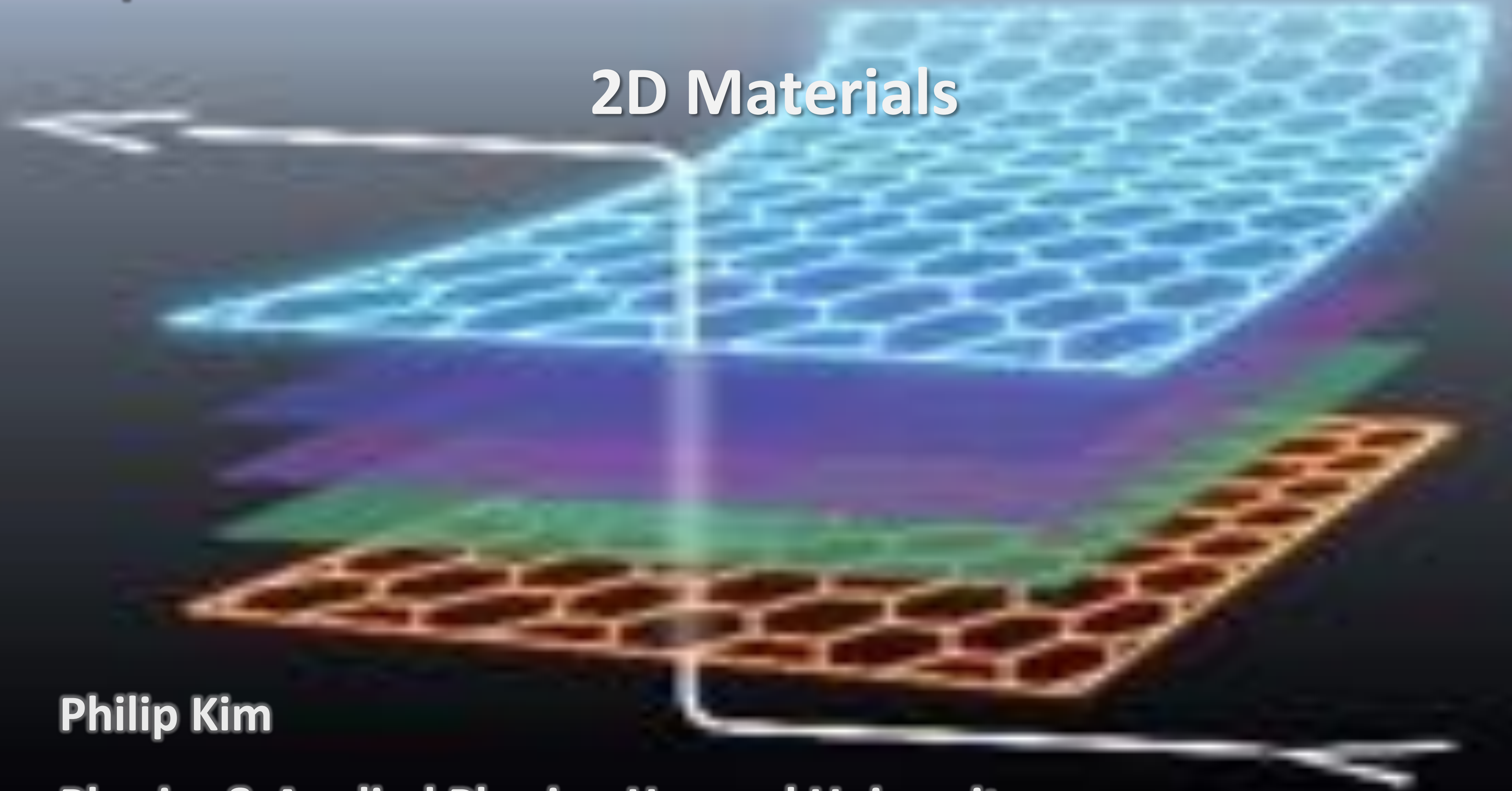
Experiments in Twisted van der Waals Interface of 2D Materials

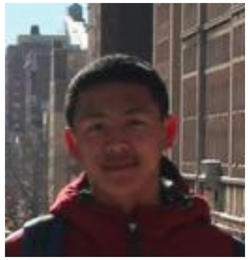


Philip Kim

Physics & Applied Physics, Harvard University

Acknowledgement

Experiments



Zeyu Hao

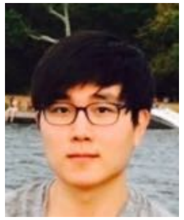
Xiaomeng Liu
(now at Princeton)

Andrew
Zimmerman

Isabelle Phinney

Theory

Ashvin Vishwanath

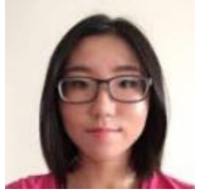


Jong Yeon Lee

Eslam khalaf

Patrick Ledwith

Efthimios Kaxiras



Zoe Zhu

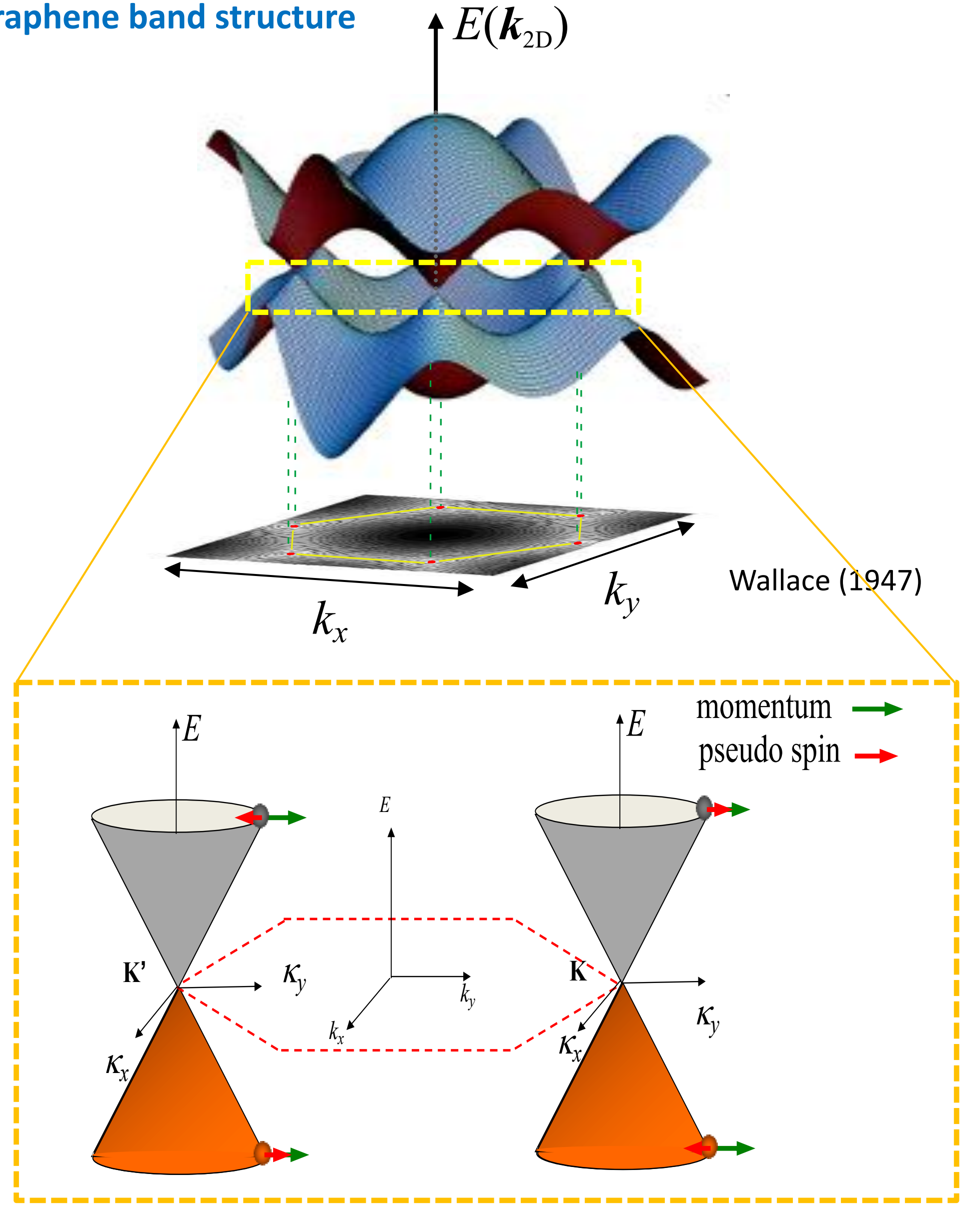
hBN

T. Taniguchi, K. Watanabe

Funding:

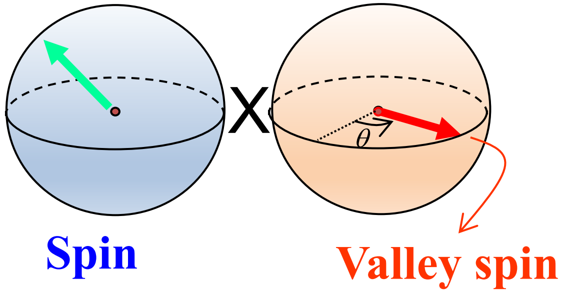
Graphene and SU(4) Spin

Graphene band structure



Graphene: possibility of SU(4) magnetism

$$SU(2) \otimes SU(2)$$



$$SU(4)\quad \begin{bmatrix} \psi_{K\Uparrow} \\ \psi_{K\Downarrow} \\ \psi_{K'\Uparrow} \\ \psi_{K'\Downarrow} \end{bmatrix}$$

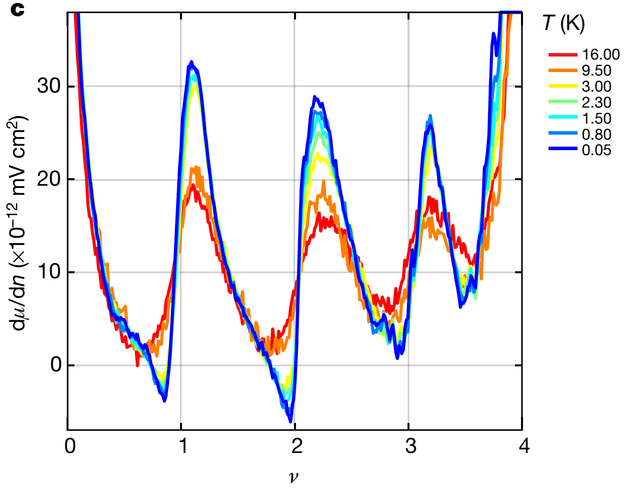
SU(4) **symmetry is conjectured to produce rich new set of ground state wavefunctions in graphene**

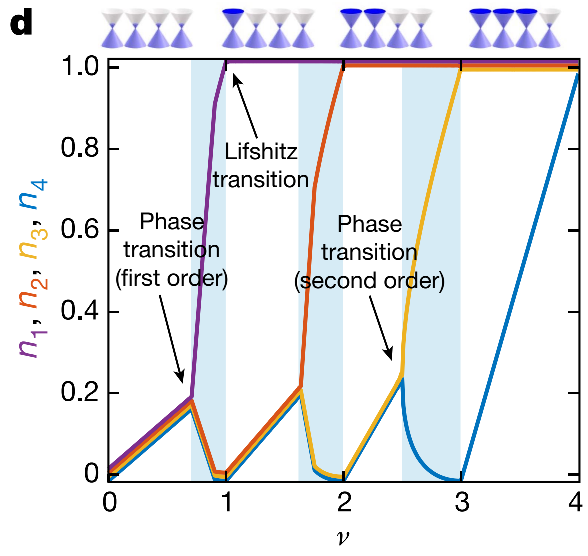
Yang, Das Sarma and MacDonal, PRB (2006);

Graphene Moire Chern Insulators: Broken Symmetry

Scanning SET

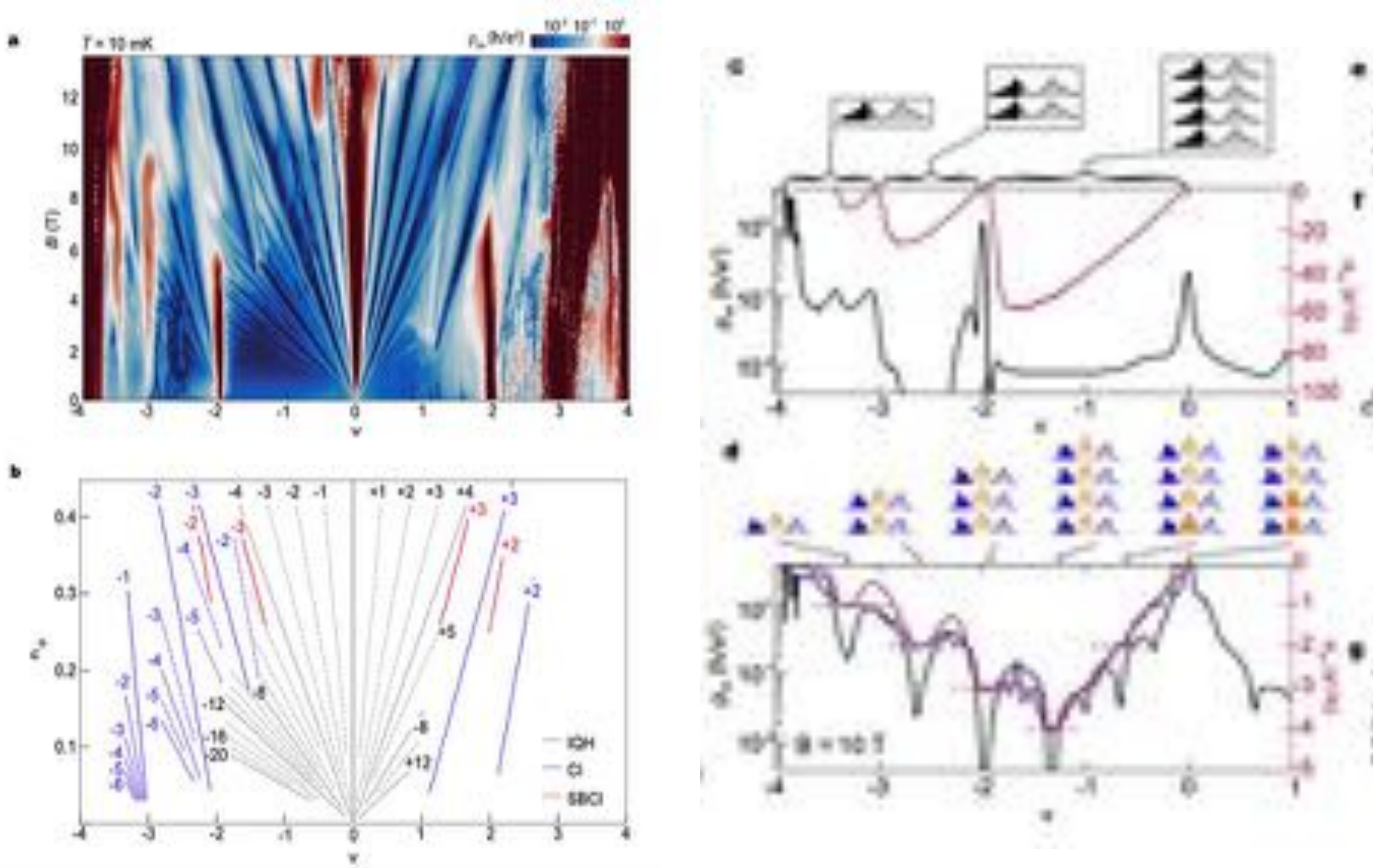
U. Zondiner *et al*, Nature 582, 203 (2020)





Cascade Transition: broken flavour symmetry

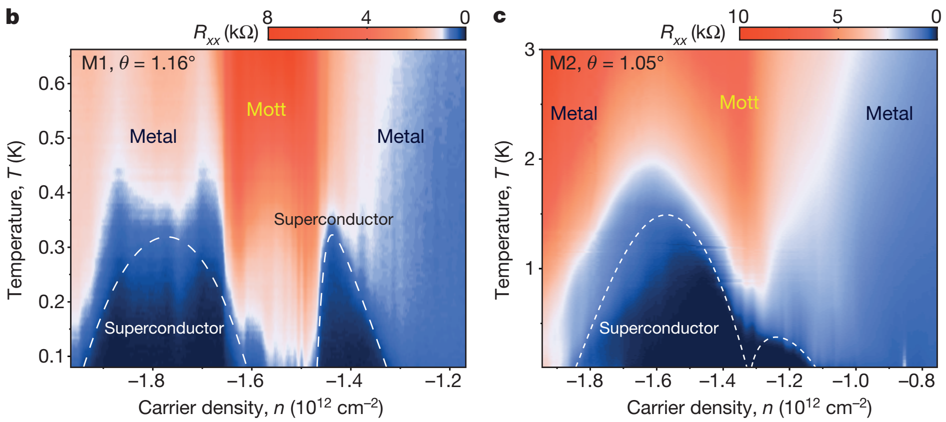
Y. Saito et al, Nature Physics 17, 478 (2021)

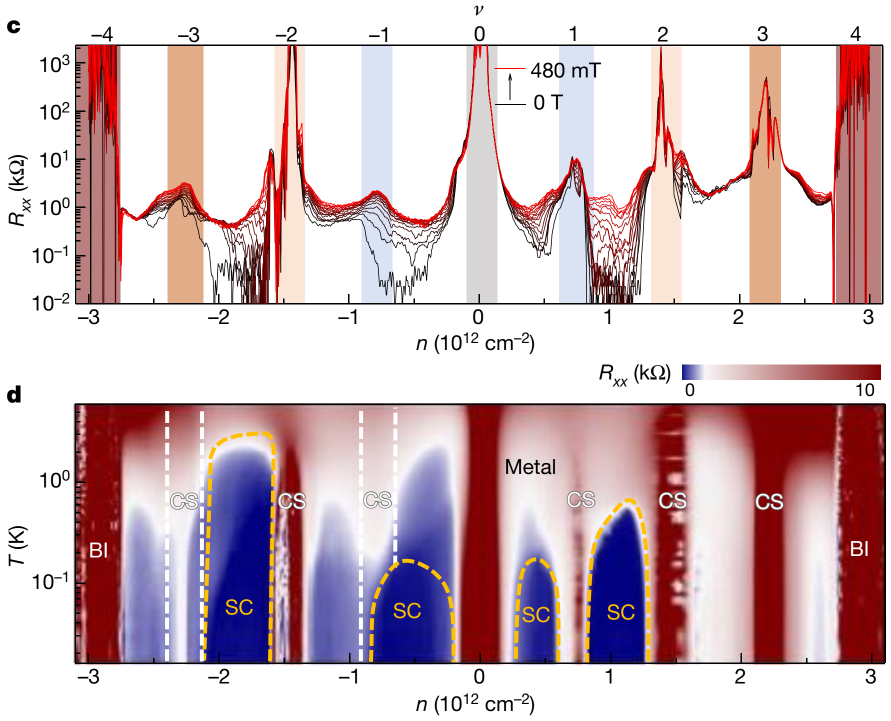


Unusual Fan Diagram: magnetic field induced Chern bands

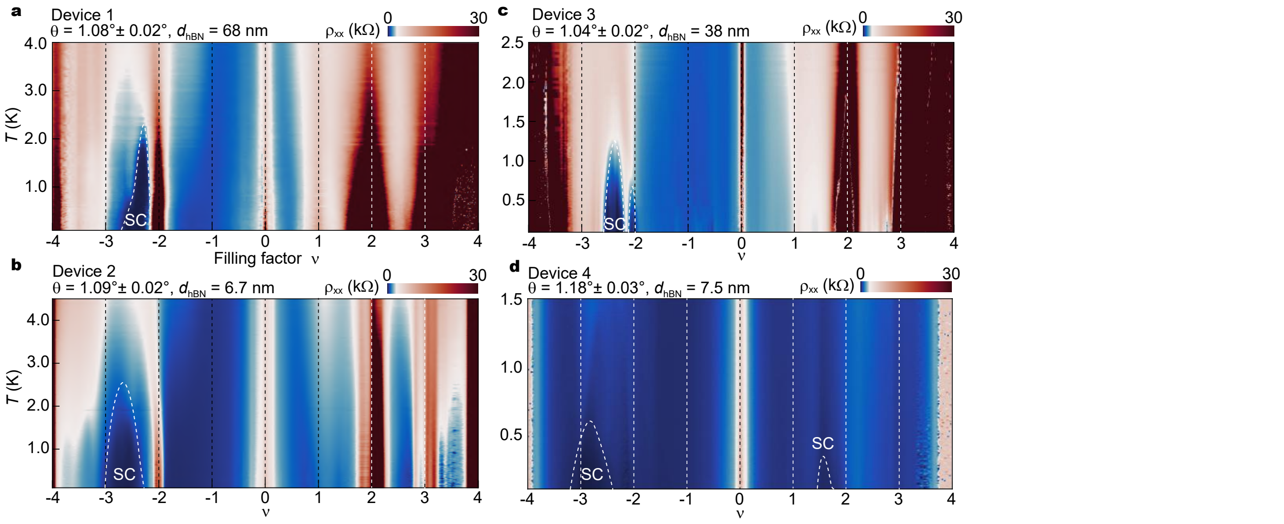
Superconducting Phase Diagram for MA TBG

Cao et al., Nature 556, 43–50 (2018).

