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Michele Masseroni
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hBN supply:
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Theory:
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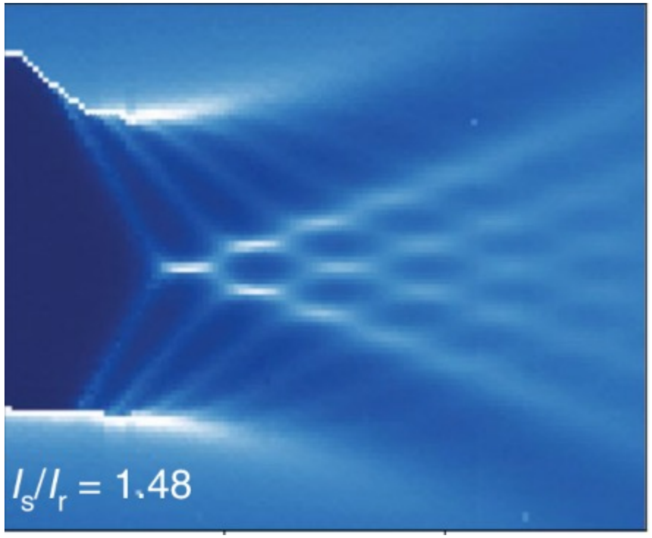
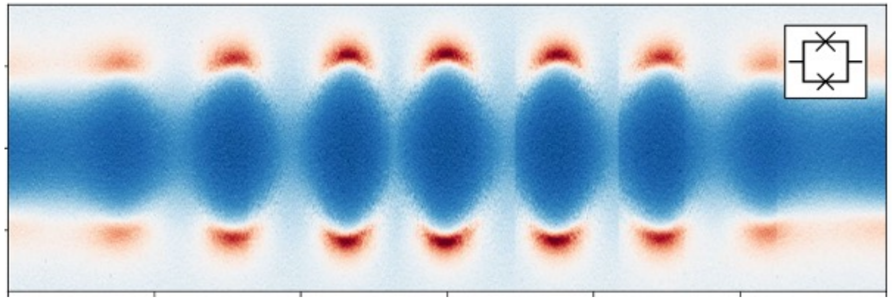
Vladimir Fal'ko
Jihang Zhu
Allan McDonald
Ming-Hao Liu
Marcin Kurpas
Klaus Richter

Graphene-based nanostructures

Part II: Twisted bilayer graphene

Thomas Ihn



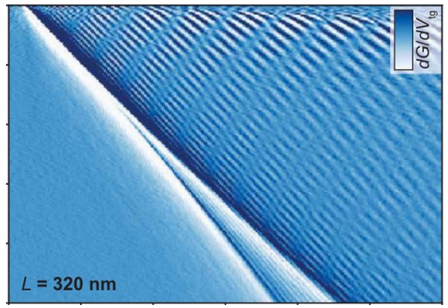


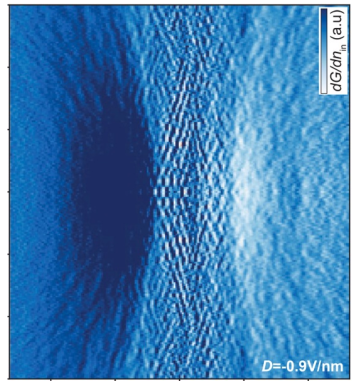
QSIT Quantum Science and Technology
National Centre of Competence in Research

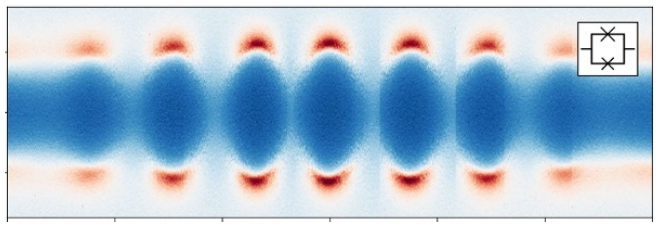
ETH Eidgenössische Technische Hochschule Zürich

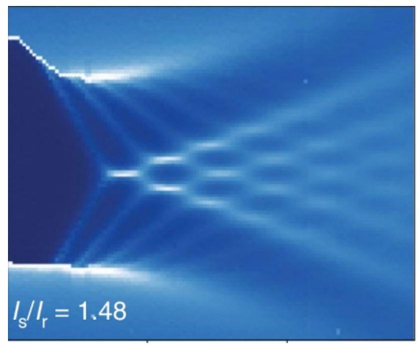
Outline

- Twisted bilayer graphene: brief historical introduction
- Fabrication
- Large twist angles
- Small twist angles
- Magic angle
- Twisted double bilayer graphene







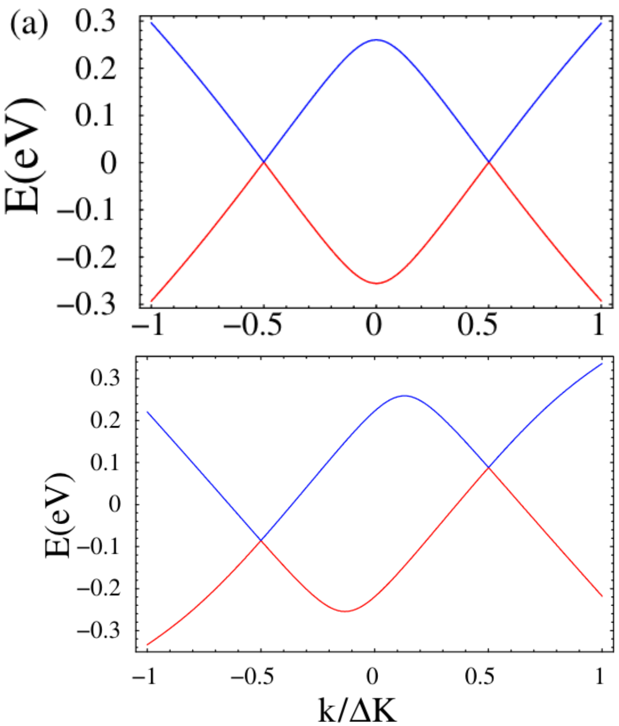


Twisting graphene

Graphene Bilayer with a Twist: Electronic Structure

J. M. B. Lopes dos Santos,[1] N. M. R. Peres,[2] and A. H. Castro Neto[3]

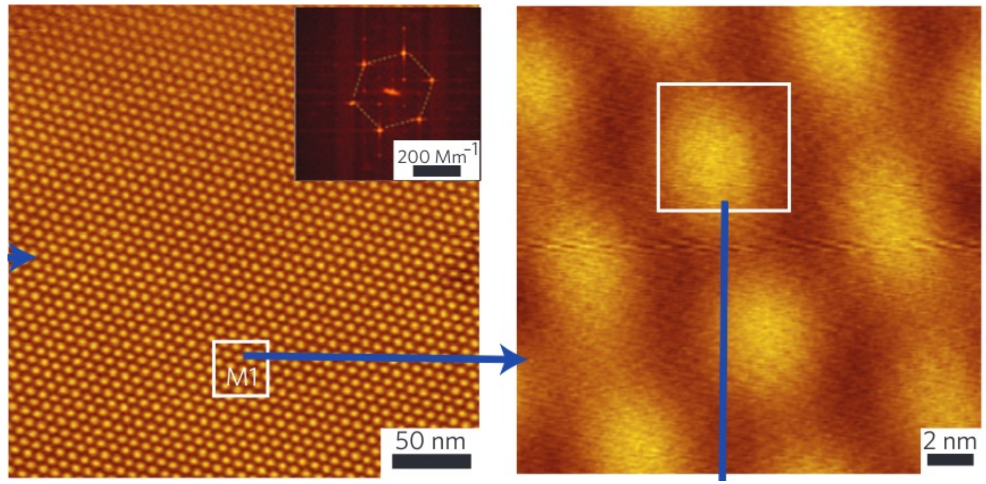
Phys. Rev. Lett. **99**, 256802 (2007).



zero displacement field

nonzero displacement field

Moiré pattern – period 7.7 nm

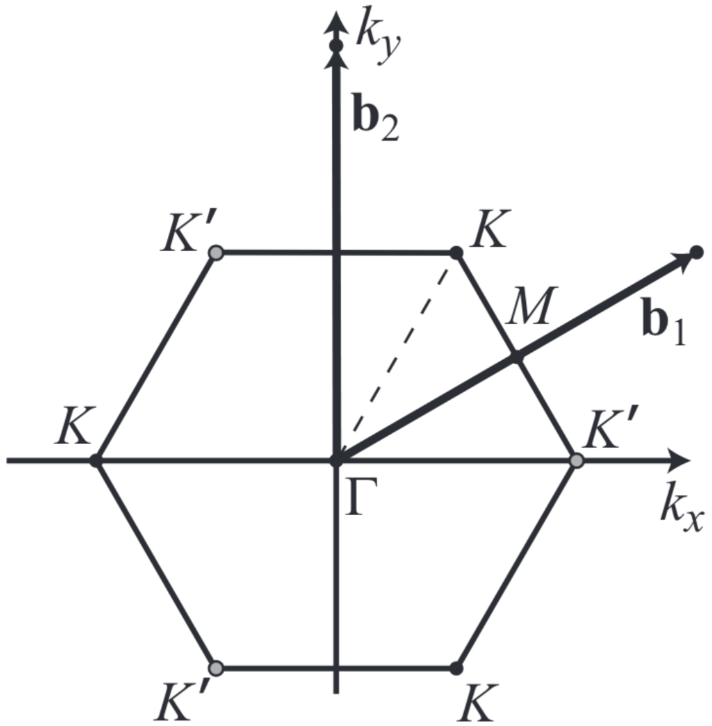


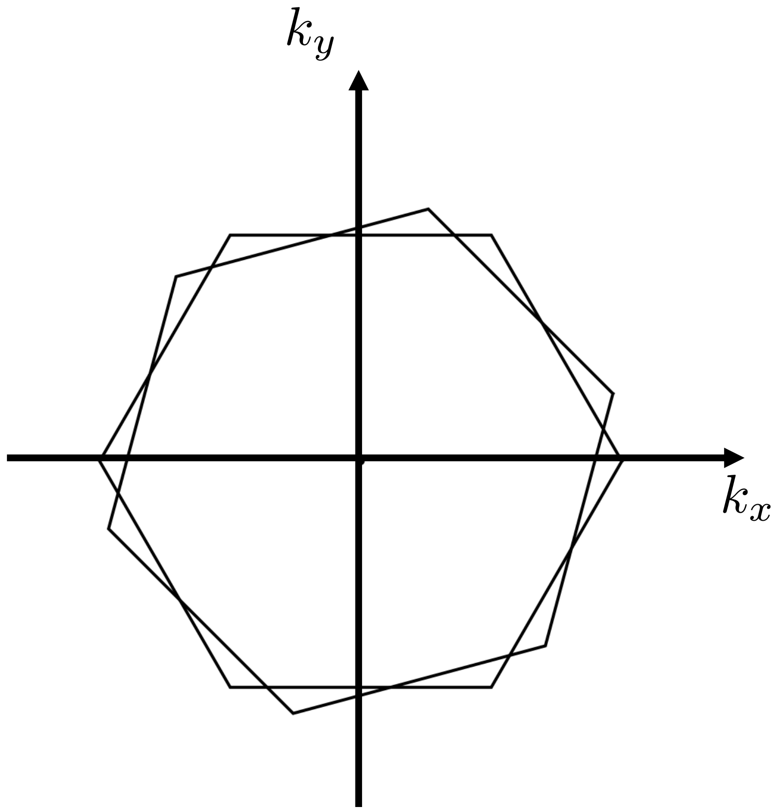
Observation of Van Hove singularities in twisted graphene layers

Guohong Li[1], A. Luican[1], J. M. B. Lopes dos Santos[2], A. H. Castro Neto[3], A. Reina[4], J. Kong[5] and E. Y. Andrei[1]*

Nature Physics **6**, 109 (2010).

Twisting graphene

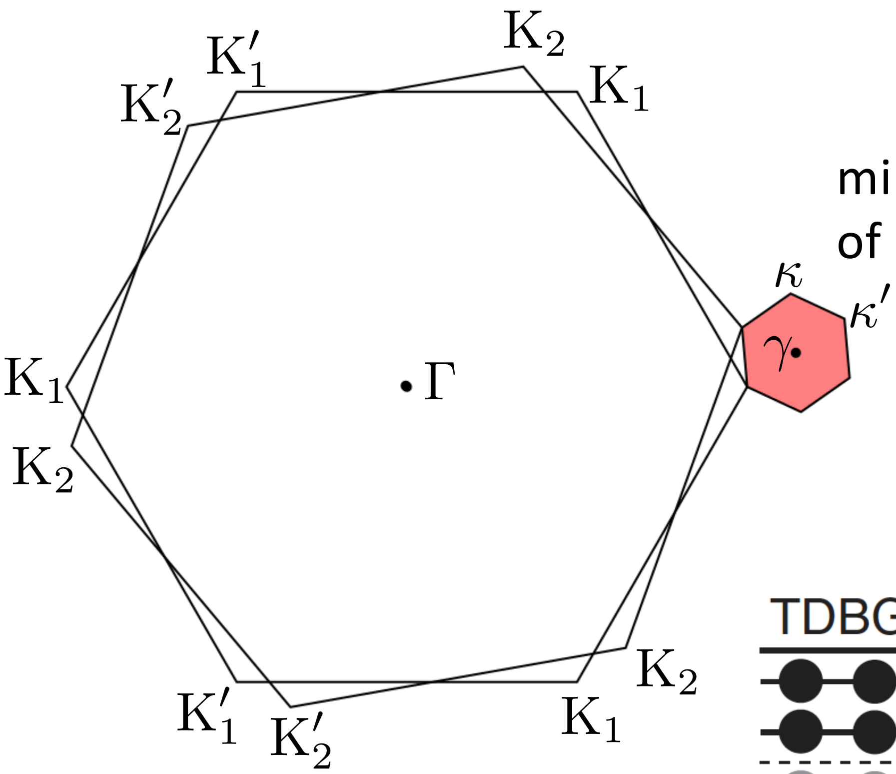




Two copies of the 1st Brillouin zone are rotated relative to each other

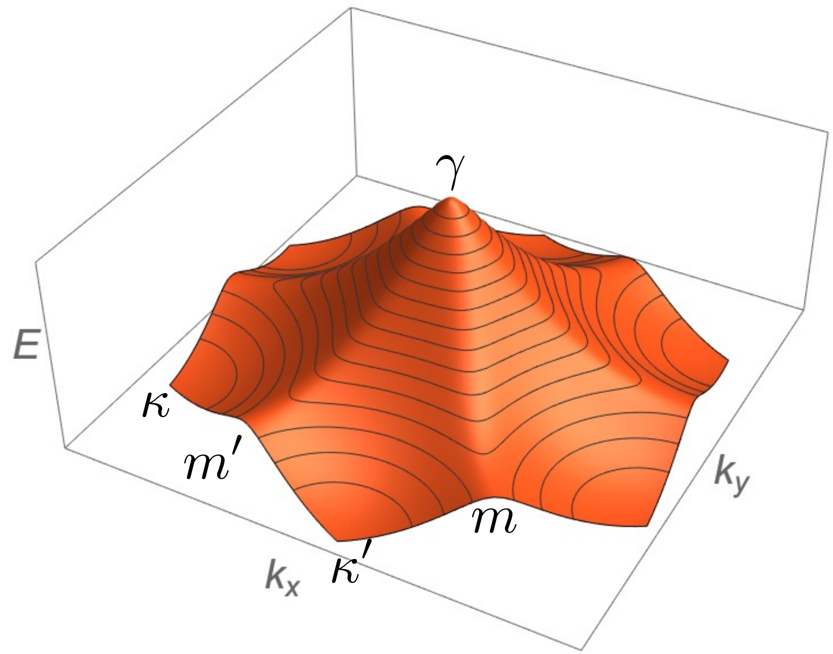
The mini Brillouin zone

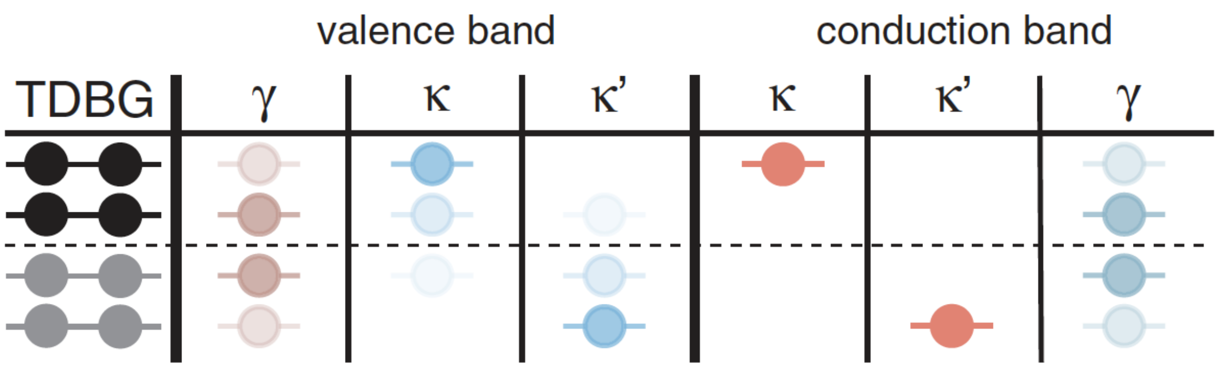
original Brillouin zones
of layers 1 and 2



