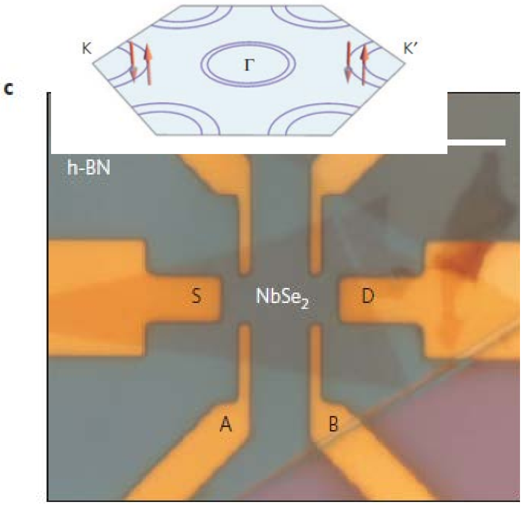
Spin valves: FM/hBN/FM layers.
Fe3GeTe2 – conducting magnets.
If the magnetization is parallel or antiparallel – different resistance.
They can be switched separately (e.g. due to difference in thickness).

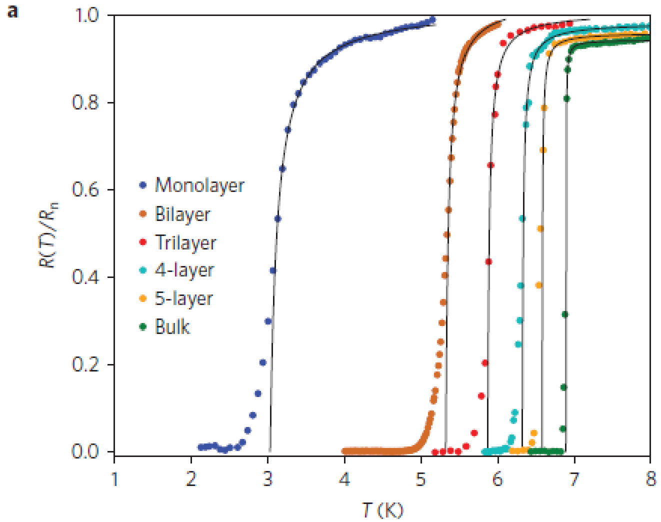
These thing magnets are electrically tuneable, e.g. Switching field, Tc can be changed, or even can be switched.

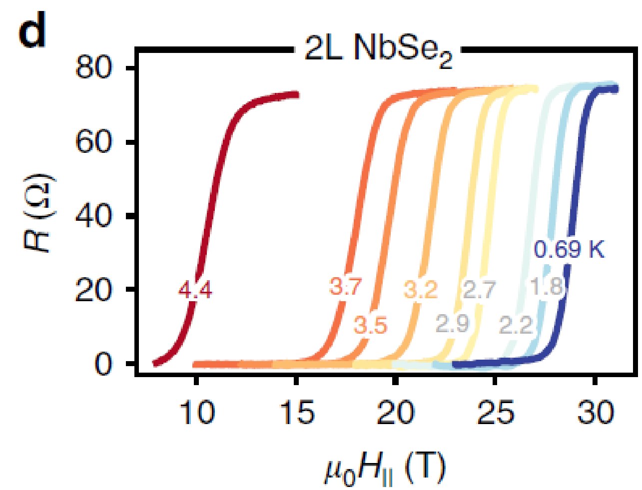


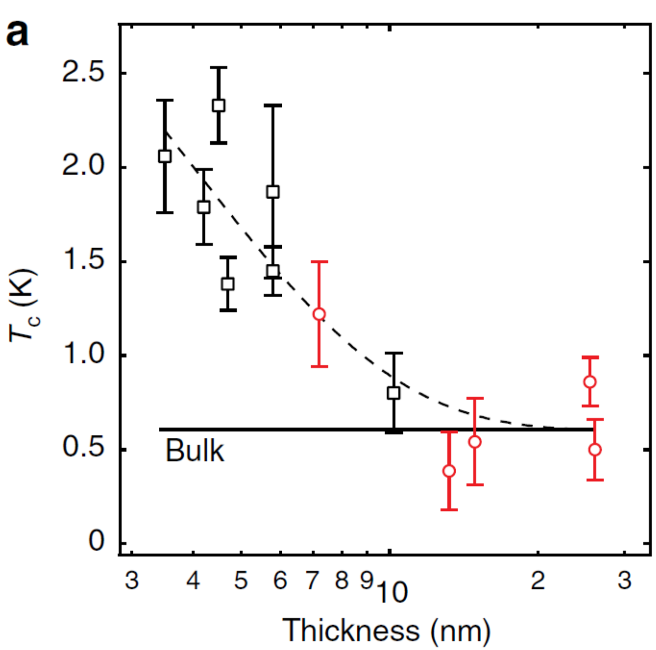
Kin Fai Mak et al., Nat. Rev. Phys., 1, 646 (2019)

Superconducting materials





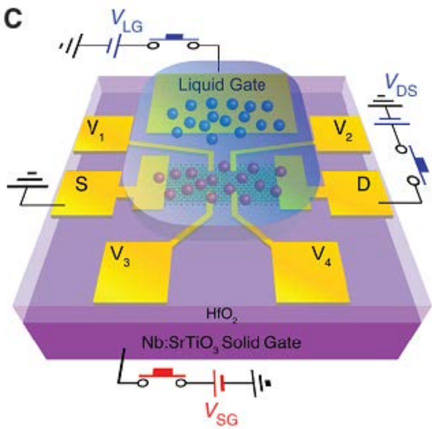




2D superconductors like: NbSe2, TaS2, down to monolayer
For NbSe2 Tc decreases with thickness
For TaS2 it increases
Test for 2D superconducting theory
Strong spin orbit (Ising pairing) – pairing in opposite valleys →
SC states survives up to large in plane magnetic fields

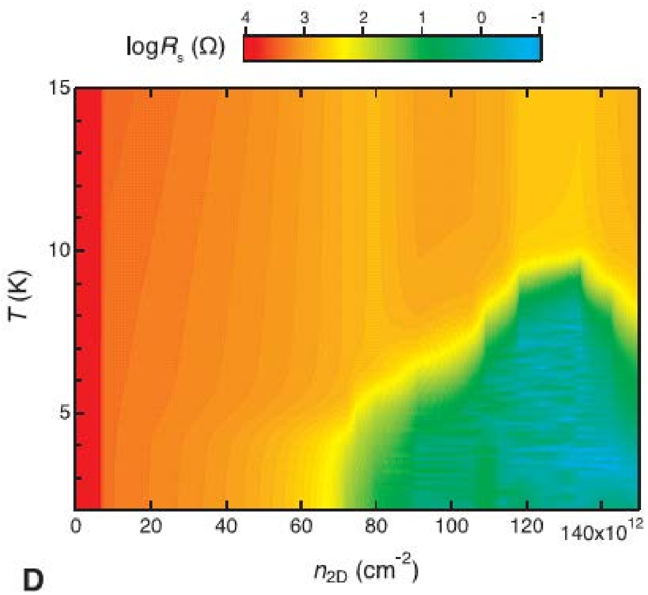
S. C. de la Barrera et al., Nat. Comm., 9, 1428 (2018)
X. Xi et al., Nature Phys. 12, 139 (2016)
E. Navarro-Moratalla et al., Nature Comm. 7, 11043 (2016)

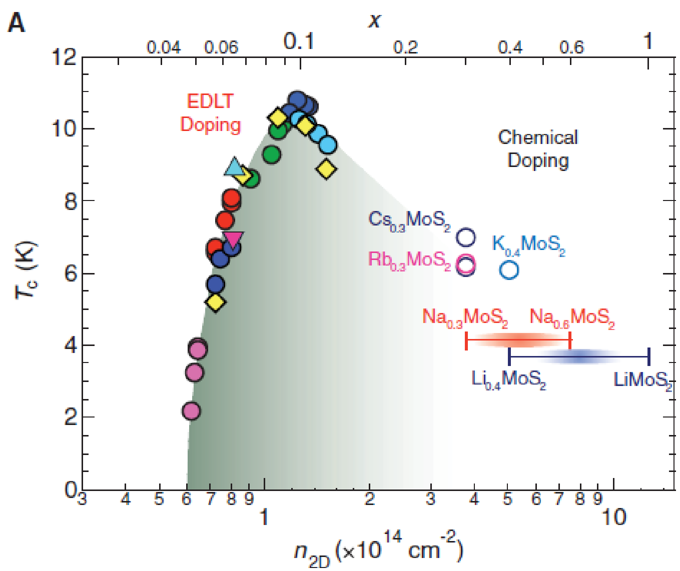
Induced superconductivity: MoS2

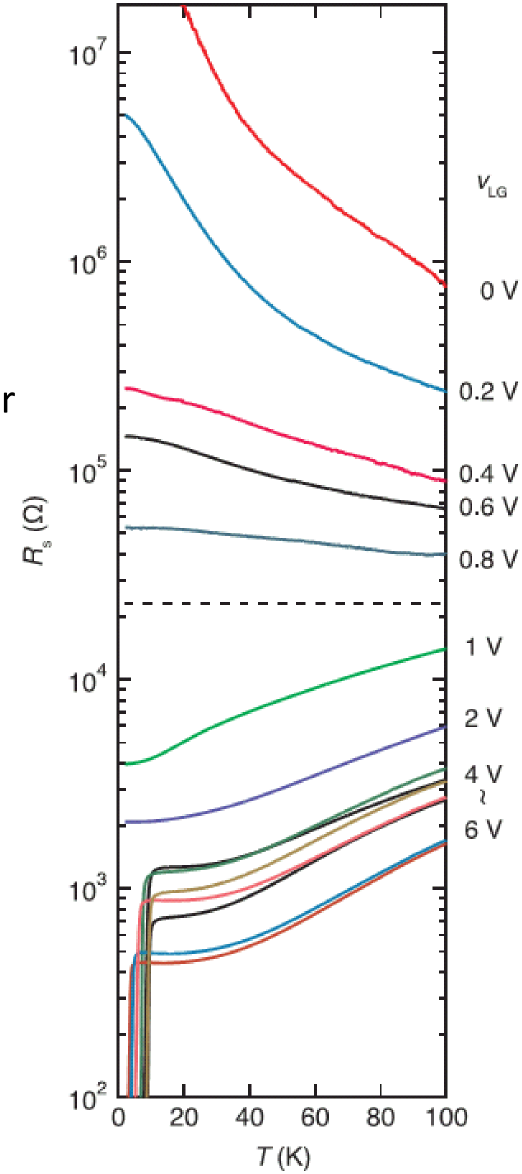


Use electrolyte gating to achieve high density: the electrons sit directly on the surface

For low gate voltages negative temp. derivative – semiconducting behaviour
For higher it changes to metallic behaviour, and for even higher voltage SC transition observed
From hall measurement – critical density for metal-ins. transition: $6.7\ 10^{-12}\ cm^{-2}$



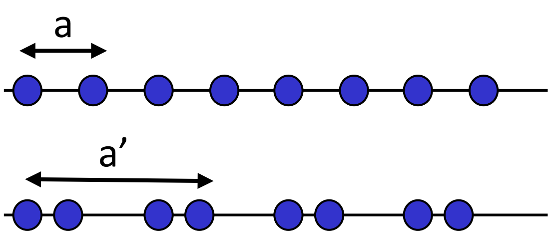


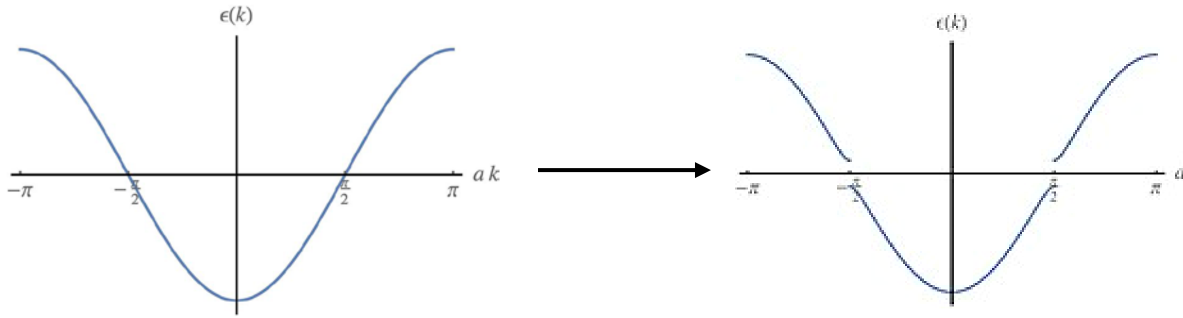


Superconducting T_c could be tuned with gate voltage
It is much simpler than real doping with other materials, since it is gate tuneable, and does not induce disorder