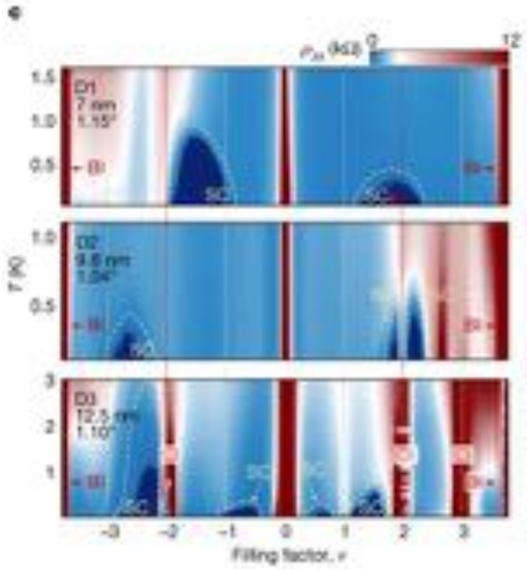




Lu et al, Nature 574, 653–657 (2019).



Saito et al., Nature Physics 16, 926-930 (2020)(see also Singh et al., Nature 583, 379 - 384 (2020))



Stepanov et al., Nature 583, 375 (2020)

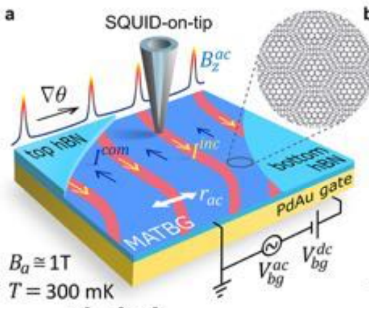
What is relation between superconductors and correlated insulators?

What is the nature correlation in the insulator?

Technical Issues: Local Variation of Twisting Angles

Scanning SQUID Measurement

Uri et al., Nature 581, 47 (2020)



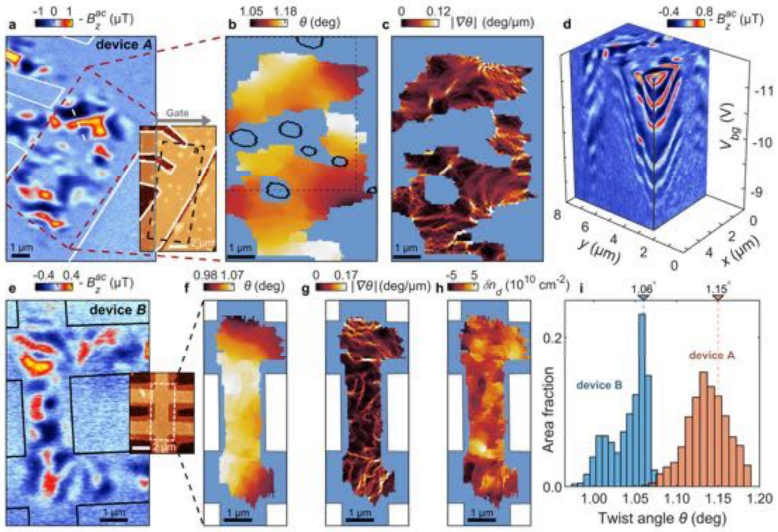
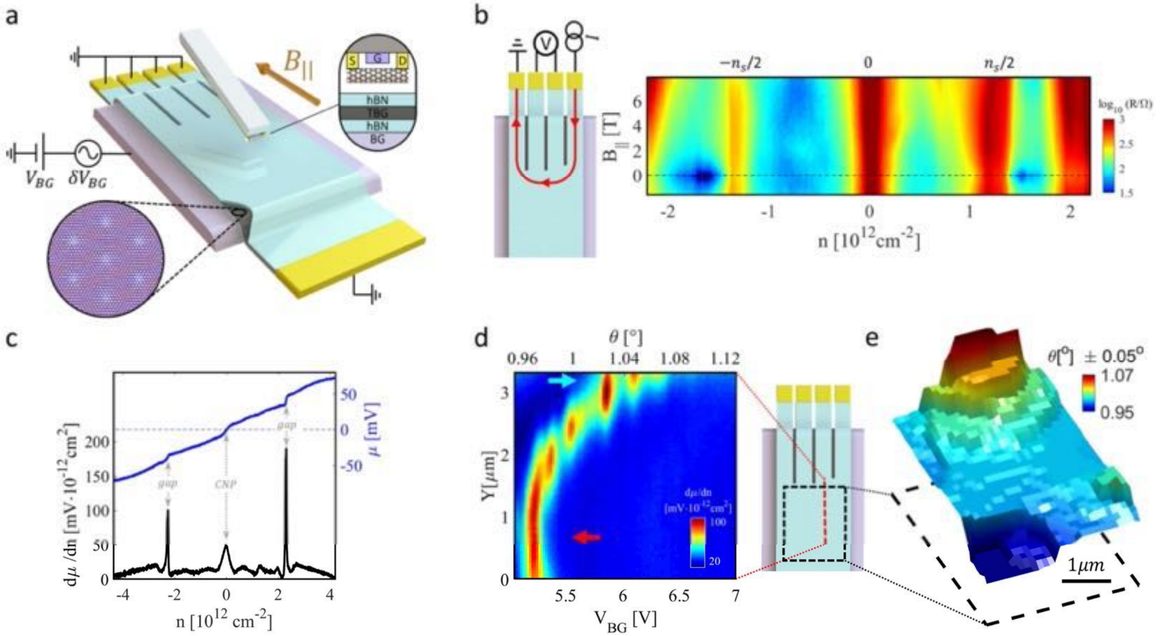


Fig. 3. Mapping the twist angle and Landau levels in MATBG. (a) B_z^{ac} image of the dashed area in the



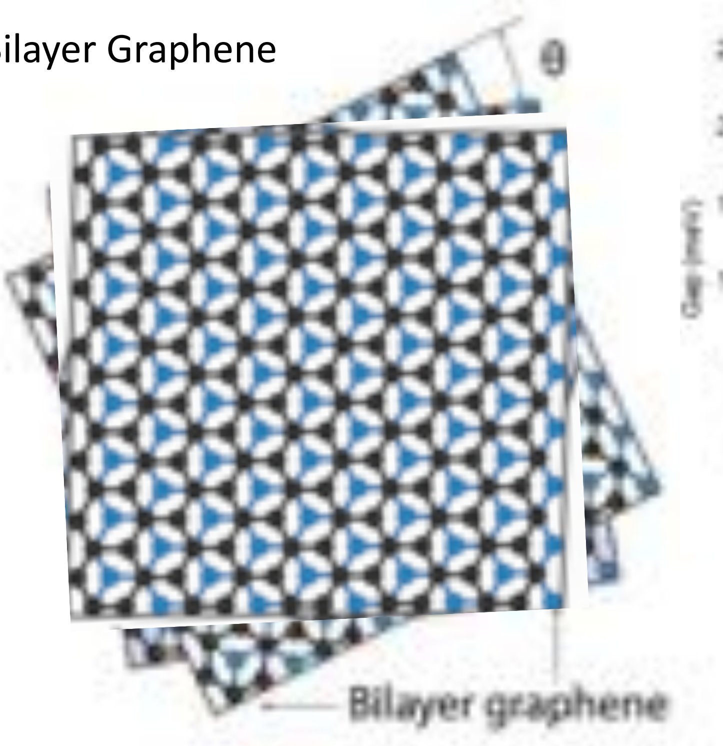
Scanning nanotube SET

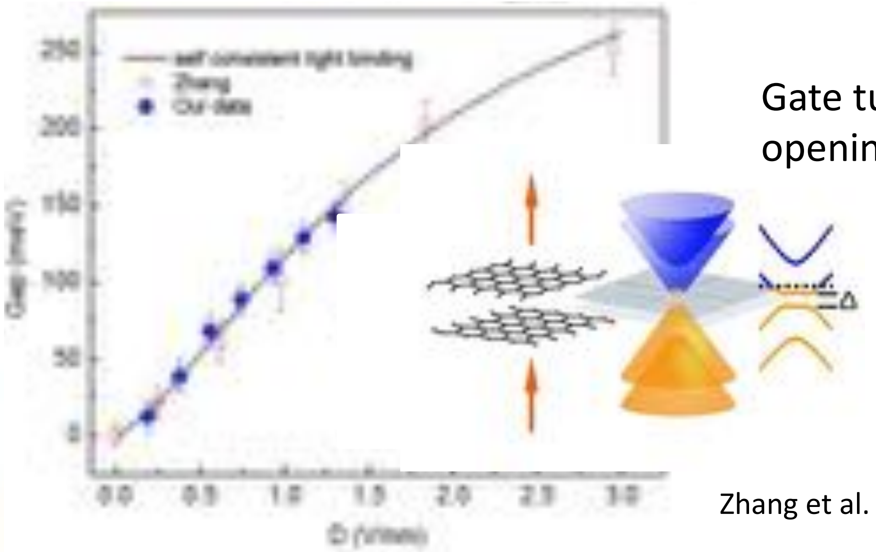
Zondiner et al., Nature, 582,203, (2020)

Every sample is different!!

Twisted Double Bilayer Graphene

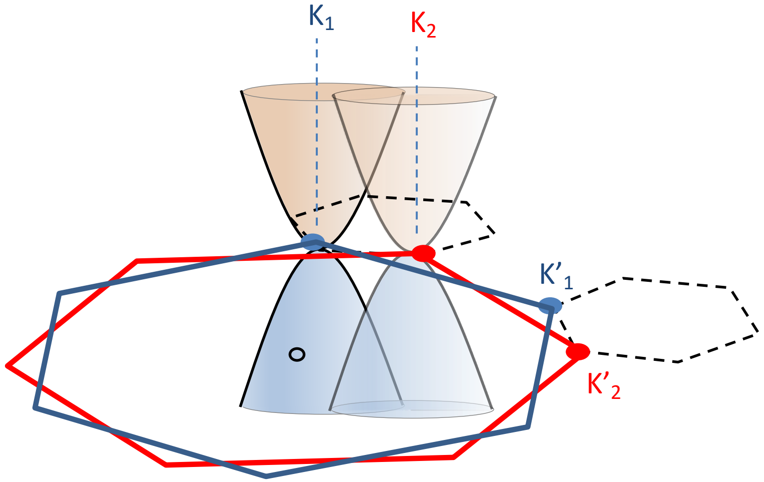
Bernal Stacked Bilayer Graphene



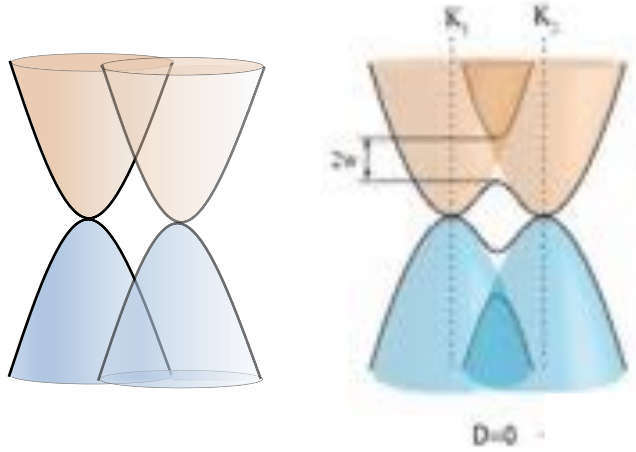


Gate tunable Gap opening in bilayer

Zhang et al. Nature (2008)

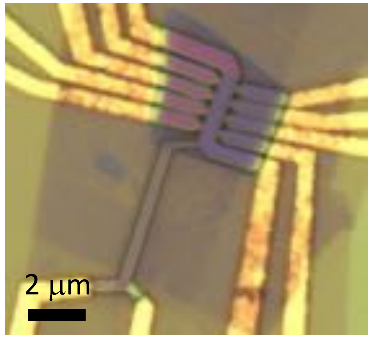


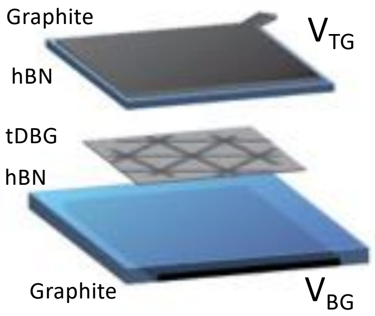
Twisted Double Bilayer Graphene (tDBG)



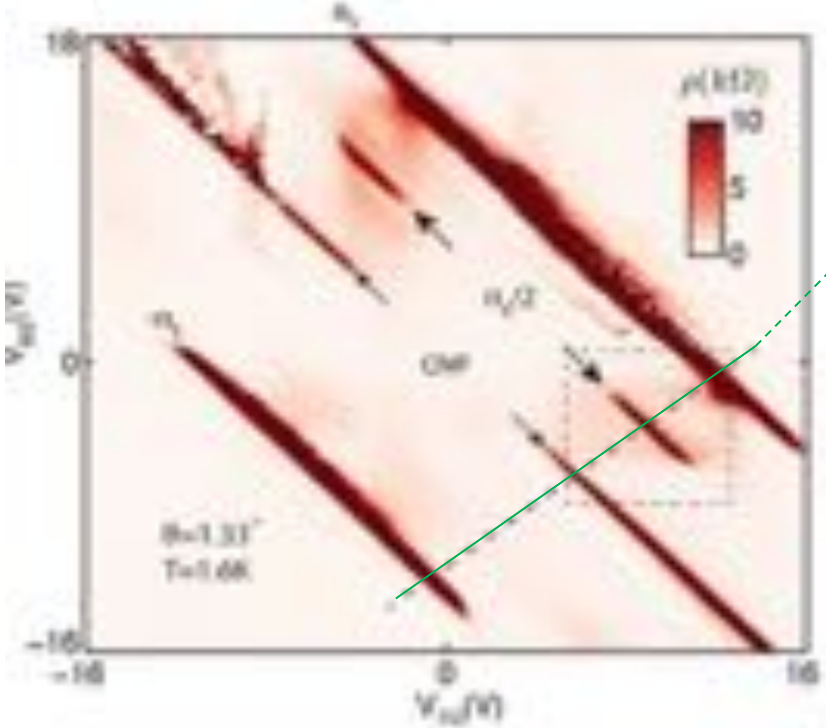
Gate tunable flat bands, no exact angle control needed!

Mott Insulators in tDBG: $\theta = 1.33^\circ$

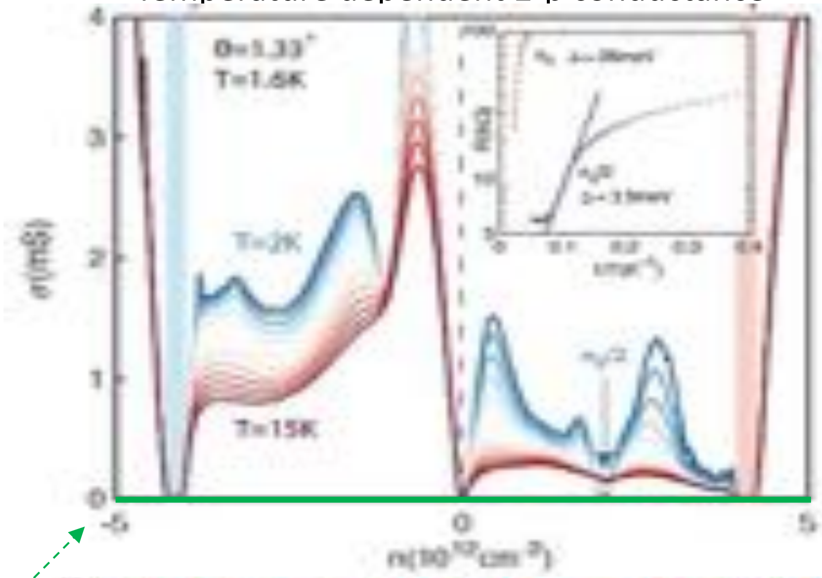


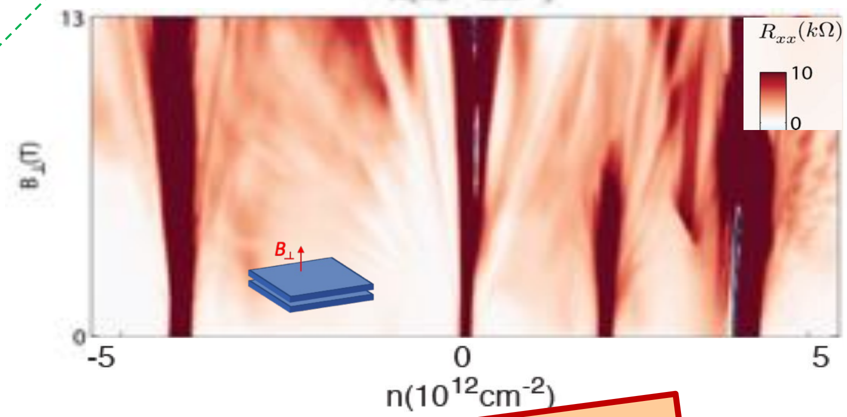


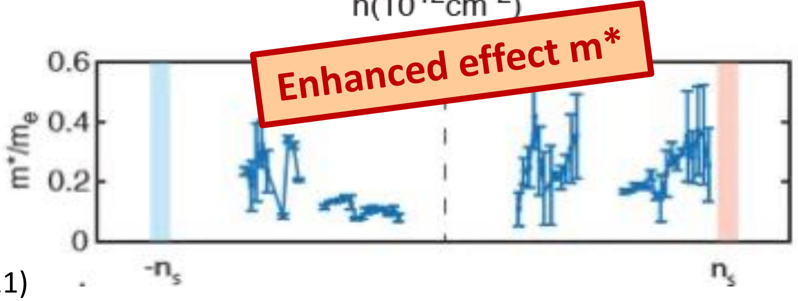
Top and bottom Gate dependent 4-terminal ρ