mini-Brillouin zone
of Moiré superlattice

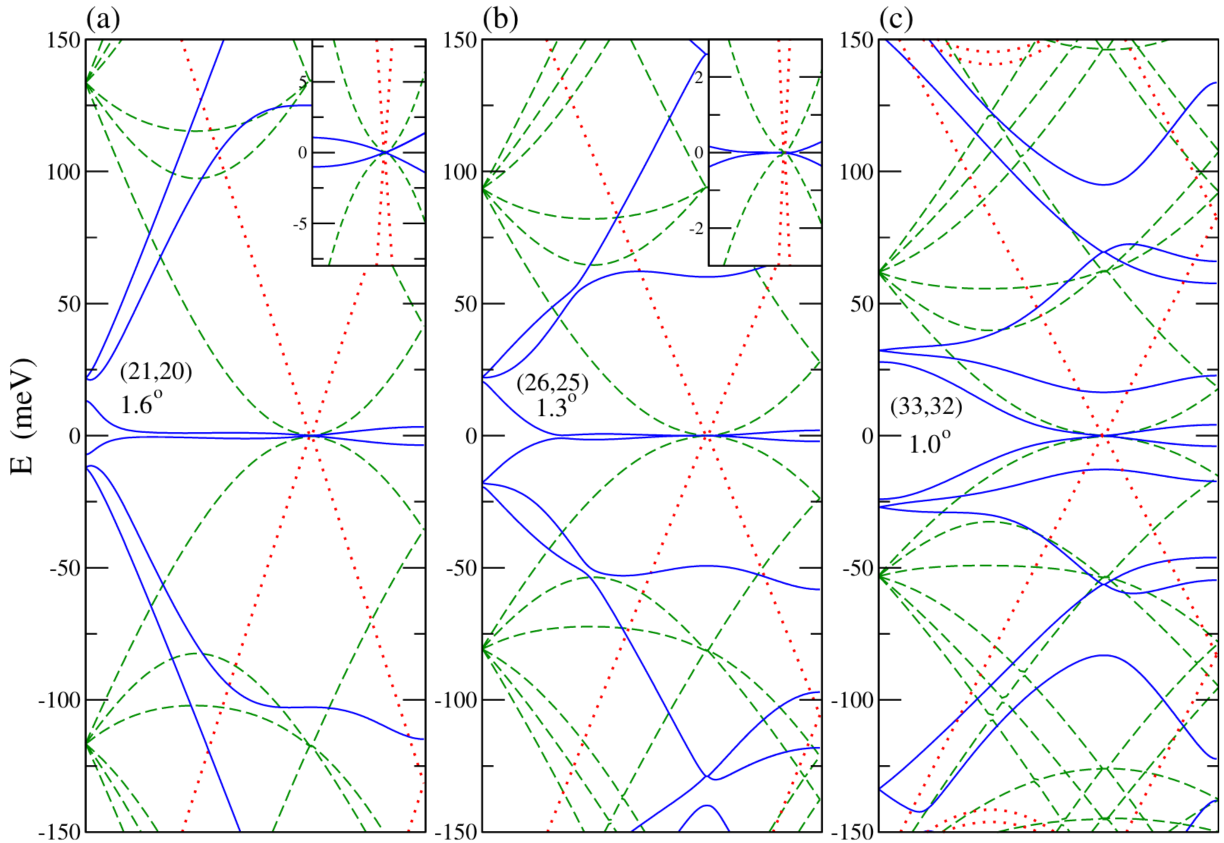
dispersion relation within
the mini Brillouin zone





Flat bands emerge for small twist angles

E. Suárez Morell *et al*, Phys. Rev. B **82**, 121407 (2010).



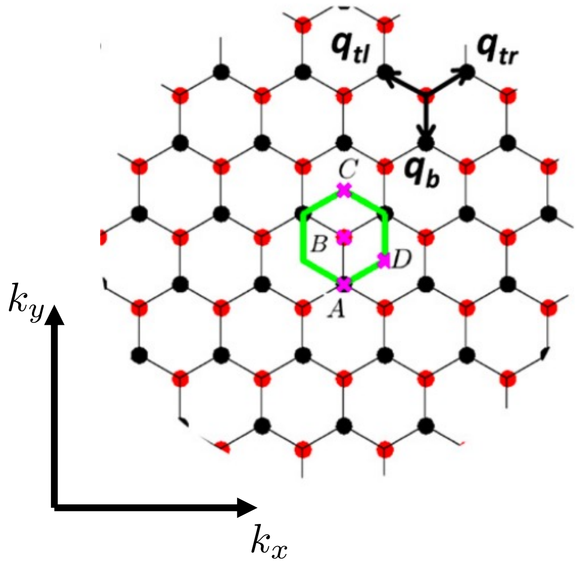
Tight binding model

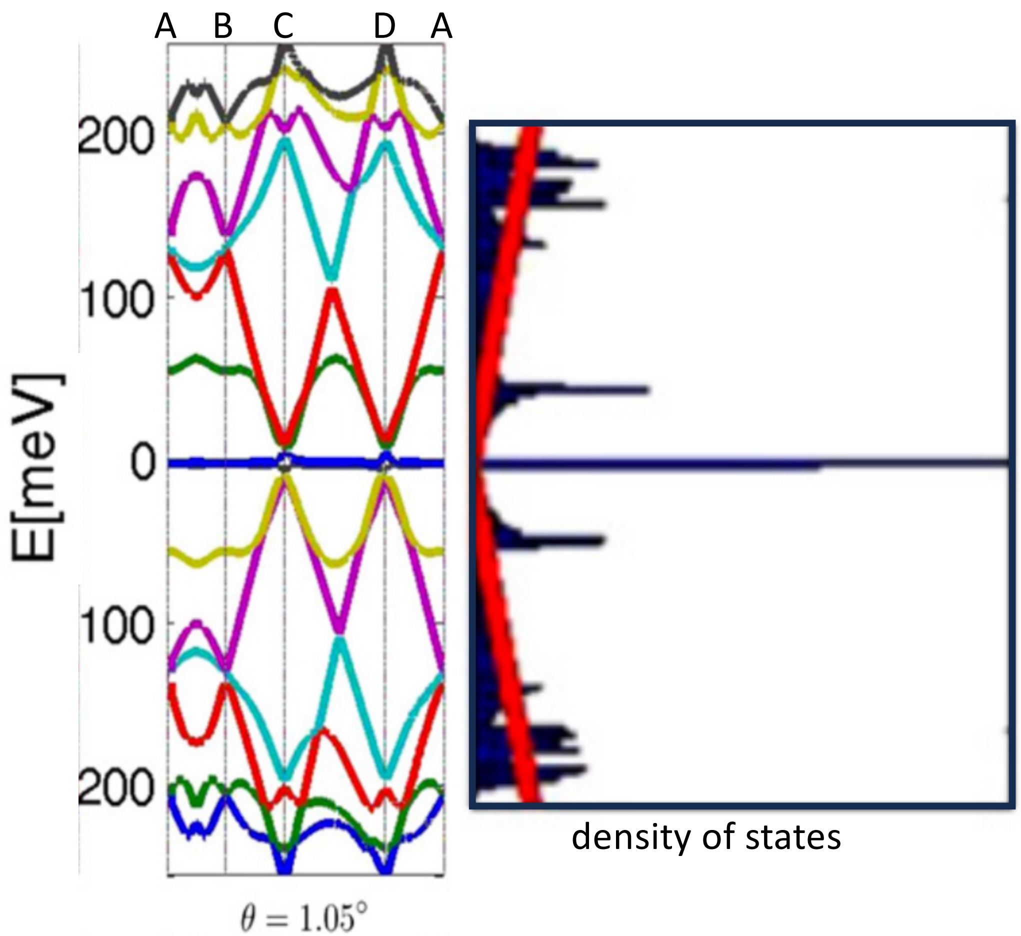
also based on comparison to DFT calculations

Magic angles: the key to generate flat bands

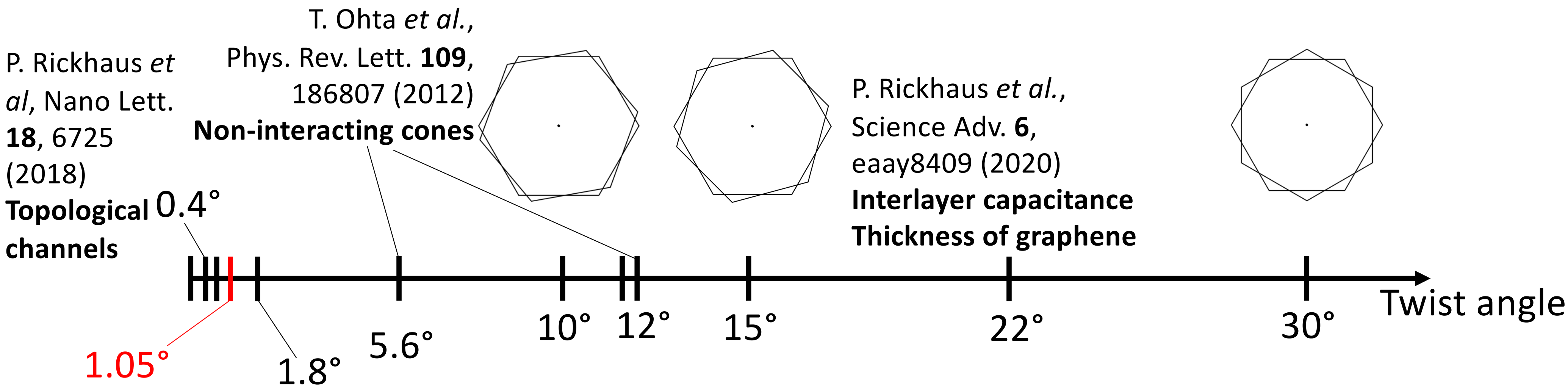
R. Bistritzer and A. H. MacDonald, Proc. Natl. Acad. Sci. U.S.A. **108**, 12233 (2011).

Continuum model
for interlayer tunneling





Twist angle as a knob to tune the dispersion relation



Y. Cao *et al.*, Nature **556**, 43 (2018)
Superconduc-tivity,
ibid 80 (2018)
Correlated insulators

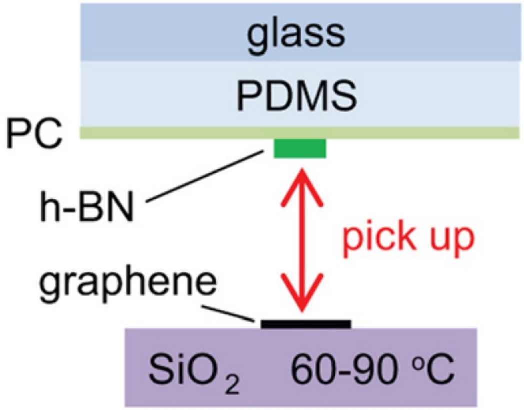
Y. Cao *et al.*, Phys. Rev. Lett. **117**, 116804 (2016)
Insulating states

The twist angle is a knob to tune the interlayer coupling:
large angles: decoupled layers, strongly dispersive bands

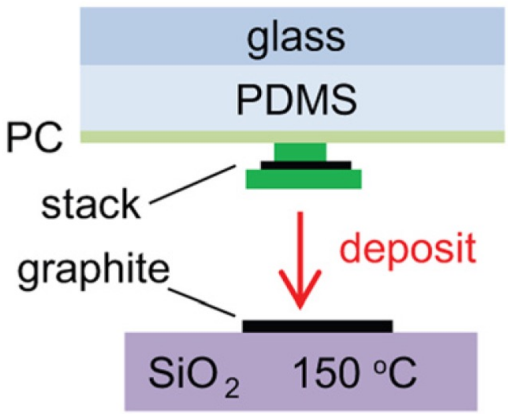
small angles: strongly coupled layers, flat bands, correlated states

intermediate angles: interlayer coupling tunable by gate voltages!