*J. T. Ye, Science **338** 1193 (2012)*

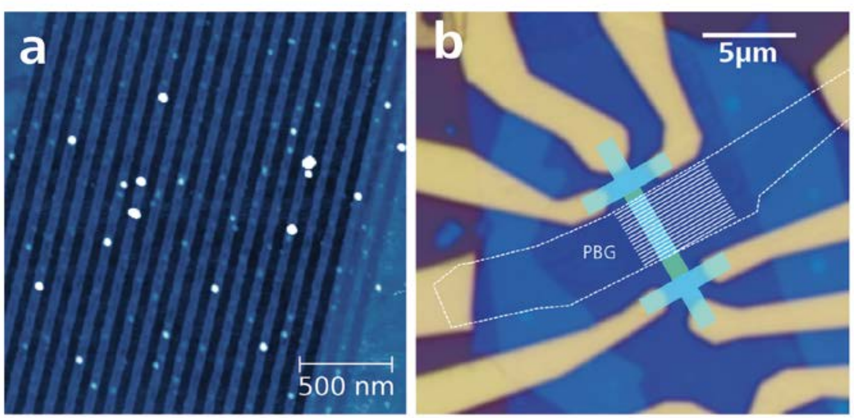
Graphene/hBN Moiré

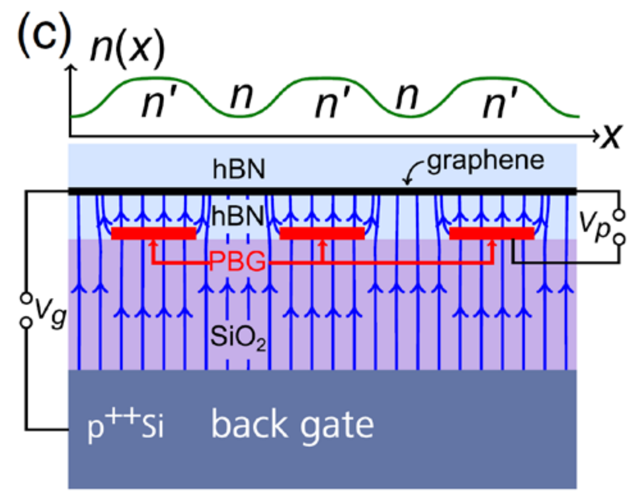




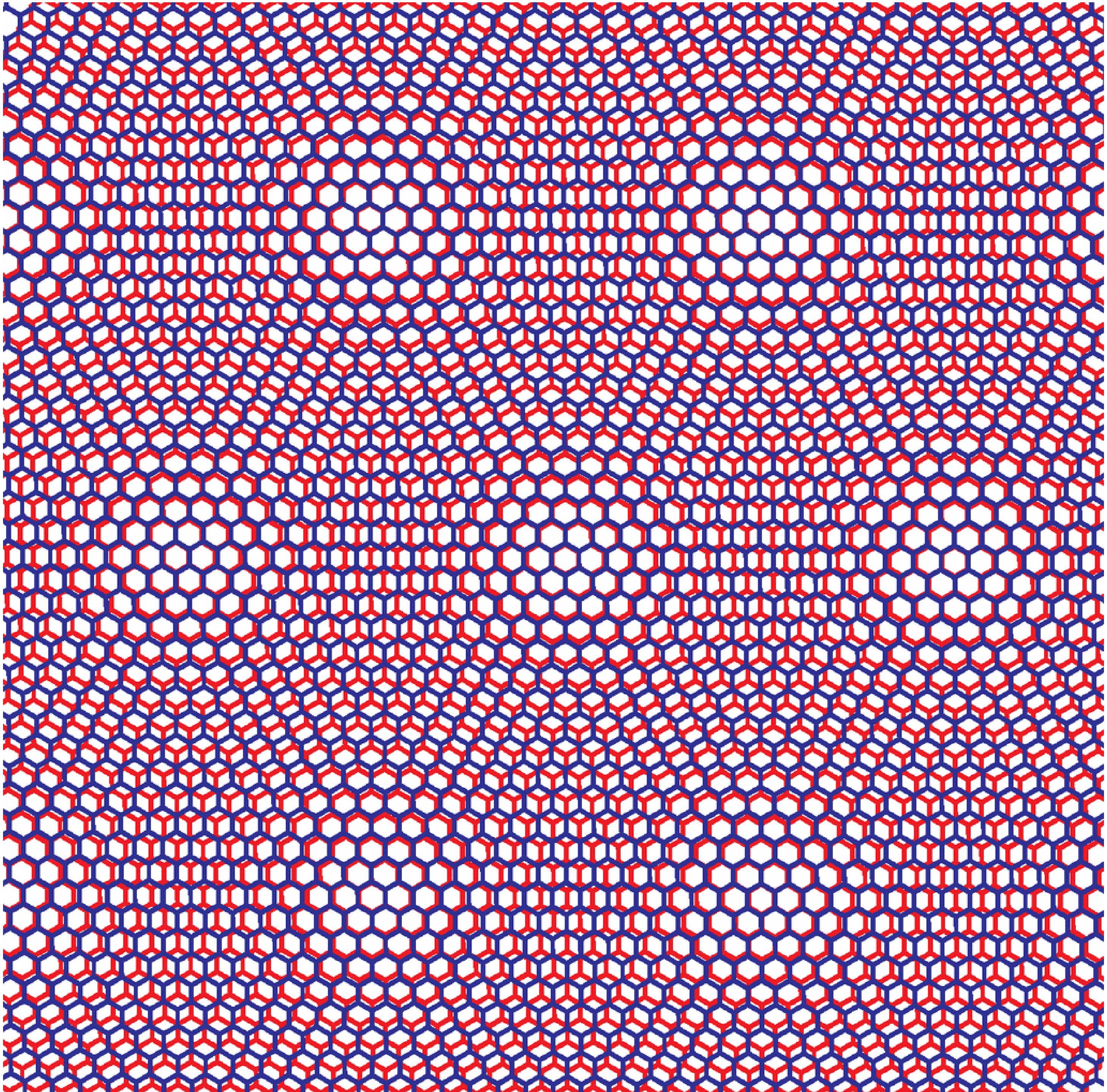
Peierls distortion: 1D simple model with lattice distortion (can be additional periodicity in the potential)
New unit cell introduced → BZ boundary changes, gap opens on the new boundary – for half filling metal-insulator transition.
With graphene?

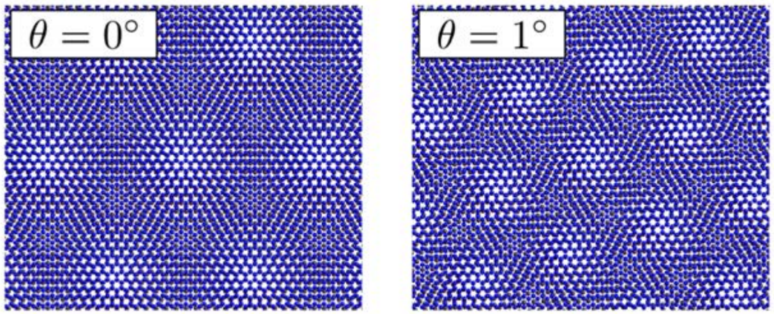
First idea: using a periodic gate array – hard to fabricate



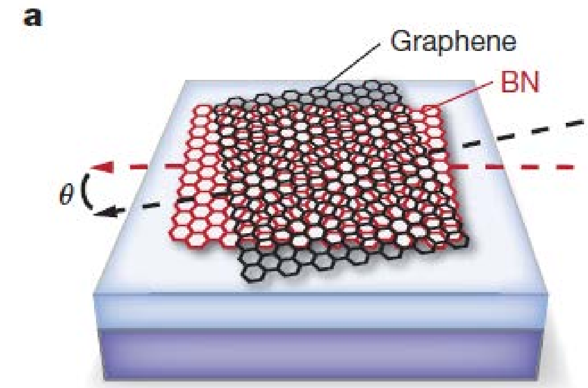


M. Drienovsky et al., Phys. Rev. Lett., 121, 026806 (2018)

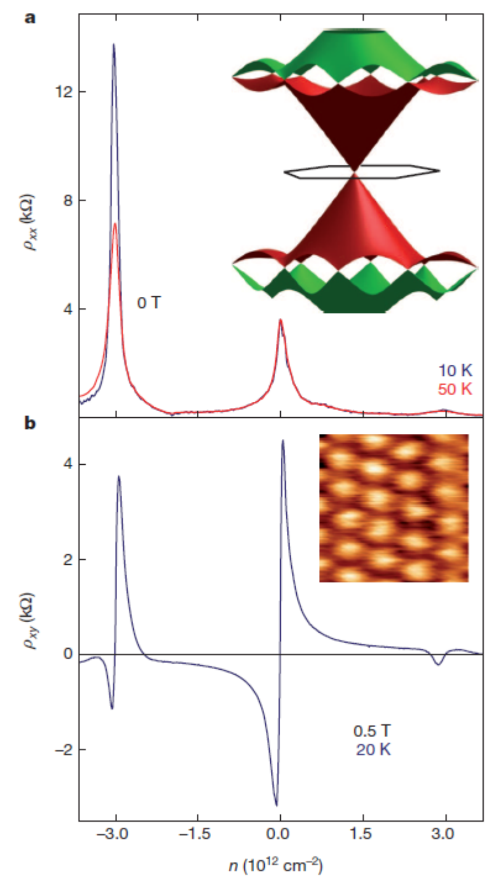




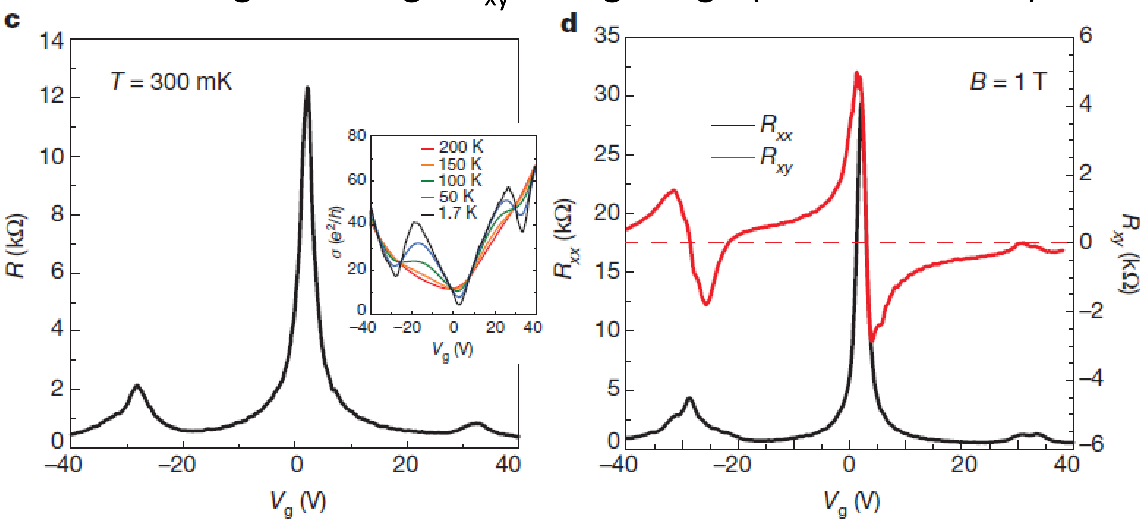
Graphene/hBN Moiré



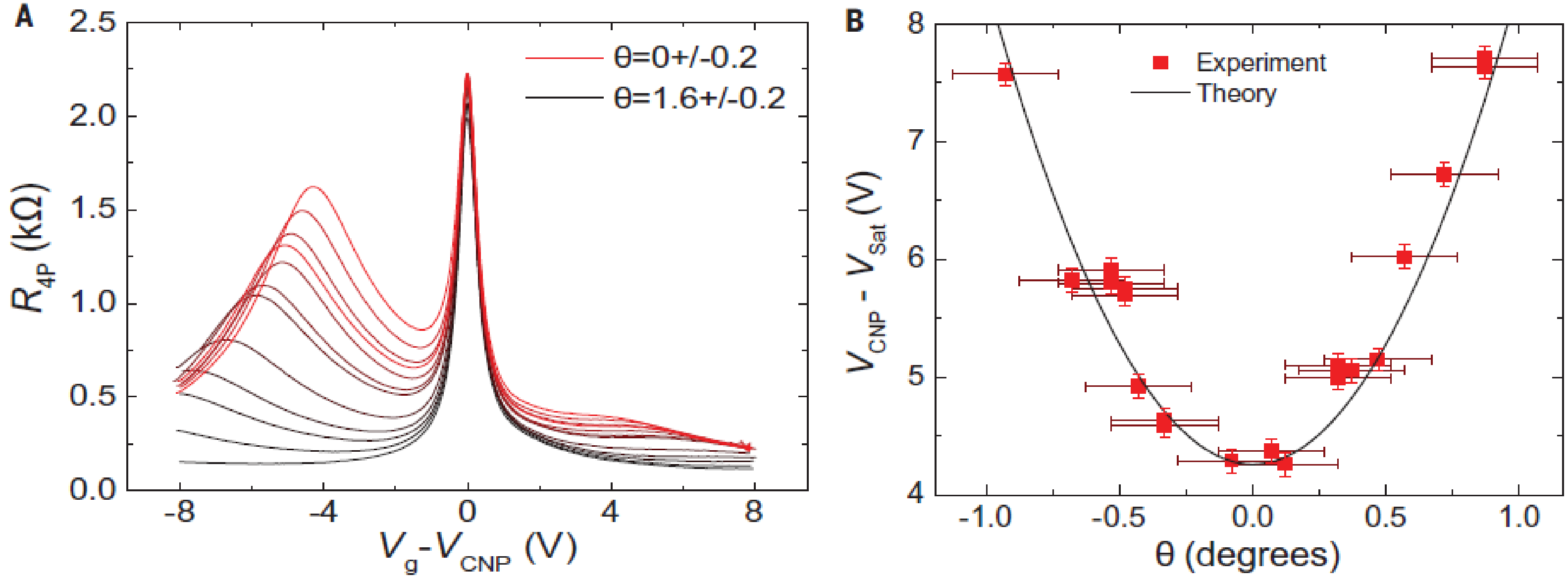
By placing graphene on hBN, a Moiré superlattice can be formed.
Similar lattice constant. hBN acts as a potential modulation – a superlat
can be formed. Wavelength depends on rotation angle.
For small rotation angle long superlattice forms – observable at reasonable low energies. Mini Brillouin zones form.