Temperature dependent 2-p conductance



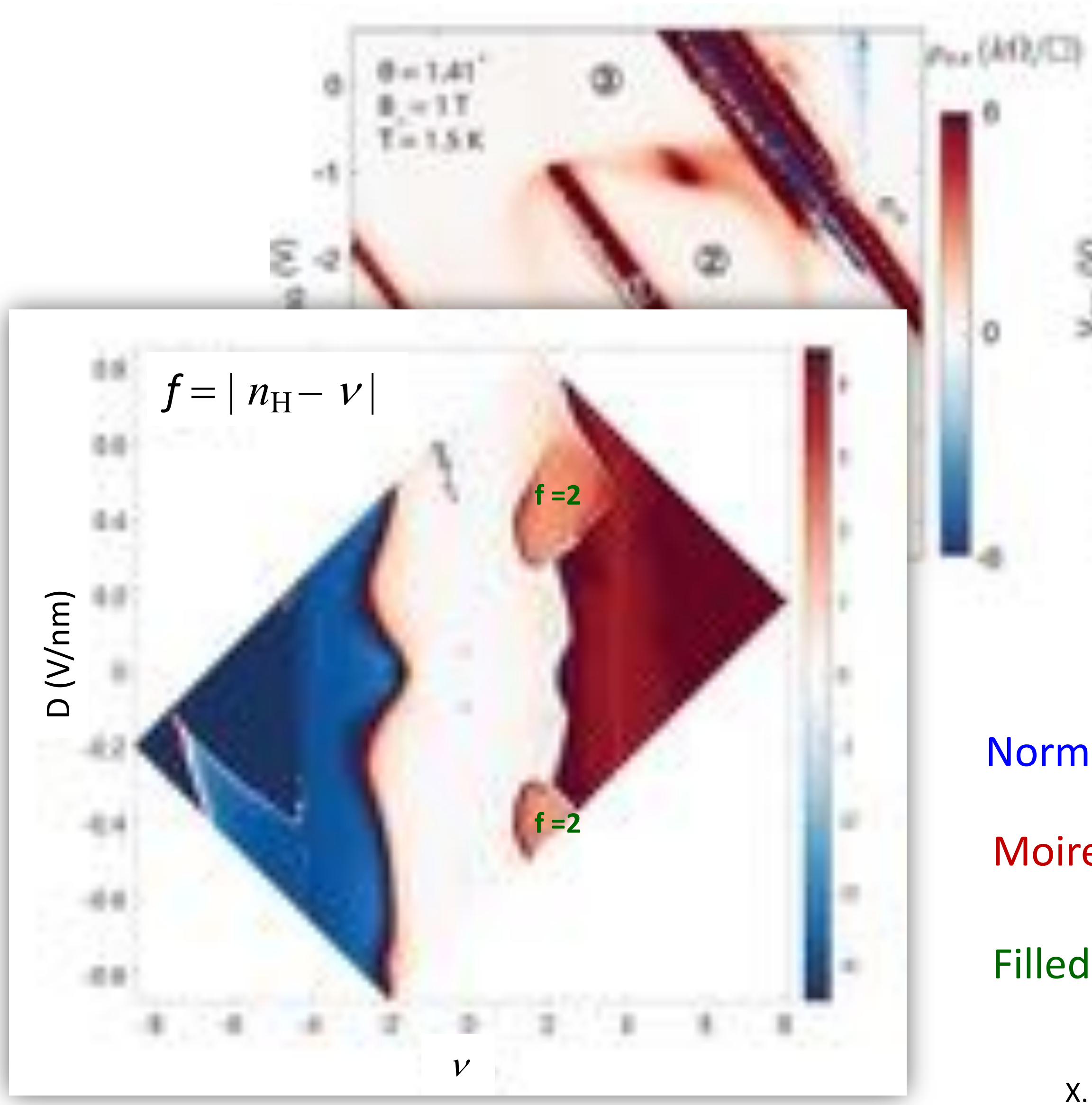




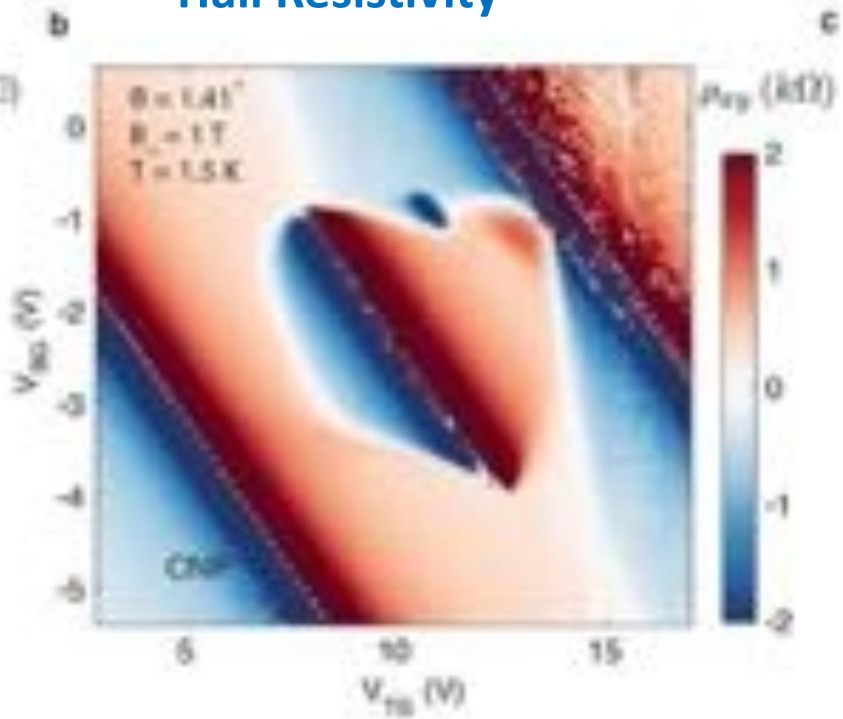
X. Liu et al., Nature (2020), similar results are also in Cao *et al*,, Nature (2020); He *et al*,, Nature Phys (2021)

Tunable Spin-Polarized Correlated States

Longitudinal resistivity



Hall Resistivity



Normalized Hall density: $n_H = B/e\rho_{xy}n_s$

Moire filling: $\nu = 4n/n_s$

Filled bands $f = |n_H - \nu|$

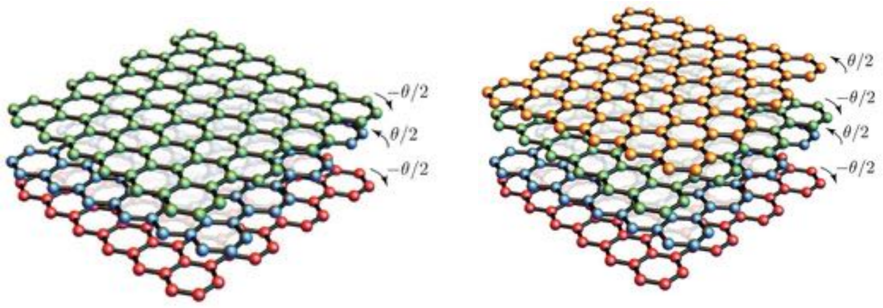
Spin-polarized correlated insulators and metals near half-filled moire flat bands

X. Liu et al., Nature (2020), similar results are also in Cao *et al*,, Nature (2020); He *et al*,, Nature Phys (2021)

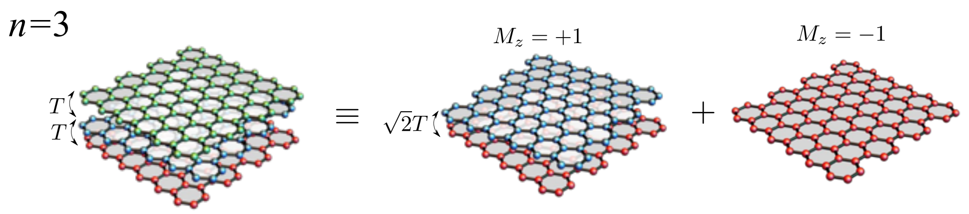
Multi-layer Graphene Moire

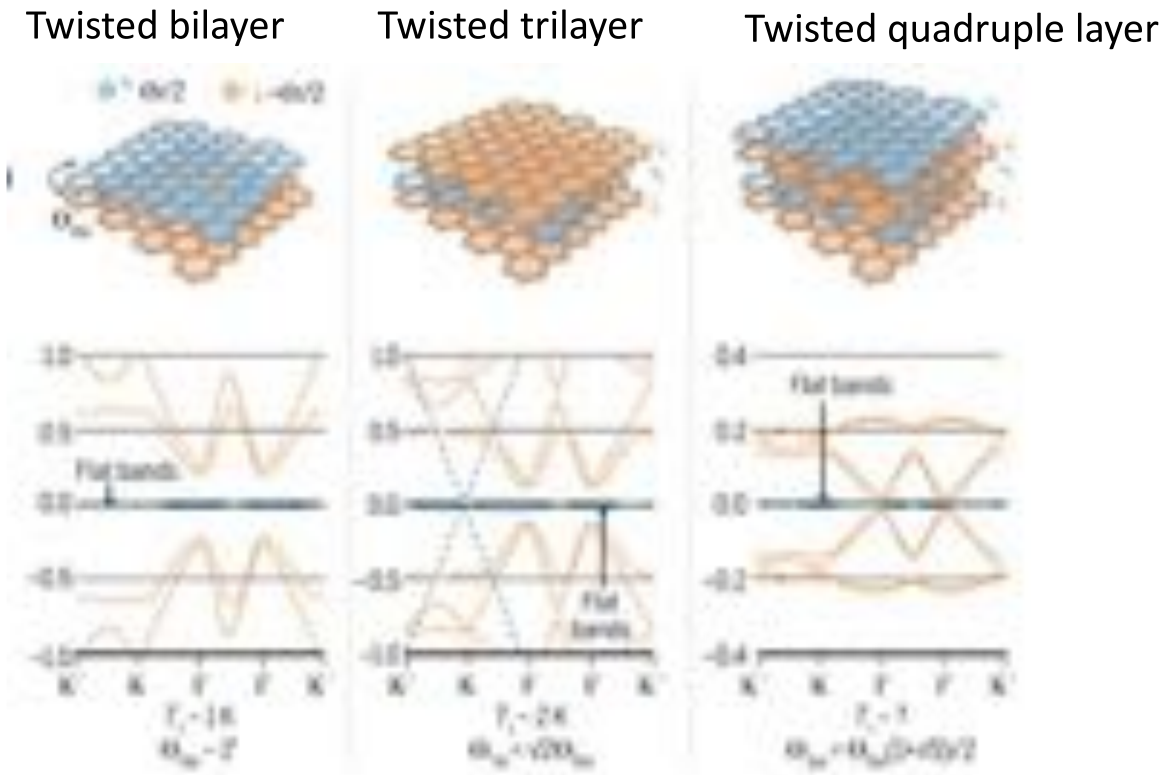
E. Khalaf, A. Kruchkov, G. Tarnopolsky, and A. Vishwanath, PRB 100, 085109 (2020)

Mulitlayer twisted stacked graphene with alternative angles



$$H(\boldsymbol{r}) = \begin{pmatrix} -iv\boldsymbol{\sigma}_+ \cdot \boldsymbol{\nabla} & T(\boldsymbol{r}) & 0 & \cdots \\ T^\dagger(\boldsymbol{r}) & -iv\boldsymbol{\sigma}_- \cdot \boldsymbol{\nabla} & T^\dagger(\boldsymbol{r}) & \cdots \\ 0 & T(\boldsymbol{r}) & -iv\boldsymbol{\sigma}_+ \cdot \nabla & \cdots \\ \cdots & \cdots & \cdots & \cdots \end{pmatrix}$$

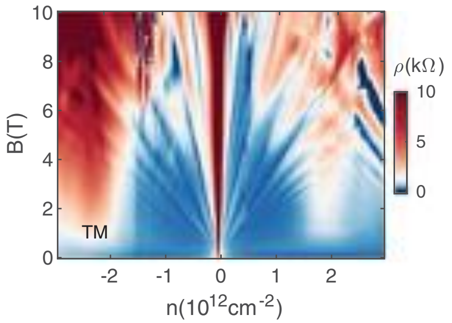


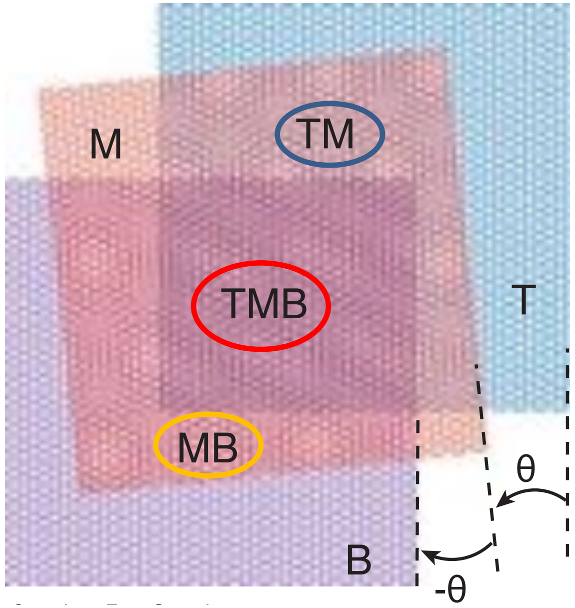


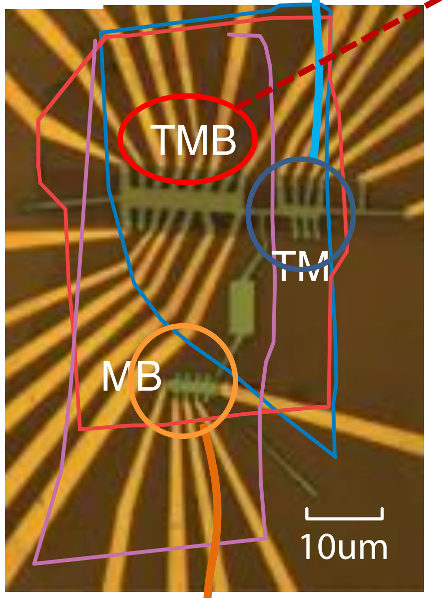
- $2n+1$ layers: single Dirac cone + n copies of TGB at different interlayer coupling
- $2n$ layers: n copies of TGB at different interlayer coupling

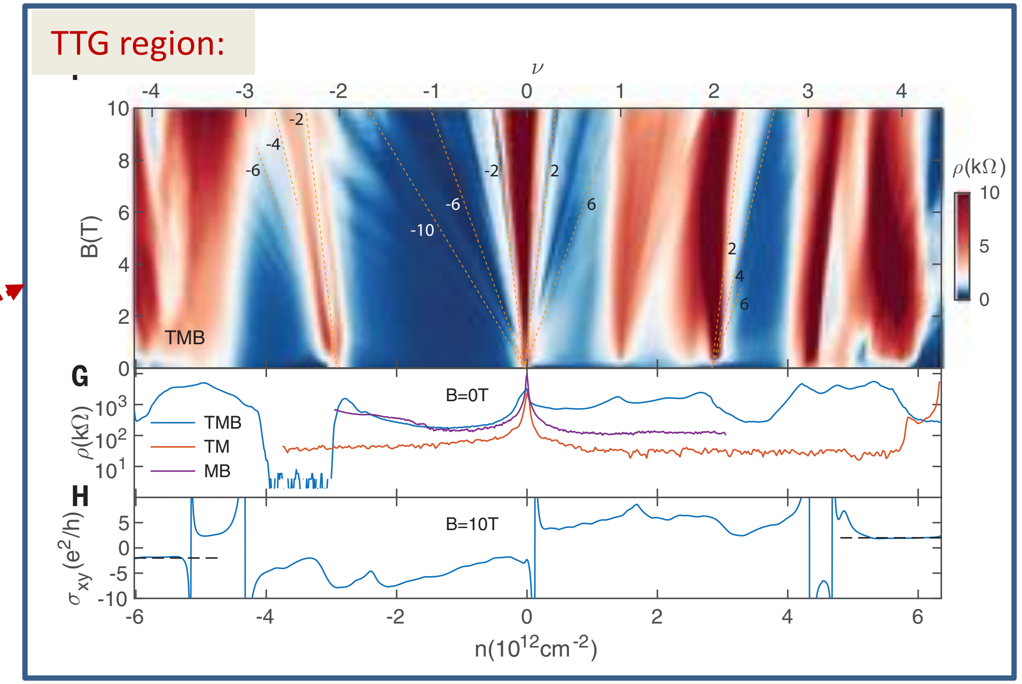
Twisted Trilayer Graphene with Alternating Angle

Top-Middle
$\theta = 1.35^{\circ}$



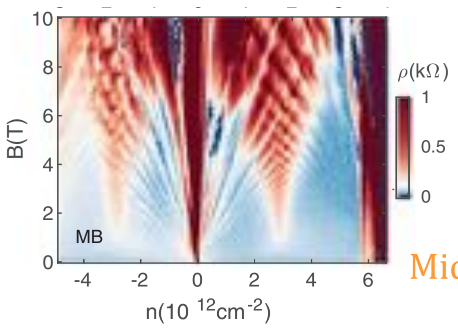






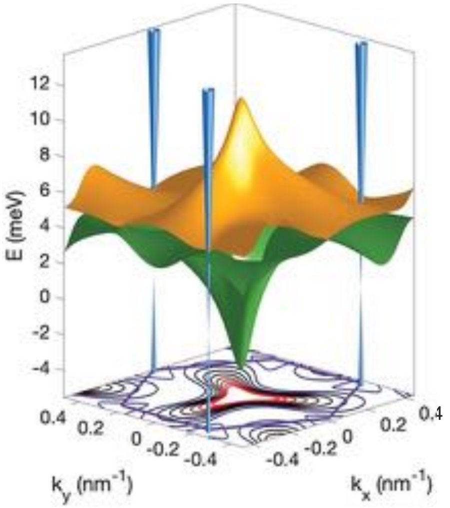
Estimated Angle $\theta = 1.55^{\circ}$

Target angle $\theta = 1.56^{\circ}$



Middle-Bottom
$\theta = 1.66^{\circ}$

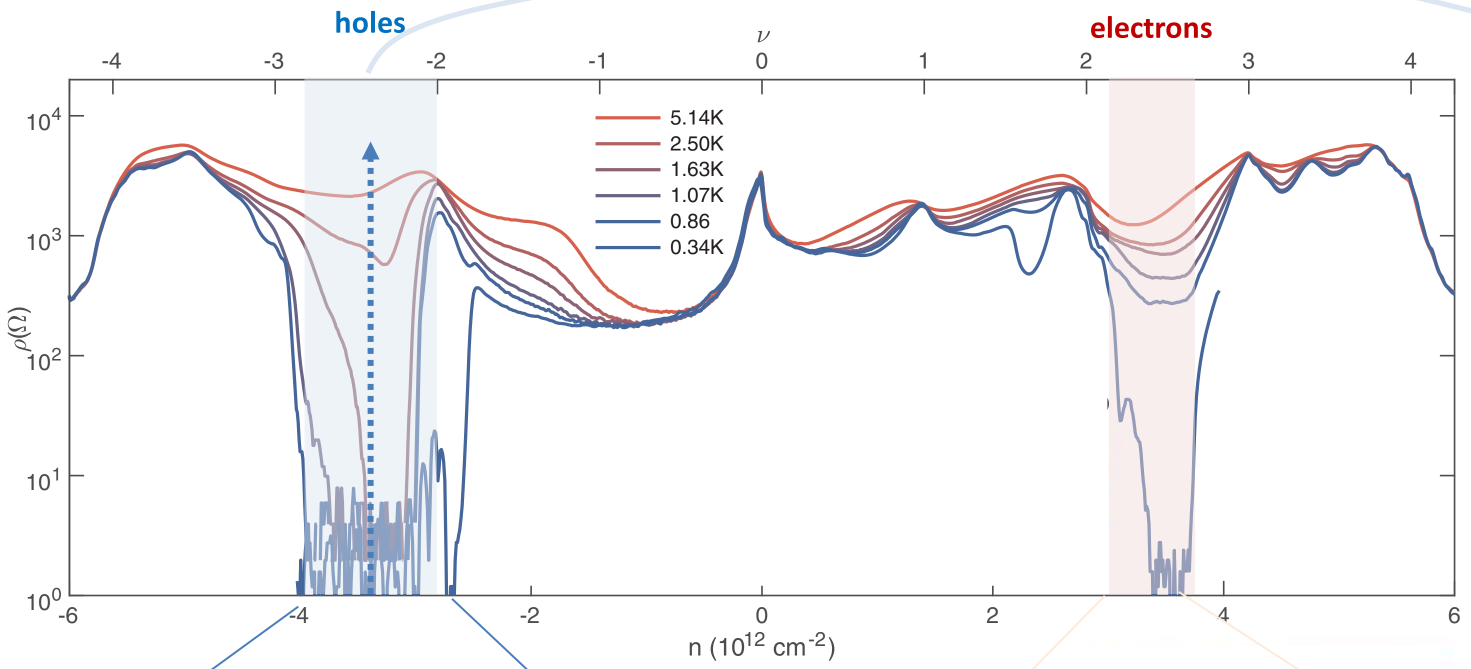
- **Consistent with expected band structures (Dirac cones and Flat bands)**