Dry transfer technique



P.J. Zomer *et al*, Appl. Phys. Lett. **105**, 013101 (2014)



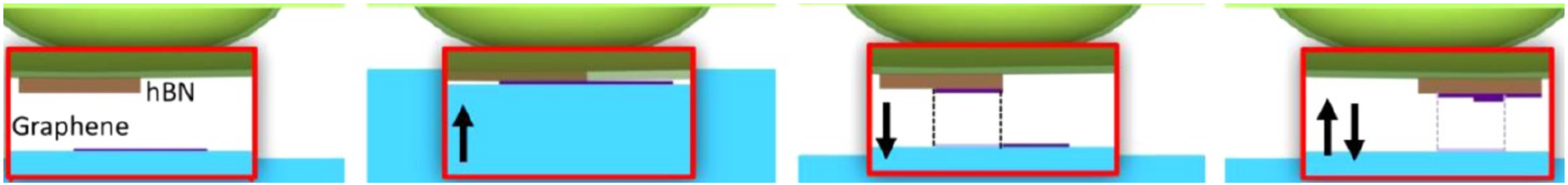
PC: Polycarbonate
PDMS: Polydimethylsiloxane

Pick-up performed in glove box with Ar atmosphere using a micromanipulator

Annealing in Ar/H_2 atmosphere at 350°C

The tear and stack method

K. Kim *et al.*, Nano Lett. **16**, 1989 (2016)



partial contact

tear
translate
rotate

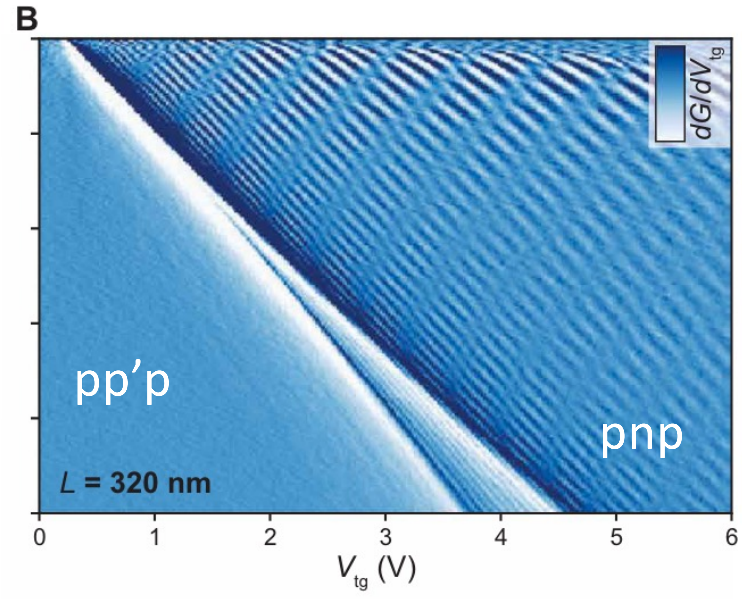
stack

Large twist angles: The electronic thickness of graphene

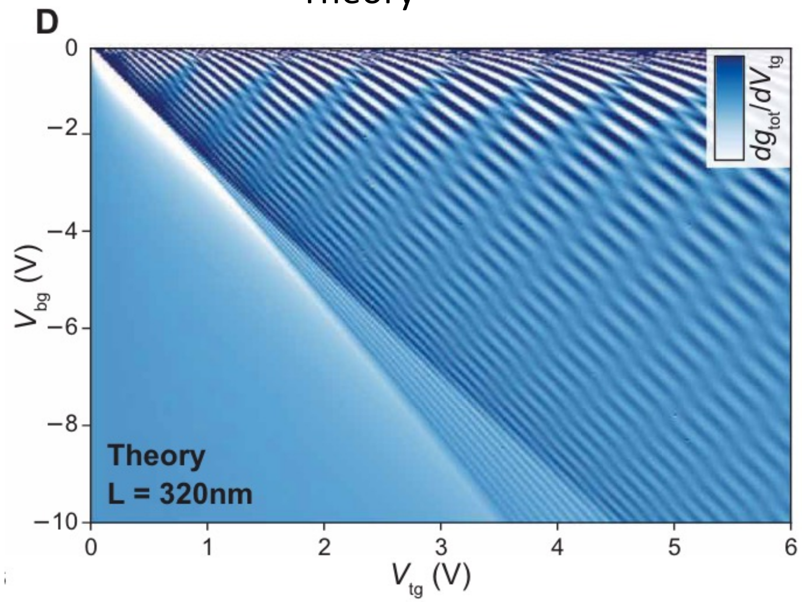
Stacked layers

twist angle 22°

Experiment



Theory



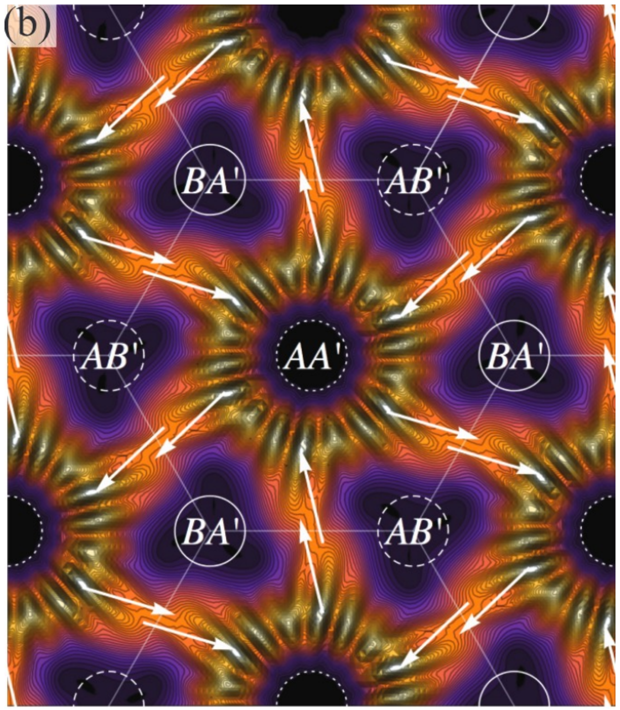
Extracted interlayer capacitance: $C_m = 7.5\,\mu\mathrm{F/cm^2}$, three times larger than expected from layer separation!

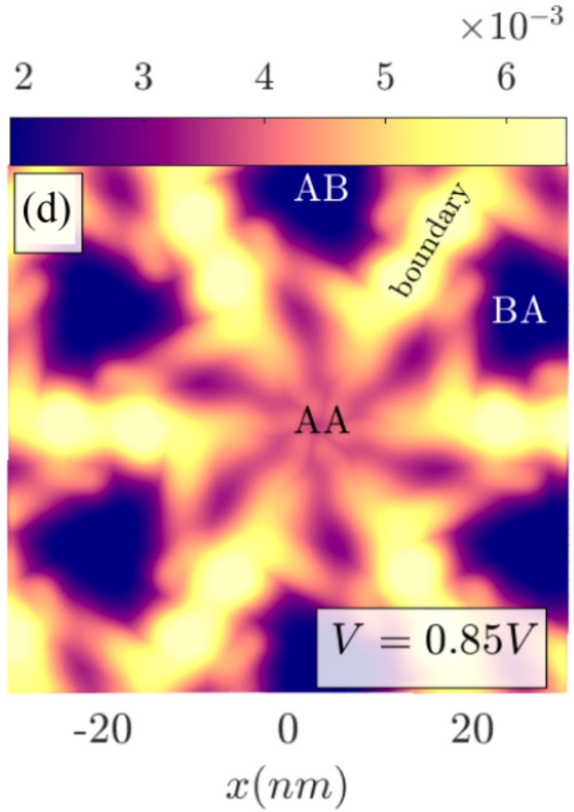
Consequence: graphene has a finite electronic thickness of $t_g = 2.6\,\text{Å}$!

P. Rickhaus *et al*, Science Advances **6**, eaay8409 (2020).

Small twist angles: Topology in twisted bilayer graphene

Probability density of a selected state (including directionality of helical curents)

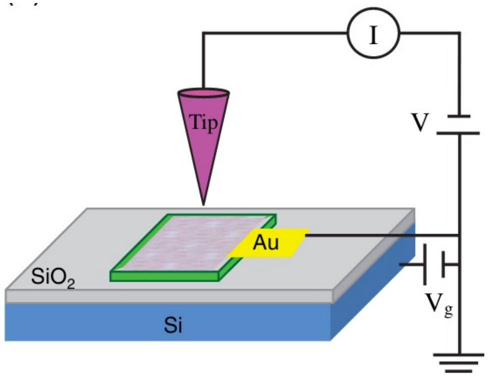




Local density of states (including lattice relaxation and disorder)

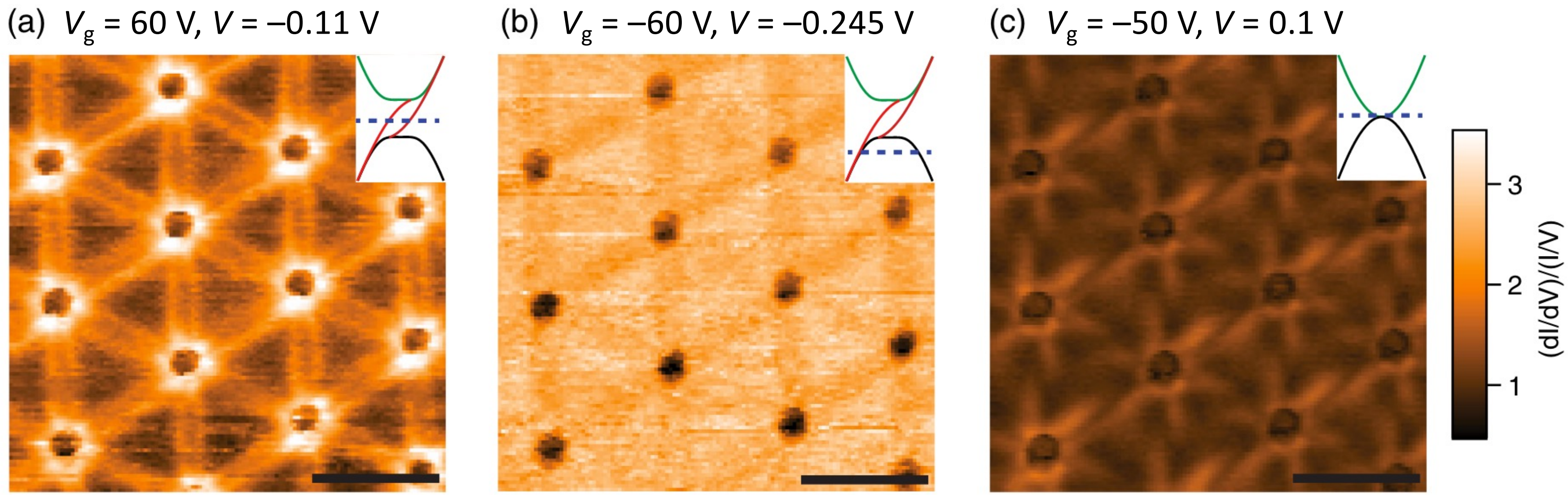
P. San-Jose and E. Prada, Phys. Rev. B **88**, 121408 (2013).

M. Andelković *et al.*, Phys. Rev. Mater. **2**, 034004 (2018).



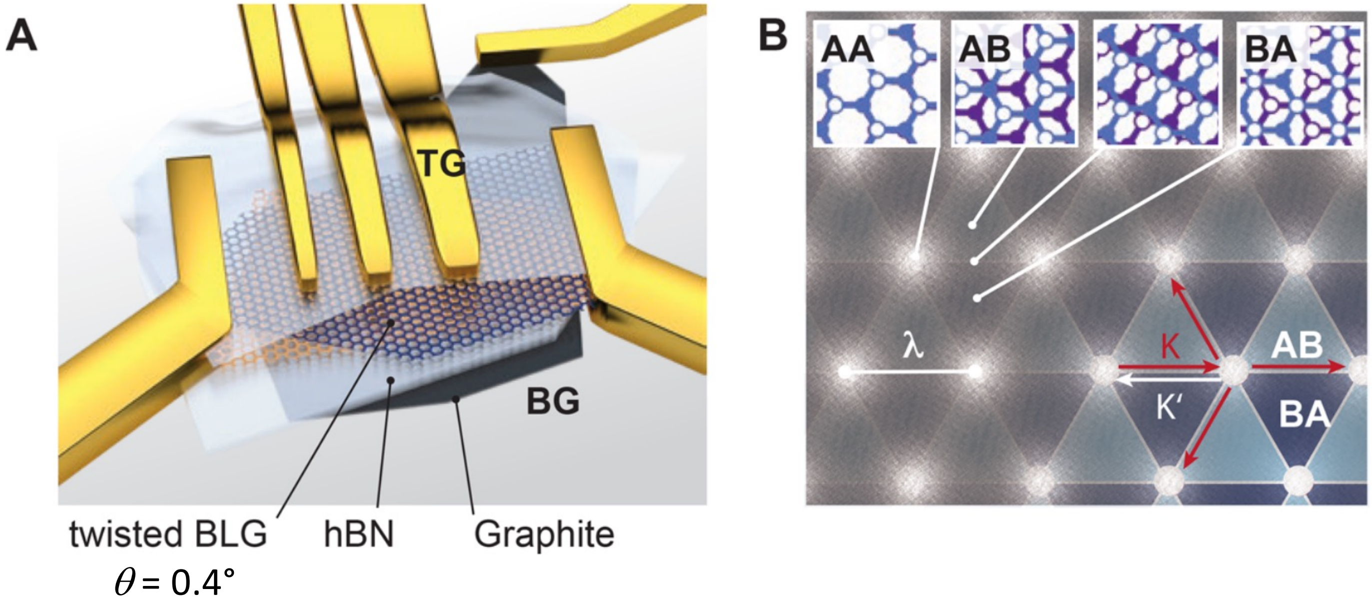
Topology in twisted bilayer graphene

Local density of states measured by STM

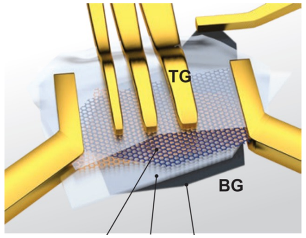


S. Huang *et al.*, Phys. Rev. Lett. **121**, 037702 (2018).

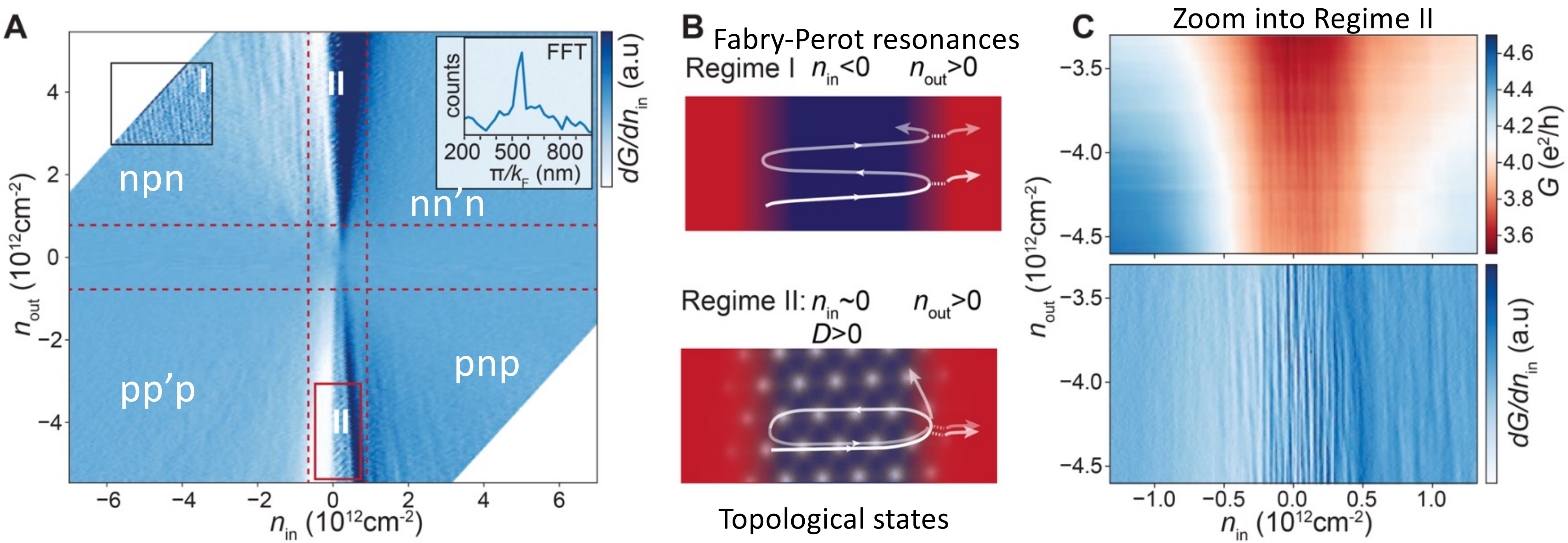
Topology in twisted bilayer graphene



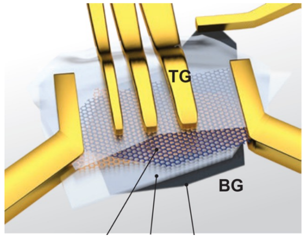
P. Rickhaus *et al*, Nano Lett. **18**, 6725 (2018).