Complex band rearangement – and secondary Dirac points form.
In resistance they show up as new peaks at high doping.
At the same gate voltages r_{xy} changes sign (electrons-holes)

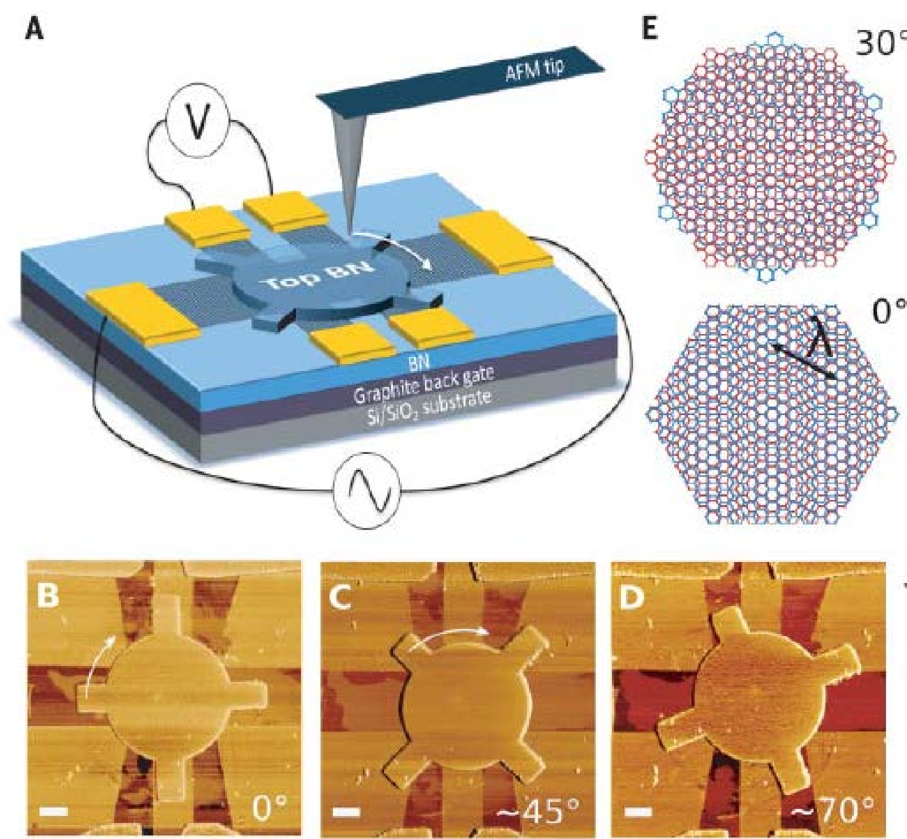


C. R. Dean et al., Nature 497, 598 (2013), L. A. Ponomarenko et al., Nature 497, 594 (2013)
J.R. Wallbank et al., Ann. Phys. (Berlin) 527, 359 (2015)



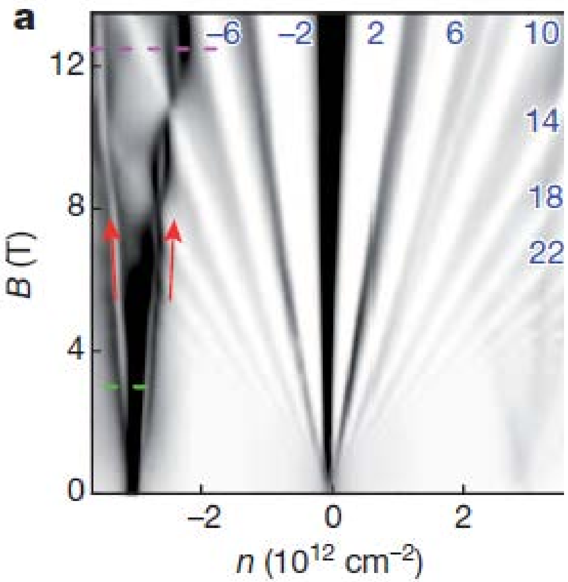
$$\lambda = \frac{(1+\delta)a}{\sqrt{2(1+\delta)[1-\cos(\theta)]+\delta^2}}$$

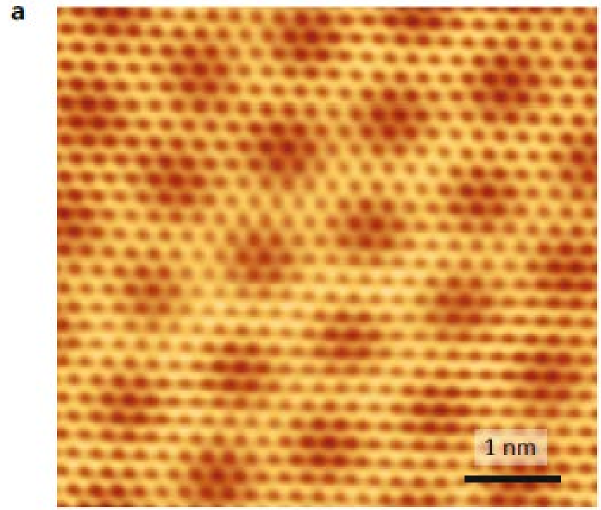
δ:Lattice constant difference
θ: rotation angle
λ: superlattice wavelenght



From simple geometric arguments superlattice peridocity → energy (density) of secondary CNP can b calculated.
Measurement: rotatable top hBN layer with AFM – an simulataneous transport measurement
V_{sat} shifts according to theory

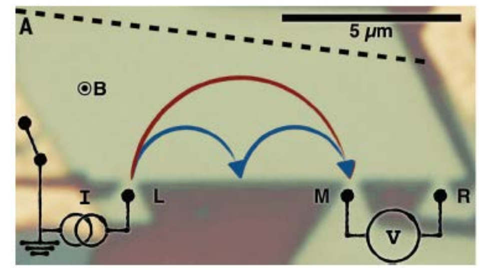
R. Ribeiro-Palau et al.,
Science 361, 690–693 (2018)





In STM measurements both the normal lattice and the superlattice is visible

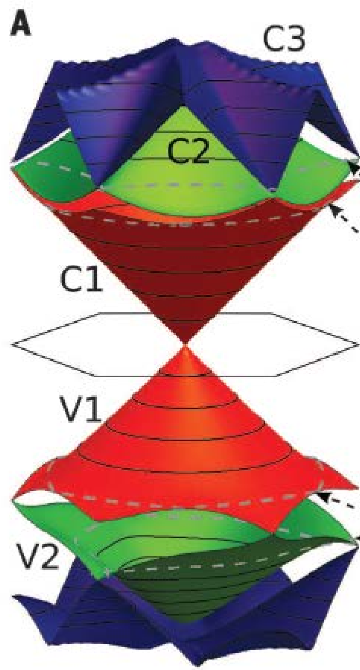
Strain patterns might appear – which can also open small bandgap in graphene at the CNP

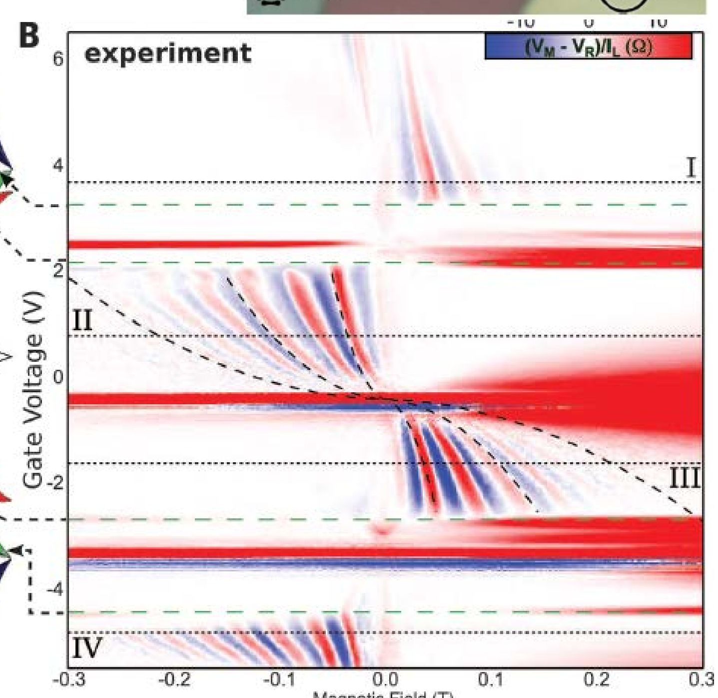


Also quantum Hall plateaus fan out from the secondary CNP
Additional magneto oscillations appear – Hofstadter physics – coexistence of two energy periodicities: Bloch and Landau

Electron focusing experiment. B field sign has to be changed at the regular CNP (e→h).

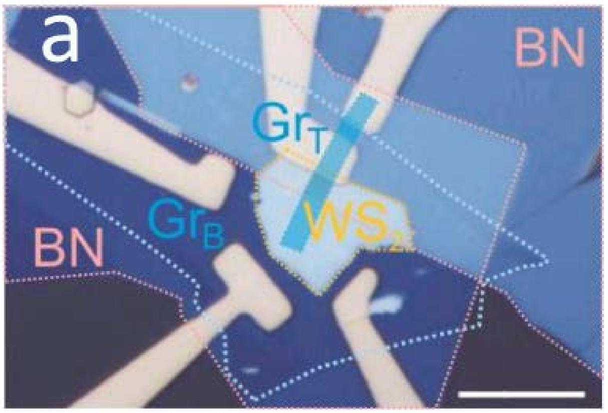
It also has to be changed at the satelite CNP!



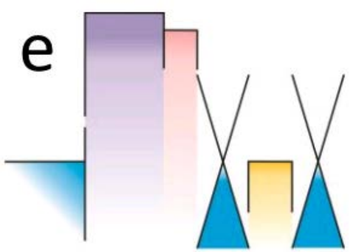


L. A. Ponomarenko et al., Nature 497, 594 (2013)
M. Yankowitz et al., Nat. Rev. Phys. 1, 112 (2019)
M. Lee et al., Science 353, 1526 (2016)

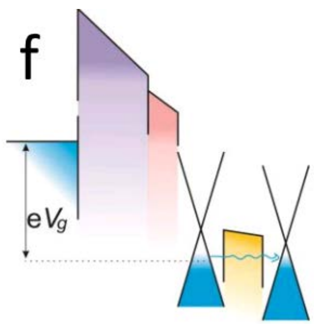
Tunnelling



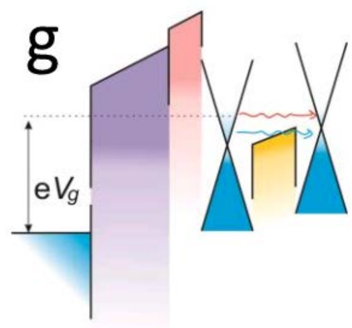
Graphene FET separated with WS_2 from topgate (graphene)
Small gap compared to BN, control of E_f on the scale of barrier height
→High on/off ratio



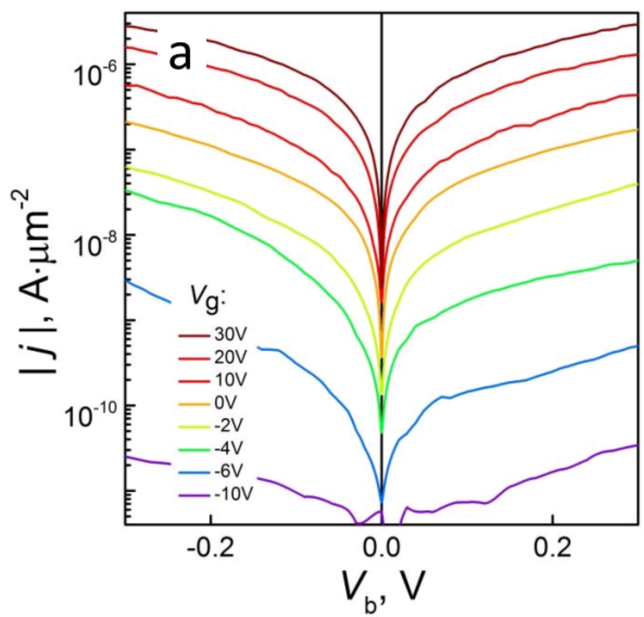
No Bg applied
Graphene aligned at bottom of WS_2 conduction band



Negative BG applied
Changes the carrier concentration
The barrier height increases
OFF state