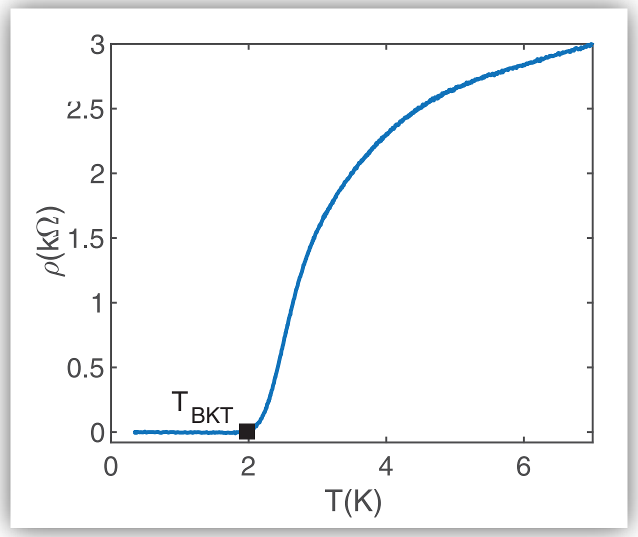


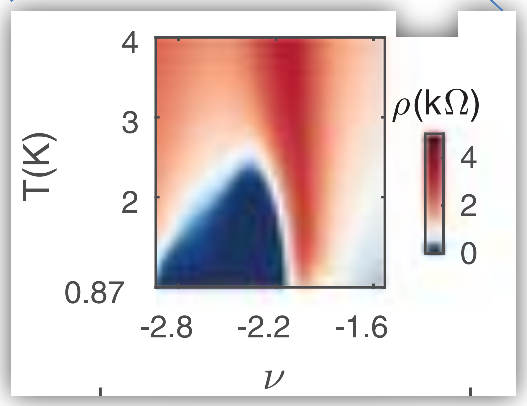
Hao et al., Science (2021); Similar results by Park et al., Nature (2021)

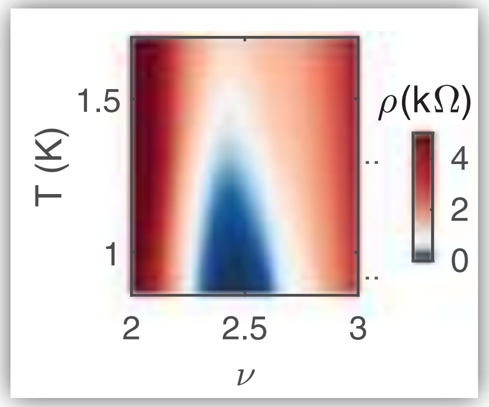
Superconductivity in TTG

$T_c(50\%) = 2.5$ K; $T_{BKT} = 2$K







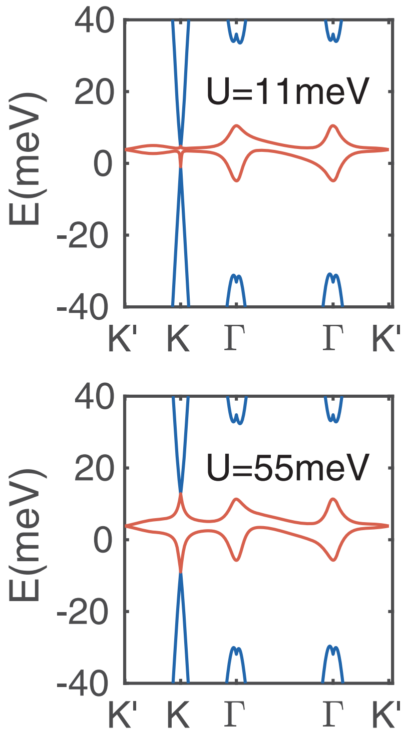


Density dependent superconducting domes near $\nu = \pm 2$

Hao et al., Science (2021); Similar results by Park et al., Nature (2021)

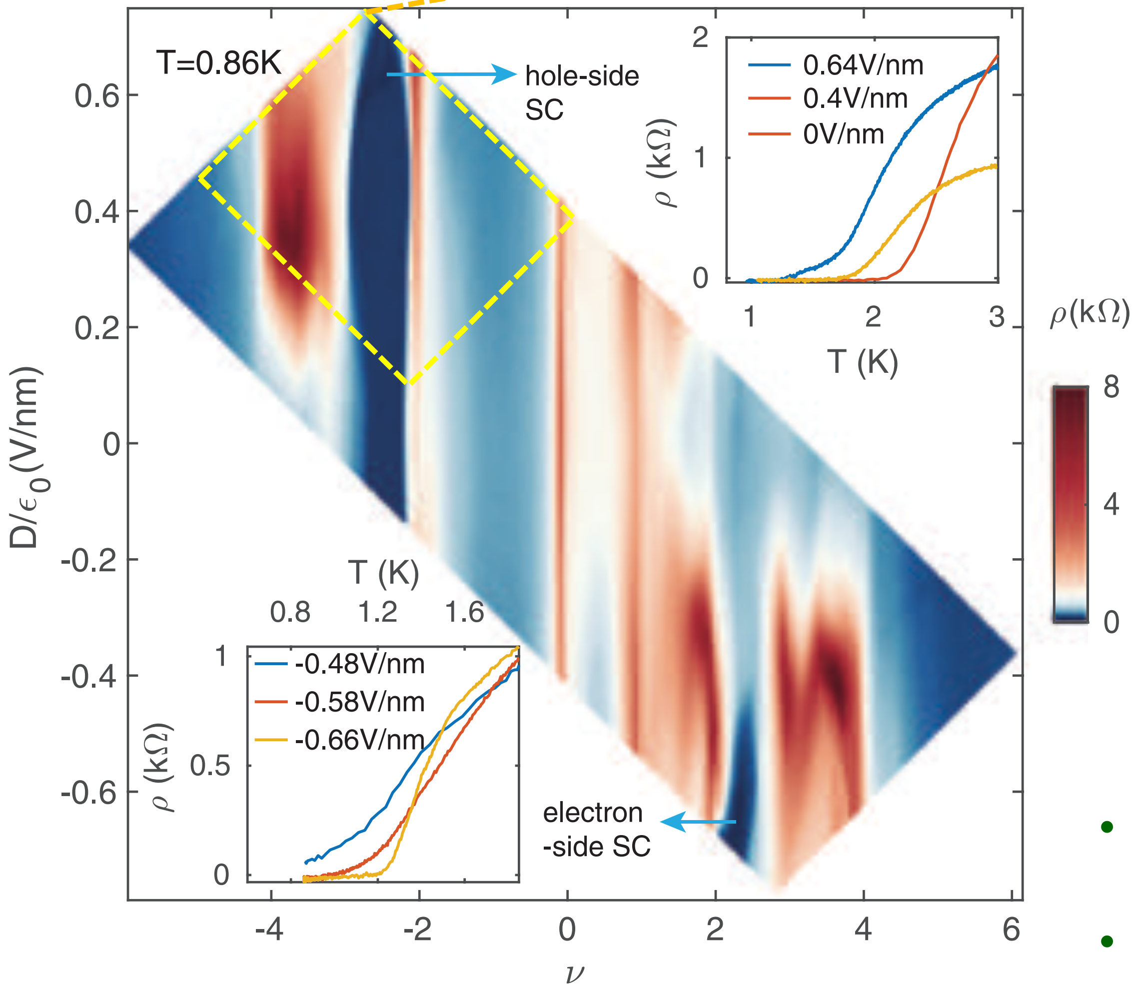
Displacement Tunable TTG Bands and Superconductivity

Field Tunable Bands



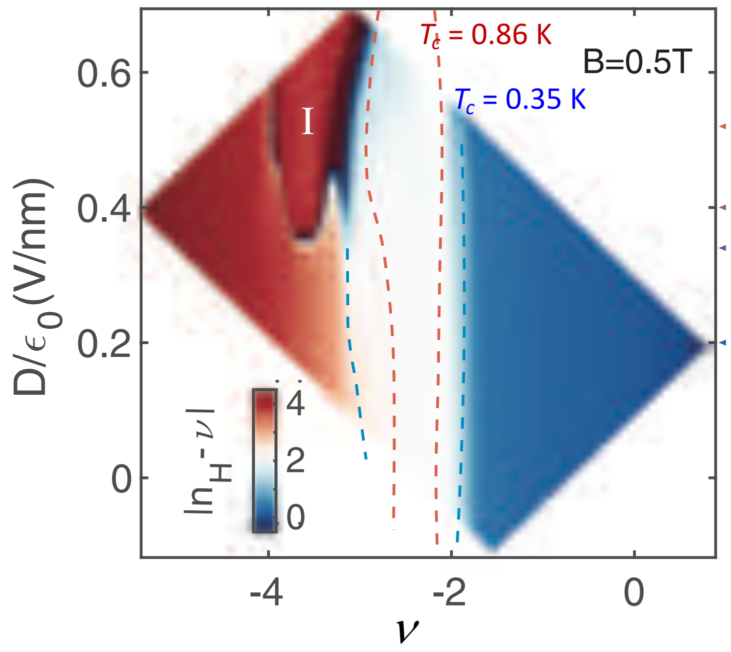
Hybridization between Dirac cones and flatbands

Tunable Superconductivity in TTG



Subtracted Normalized Hall Density

$n_H = B/e\rho_{xy}n_s$

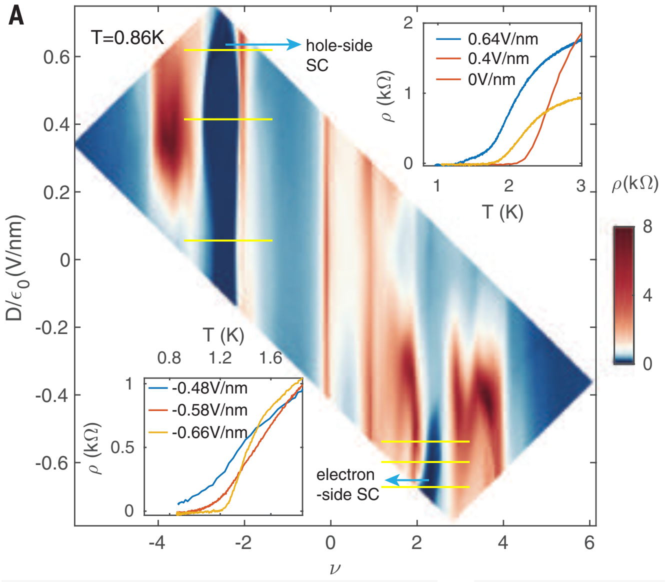


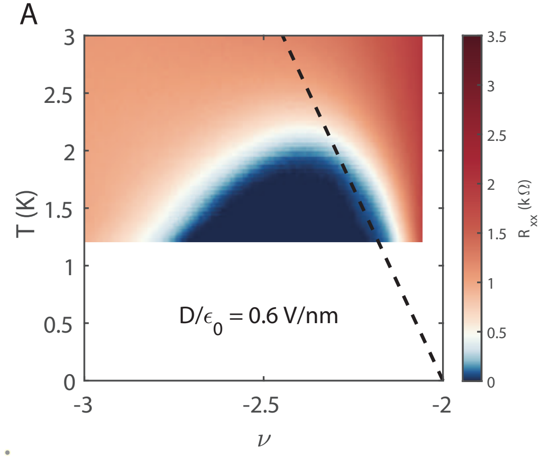
- Doping ν=-2 states is deeply related to SC
- Superconductivity becomes even weaker near the van Hove singularity

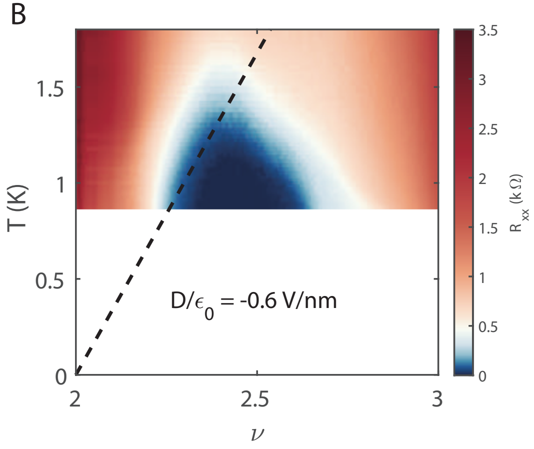
Hao et al., Science (2021); Similar results by Park et al., Nature (2021)

Displacement Field Tunable Superconducting Domes

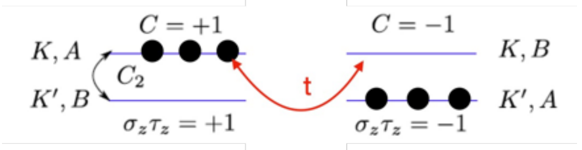
Temperature dependent domes

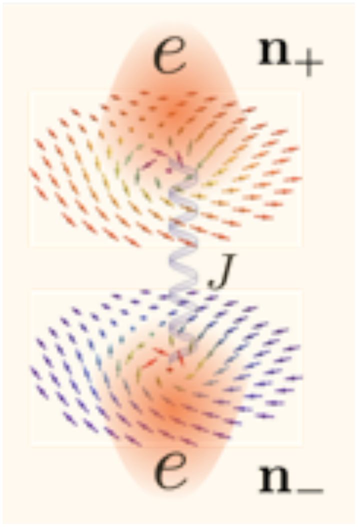




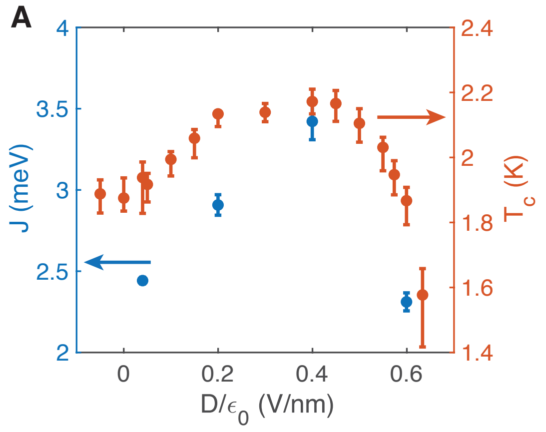


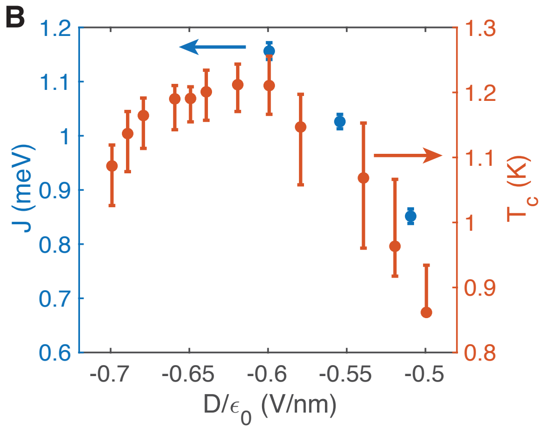
$$\Delta E = J\hat{n}_+ \cdot \hat{n}_-$$





$$k_B T_c = \frac{\nu_{2e}\pi\hbar^2}{2A_M M_{2e}} = \frac{\nu_{2e}J}{2}$$





Pseudo-spin Skyrmion mediated superconductivity

Khalaf et al., arXiv:2004.00638; Christos et al., PNAS 117, 29543 (2020);

Beyond Twisted Trilayer: n=4 and n=5 Twisted Grapehene Multilayers

arXiv:2112.09270

Ascendance of Superconductivity in Magic-Angle Graphene Multilayers

Yiran Zhang[1,2,3*], Robert Polski[1,2*], Cyprian Lewandowski[2,3], Alex Thomson[2,3,4], Yang Peng[5], Youngjoon Choi[1,2,3], Hyunjin Kim[1,2,3], Kenji Watanabe[6], Takashi Taniguchi[6], Jason Alicea[2,3], Felix von Oppen[7], Gil Refael[2,3], and Stevan Nadj-Perge[1,2†]

arXiv:2112.10760

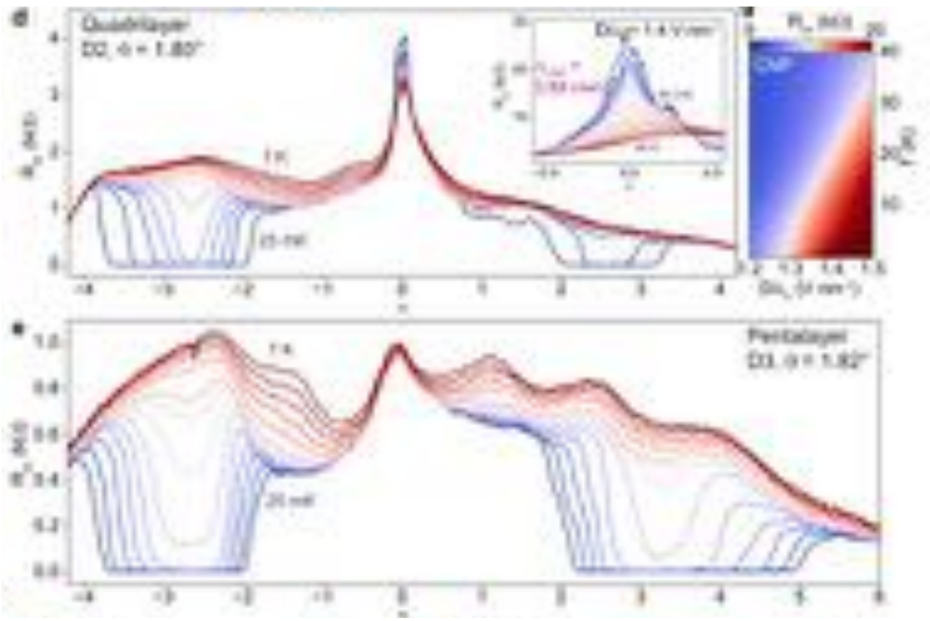
Magic-Angle Multilayer Graphene: A Robust Family of Moiré Superconductors

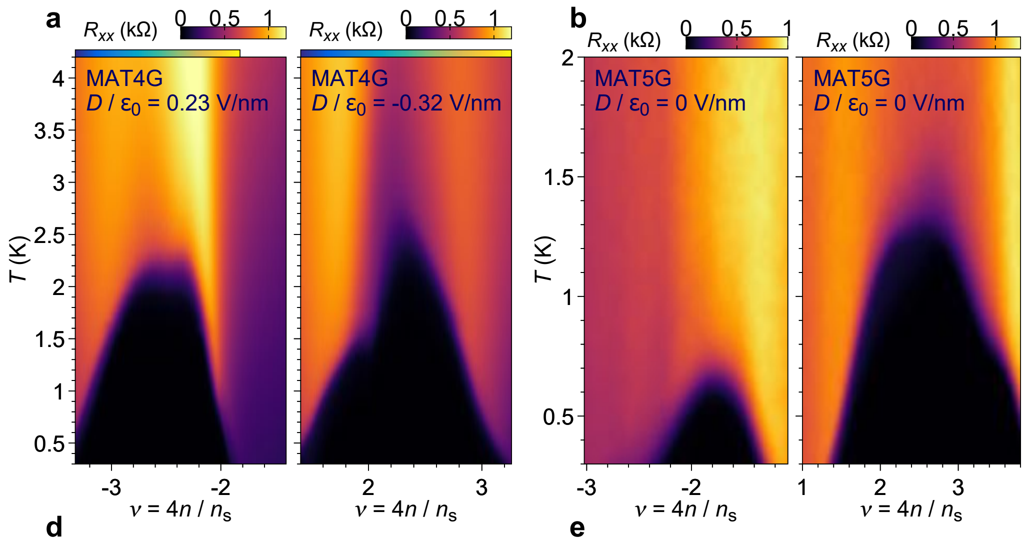
Jeong Min Park,[1,*,†] Yuan Cao,[1,2,*] Liqiao Xia,[1] Shuwen Sun,[1]
Kenji Watanabe,[3] Takashi Taniguchi,[3] and Pablo Jarillo-Herrero[1,†]

arXiv:2201.01637

Emergence of Correlations at the Edge of the Magic Angle Regime in Alternating Twist Quadrilayer Graphene