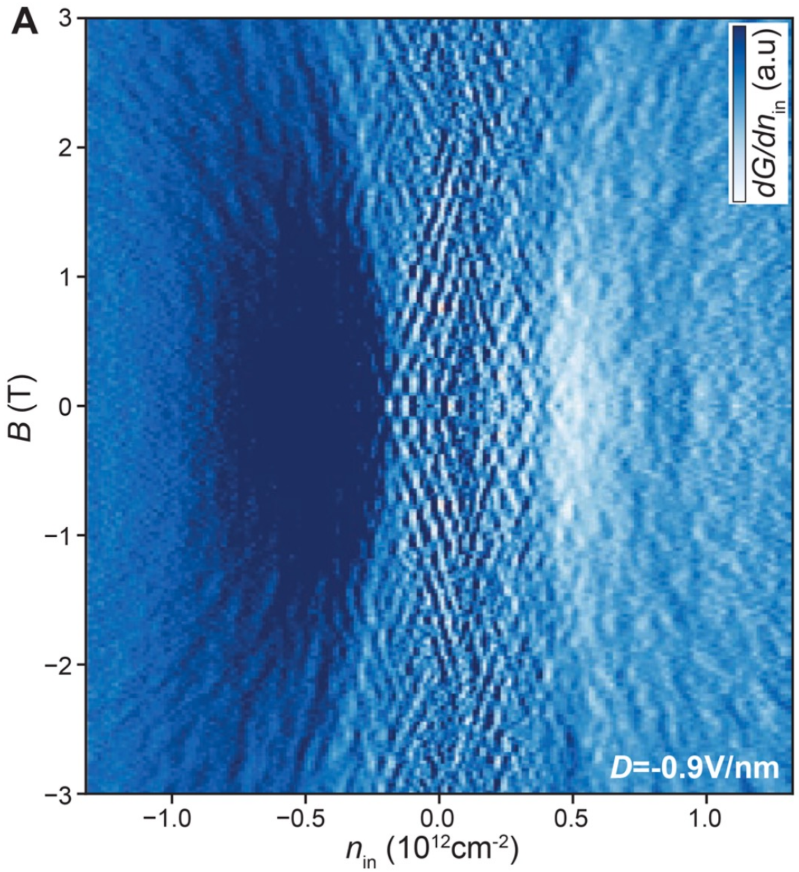
Topology in twisted bilayer graphene



P. Rickhaus *et al*, Nano Lett. **18**, 6725 (2018).



Topology in twisted bilayer graphene



P. Rickhaus *et al*, Nano Lett. **18**, 6725 (2018).

in 1D: $k_{\mathrm{F}} \propto n_{\mathrm{in}}$

Moiré periodicity: $\lambda = 34\,\mathrm{nm}$

Height of moiré unit cell:

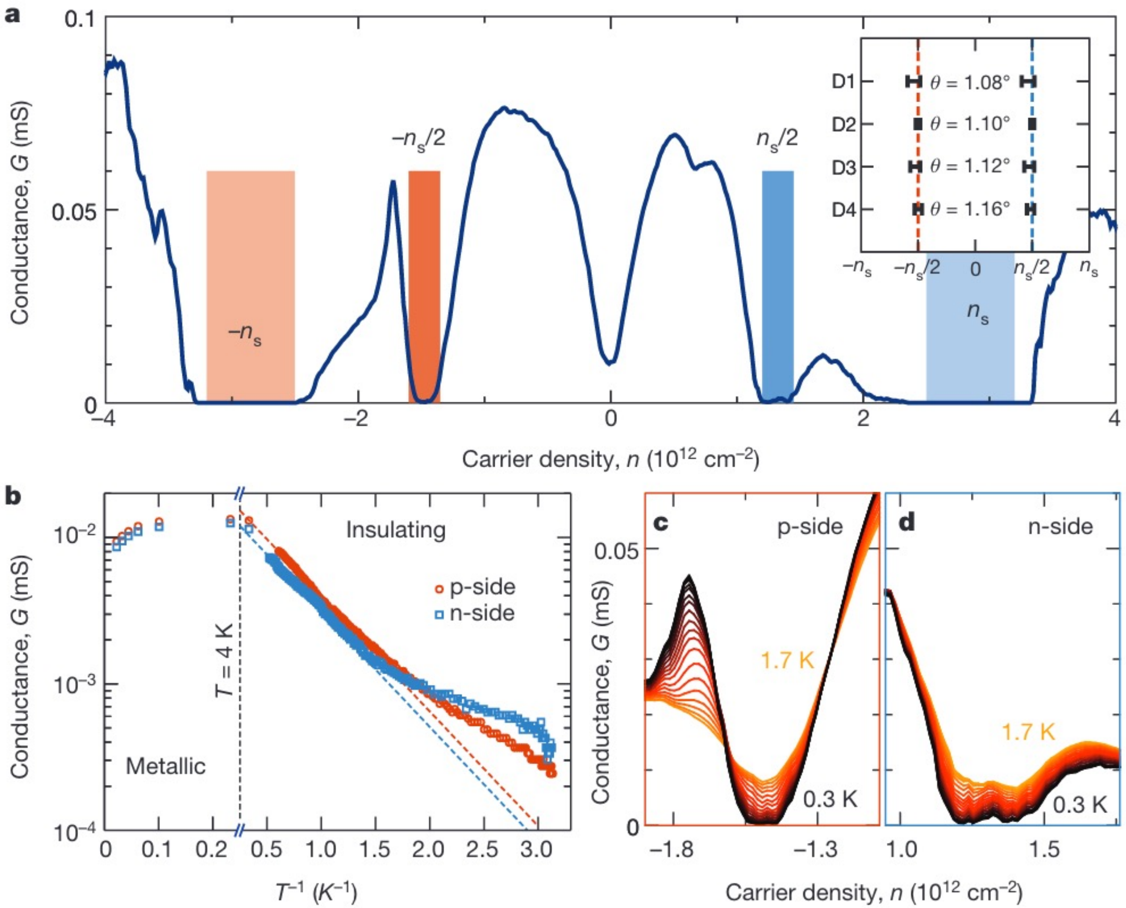
$$h = \frac{\sqrt{3}\lambda}{2} = 29\,\mathrm{nm}$$

Cavity length: $L = 400\,\mathrm{nm}$

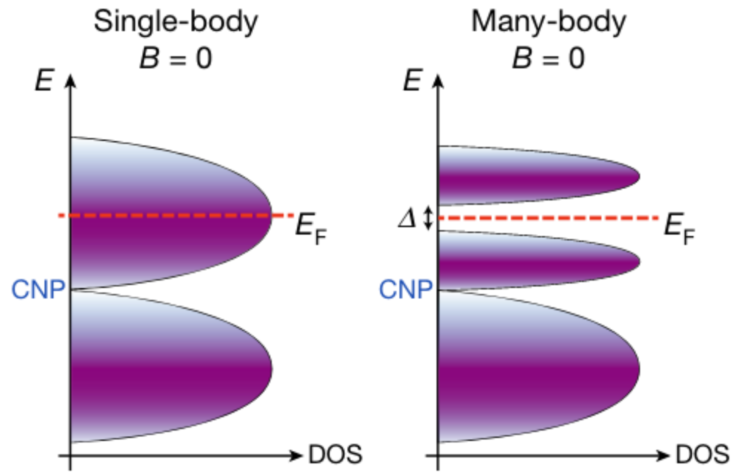
Magnetic field periodicity fits to

$$\Delta B = \frac{\Phi_0}{Lh} = \frac{h}{e}\frac{1}{Lh}$$

Magic angle twisted bilayer graphene: correlated insulators

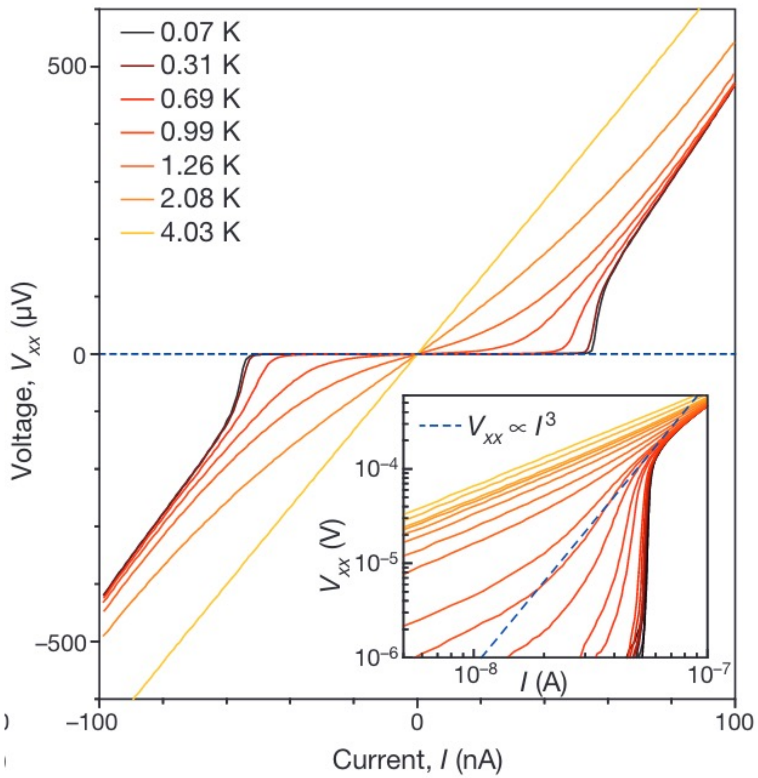


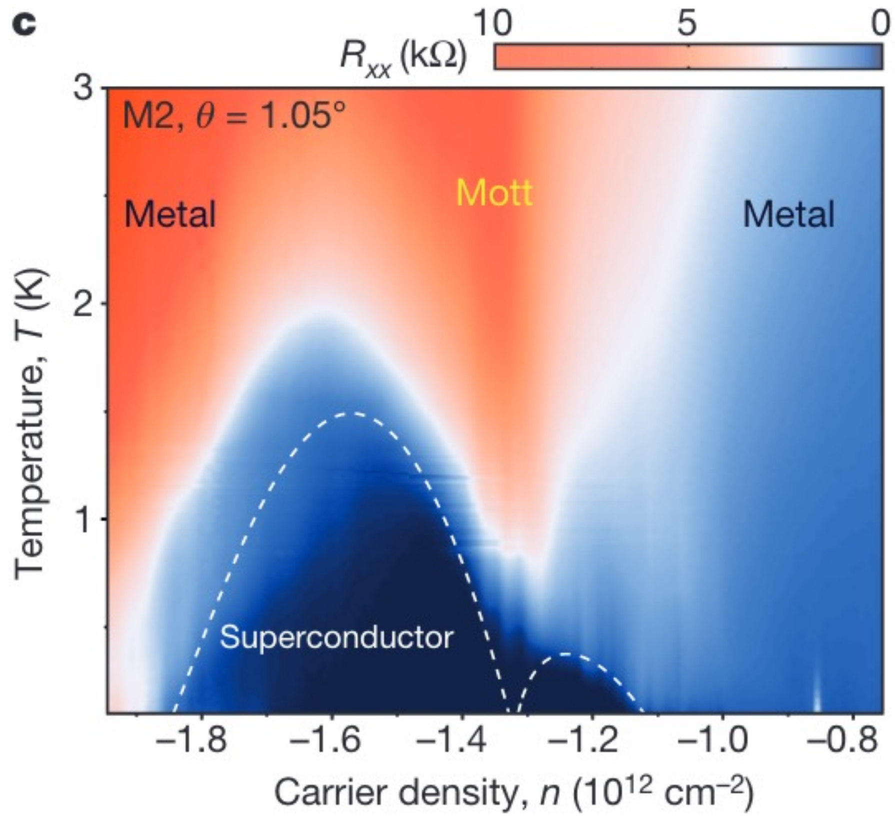
Mott-insulator at half filling



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

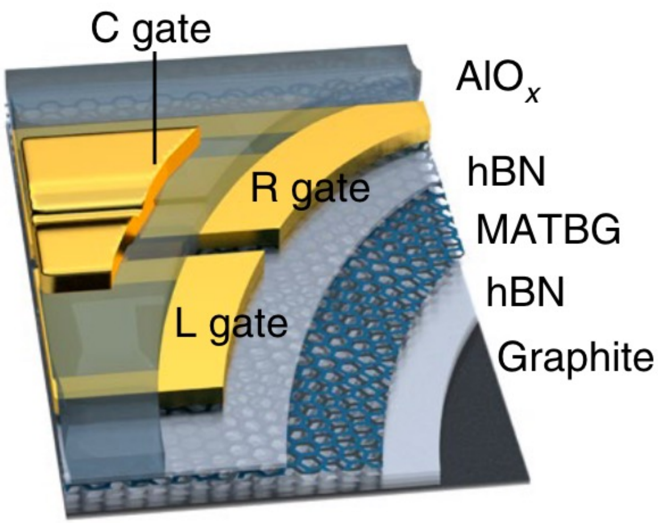
Magic angle twisted bilayer graphene: superconductivity

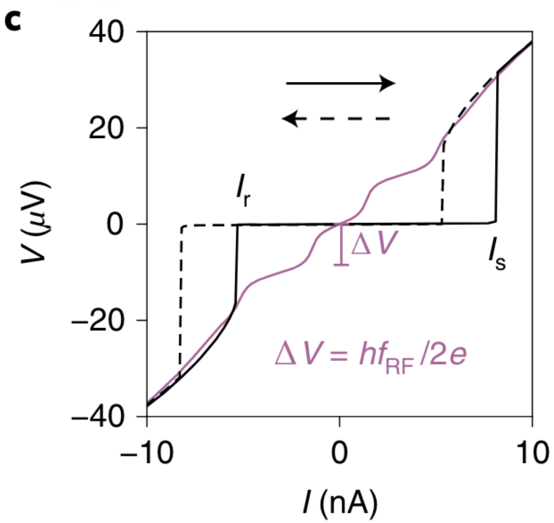




Y. Cao *et al*, Nature **556**, 43 (2018).

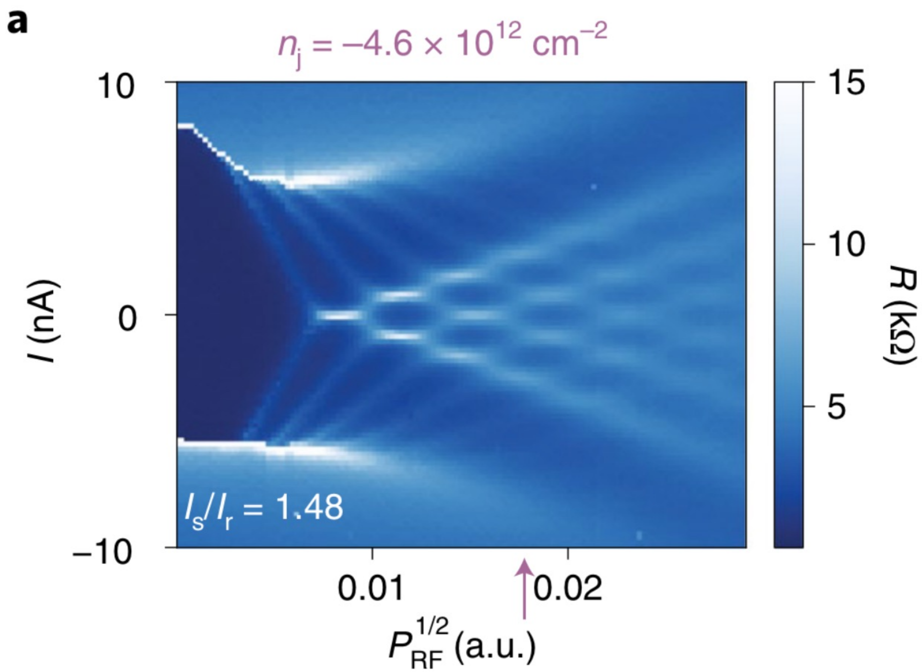
Magic angle twisted bilayer graphene: gate tunable Josephson junction