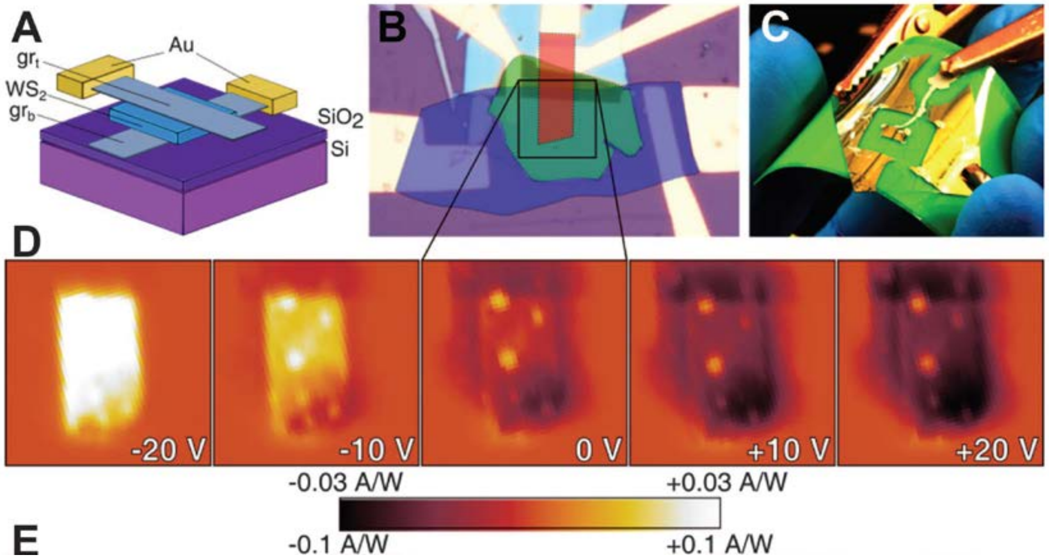
Positive BG applied
Populates conduction band
The barrier height is reduced and thermionic current appears
ON state



T.Georgiou et al., Nature Nano. **8**, 100 (2013)

Photovoltaic structures

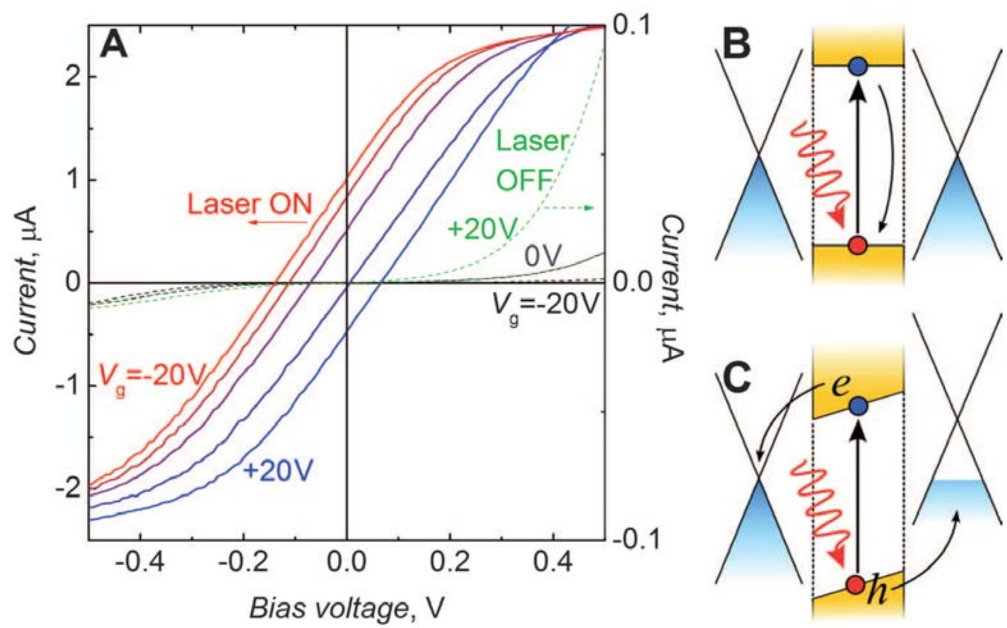
TMDC tunnelling devices



Photovoltaic structure:
hBN/G/WSe2/G/hBN
WSe2: active material
G: electrode
Under illumination photocurrent is generated where the G and WSe2 layers overlap
Photocurrent gate tuneable (e.g. with SiO2 backgate)
Also works on flexible substrate

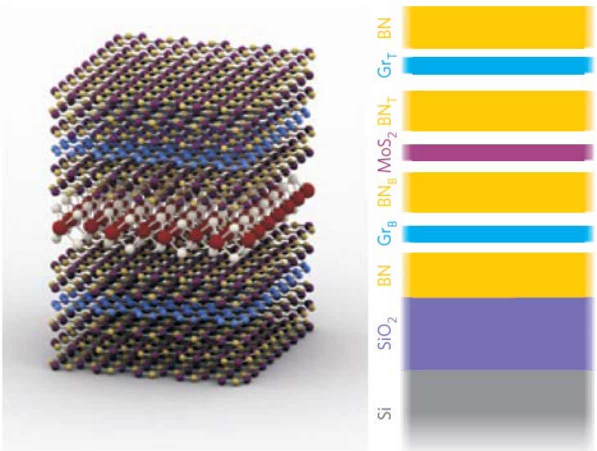
Light generates electron hole pairs
Without electric field device is symmetric – no photocurrent present

If built in electric field is present (either gate or source-drain voltage) than finite photocurrent is generated
Large response due to strong absorption



L. Britnell et al. Science 340, 1311 (2013)

Light Emitting Diodes

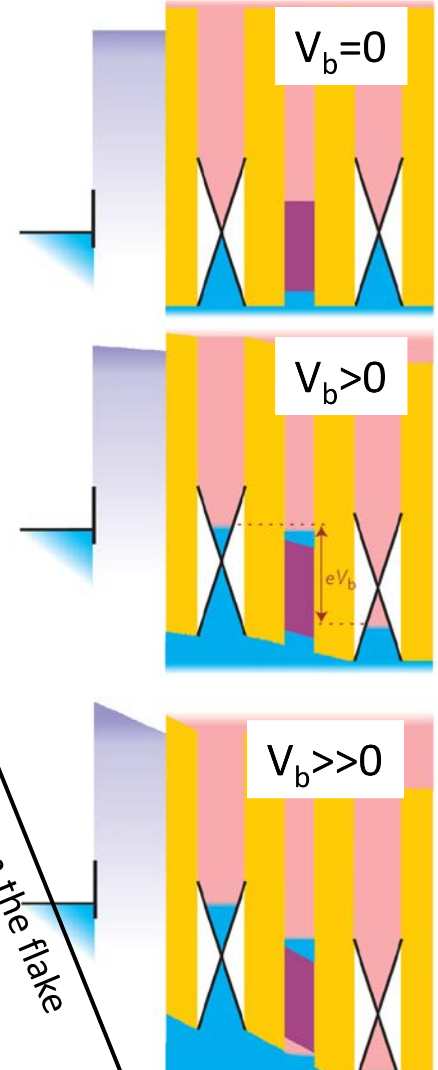


For V_b=0 symmetric band alignment (In PL spectrum neutral A exciton seen)

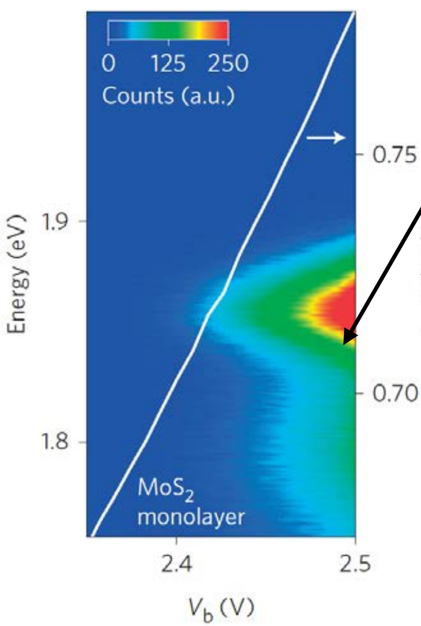
For increase V_b tunnel current through the device

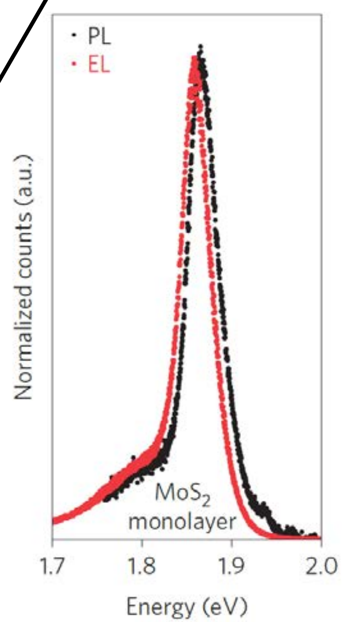
At a threshold V_b conduction band of MoS2 starts to be populated (trion appears in PL)

Even higher voltage holes can be injected to valence band: electrons and holes can be recombined and electroluminescence appears



EL spectra





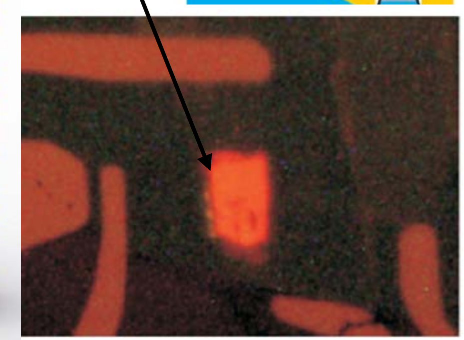
Matches PL – recombination of trions

Yield – 1%

Using multiple structure yield can be increased

Can be boosted to 100%

emission from the flake

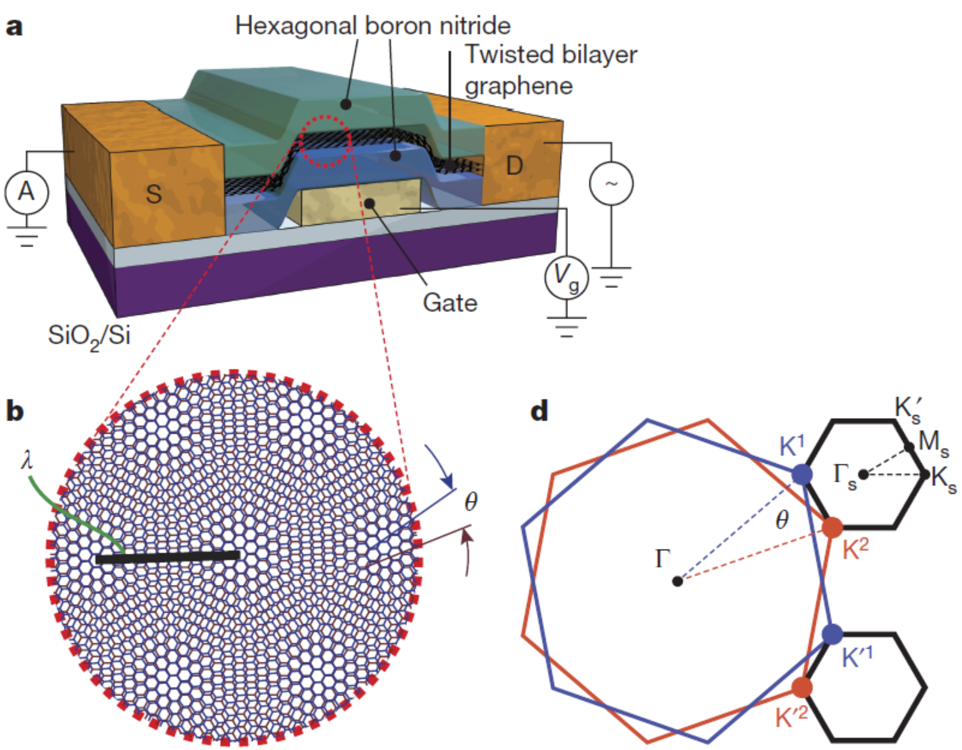


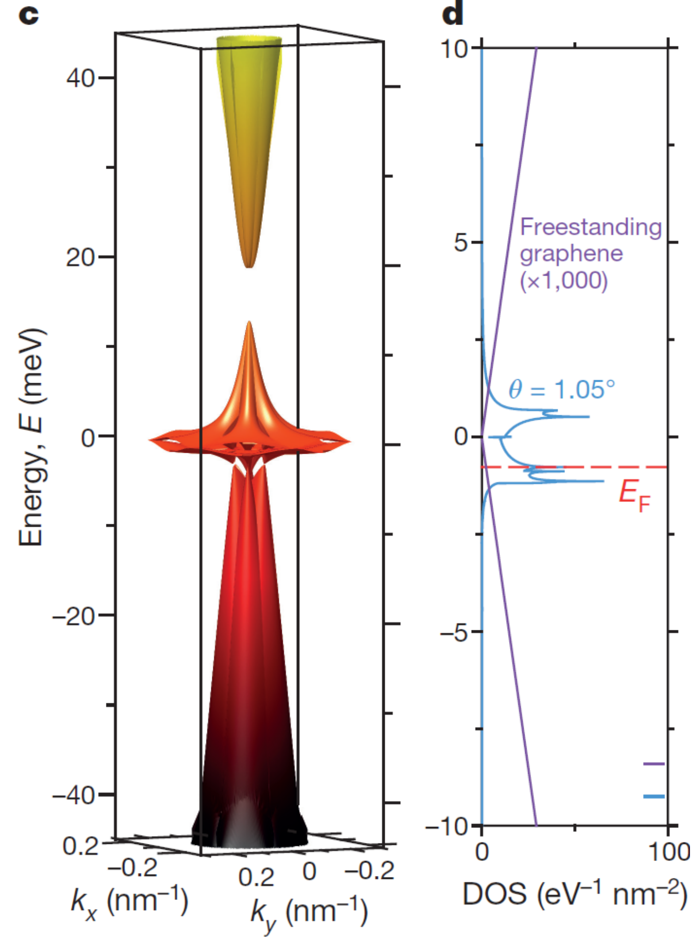
F. Withers et al. Nat. Materials. 14, 301 (2015)

Interactions in flat bands - TBG

Usually electron-electron interactions can be neglected
Kinetic energy usually dominates and only corrections are given to Fermi liquid theory from e-e interaction
However if narrow bands form (heavy system), correlations might become important
E.g. Mott transition (small t, large U in Hubbard picture)

Twisted BLG: Two layers can form a Moiré →band reconstruction. For certain *magic* angles the bands become flat.