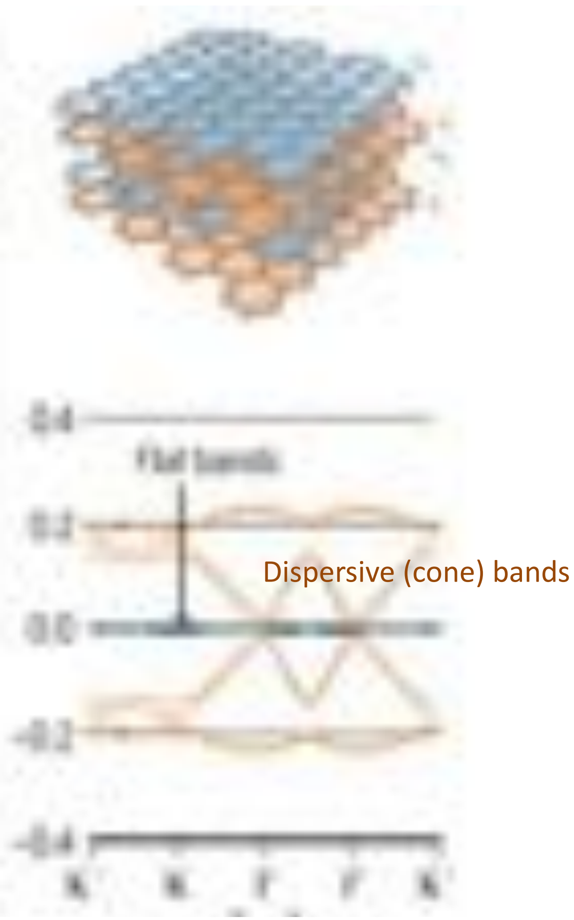
G. William Burg[1], Eslam Khalaf[2], Yimeng Wang[1], Kenji Watanabe[3], Takashi Taniguchi[4], Emanuel Tutuc[1]



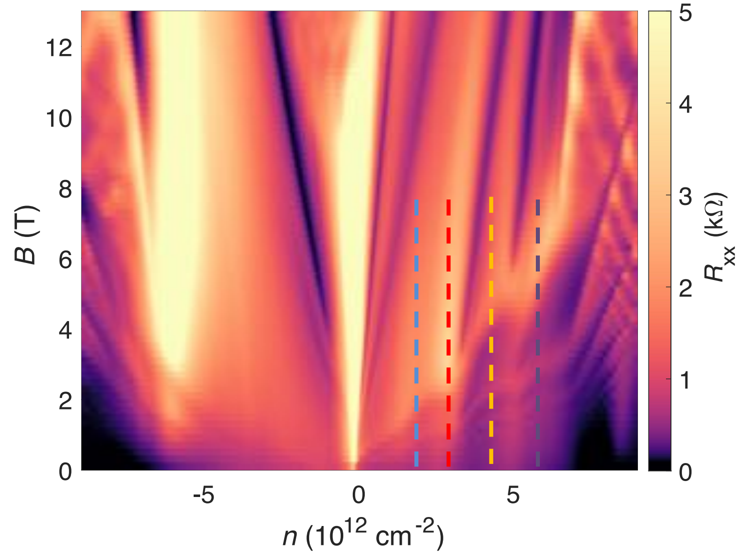


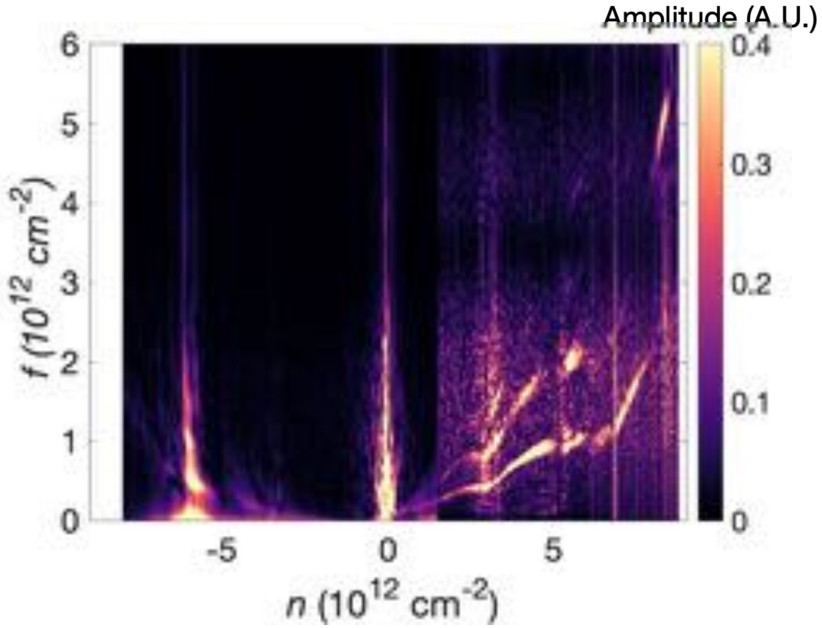
Twisted Quadruple Layer Graphene: Dispersive versus Flat Bands

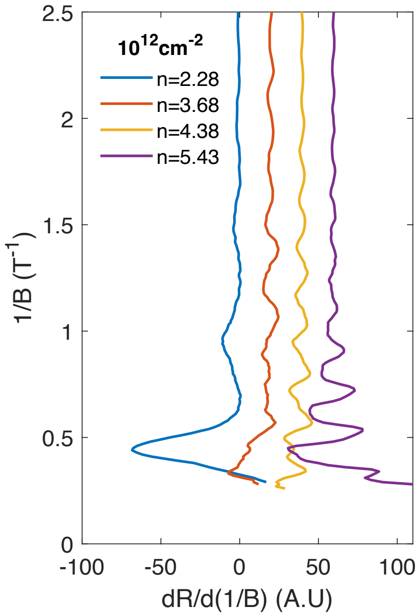
Phinney et al., in preparation



E. Khalaf, et al., PRB 100, 085109 (2020)







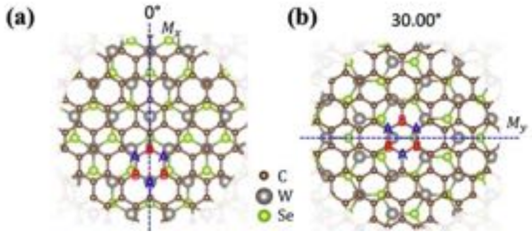
SdH Oscillation from Dispersive (cone) bands

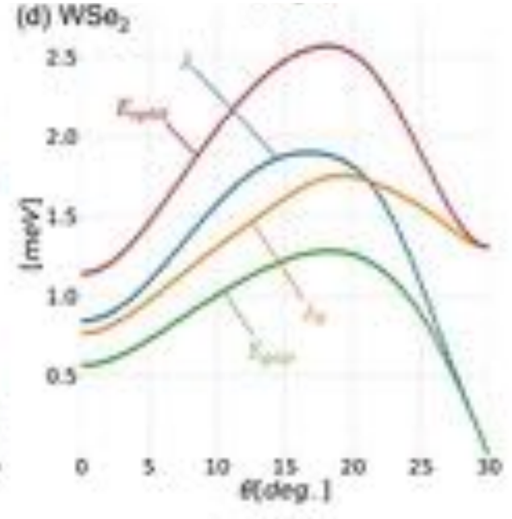
$$n_{cone} = 8\left(\frac{e}{h}\right) f_B$$

$$v_{\text{flat}} \approx 0.85\, v$$

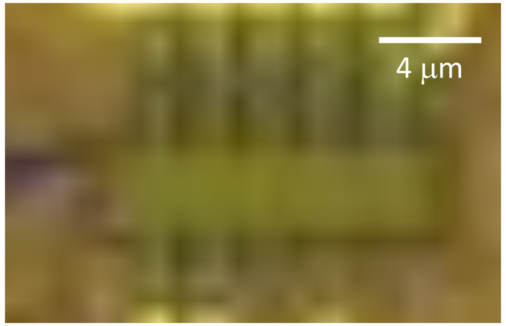
Superconductivity with proximity induced SOC

Angle dependence of SOC

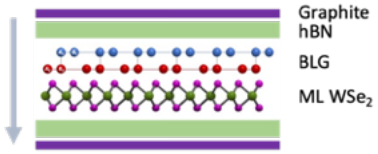




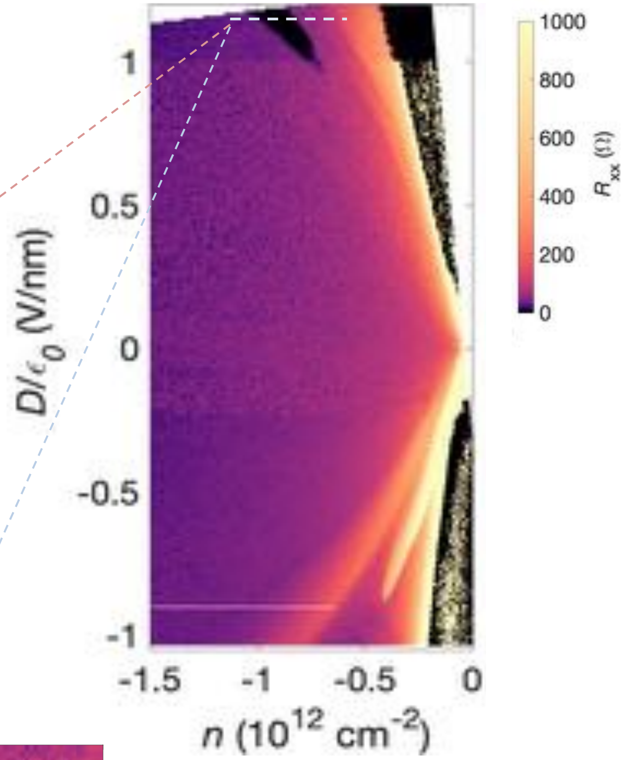
Aligned Bilayer graphene/WSe2



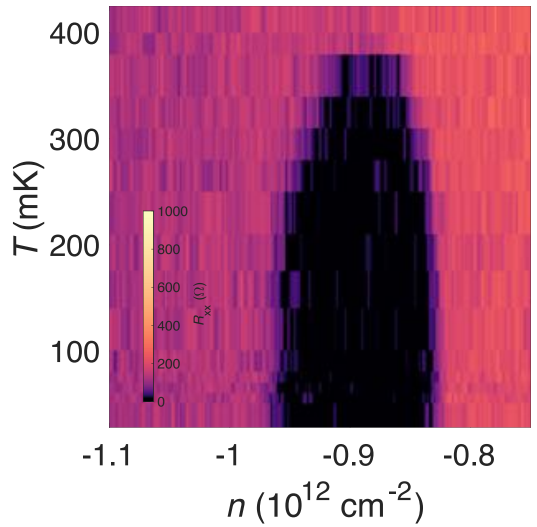
With corner NbN JJs



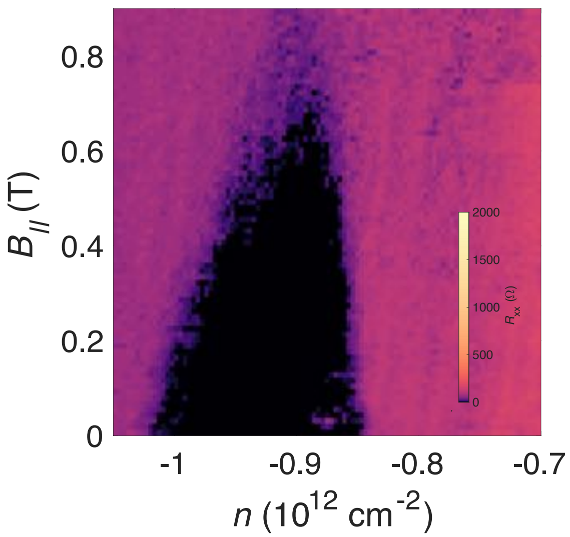
Displacement Field controlled SOC



T_c^{max} ~ 350 mK



Pauli limit violation



Y Li, M Koshino, PRB (2019)
S Lee, et al. arXiv:2206.09478 (2022)
A Veneri, et al. PRB (2022)

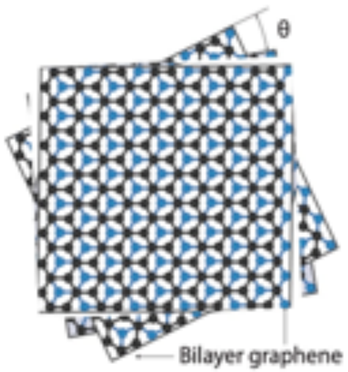
Phinney et al., in preparation

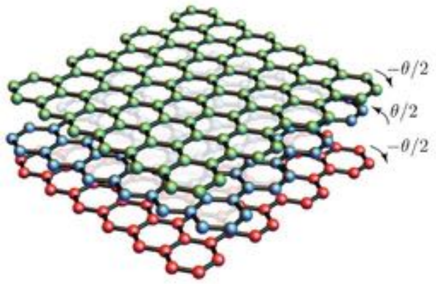
Summary

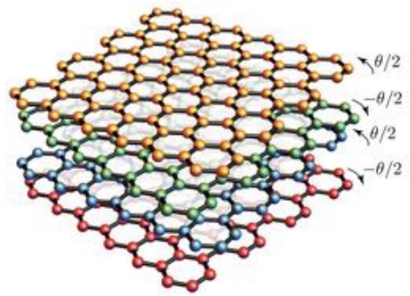
- SU(4) flavor polarization can create Chern bands in twisted graphene

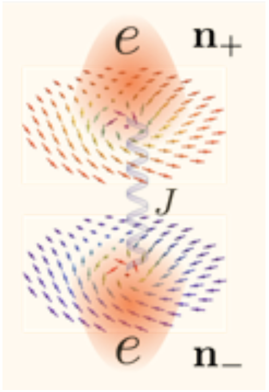
- Superconductivity in twisted graphene is deeply connected to flavor polarization

- Multilayer twisted stacked graphene systems provide various correlated Chern insulators that can provide flavour polarized metals and potentially unconventional superconductivity









Acknowledgements

Experiment

Rebecca Engelke, Richard Allen, Kevin Xu, Hoseok Heo, Hongkun Park (Harvard),

Hyobin Yoo, Ayoung Yuk, Kahyun Ko, Junhyung Kim (Seogang Univ, Korea)

Elliot Padgett, Zhen Chen, David Muller (Cornell)

Takashi Taniguchi, Kenji Watanabe (NIMS)

Theoretical

Stephen Carr, Efthimios Kaxiras (Harvard); Jung Hoon Han (SKKU)

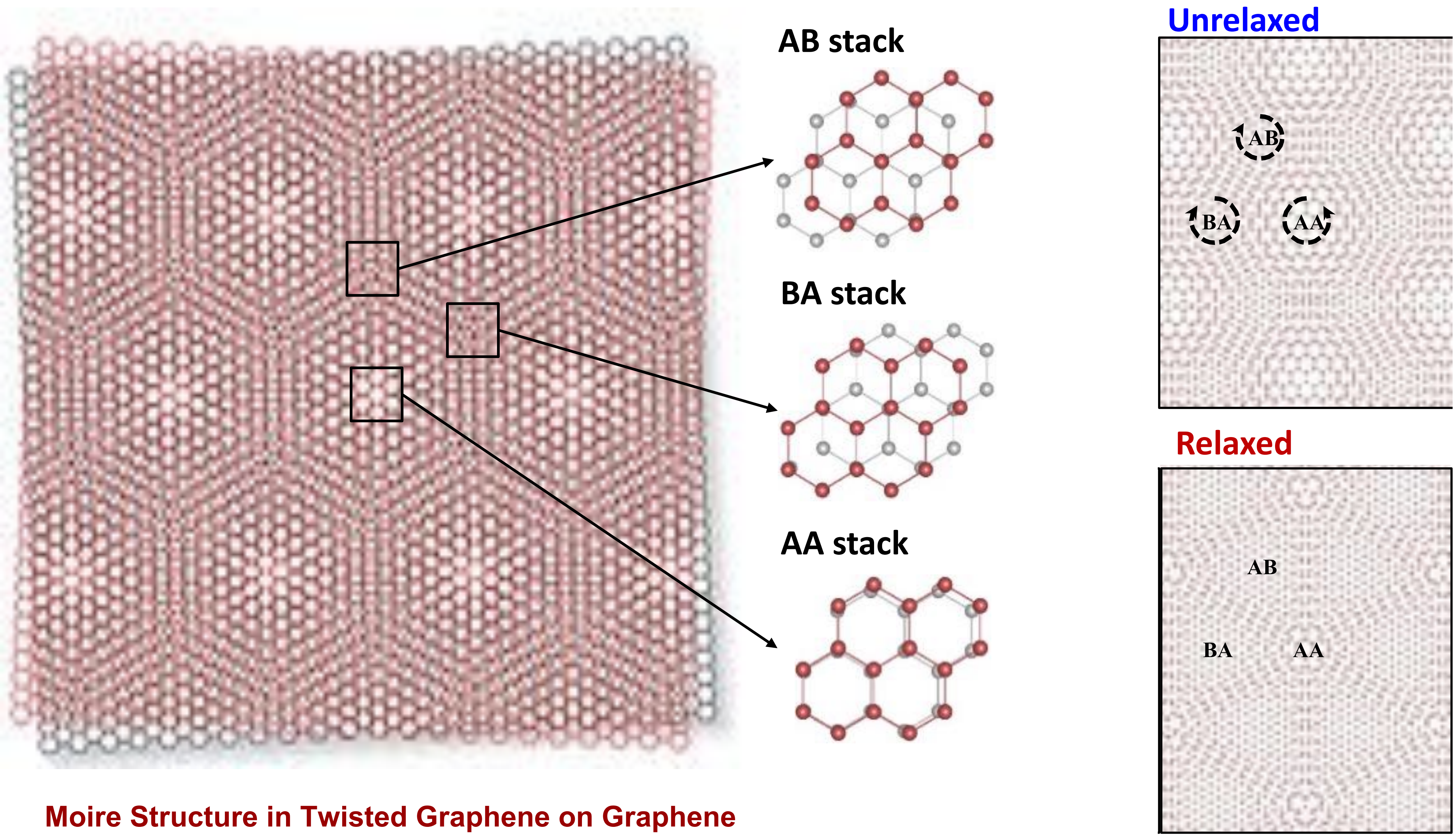
Mathematics

Paul Cazeaux, Mitchell Luskin (U of Minnesota); Minhyoung Kim (Oxford)

Funding:

National Research Foundation of Korea

Graphene: Moire Superlattice



Moire Structure in Twisted Graphene on Graphene

Imaging AB/AC domain in Bernal stacked bilayer graphene

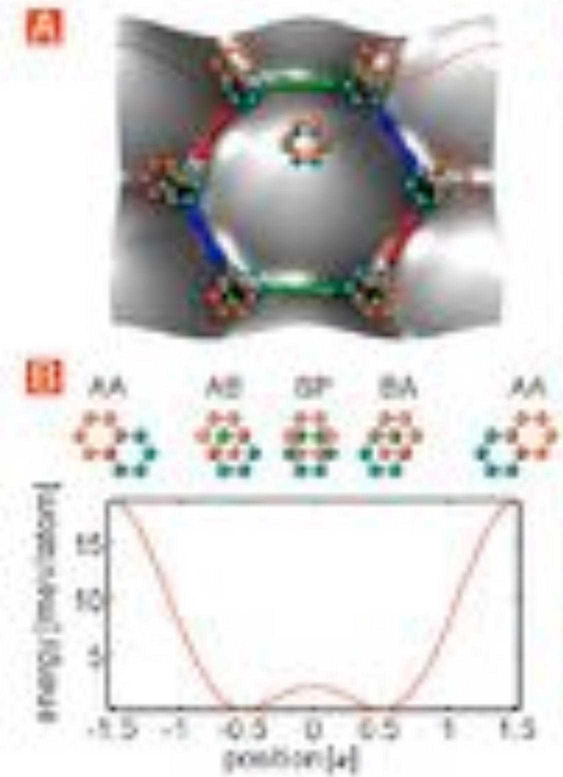
PNAS

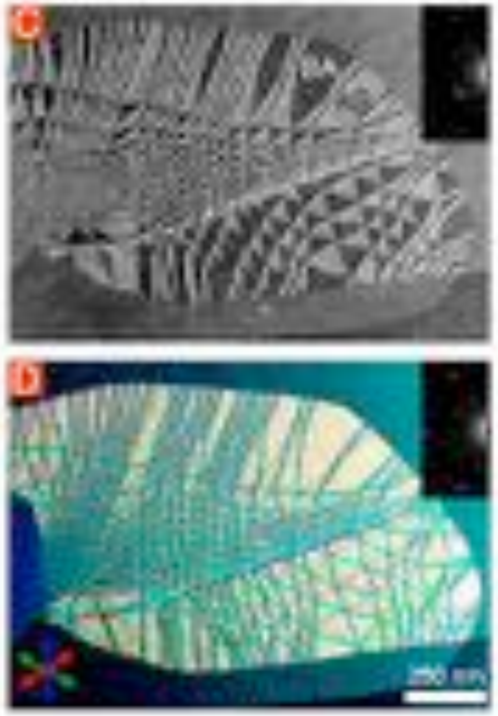
Strain solitons and topological defects in bilayer graphene

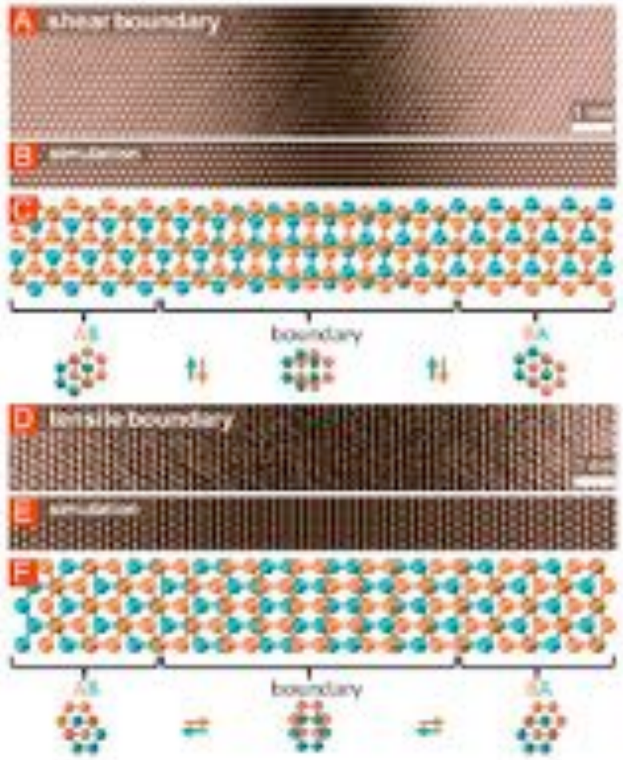
Jonathan S. Alden[a], Adam W. Tsen[a], Pinshane Y. Huang[a], Robert Hovden[a], Lola Brown[b], Jiwoong Park[b,c], David A. Muller[a,c], and Paul L. McEuen[c,d,1]

[a]School of Applied and Engineering Physics, [b]Department of Chemistry and Chemical Biology, [c]Kavli Institute at Cornell for Nanoscale Science, and [d]Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, NY 14853

Contributed by Paul L. McEuen, May 23, 2013 (sent for review April 28, 2013)



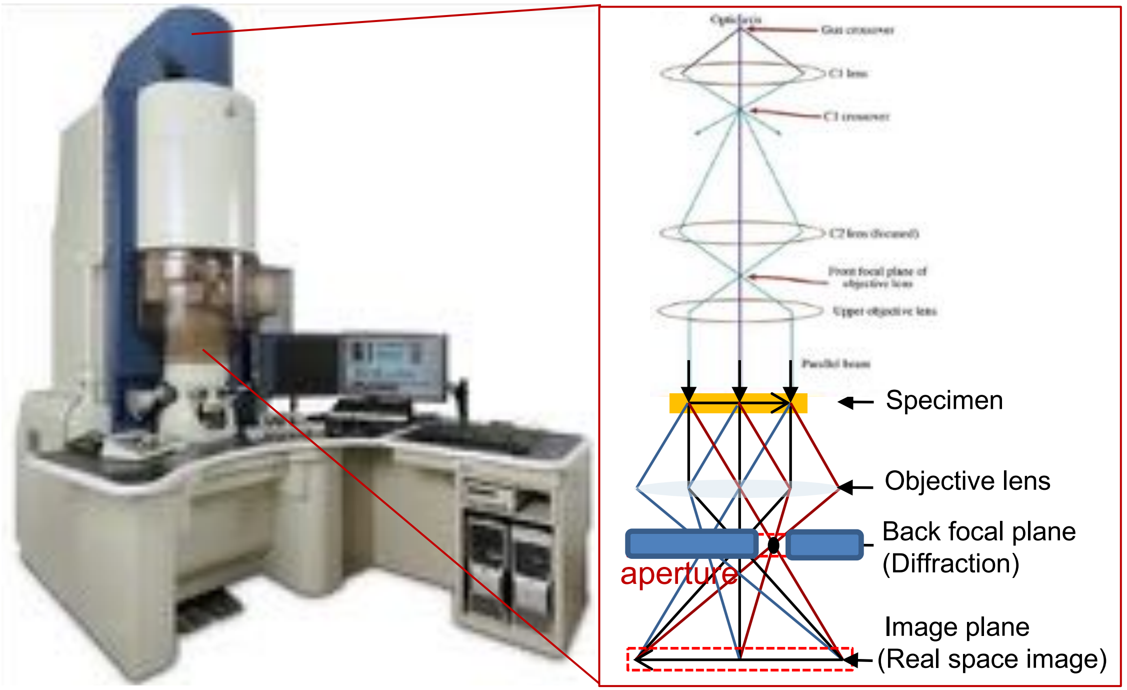




Dark Field Imaging Graphene Domains

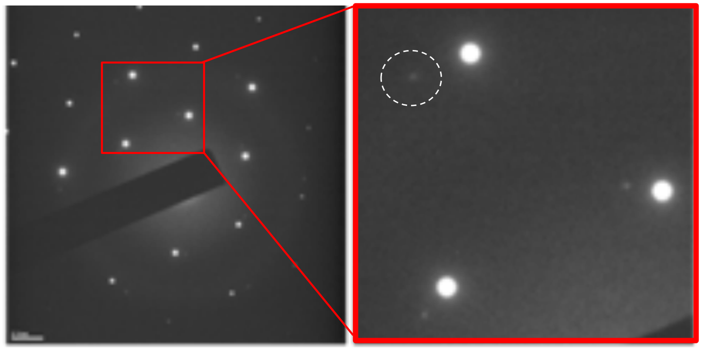
Transmission Electron Microscopy

Instrument and ray diagram

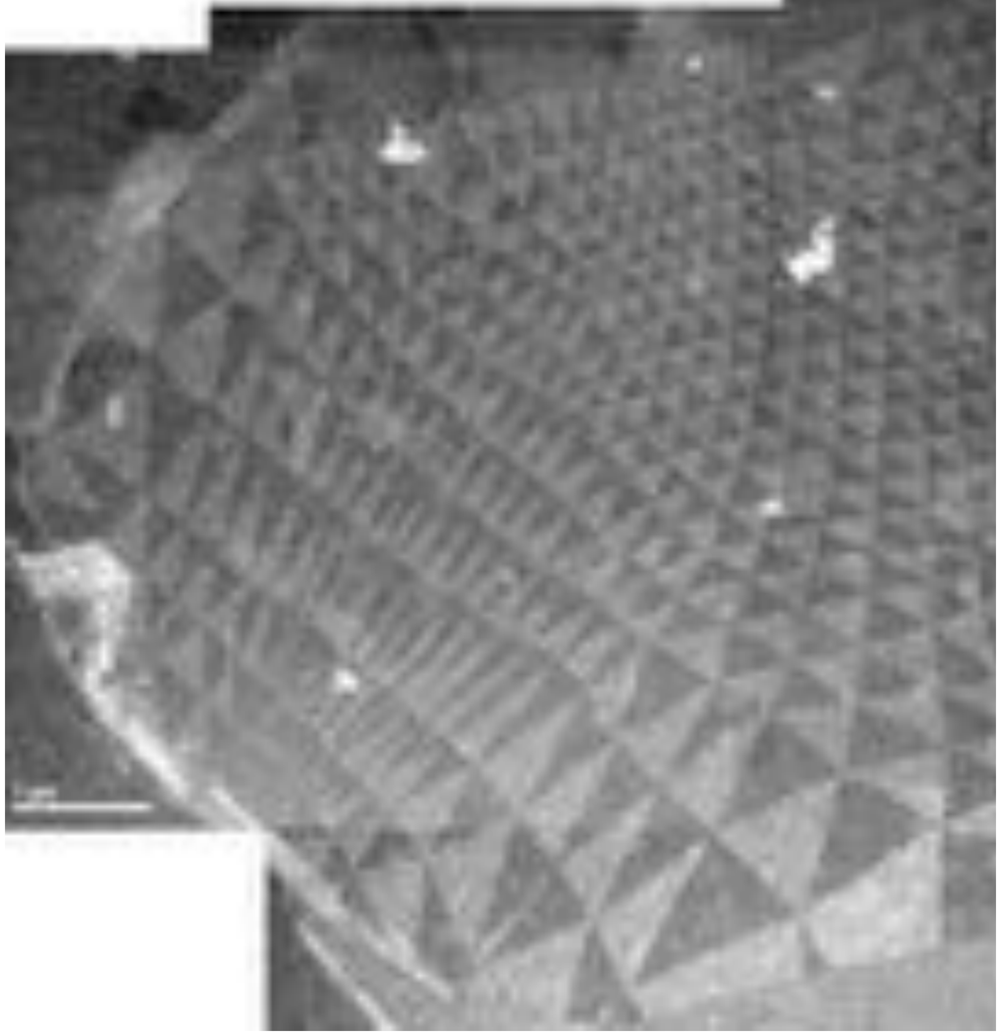


Dark field imaging

Graphene/graphene/hBN



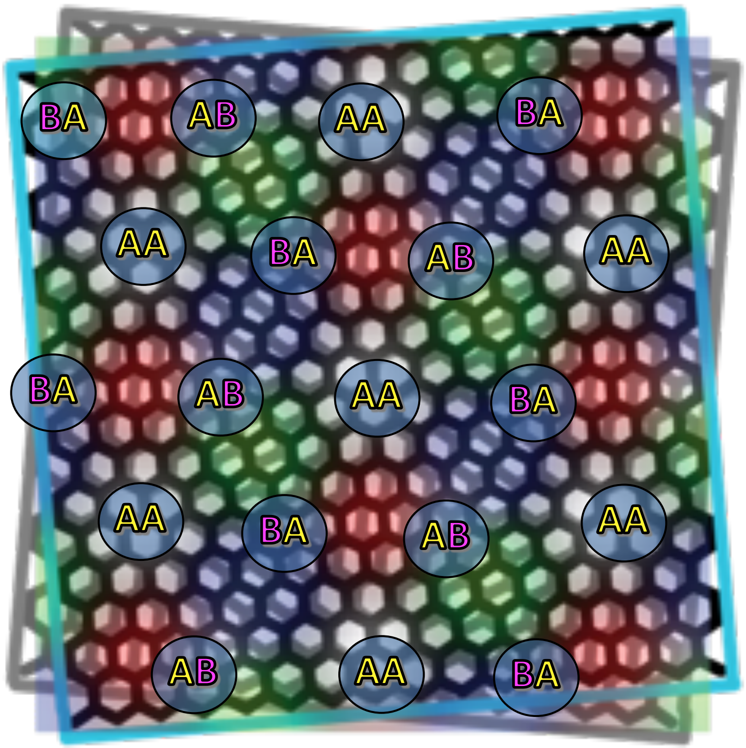
Real space image of reconstructed AB/BA moire domains



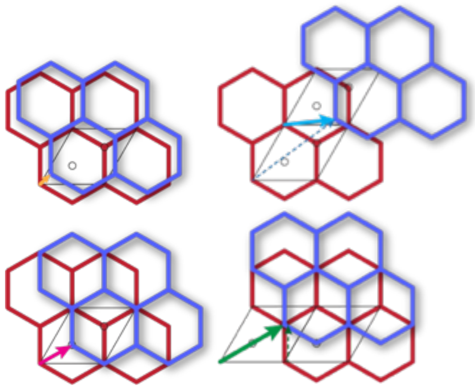
Alden et al., PNAS (2013)

Lattice Shift Vector: Order Parameter for Relaxation Process

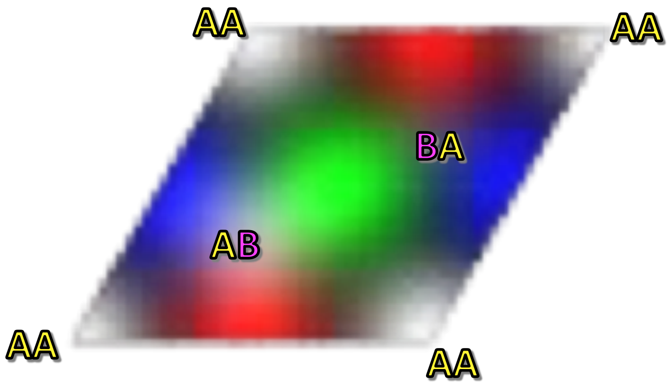
Unrelaxed



Local Relative Displacement



Order parameter maps of unrelaxed lattice

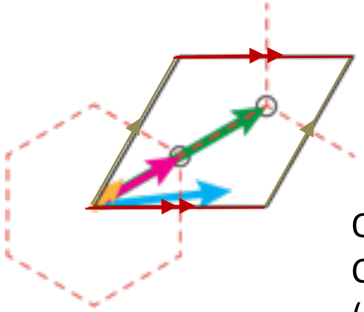


Intensity corresponds to distribution densities

Define Lattice Shift Vector:

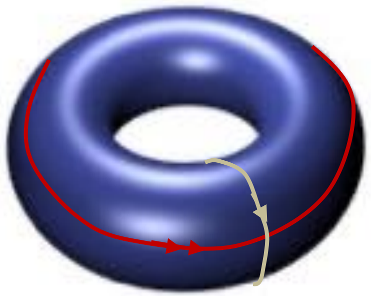
$$\boldsymbol{u} = [\mathbf{R}(\text{upper layer}) - \mathbf{R}(\text{lower layer})]_{\text{unit cell}}$$

Shift vector: $\boldsymbol{u}$



Order parameter
Configuration space
(periodic unit cell)

2D Periodicity in configuration space

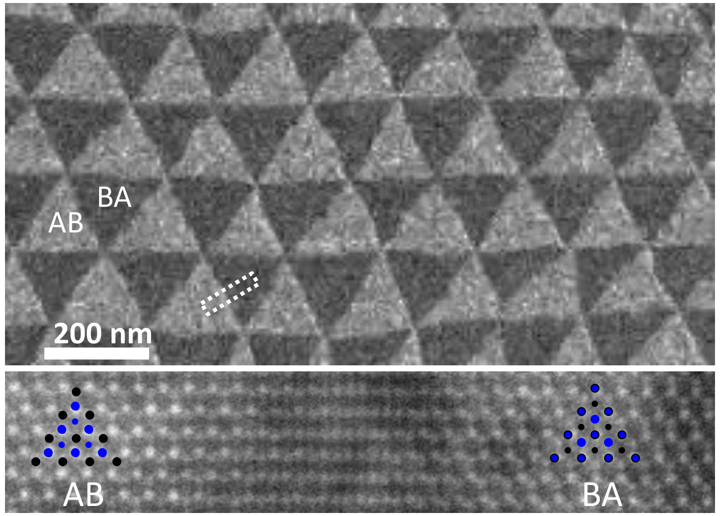


We also define $\boldsymbol{u}$ = 0 for untwisted sample and **u** = 0 for a AA site as a rotational center.

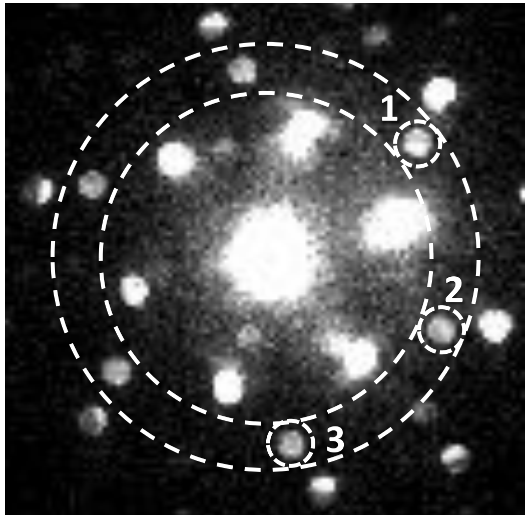
Following Alden et al., PNAS (2013)

Burgers Vector for Moire Boundaries in TBG

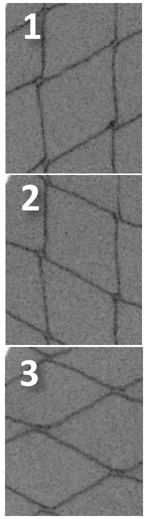
Dark field TEM/ Atomic-resolution Scanning TEM



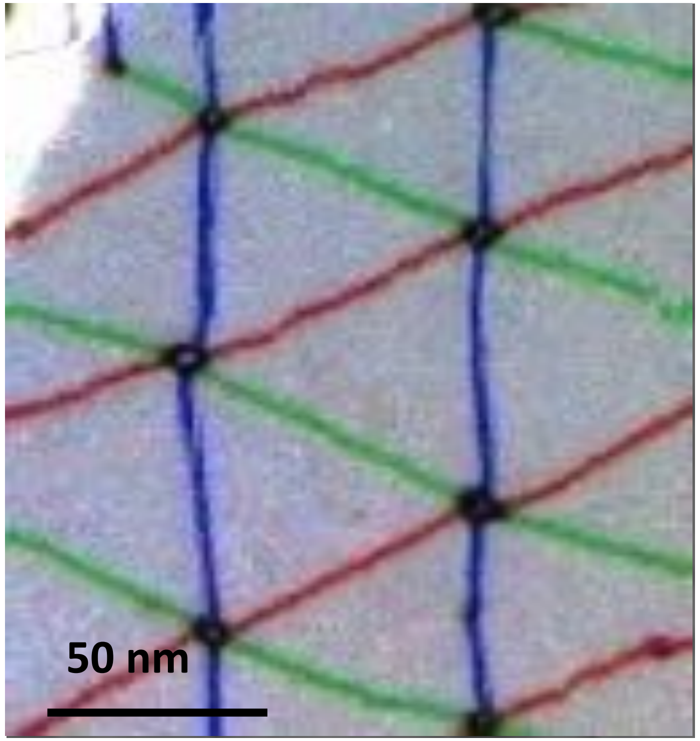
Electron diffraction



Second Order Brag Peak
Dark field imaging



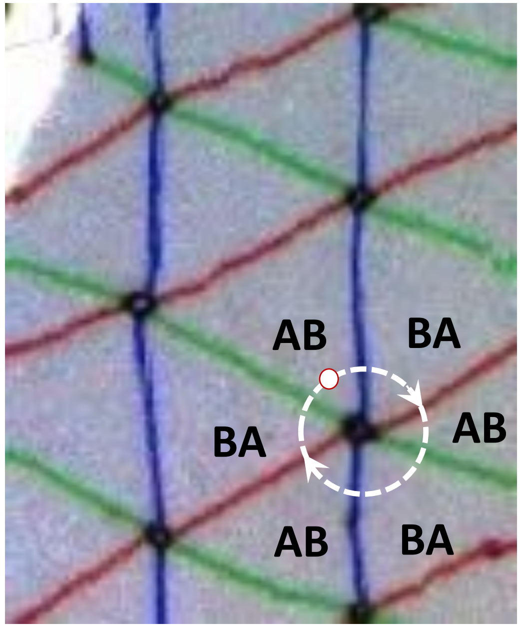
Domain Boundary Coloring by Burgers Vector



After the lattice relaxation, AA site is a junction for three dislocation lines intersect!