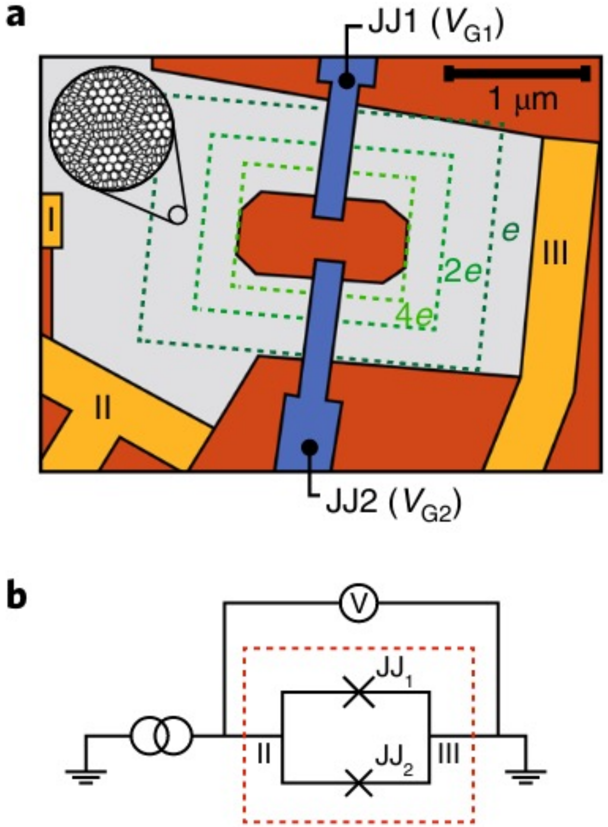
DC Josephson effect and Shapiro steps

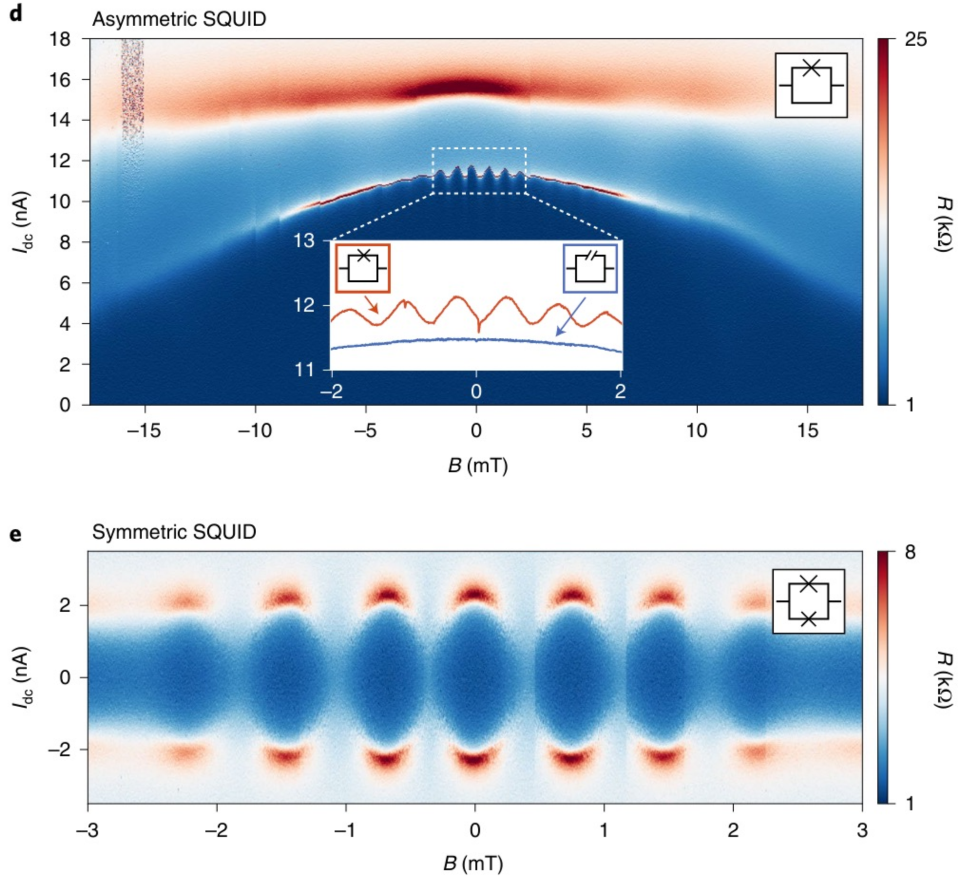
Shapiro steps



F. K. de Vries *et al*, Nat. Nanotechnol. **16**, 760 (2021).

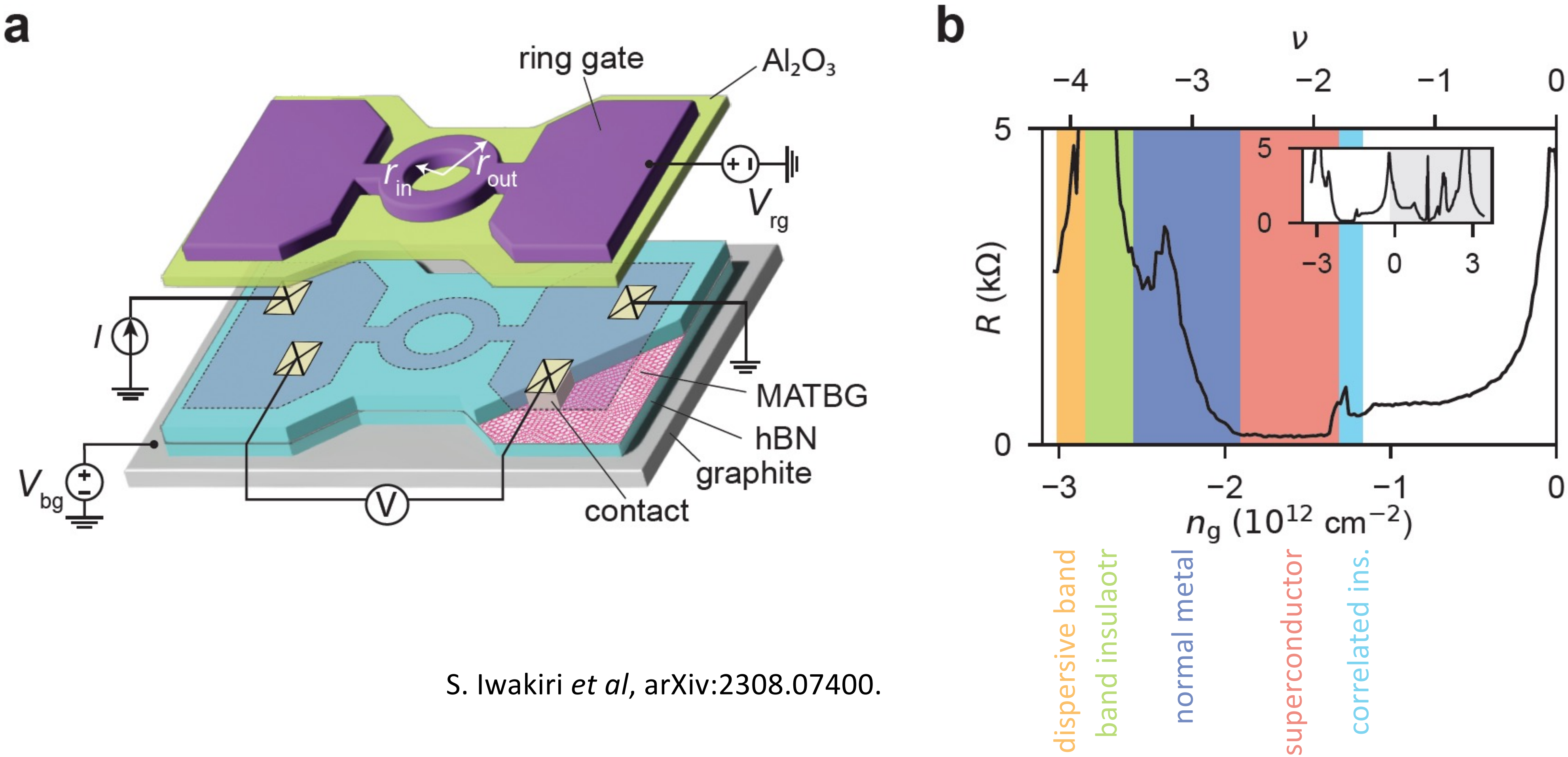
Magic angle twisted bilayer graphene: gate-tunable SQUID





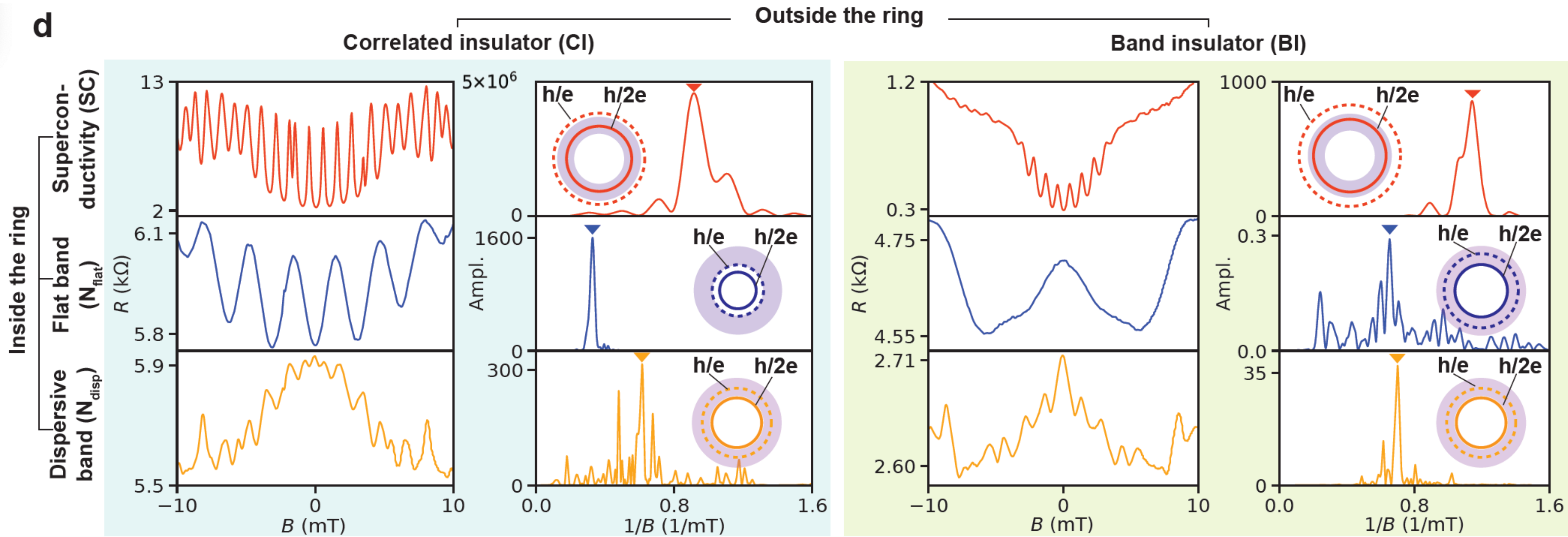
E. Portolés *et al*, 2D Mater. **9**, 014003 (2022).

Magic angle twisted bilayer graphene: Little-Parks effect



S. Iwakiri *et al*, arXiv:2308.07400.

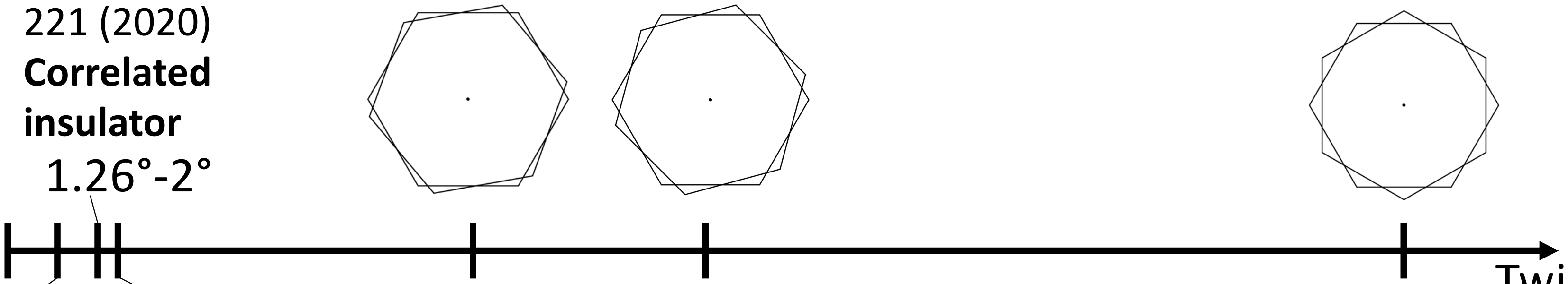
Magic angle twisted bilayer graphene: Little-Parks effect



S. Iwakiri *et al*, arXiv:2308.07400.

Twisted double bilayer graphene

X. Liu *et al.*, Nature **583**, 221 (2020)
Correlated insulator



G. W. Burg *et al.*, Phys. Rev. Lett. **123**, 197702 (2019)
Correlated insulator
C. Shen *et al.*, Nature Phys. **16**, 520 (2020).