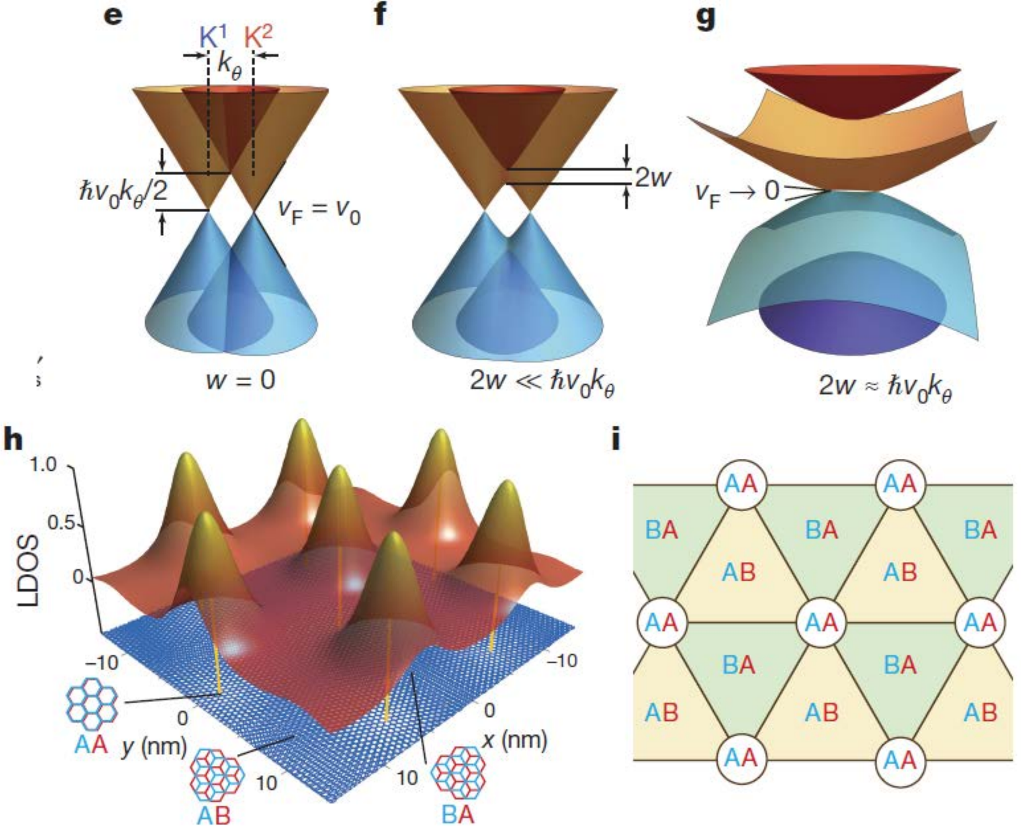
Precize alignment is needed.

Take a flake, tear part of it off, rotate and place down. Encapsulate than fully.

Y. Cao et al., Nature 556, 43 (2018)
Y. Cao et al., Nature 556, 80 (2018)

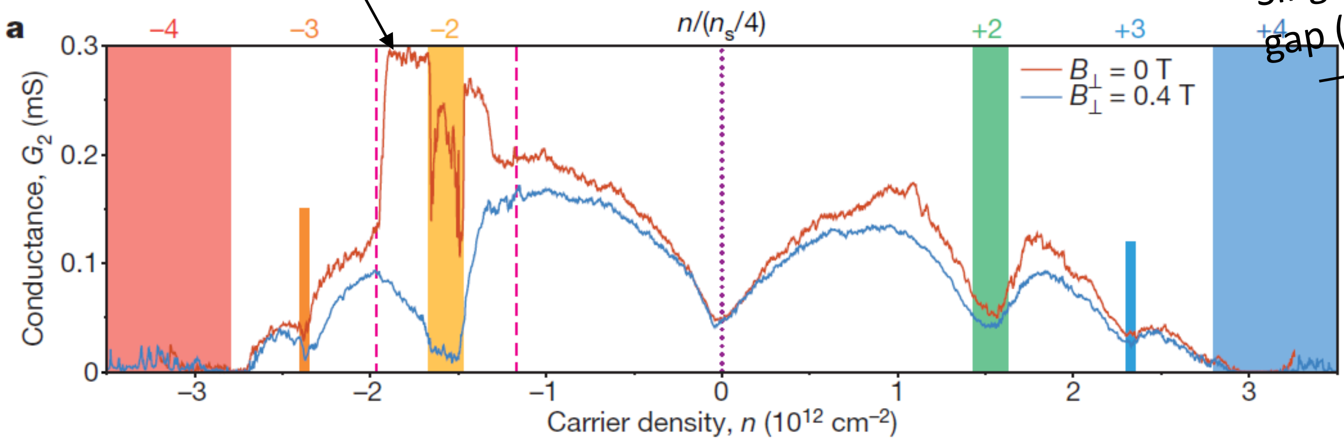
TBG

Two overlapping cones in momentum spa[c]
Interaction between the layers gives small avoided crossing (2w). For magic angles th[e] interaction dominates and flattens out the bands.
In real space AB/BA stacking regions oscill[ate] and small AA stacking parts appear. Here the wavefunction localizes

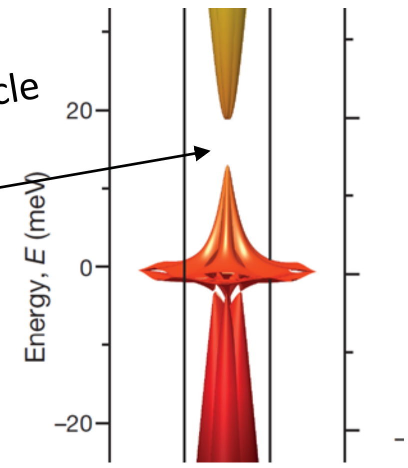


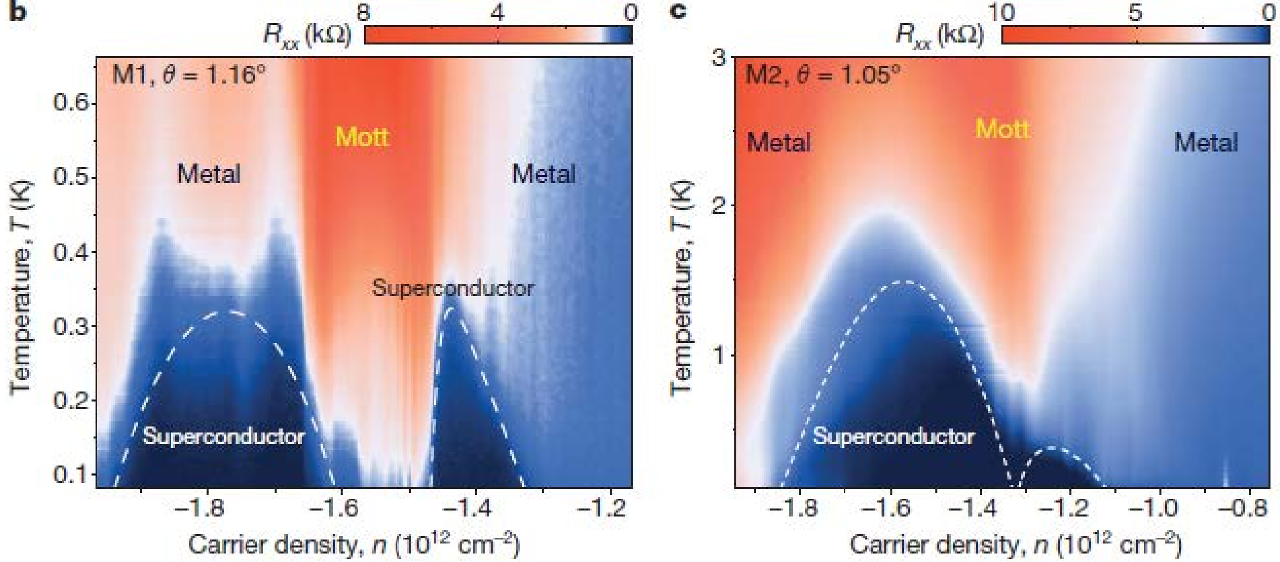
Measurement:
Blue/purple regions: single particle gaps
@0.4 T gaps (low conductance) appear at several densities (yellow, green box)
→Mott phase
@0T large increase in conductance at negative voltages → SC phase

SPG



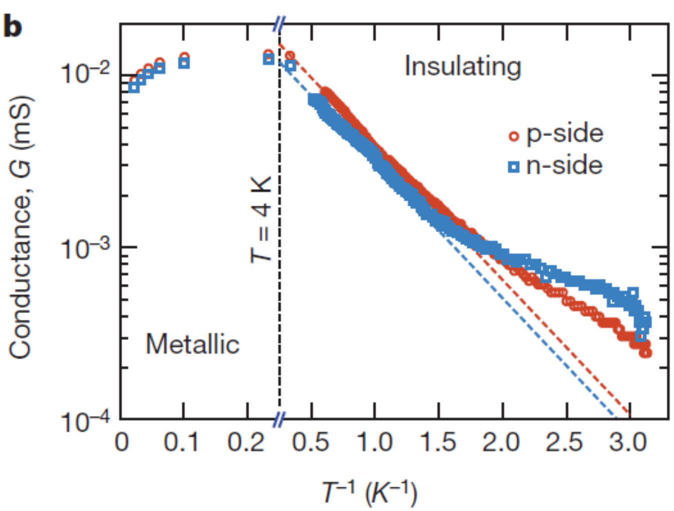
Single particle gap (SPG)

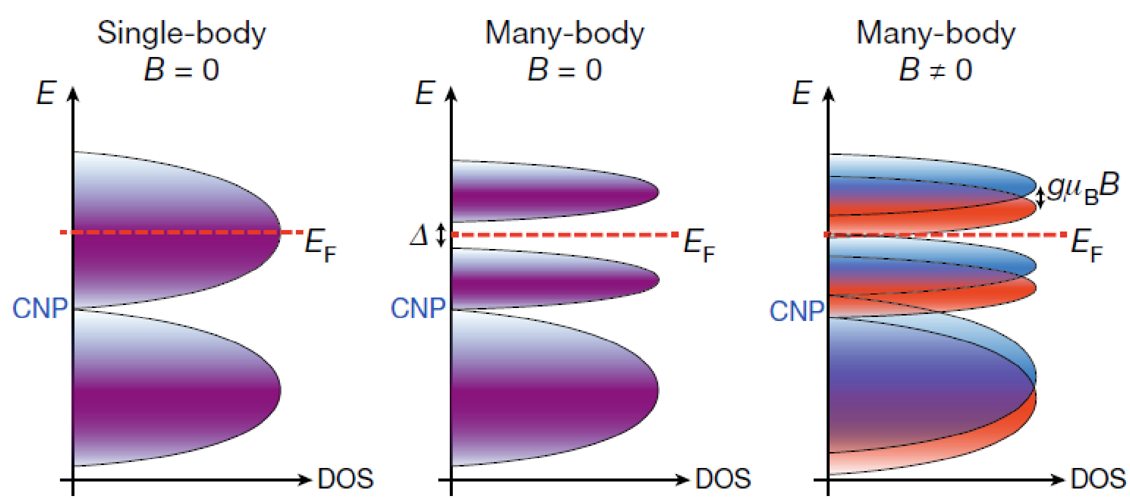




SC phase appears with zero resistance with Tc up to 1-2 K.
It is sourranded by insulating phases – „dome" structures
Resambles the phase diagram of high Tc SCs, however now this is gate tunable.
Around 0.4 T, the SC phase vanishes.

Insulating phase is a Mott phase.
Only appears below 4K.
Many-body gap opens.
It disappears around 6T.