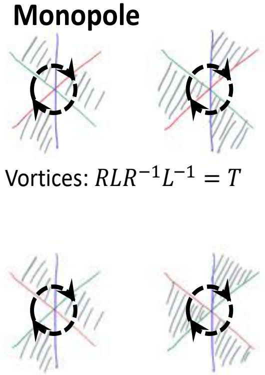
Topology of Moire Network

Second Order Dark Field Image:
Highlighted dislocation lines



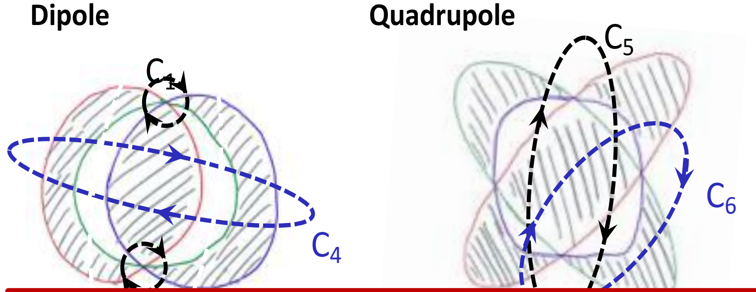
Nonabelian vortices (NAV)

Topology of F2 Free Group Elements



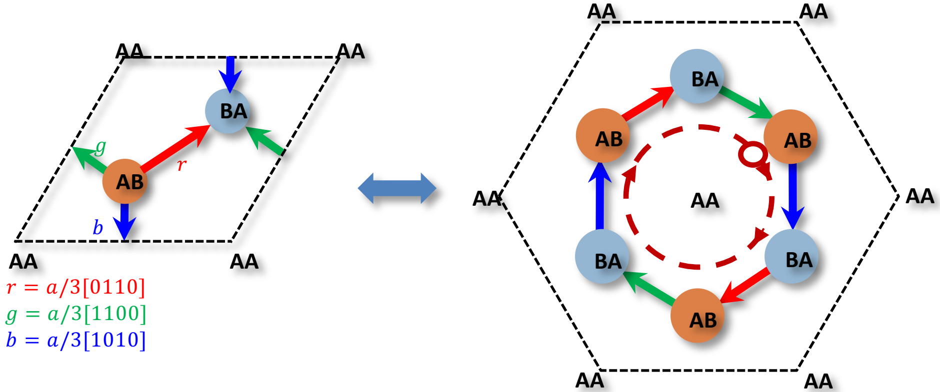
Vortices: $RLR^{-1}L^{-1} = T$

Anti-vortices: $LRL^{-1}R^{-1} = T^{-1}$



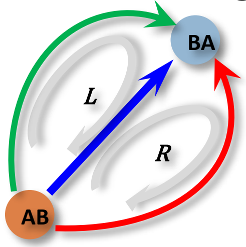
- **NAV are topological objects. They can be only removed through the boundaries of the samples.**
- **NAV density is proportional to local twisting angles**

Order parameter (shift vector) space



$r = a/3[0110]$
$g = a/3[1100]$
$b = a/3[1010]$

Topological Fundamental group of order parameter space



Free group on 2 generators
→ $RL \neq LR$

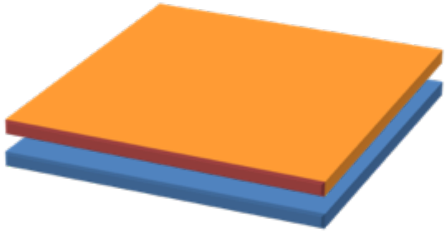
$\boldsymbol{L} = gb^{-1}$
$\boldsymbol{R} = br^{-1}$

A loop around AA site corresponds to
$\boldsymbol{RLR^{-1}L^{-1}} = br^{-1}gb^{-1}rg^{-1}$

R. Engelke et al., arXiv:2207.05276,

Vortex and Anti-Vortex Pair

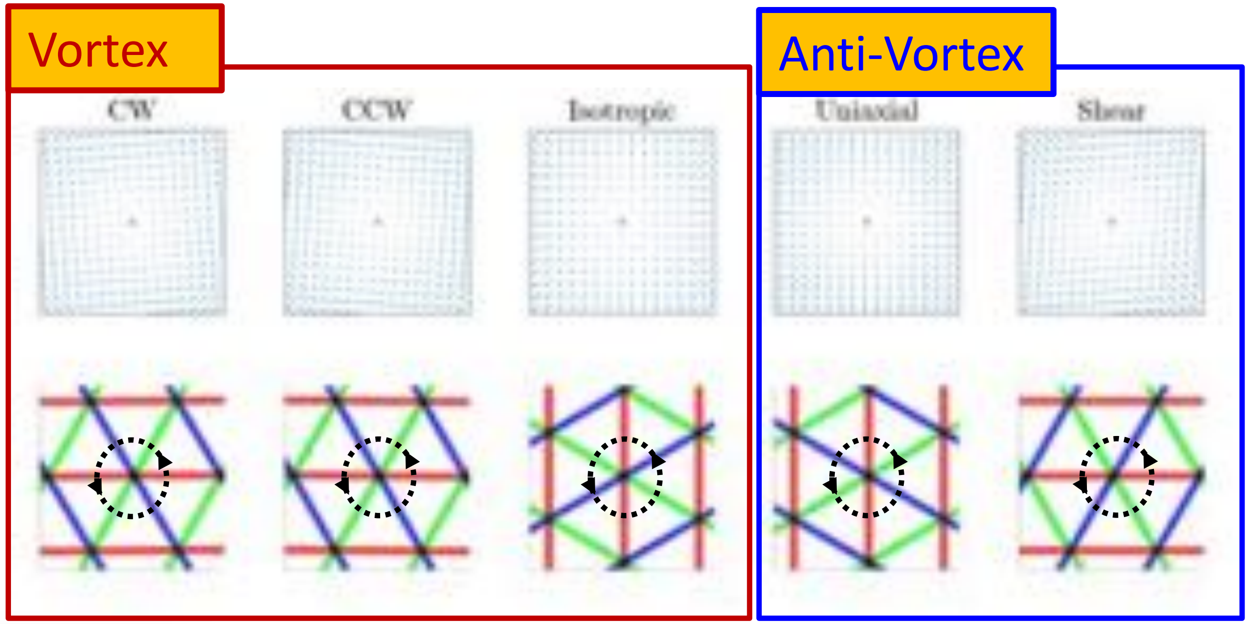
Interlayer relative elastic deformation



$$\boldsymbol{u}(\boldsymbol{r}) = \left(\boldsymbol{1} + \frac{\partial u_i}{\partial r_j}\right)\boldsymbol{r} = \begin{cases} \begin{pmatrix} \cos(\theta)-1 & -\sin(\theta) \\ \sin(\theta) & \cos(\theta)-1 \end{pmatrix} & \text{Rotation} \\ \begin{pmatrix} \alpha & 0 \\ 0 & \alpha \end{pmatrix} & \text{Isotropic} \\ \begin{pmatrix} \beta_1 & 0 \\ 0 & -\beta_1 \end{pmatrix} & \text{Uniaxial} \\ \begin{pmatrix} 0 & \beta_2 \\ \beta_2 & 0 \end{pmatrix} & \text{Shear} \end{cases}$$

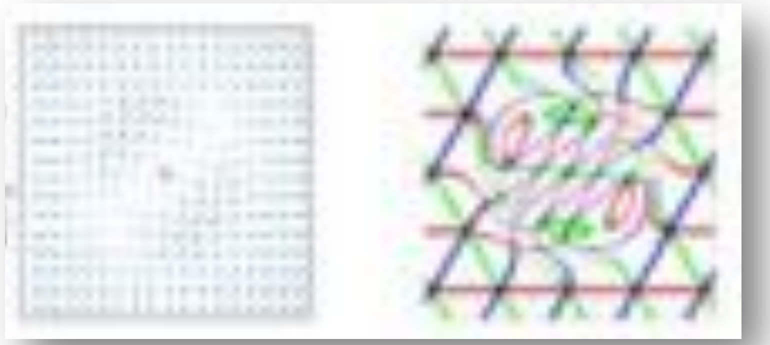
u(r) map in different elastic deformation

Corresponding dislocation network



$\boldsymbol{RLR^{-1}L^{-1}} = br^{-1}\,gb^{-1}rg^{-1}$

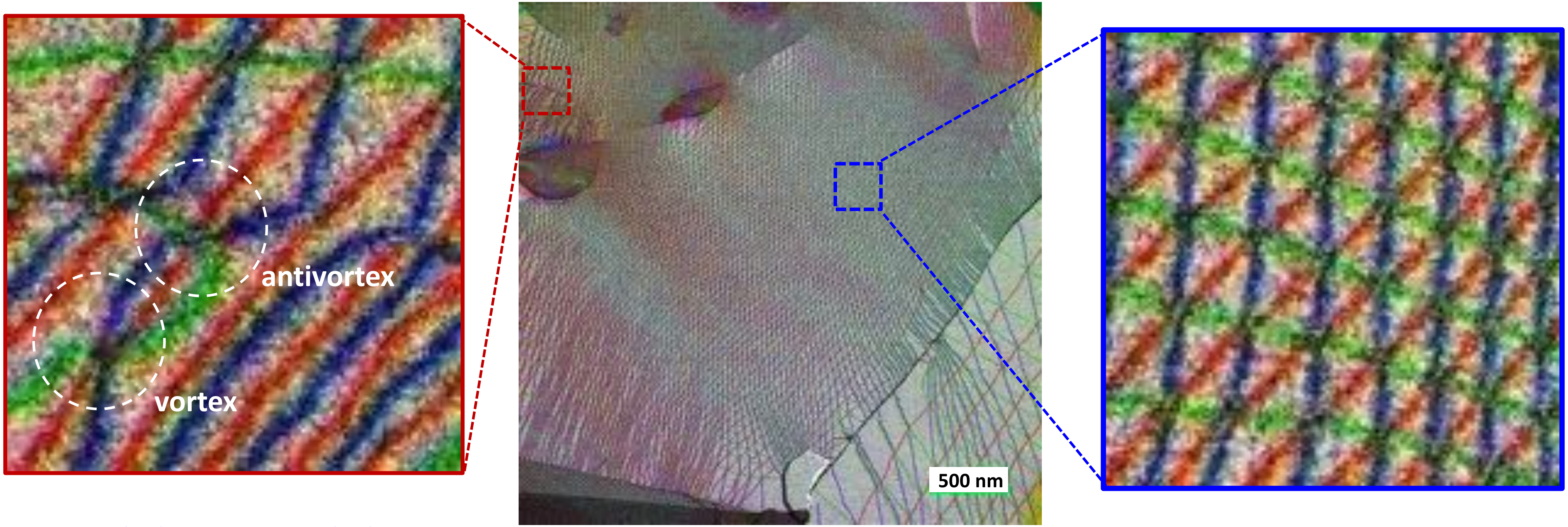
$\boldsymbol{LRL^{-1}R^{-1}} = gr^{-1}bg^{-1}rb^{-1}$



Vortices and Antivortices in mixed (shear + rotation) elastic deformation

R. Engelke et al., arXiv:2207.05276,

Realization of Vortex and Anti-Vortex Pair



Vortex: rgbrgb,   antivortex: rbgrbg

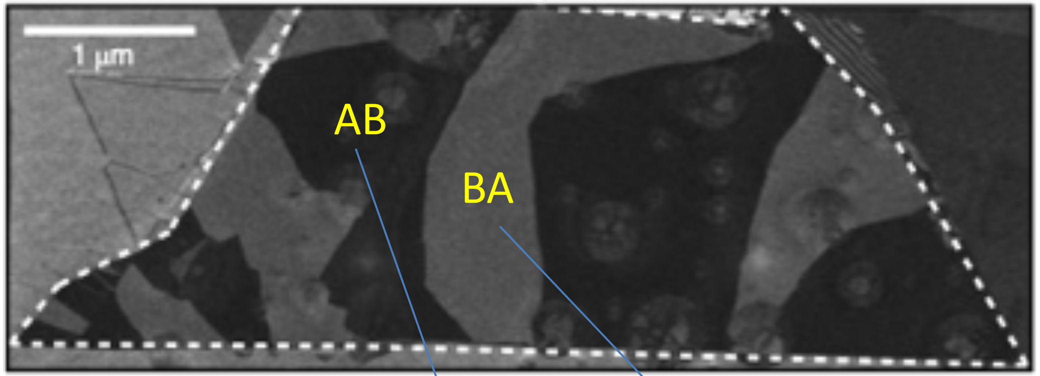
For uniformly twisted region:
Vortex density ~ twisting angle

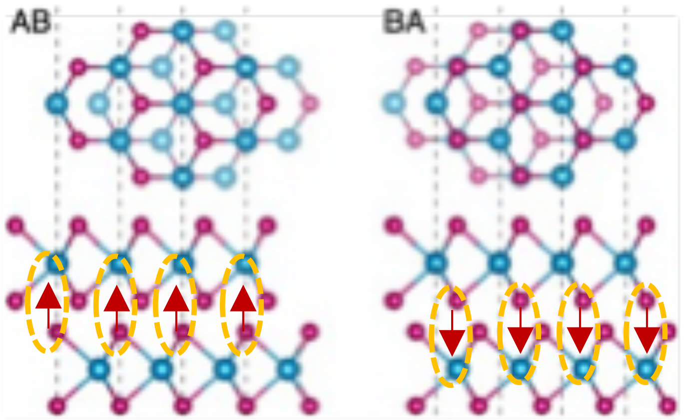
Vortice-anti vortices may coexist when there is competition between different strain components

R. Engelke et al., arXiv:2207.05276,

Broken Mirror Symmetry and Spontaneous Dipoles

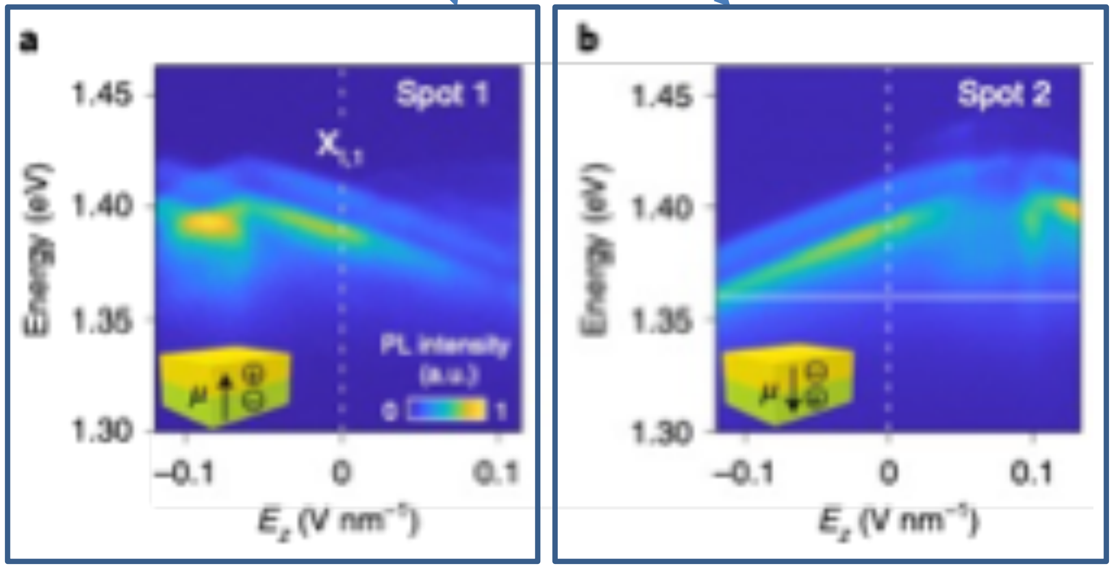
$MoSe_2/MoSe_2$ near 0 degree



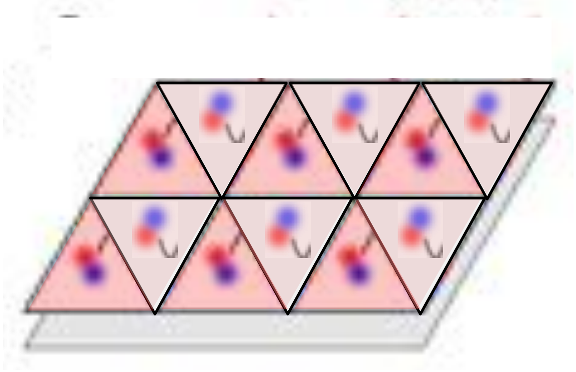


Interlayer dipole moment due to charge transfer!

Electric Field Dependent Photoluminescence



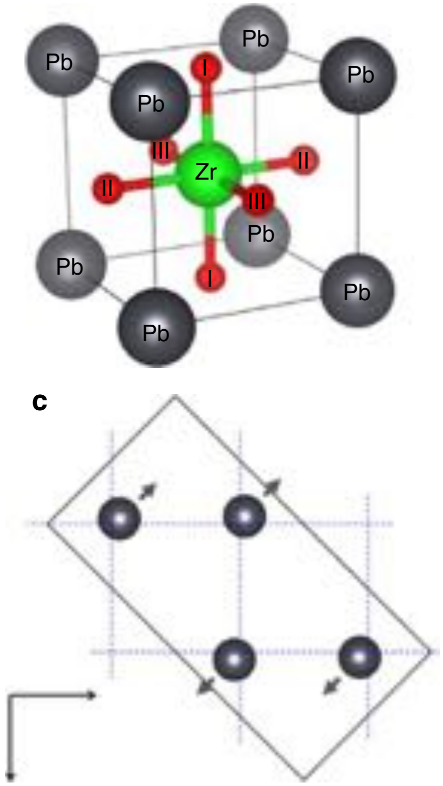
Arrays of alternating dipole moment interlayer to broken mirror/inversion symmetry in the AB and BA domains.