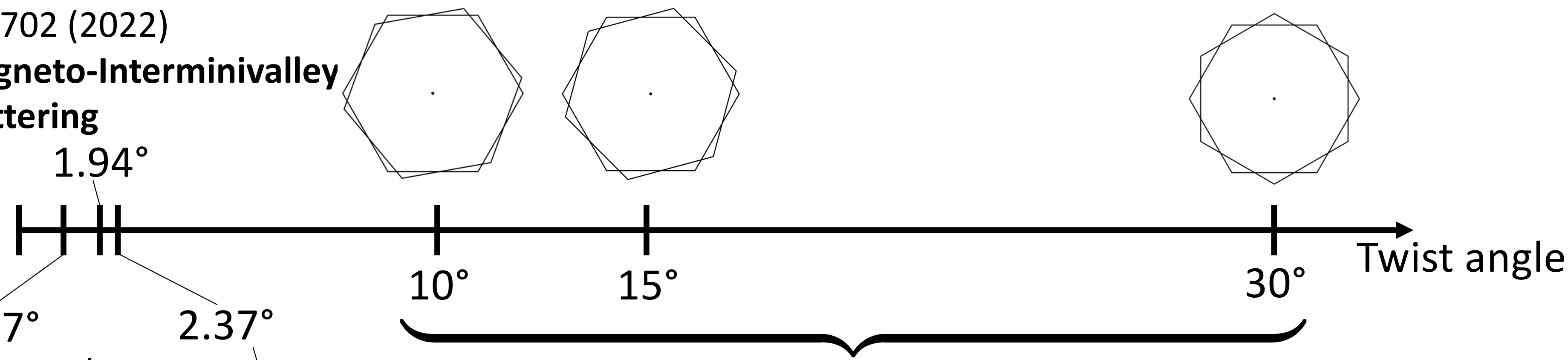
The twist angle is a knob to tune the interlayer coupling:
large angles: decoupled layers, strongly dispersive bands

small angles: strongly coupled layers, flat bands, correlated states

intermediate angles: interlayer coupling tunable by gate voltages!

Our work on twisted double bilayer graphene

P. Tomić *et al.*,
Phys. Rev. Lett. **128**,
057702 (2022)
Magneto-Interminivalley scattering

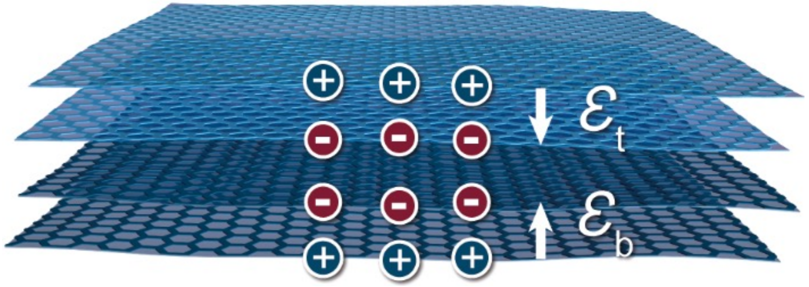


E. Portolés *et al.*,
2D Mater. **9**,
014003 (2022)
Fabry–Perot QD Physics

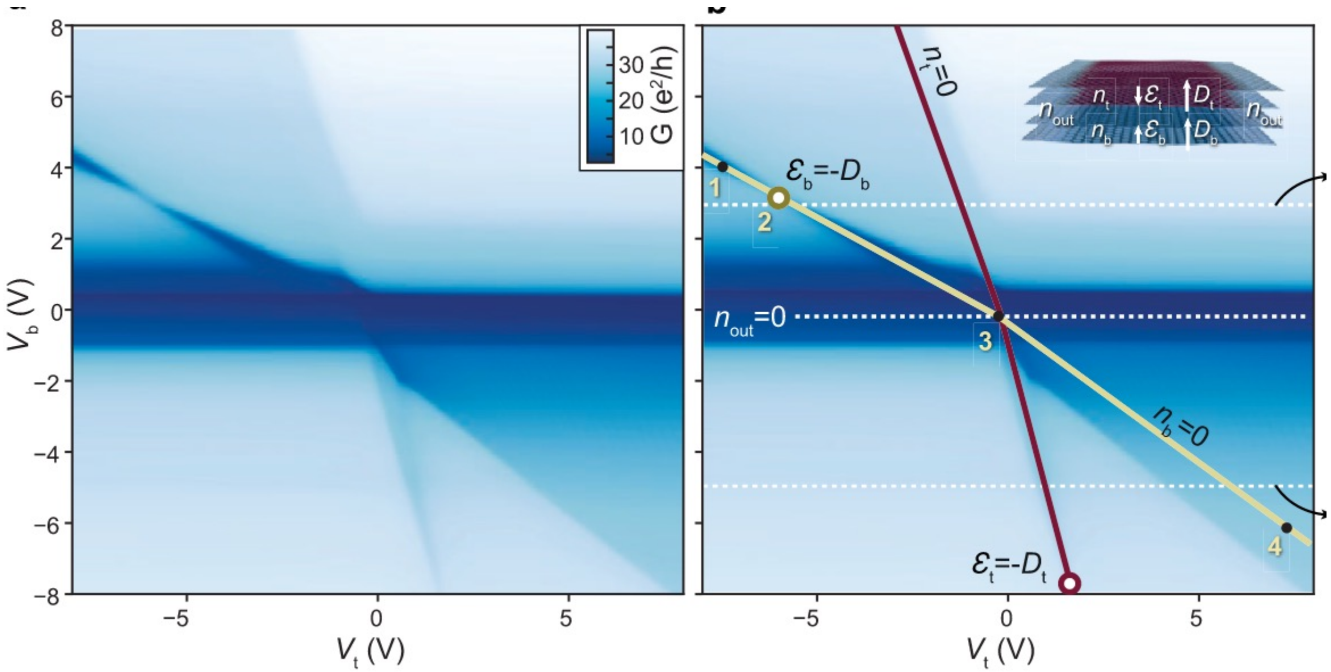
P. Rickhaus *et al.*, Nano Lett. **19**, 8821 (2019)
Crystal fields, decoupled layers

F. K. de Vries *et al.*,
Phys. Rev. Lett. **125**, 176801 (2020)
Minivalley control
P. Rickhaus *et al.*,
Science **373**, 1257 (2021)
Correlated electron-hole state

Decoupled layers:
Band gap in twisted double bilayer graphene by crystal fields

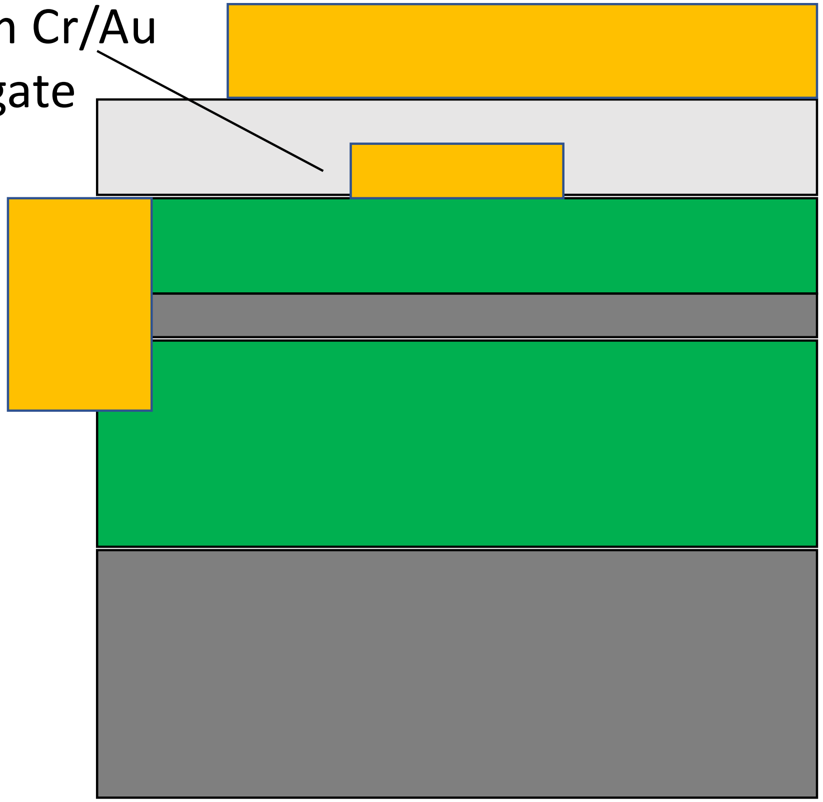


P. Rickhaus *et al*, Nano Lett. **19**, 8821 (2019).



The $\theta = 2.37°$ stack

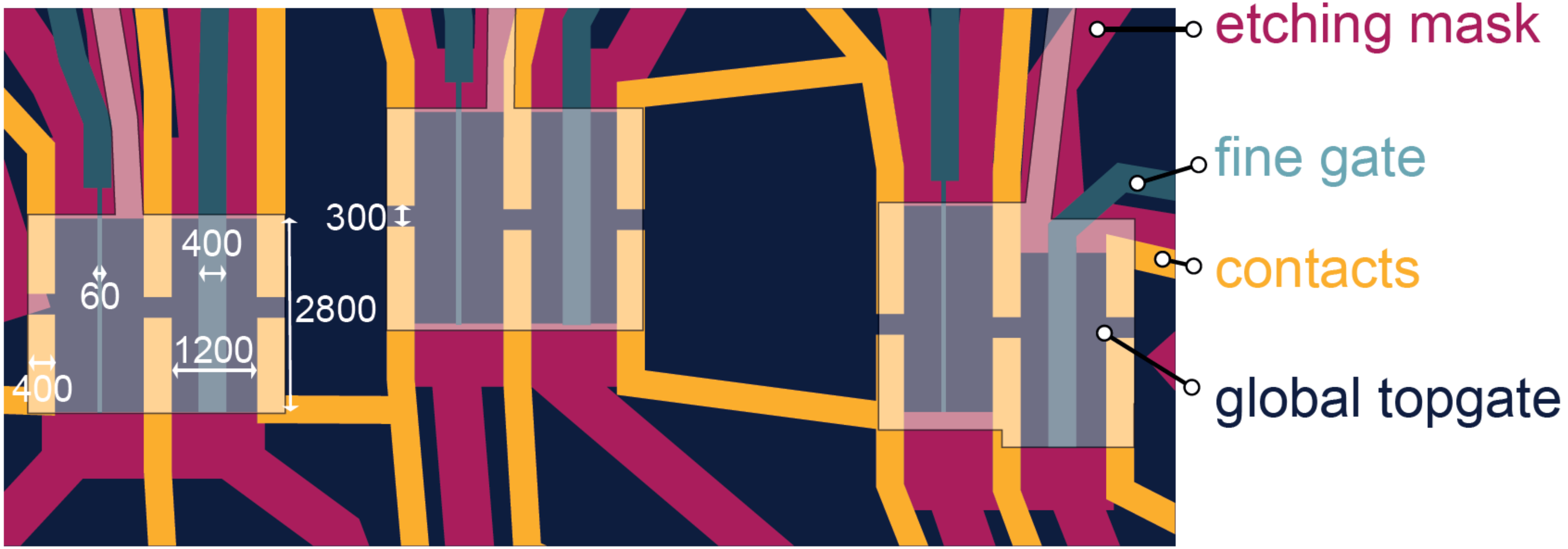
F. K.de Vries *et al.*,
Phys. Rev. Lett. **125**, 176801 (2020)



Here: the same gate voltage is applied to the fine gate and the top gate

Lateral device layout

typical carrier mobility: 25’000 cm^2/Vs
mean free path: ~ 350 nm
Moiré lattice constant: ~ 6 nm