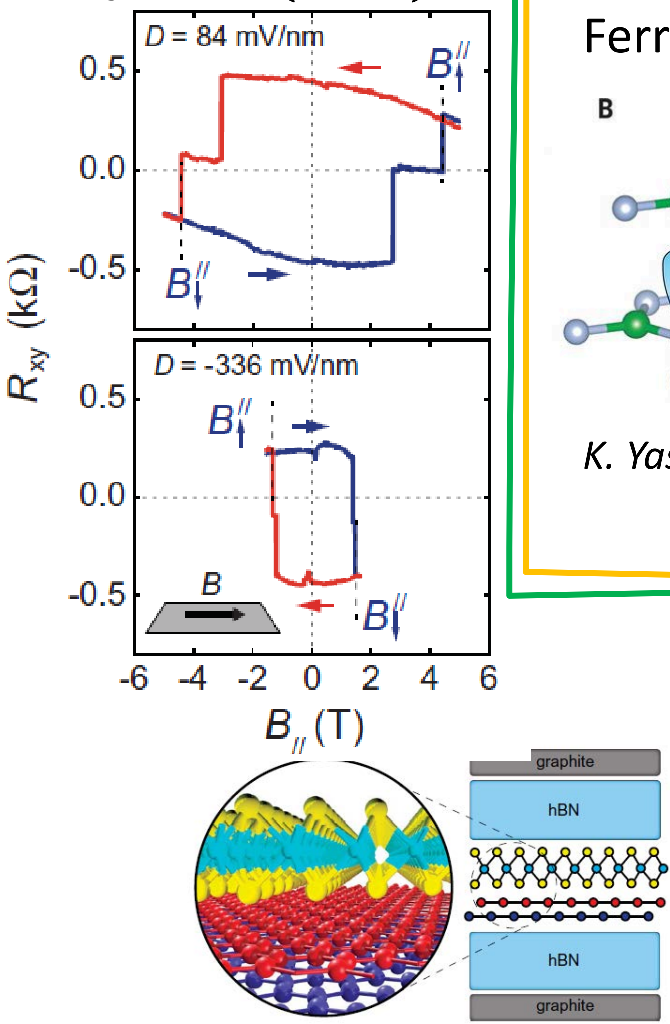




Twisted…

TBG/WSe2

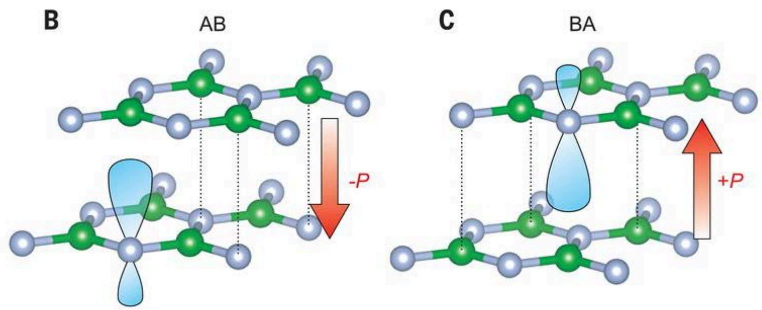
Chern insulating at half filling of TBG ($\nu = 2$)



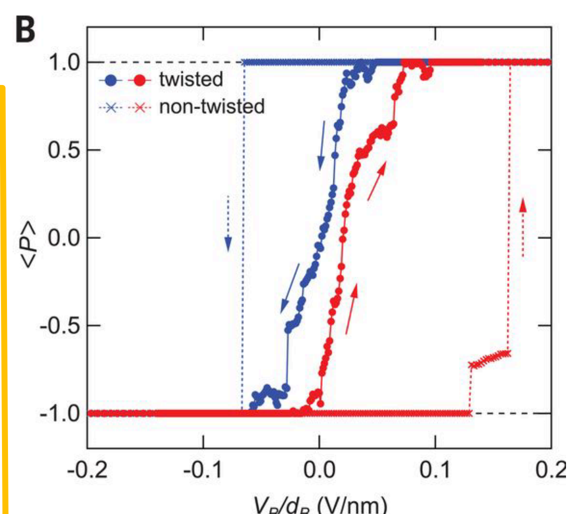
Lin et al., Science 375, 437–441 (2022)

Twisted hBN

Ferroelectricity

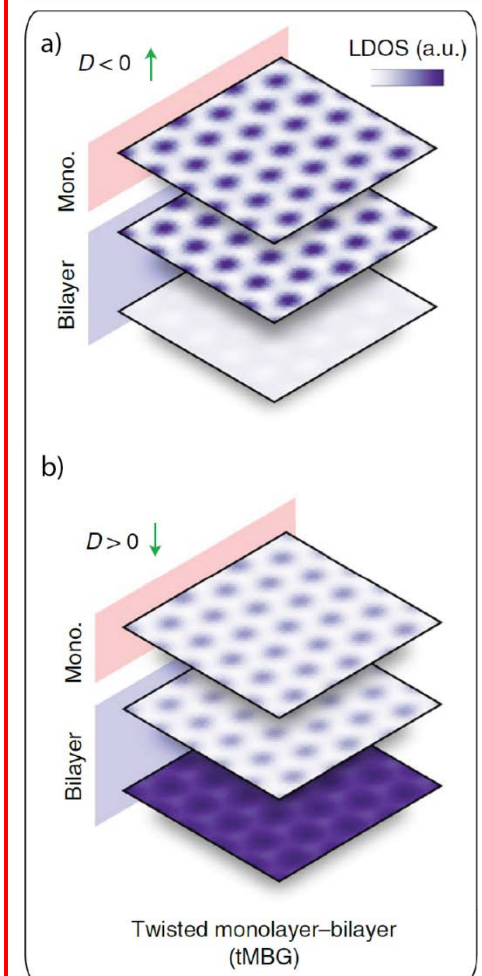


K. Yasuda et al., Science 372, 1458 (2021)



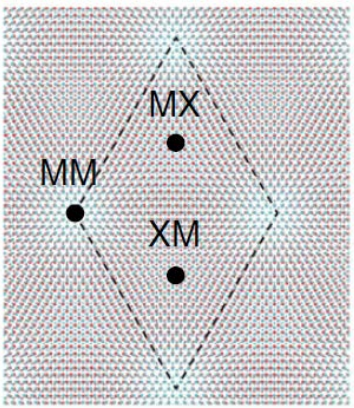
Mono-bilayer graphene

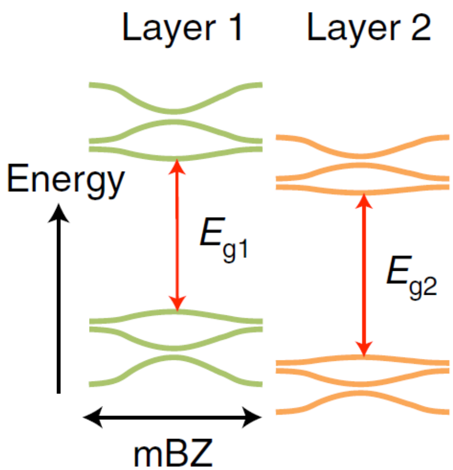
Spin polarized states, superconductivity

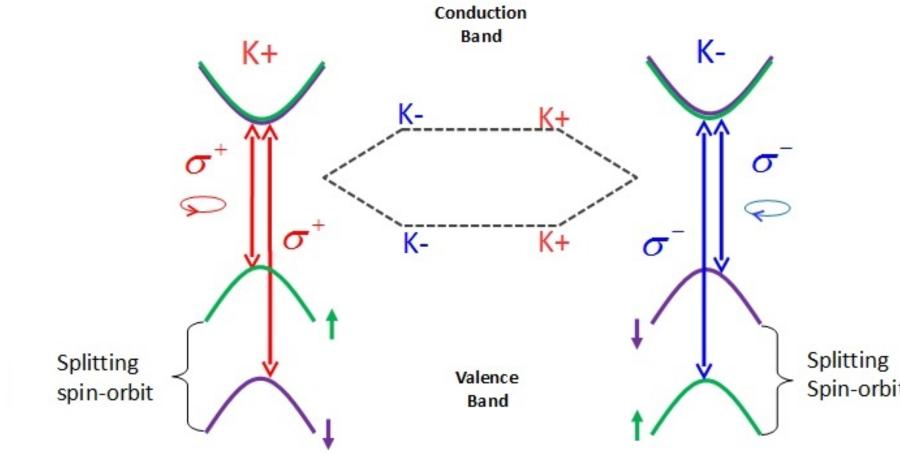


S. Chen et al., Nature Phys. ***17****, 374–380 (2021)*

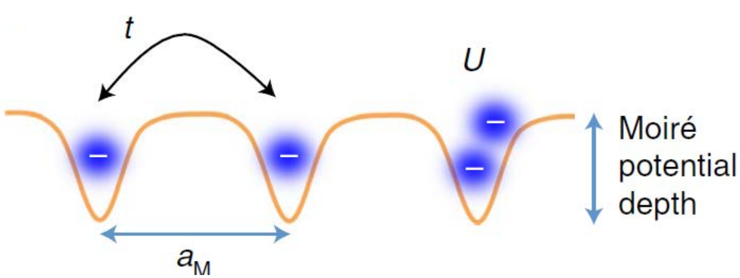
Twisted TMDs





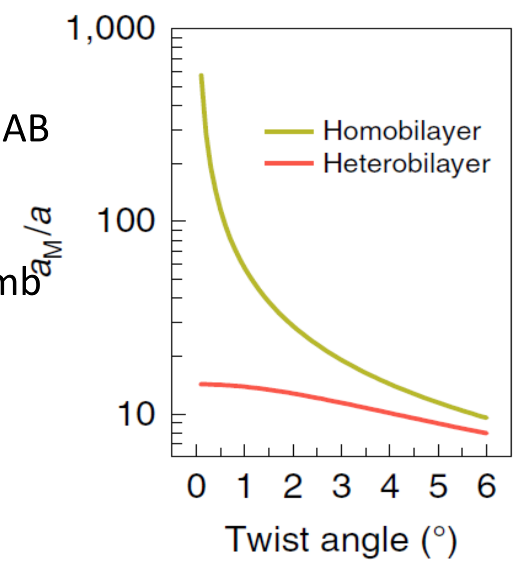


$$W \approx 1\text{–}100\ \text{meV}$$
$$U \approx 100\text{–}200\ \text{meV}$$



- Homo or heterobilayers (same or different materials)
- No magic angle condition
- Usually local description works and Hubbard type physics arises
- Description like movement and localization on moiré potential

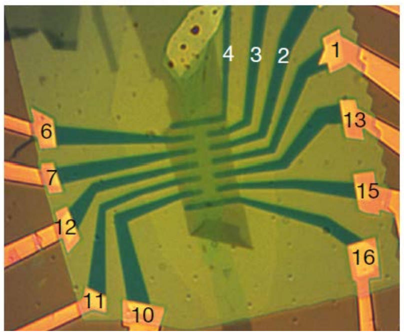
- Different stacking configurations: AA & AB
- Strongly correlated regime (U>>W) usally (even intersite Coulomb is large)

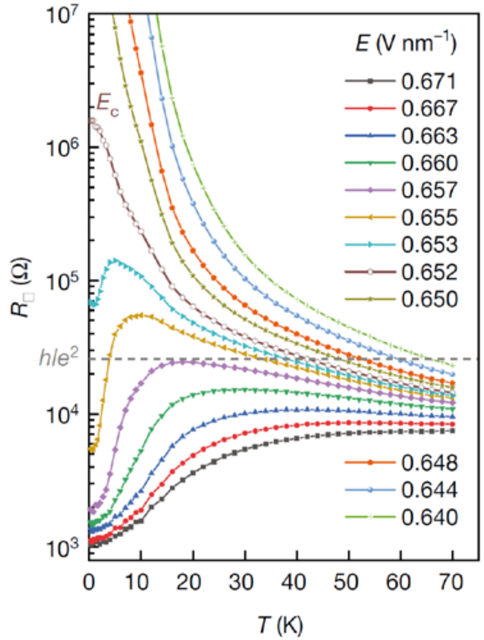


- For layer asymmetry (hetero bilayers or homob. with displacement field) the layer degeneracy is broken. Spin is broken by SOC in TMDs. Only two-fold valley degen. remains for flat bands

K. F. Mak et al., Nat. Nano 17, 686 (2022)

Mott transition in twisted TMDs

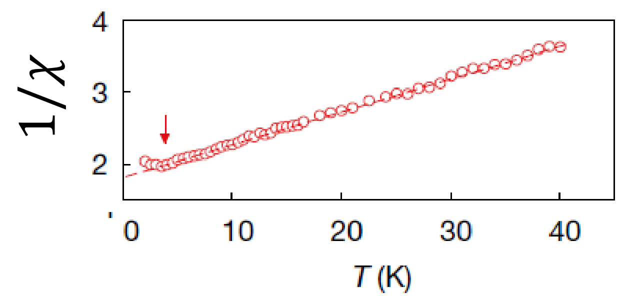


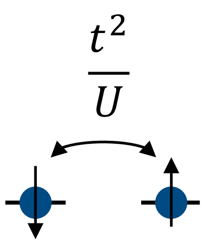


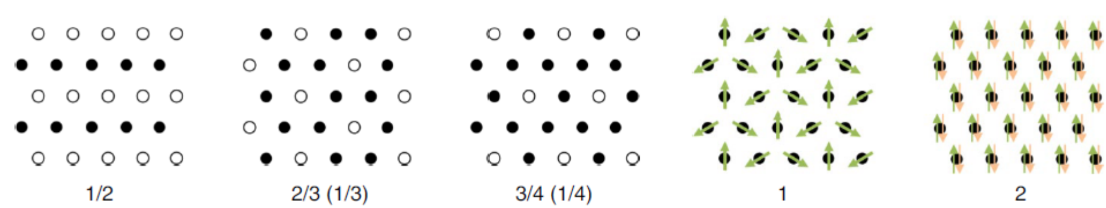
WSe2/MoTe2 structures with dual gating

- Measurements at half filling shows a large gap for certain displacement field.
- According to single particle physics it should be conducting
- Mott gap appears and insulates
- With displacement field band alignment and moiré depth is tuneable – can reach U~W condition and can go through transition with E-field.
- Can investigate scaling, gaps etc.