J. Sung et al., Nature Nano 2020 (Collaboration with Park and Falko's Groups)

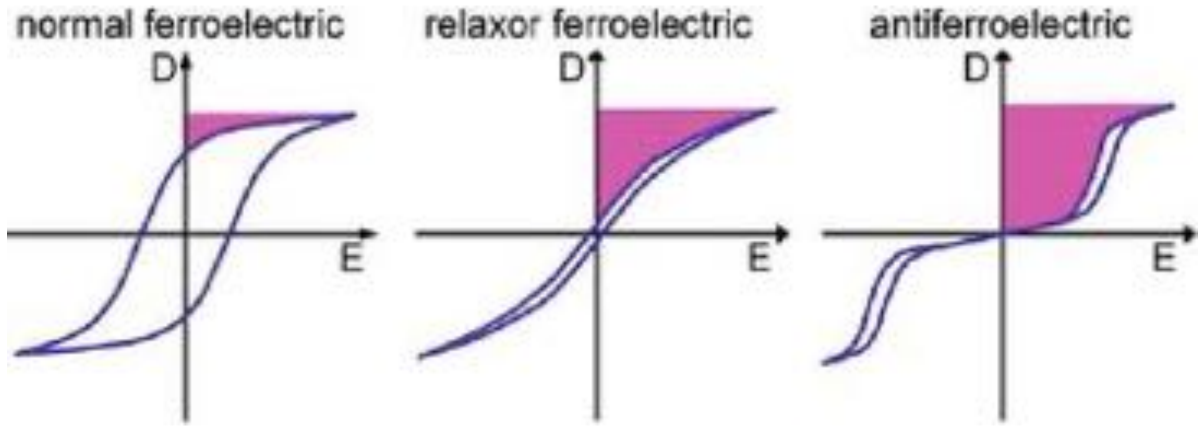
Ferroelectric and Anti-ferroelectric

A. K. Tagantsev et al., Nature Com 2229 (2013)



Antiferroelectric state of $PbZrO_3$

Various Ferroelectric Behaviors

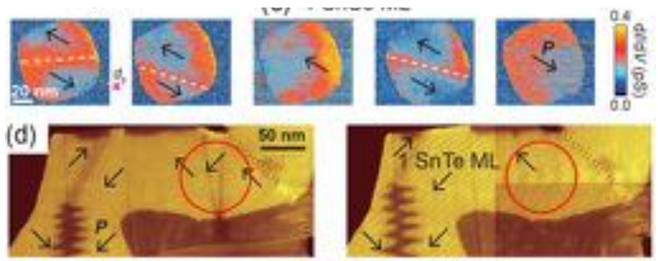


K. Rabe, https://doi.org/10.1002/9783527654864.ch7

Atomic scale anti-ferroelectric versus Moire scale anti-ferroelectric domains?

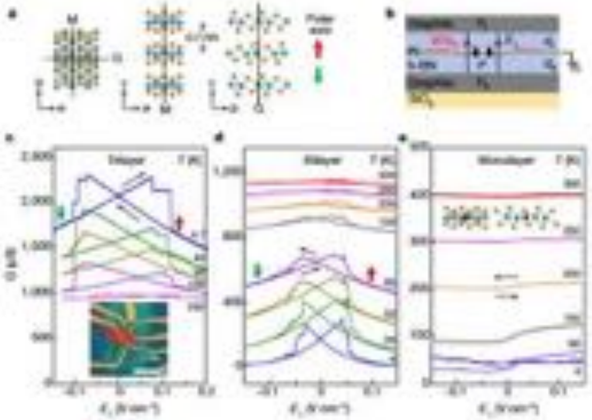
Ferroelectricity in vdW Hetero/homostructures

Atomically thin group-IV monochalcogenide



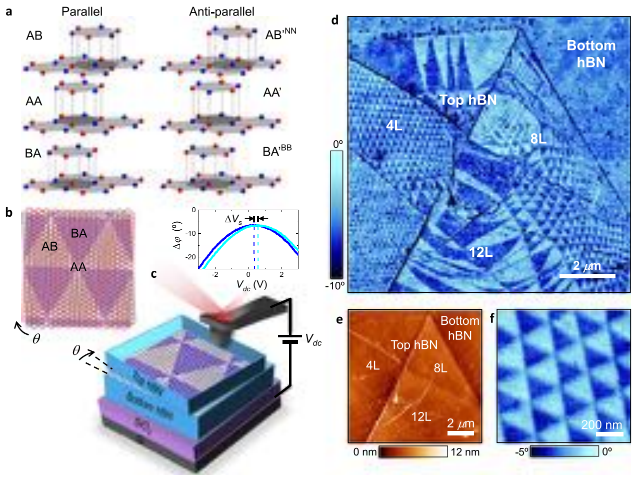
K. Chang et al., Science 353, 6296 (2016)

A few layer WTe_2

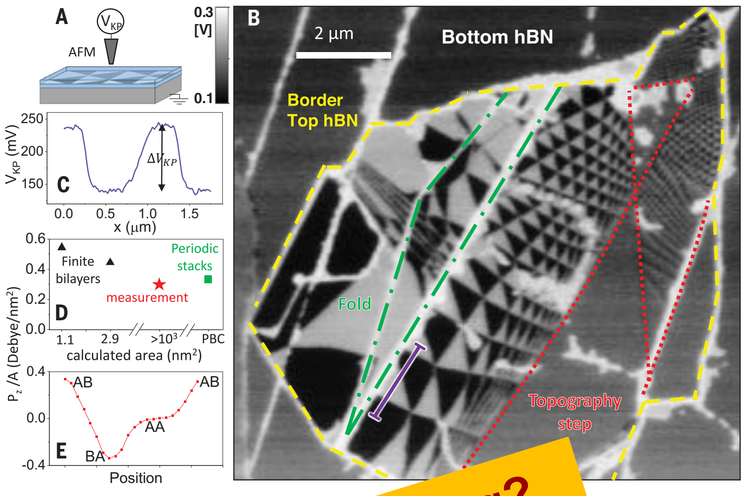


Z. Fei et al., Nature 560, 336 (2018)

Twist Bilayer hBN

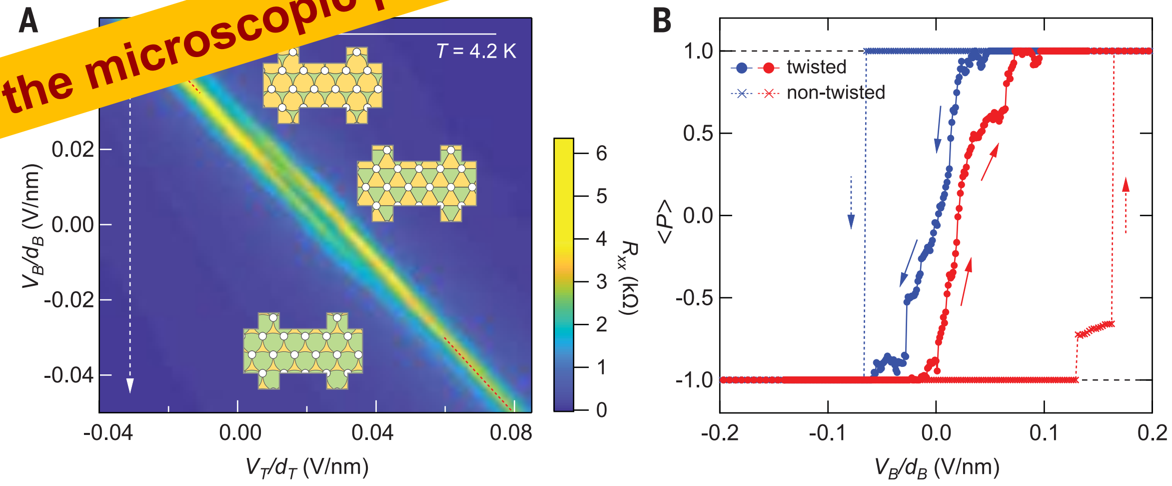


C. R. Wood et al., Nature Com 12:347 (2021)



M Vizner Stern et al., ...162 (2021)

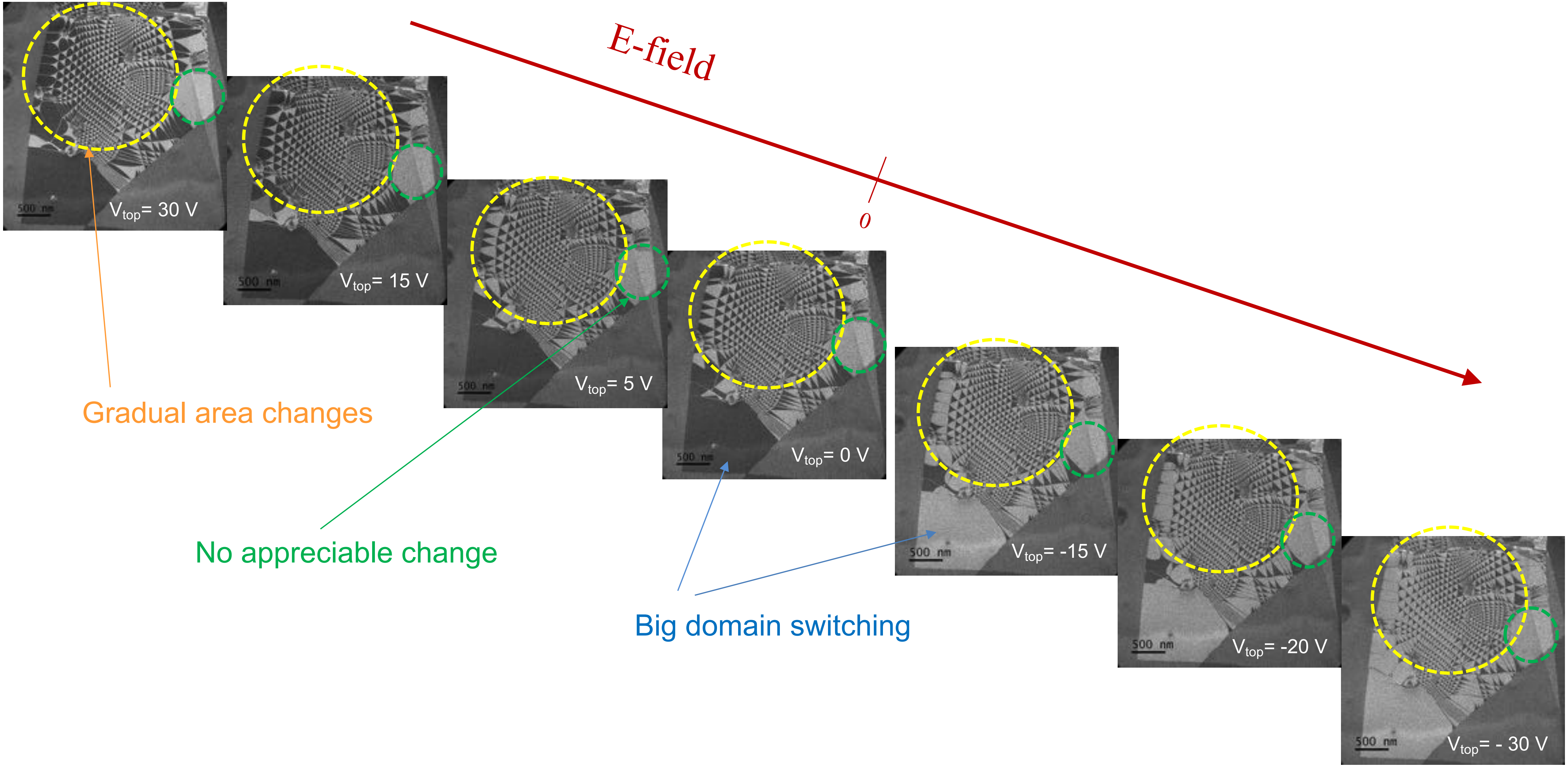
What is the microscopic picture of domain reversing?



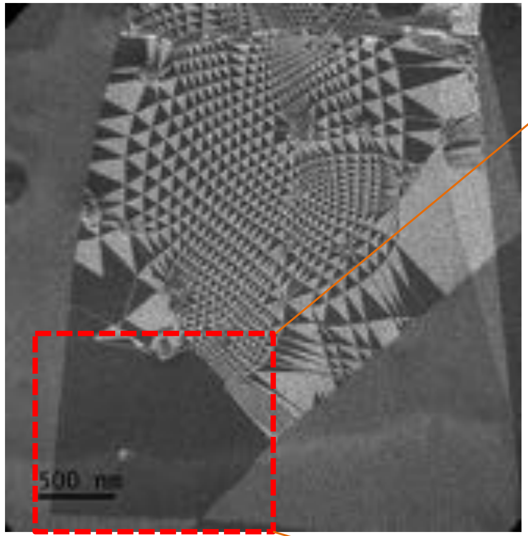
K. Yasuda et al., Science 372, 1458 (2021)

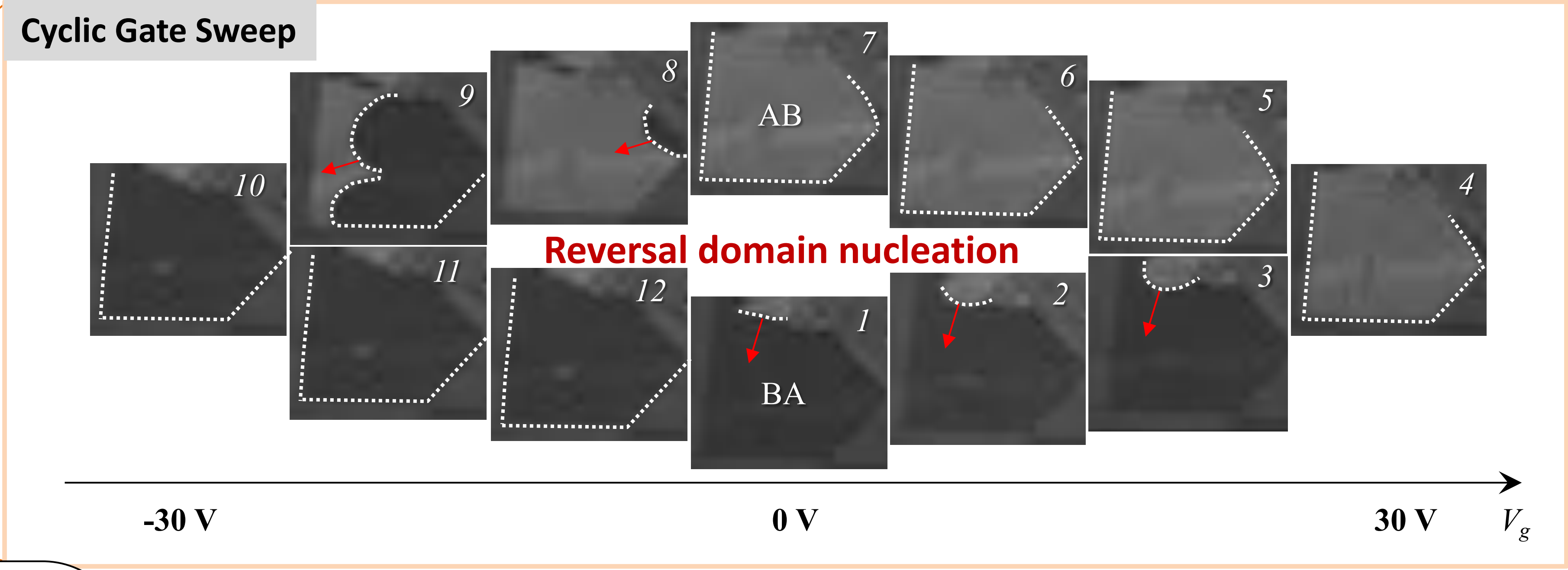
- Noncentrosymmetric
- Formation of polar domains

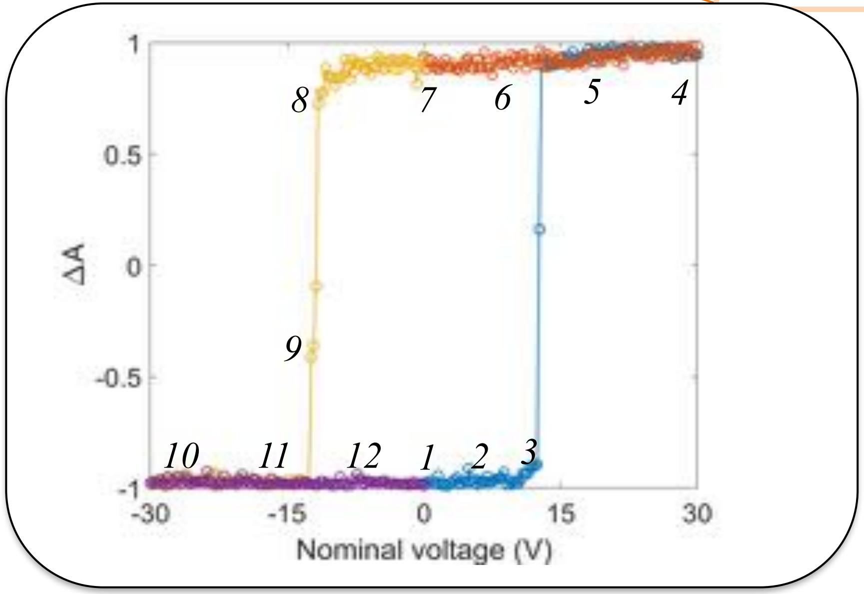
Dynamics of Domain Polarization Switching



Hysteretic domain dynamics





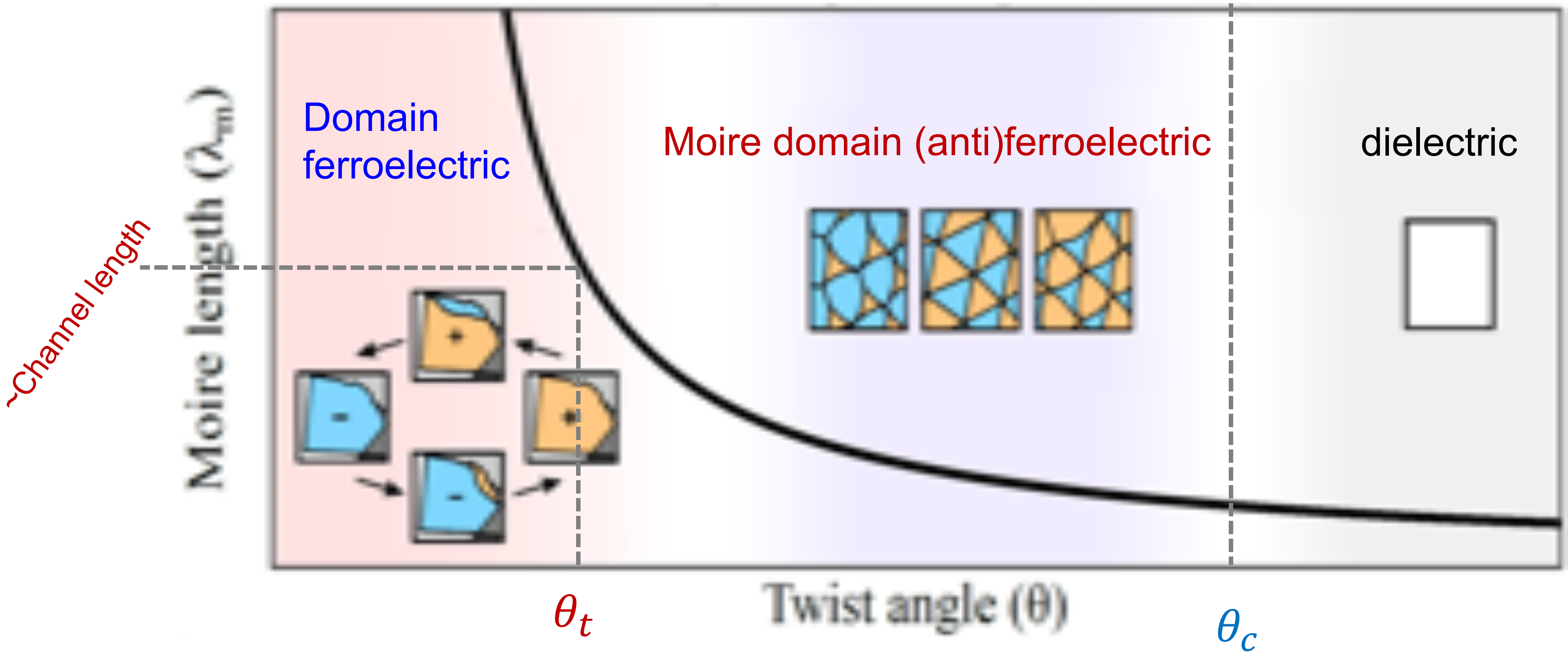


Hysteresis of polarization due to full polarization reversal

$P = 4.3 \times 10^{11}\ e\ \mathrm{cm}^{-2}$

H. Yu et al., submitted

Summary and Outlook



Domain nucleation
Polarization reversal

Atomic relaxation:
Commensurate domains
and domain boundary formation

Domain engineering for topological transition between ferroelectric and anti-ferroelectric might be possible.