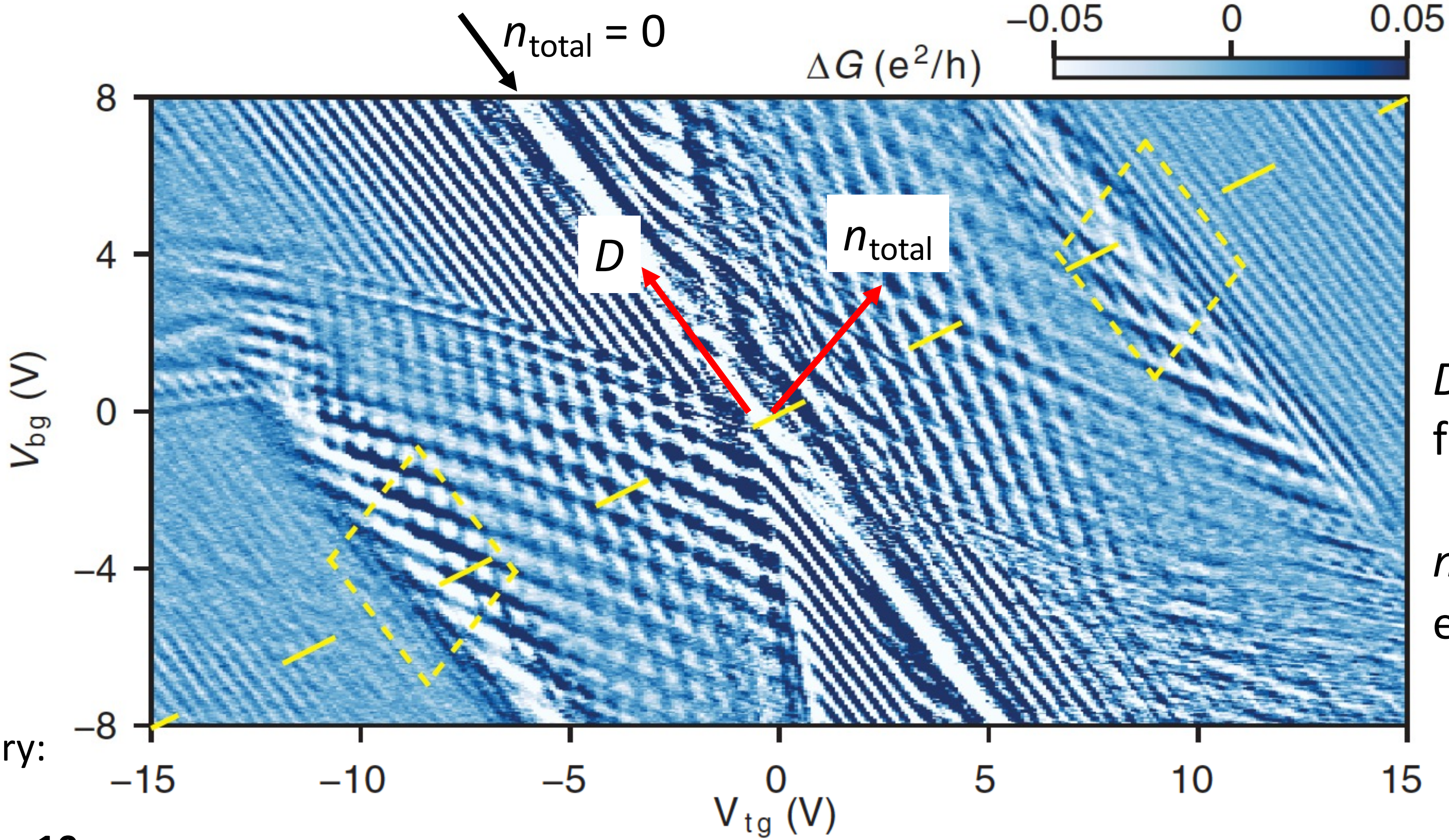


all lengths in nanometers

Here: the same gate voltage is applied to the fine gate and the top gate

Shubnikov–de Haas oscillations

F. K. de Vries *et al*,
Phys. Rev. Lett. **125**, 176801 (2020).



T = 70 mK
$B_\perp$ = 2 T

D: displacement field

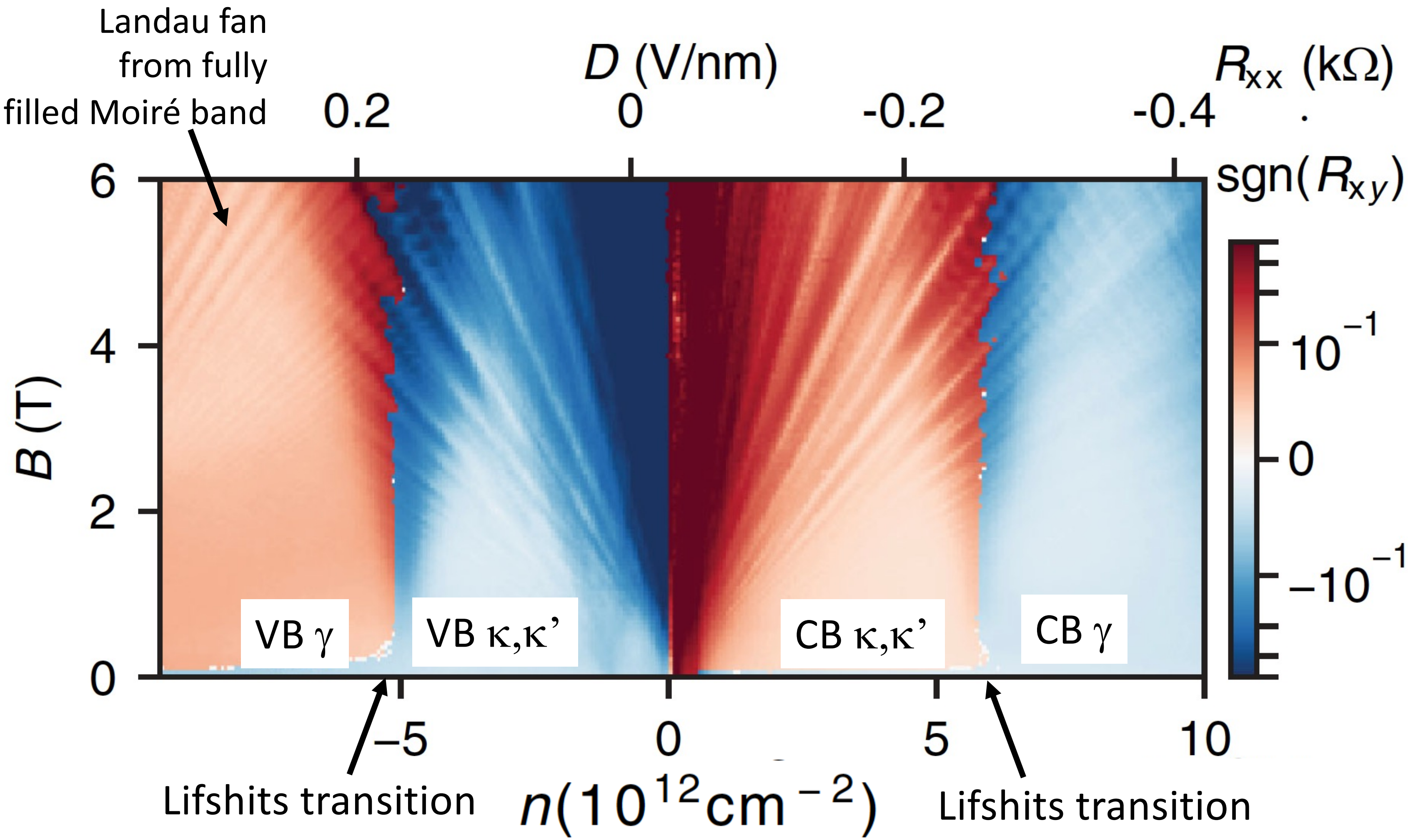
n_{total}: total electron density

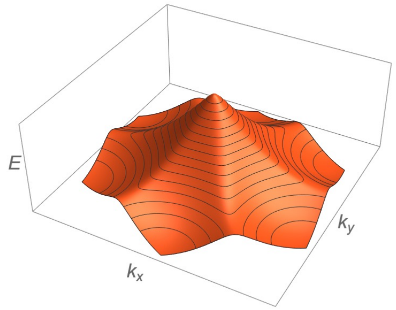
e-h asymmetry:
J.Y. Lee *et al.*,
Nat. Commun. **10**, 5333 (2019)

ΔG: Smooth background subtracted from raw data

Shubnikov–de Haas oscillations

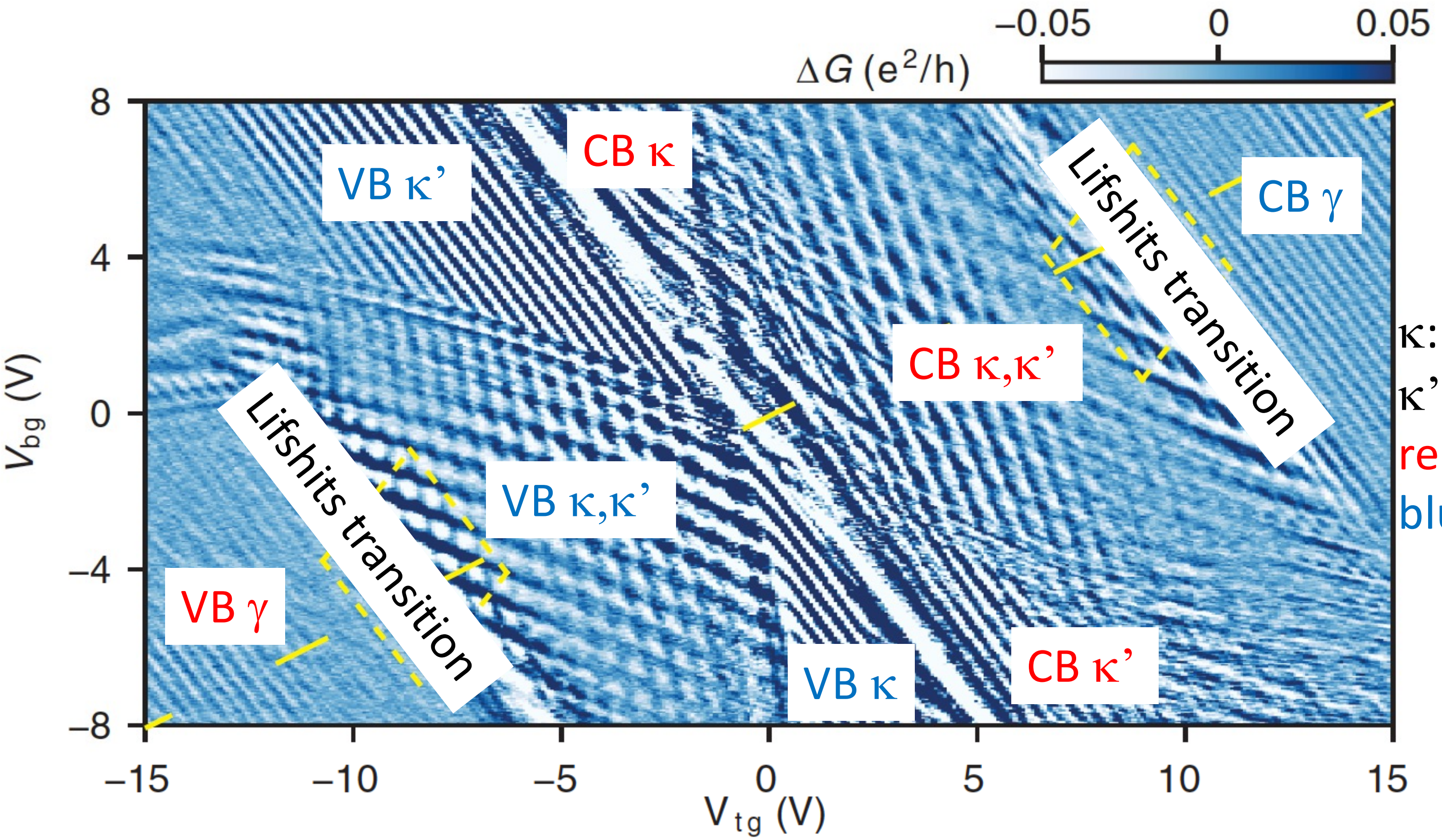
F. K. de Vries *et al*,
Phys. Rev. Lett. **125**, 176801 (2020).





Shubnikov–de Haas oscillations

F. K. de Vries *et al*,
Phys. Rev. Lett. **125**, 176801 (2020).



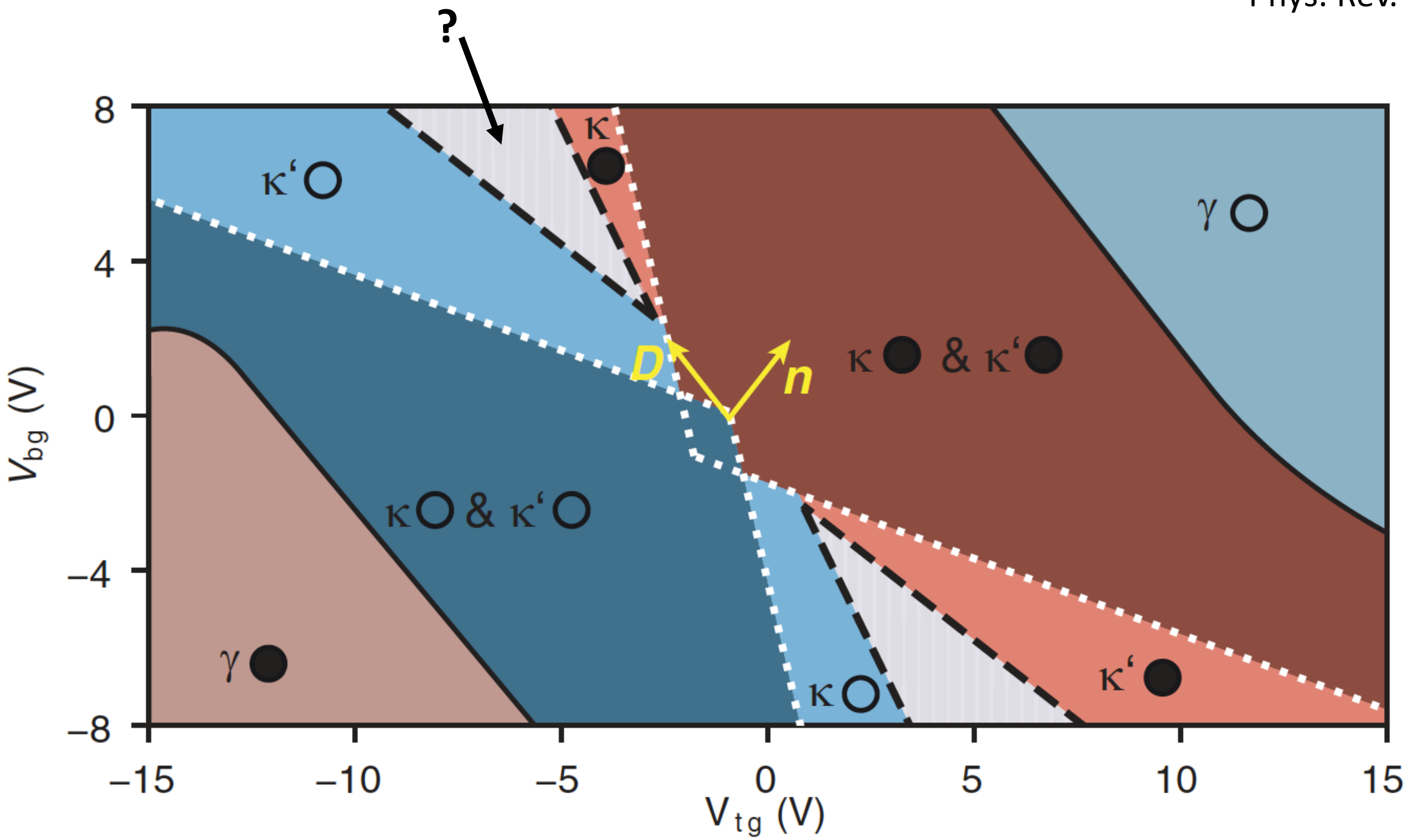
T = 70 mK
$B_\perp$ = 2 T

κ: bottom bilayer
κ': top bilayer
red: electron-like
blue: hole-like

ΔG: Smooth background subtracted from raw data

A rich phase diagram

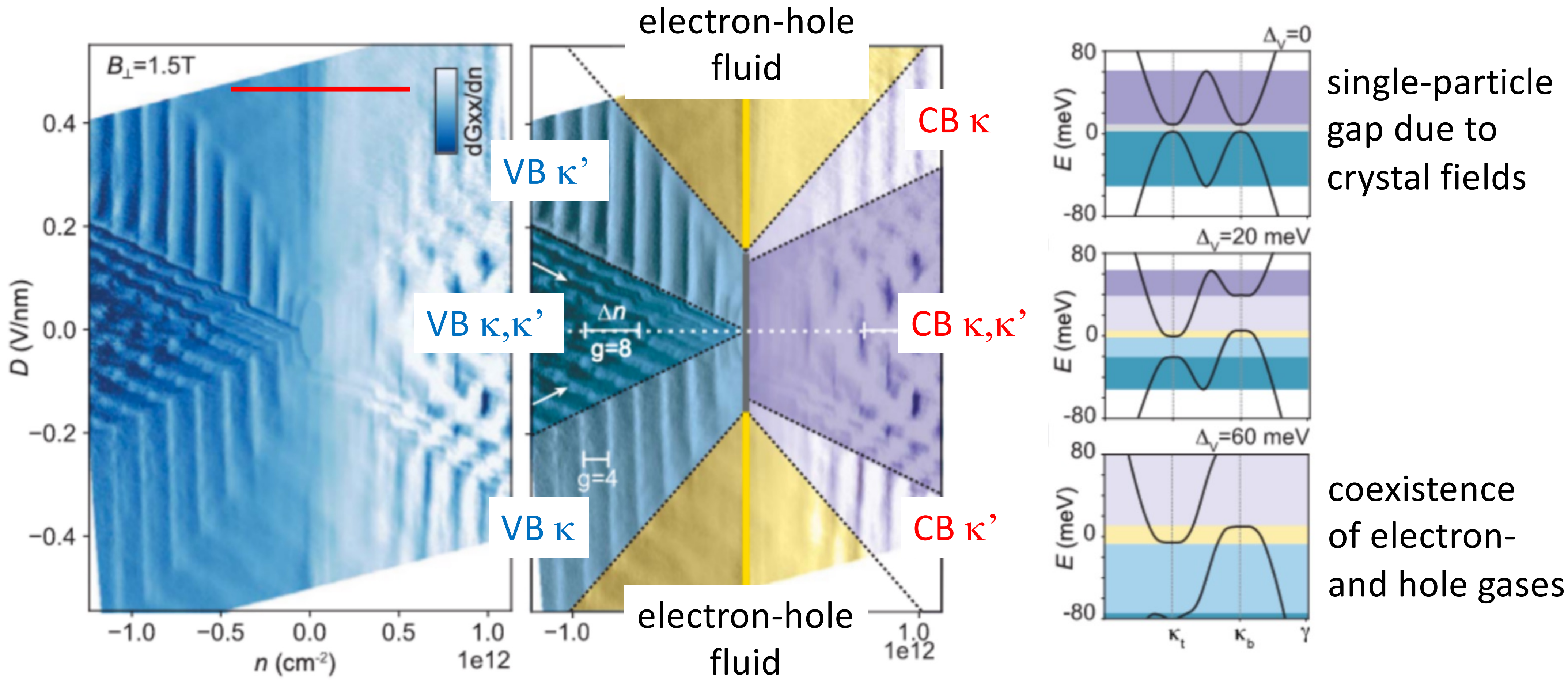
F. K. de Vries *et al*,
Phys. Rev. Lett. **125**, 176801 (2020).