- Suspected order: AFM (canted on triangle lattice)
- AFM coupling should give Curie-Weiss law with negative T
- Susceptibility measurements with optics (MCD) shows CW-law

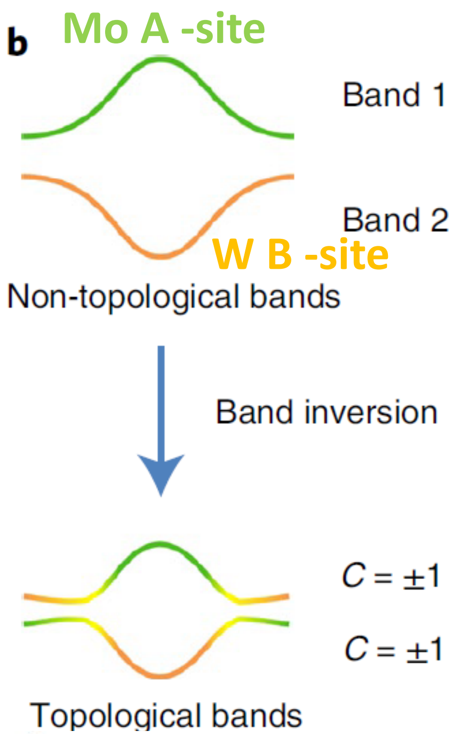


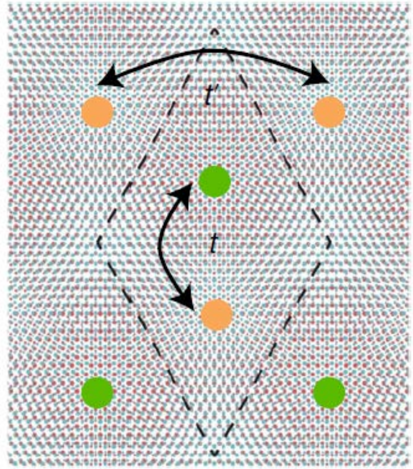




T. Li et al., Nature 597, 350 (2021)

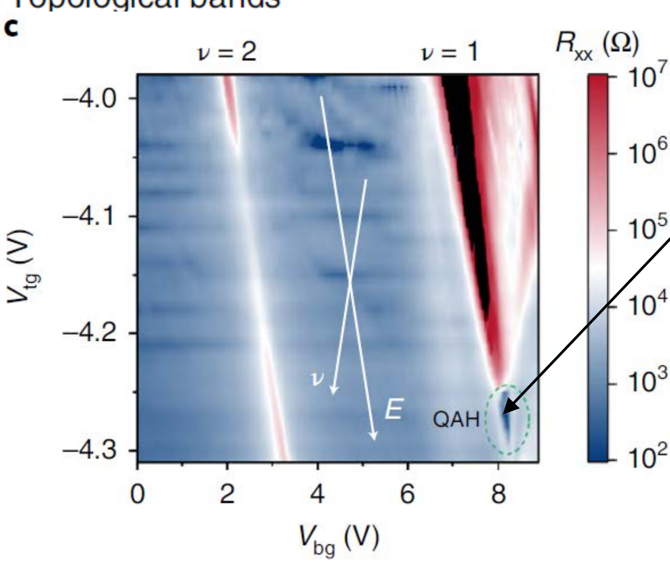
Topology in twisted in WSe2/MoTe2

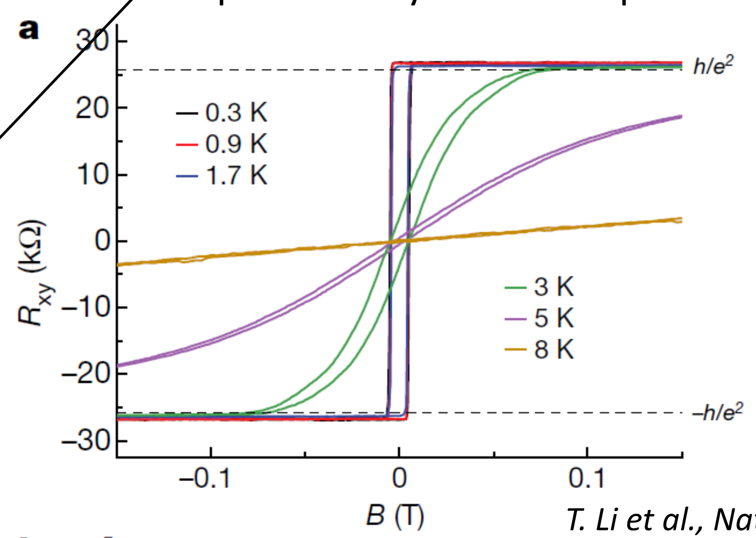




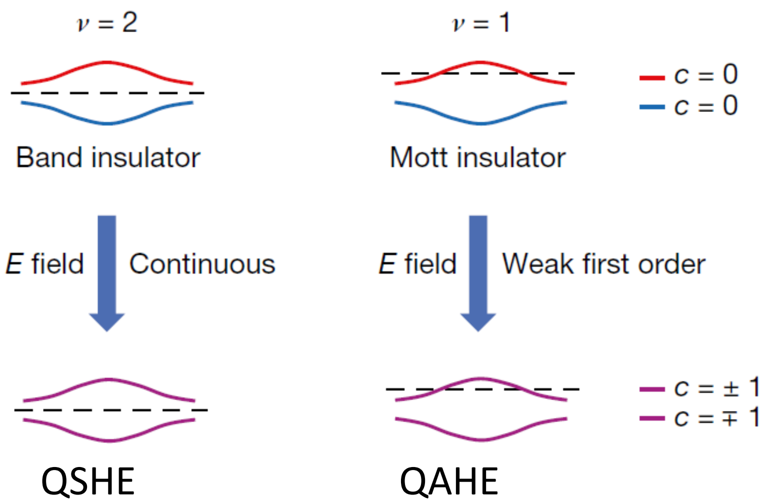
$$\sigma_{xy} = \frac{e^2}{h} C$$

- AB-rotated MoTe2/WSe2
- Valence bands of Mo and W are forming a triangular lattice with t' hopping
- Altogether a hexagonal lattice, with A and B on opposite layers
- t interlayer (nearest neighbour) hopping smaller than t'
- Displacement field exchanges them
- t hybridizes them and opens up a crossing
- Finite Chern number appears (topological number)
- Quantized conductance and edge state are expected
- At half filling a transition from a Mott like state to a C=1 state (Quantum Anomalous Hall is seen) – quantized hysteresis loops in conductance

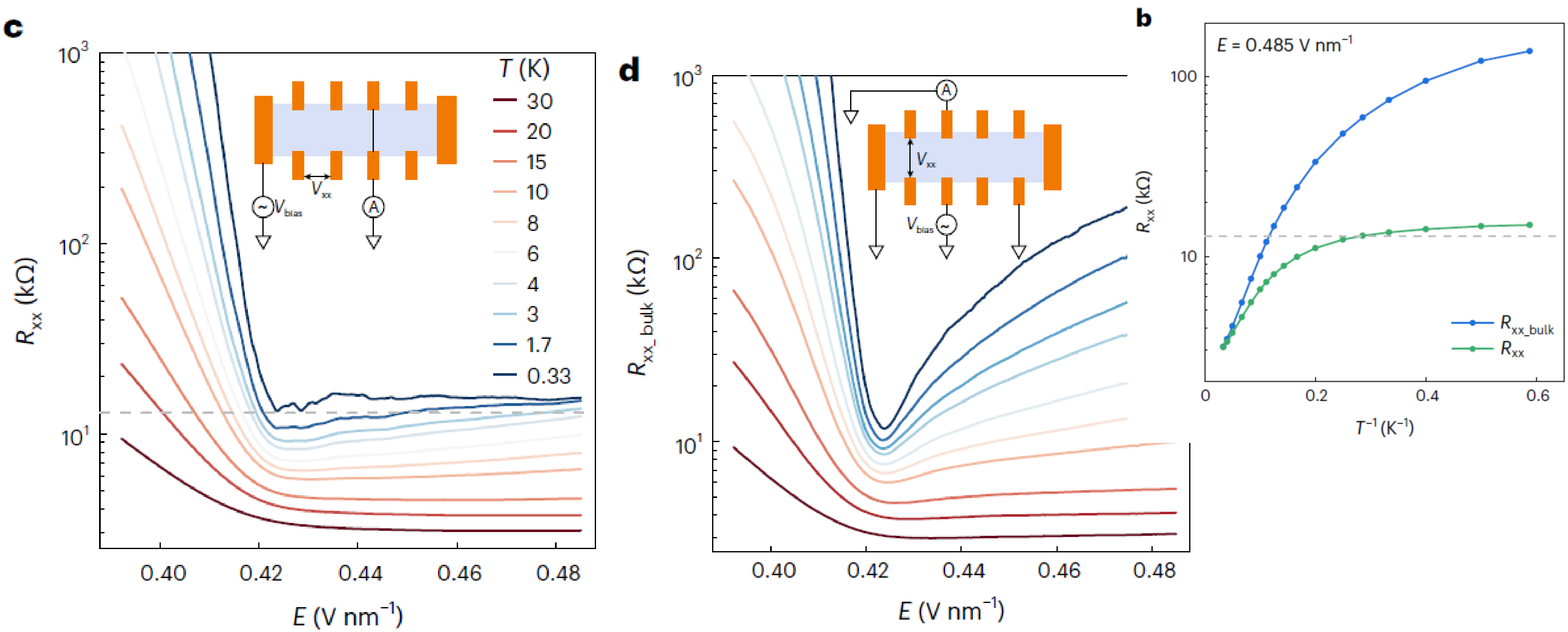




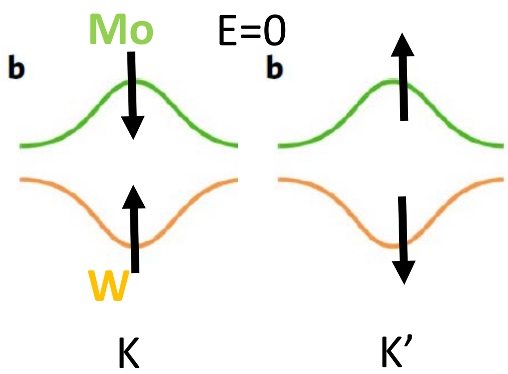
T. Li et al., Nature 600, 641 (2021)

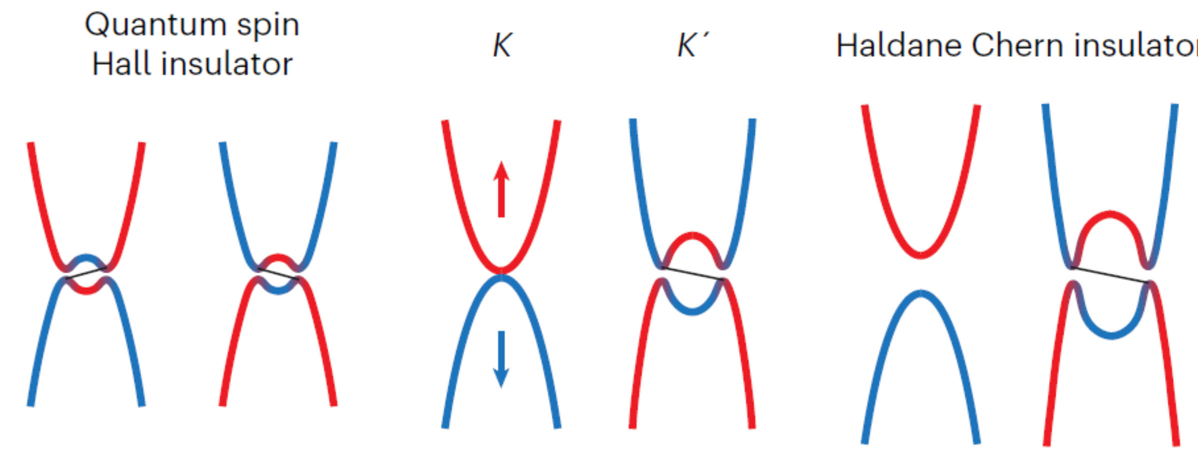


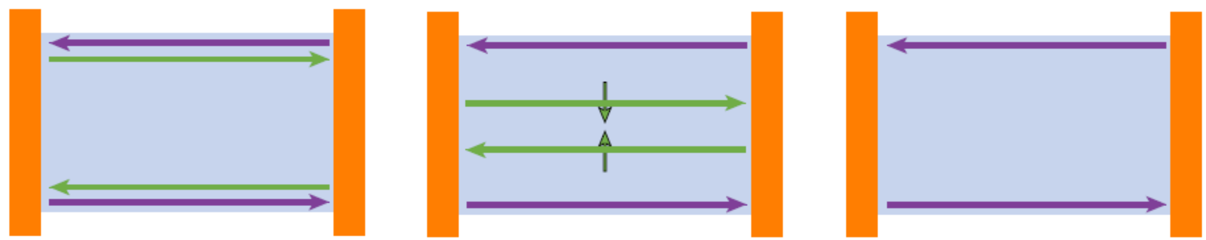
- For full filling ($\nu = 2$) E field drives transition to quantum spin-Hall insulator
- Small E-fields insulating behaviour, for large at low temperature saturates to $2e^2/h$
- Bulk measurements are gapped for large E-field – edge states circulate in the device
- Temperature dependence also supports this



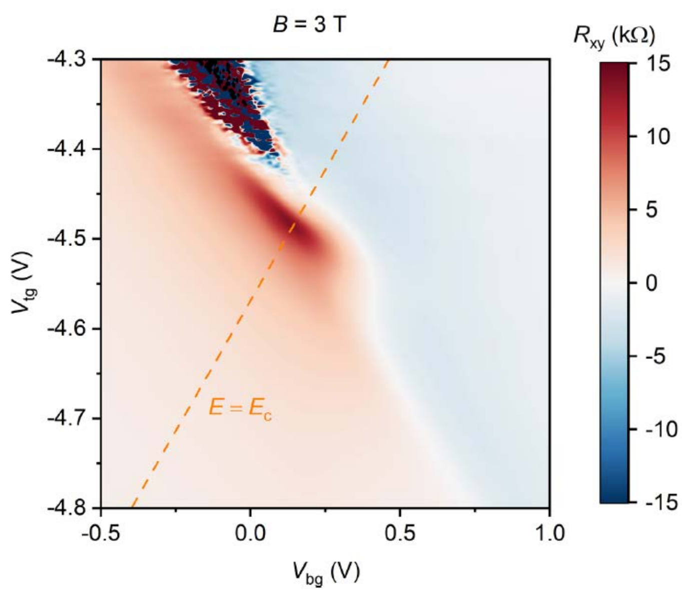
W. Zhao et al., Nature Phys. 20, 275 (2024)