κ: bottom bilayer
κ': top bilayer

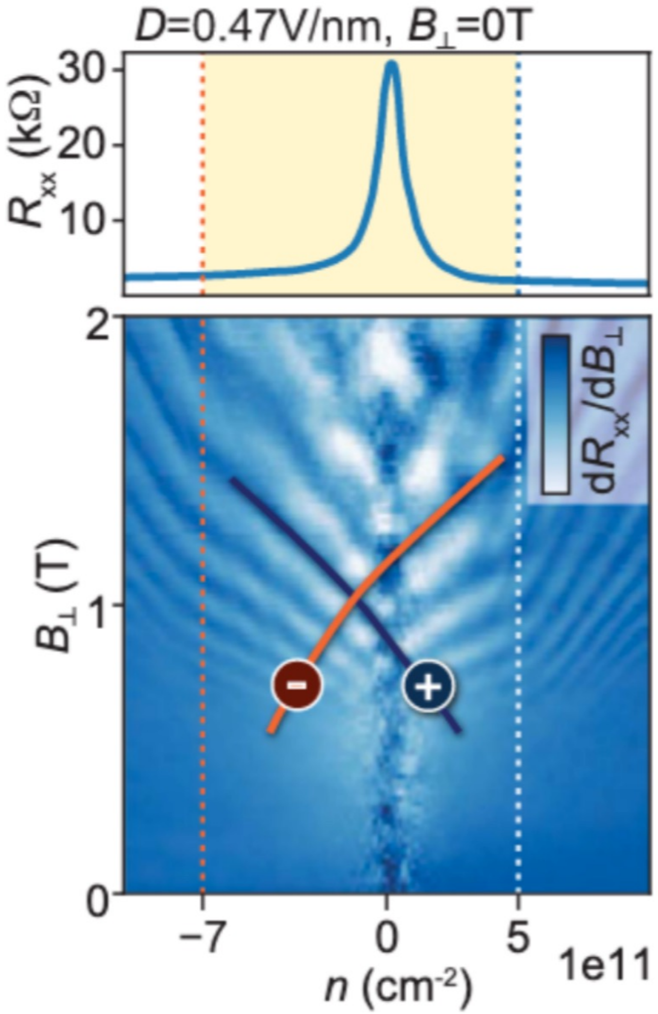
red: electron-like
blue: hole-like

Coexistence of electrons and holes in the two layers (new device!)



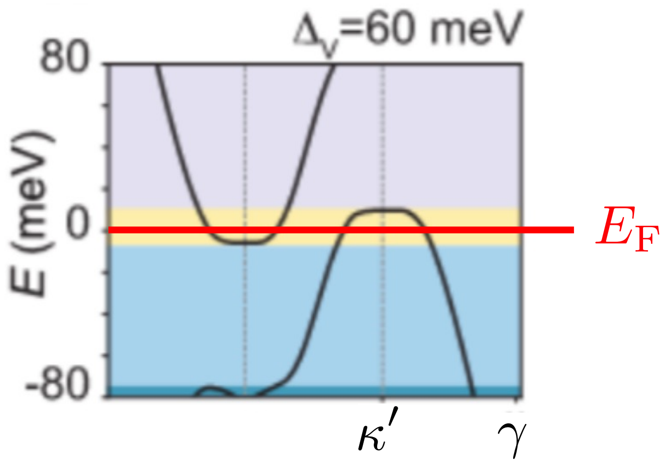
P. Rickhaus *et al.*, Science **373**, 1257 (2021)

Coexistence of electron and hole gases



Resistance peak at $B_\perp = 0$:
indicative of an energy gap
brought about by electron-hole correlations

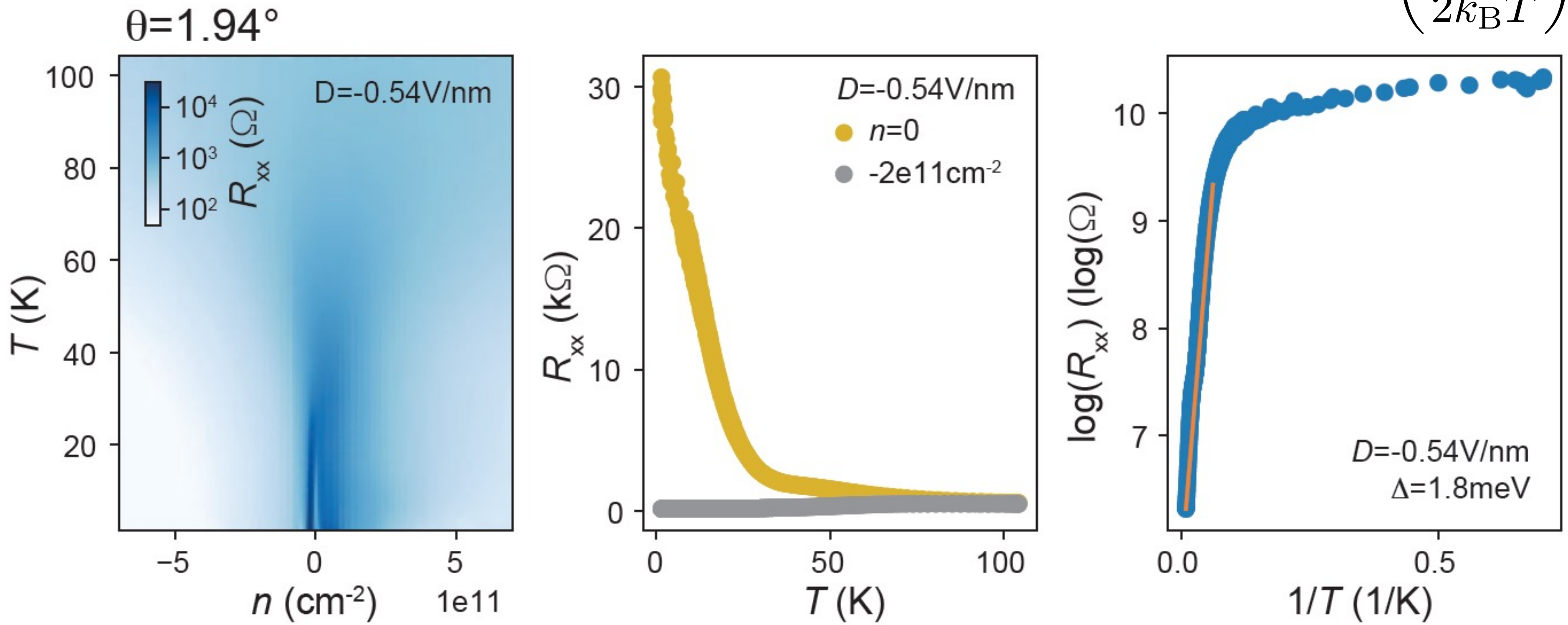
in-plane separation of carriers: ~ 10 nm
separation of e/h layers: ~ 4 Å



P. Rickhaus *et al.*, Science **373**, 1257 (2021)

Quantifying the energy gap

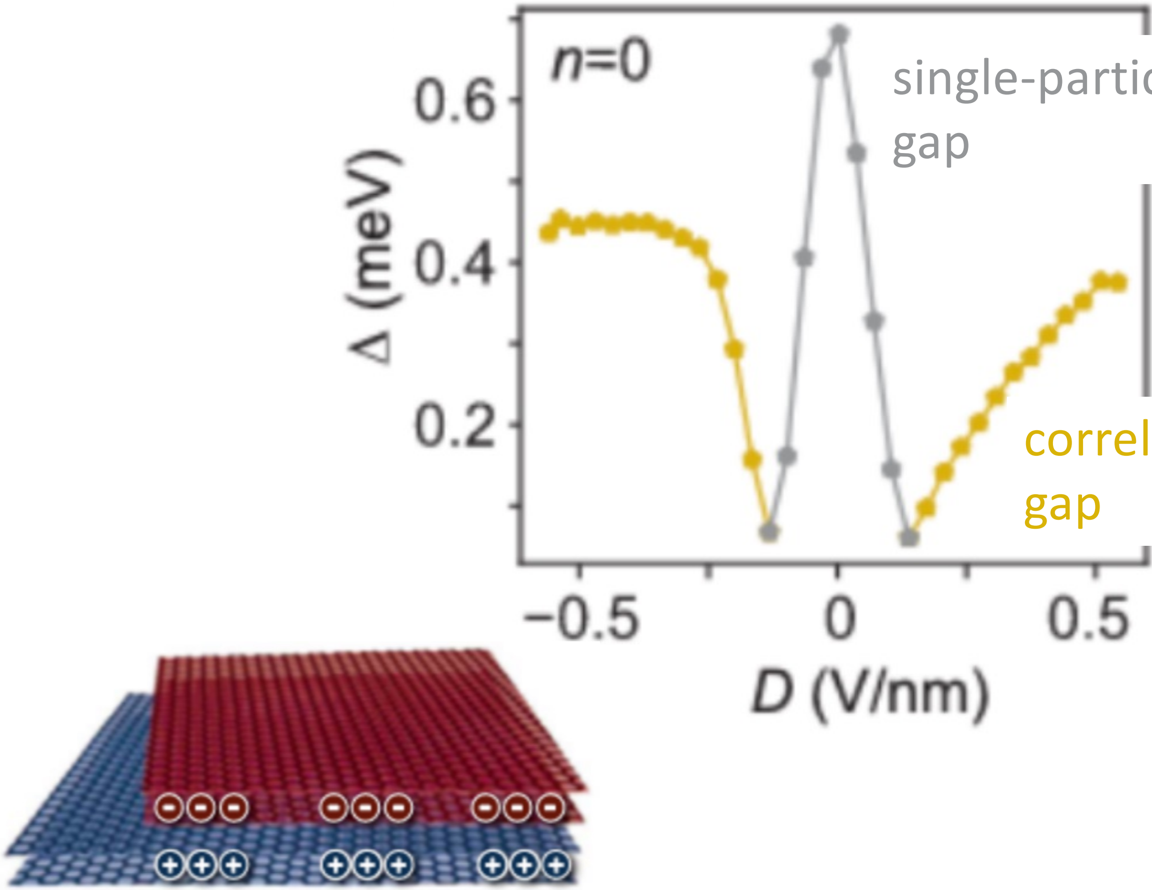
$$R_{xx} \propto \exp\left(\frac{\Delta}{2k_\mathrm{B}T}\right)$$



P. Rickhaus *et al.*, Science **373**, 1257 (2021)

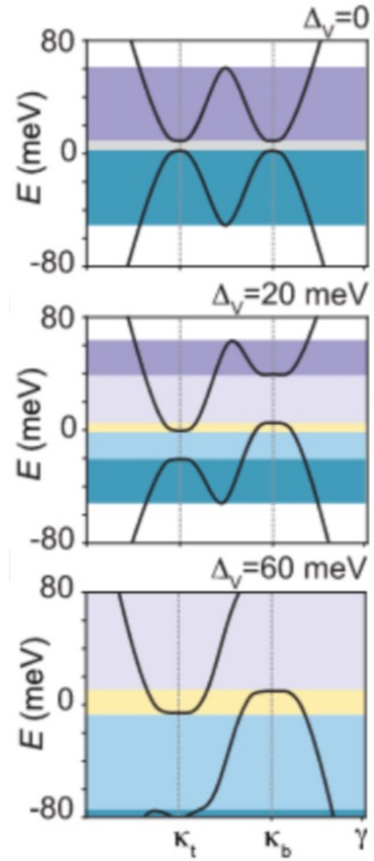
Energy gap as a function of displacement field

measured at charge neutrality from activation



single-particle gap

correlation gap

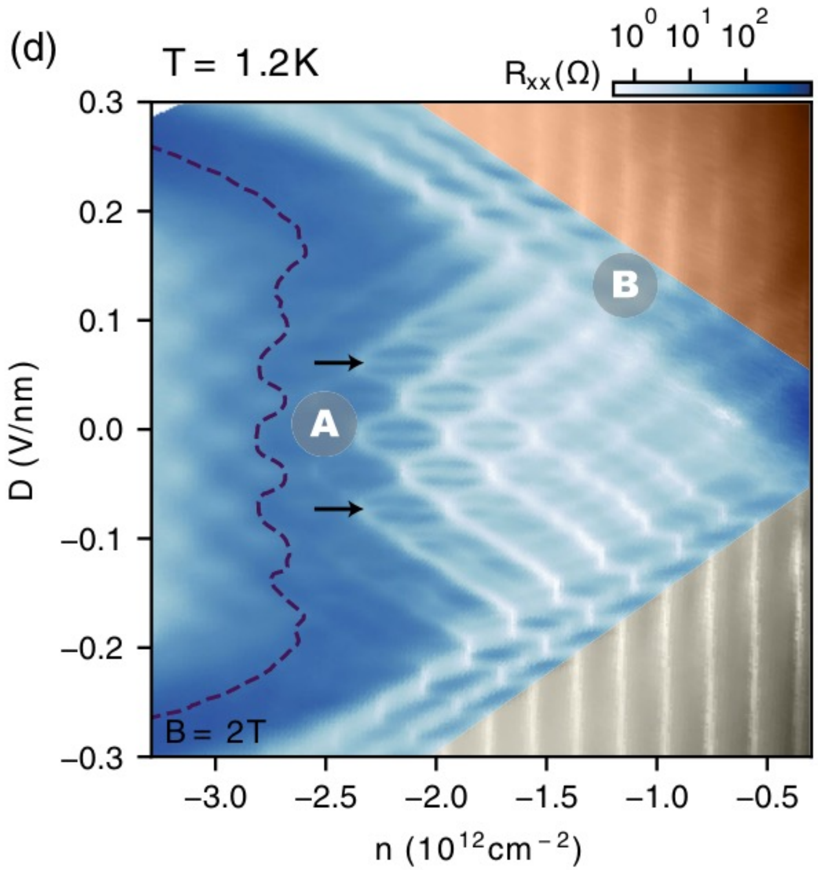


single-particle gap due to crystal fields

coexistence of electron- and hole gases

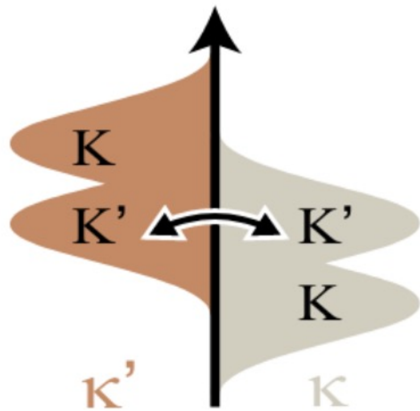
P. Rickhaus *et al.*, Science **373**, 1257 (2021)

Scattering between minivalleys in twisted double bilayer graphene



P. Tomić *et al*, Phys. Rev. Lett. **128**, 057702 (2022).

Resistance peaks analogous to magneto-intersubband oscillations



Conclusion

- Twisting 2D materials offers completely new ways of tailoring materials
- The resulting physics is rich and full of surprises
- Twisted materials can be the basis for monolithic devices, in which multiple phases of matter are combined