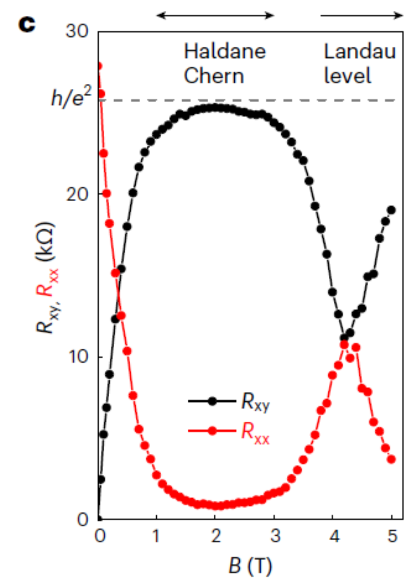






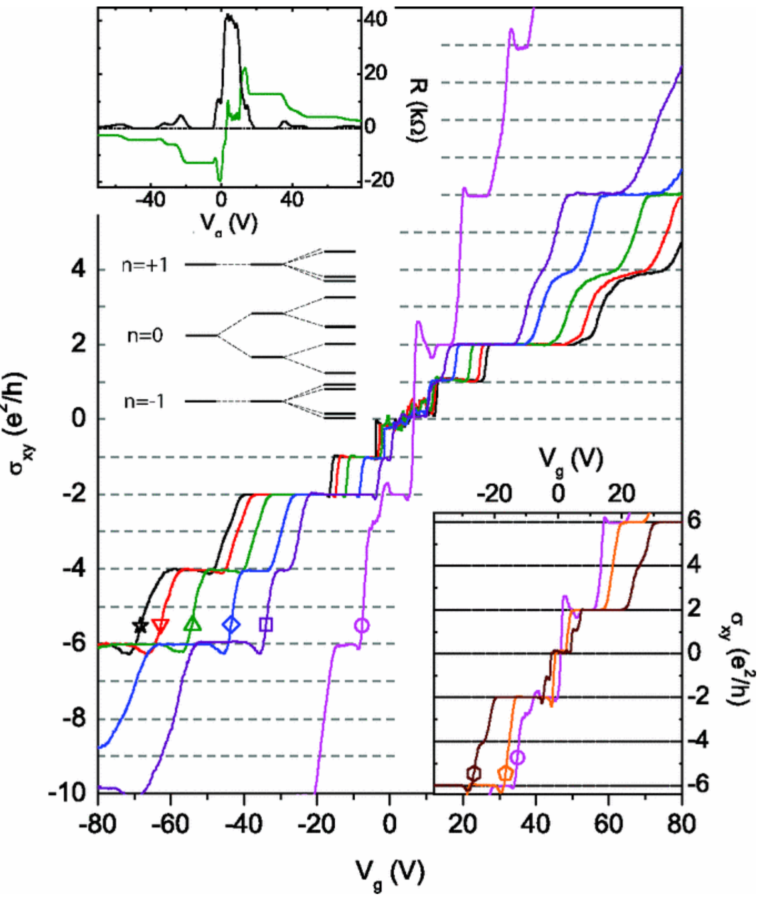
- Opposite spin alignment in the two bands and valleys (without E-field)
- With E-field a QSH state is generated.
- A Zemman field will close and reopen the gap in one valley (trivial gap).
- In the opposite valley it increases the gap.
- A QAH state formed
- It is visible between 1 and 3T magnetic fields.





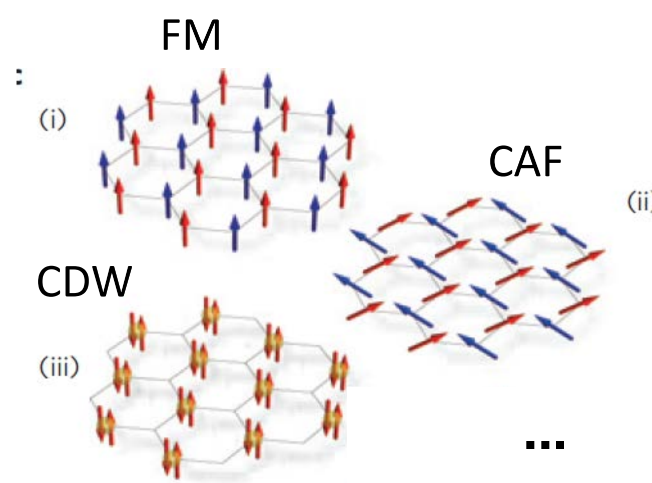
W. Zhao et al., Nature Phys. 20, 275 (2024)

Broken symmetries in QHE



Landau level degeneracies split up in high magnetic fields
4-fold degeneracy: spin and valley

Energy scales:
- Cyclotron gap
- Coulomb (Exchange interactions)
- Zeeman – energy
- Disorder scale

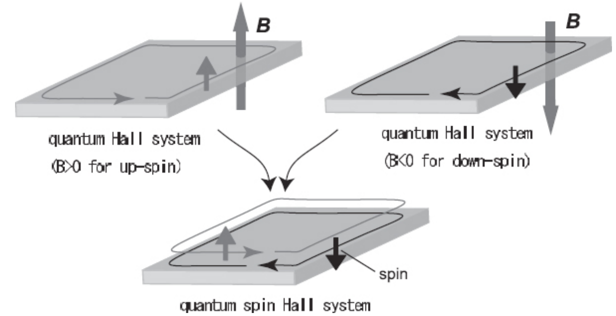


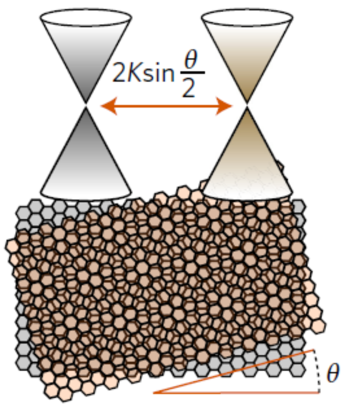
Complex phases appear (e.g. half filling, n=0):
Many possible ground states (e.g. Ferromagnet (FM), canted antiferromagnet (CAF), charge density wave)
Use tilted field measurements (only acts on Zeeman-term, no orbital contribution)
-N=0, n=0 (half filling) – not spin polarized
Canted antiferromagnetic state

A. F. Young et al., Nat. Phys. 8, 550 (2012)

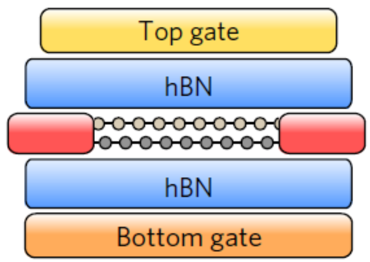
Y. Zhang et al., PRL 96, 136806 (2006)

Quantum Spin Hall - BLG

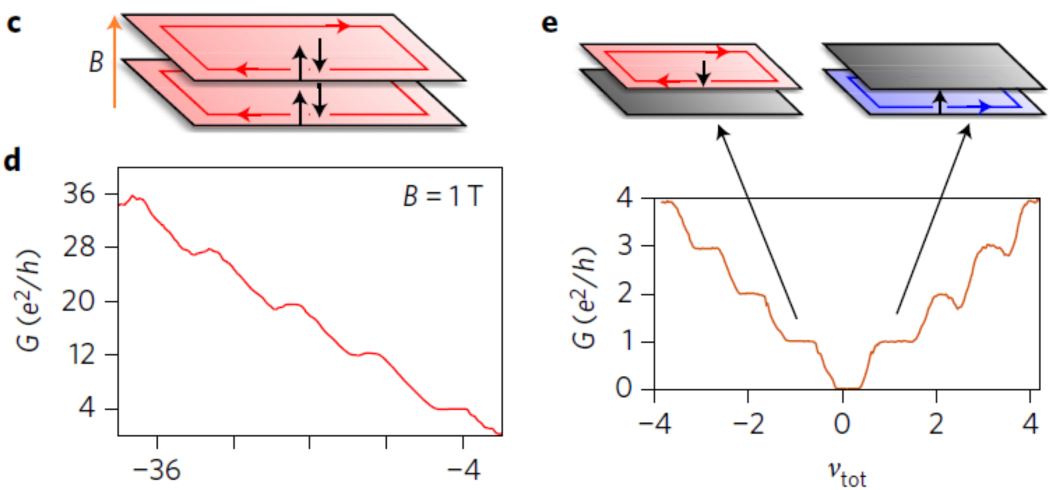




b

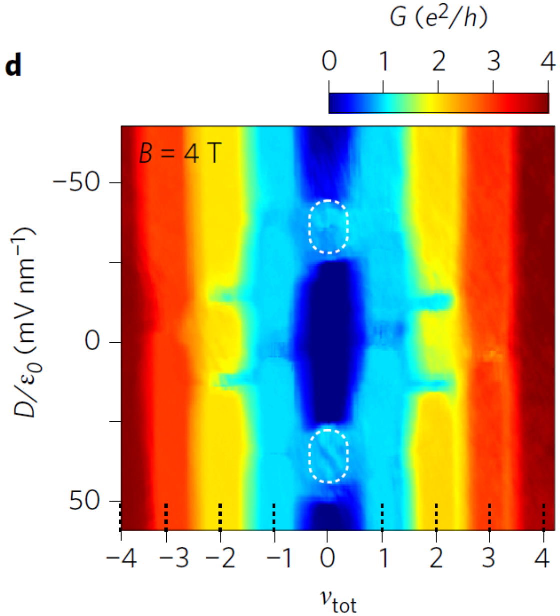


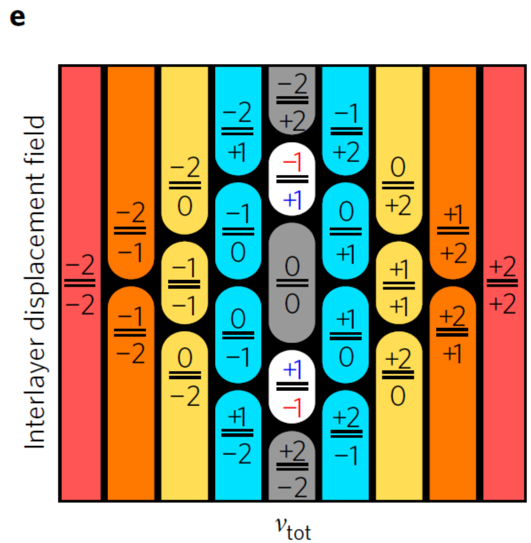
Idea: QSH is two normal QH (reversed):
Use a double layer – twisted bilayer graphene
If not aligned – layers decouple
Separate tuning of top and bottom layer filling



In unipolar regime QH is doubled:
two parallel channels
Degeneracies are split
Bipolar:
e.g. One is tuned to gap, and the other conducts

J. D. Sanchez-Yamagishi et al., Nat. Nano 12, 118 (2017)



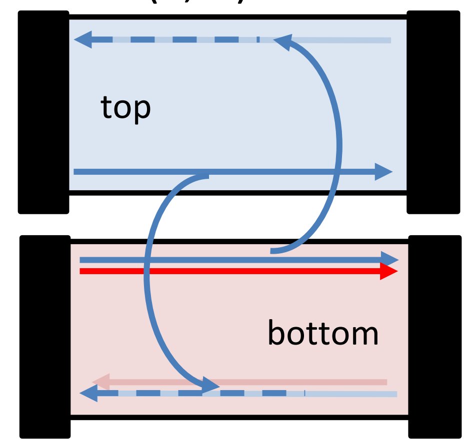


Since the layers are close – interlayer tunelling is allowed

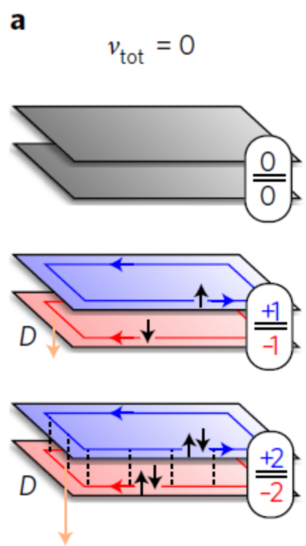
Occupied states can be scattered to unoccupied – if the pair of the state with opposite momentum is there (and not filled) – current cancels

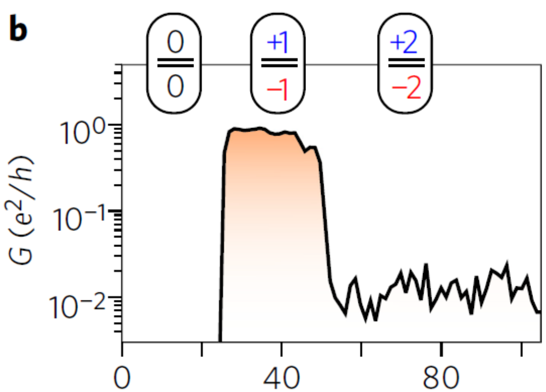
(1,-2):

$G = \nu_{tot}$



Only difference: **1/-1 configuration** which should give a gapped state, but it gives $2e^2/h$ – due to spin conservation scattering is prohibited
Artificial QSH state





J. D. Sanchez-Yamagishi et al., Nat. Nano 12, 118 (2017)