# **Experiments in Twisted van der Waals Interface of**

**2D Materials**

**Philip Kim**

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# **Acknowledgement**

#### **Experiments**

**Theory**





**Zeyu Hao**

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**Andrew Zimmerman**



**Isabelle Phinney**







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**Patrick Ledwith**





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## **Funding:**

# **Graphene and SU(4) Spin**



#### **Graphene: possibility of SU(4) magnetism**



*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**  $\overline{a}$



Cascade Transition: broken flavour symmetry state of the feature of the feature  $\mathbb{R}^n$ metal' behaviour was observed in transport33,34. This suggests that the  $T = T$  robust features of the compressibility call for a theoretical for a the  $\mathbf{y}$ , reproducing the section  $\mathbf{y}$ , reproducing experimental experimen **Cascade Transition: broken flavour symmetry** 

**Example 21 Manusual Fan Diagram: magnetic field induced Chern bands** towards the CNP1, where the CNP1, experiment

## **Superconducting Phase Diagram for MA TBG**





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



concomitant with the appearance of a correlated insulator at What is relation between superconductors complete in Device in Device in Device 4, shown in Device 4 (Fig. 2d). In Device 4, shown in Device 4, shown i and correlated insulators?

magnetoresistance over a broad range of filling factor, devoid

behavior of quantum oscillations and Hall effect in Devices

3 and 4 support the absence of significant interaction-induced What is the nature correlation in the insulator? as an intrinsic property.

 $b_{\text{max}}$  in  $b_{\text{min}}$  in finite magnetic field ( $\overline{c}$ Stepanov et al., Nature 583, 375 (2020)

## **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**

detected within the measured span of %&'. These regions correlate with the locations of bubbles (black **EM image of angle 25 inset). The AFM insert angle physics is angle physics in angle physics is approximately angle physics in approximately angle physics is approximately angle physics is approximately approximately appro** 



# **Twisted Double Bilayer Graphene**



Gate tunable flat bands, no exact angle control needed!

# **Mott Insulators in tDBG:**  $\theta$  = 1.33<sup>°</sup>



# **Tunable Spin-Polarized Correlated States**





Normalized Hall density:  $n_{\rm H}$  =  $B/e\rho_{xy}n_{\rm s}$ Moire filling:  $v = 4n/n_s$ Filled bands  $f = | n_H - \nu |$  $v = 4n/n_s$ 

#### Spin-polarized correlated insulators and metals near half**filled moire flat bands**  $\frac{1}{2}$  and metals near name the right bands in the value of  $\mu$

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**

also applies the following in the following  $\boldsymbol{\mu}$ 

#### **Twisted Trilayer Graphene with Alternating Angle** 2 2 .<br>ب 0 TM MB JI ITI  $\overline{a}$

B

10um

0

**D E**

**A B**



 $\mathsf{C}$ .<br>Sin  $\frac{1}{2}$  . Only structure and characterization. (A)  $\frac{1}{2}$ Hao et al., Science (2021); Similar results by Park et al., Nature (2021)  $A = \frac{1}{2}$  and  $\frac{1}{2}$  and  $\frac{1}{2}$ acteristics in the low current and voltage range,

0

# **Superconductivity in TTG**





in the differential resistance, domestic resistance, domestic resistance,  $\frac{1}{2}$ superconducting domes near  $\bm \nu = \pm 2$ **Density dependent** 

behavior in the low V range. The black dashed line indicates where  $H$  and  $\epsilon$ and the superconductions of the superconductivity of the superconductivi .<br>Downloaded from the December 07, 2021<br>Downloaded From December 07, 2021 pattern, demonstrating phase coherence (fig.  $\ddot{\phantom{a}}$  in Touris  $\ddot{\phantom{a}}$  $\frac{6}{3}$  -6  $\frac{6}{3}$  -4  $\frac{6}{3}$  -6  $\frac{6}{3}$  -6  $\frac{6}{3}$  -6  $\frac{1}{3}$  -6  $\frac{1}{3}$  -6  $\frac{1}{3}$  -6  $\frac{1}{3}$  -6  $\frac{1}{3}$ 

# **Displacement Tunable TTG Bands and Superconductivity** <u>l</u> un 1 -0.44V/nm interlayer interlayer interlayer interlayer interlayer interlayer interlayer interlayer interlayer 4  $\frac{h}{h}$ **Bands and Superconductivity.** the band structure is a splitting at in TTG can be ascribed to the tuning of single- $\hbox{\bf tiv}$ itv $\hbox{\bf iv}$  $t_{\text{max}}$



from each other, increasing the combined band $h$ , before  $(2021)$ , bitting tesuts by t and to an, ivaliate  $(202$ . Science ( aı Hao et al., Science (2021); Similar results by Park et al., Nature (2021)

#### **Displacement Field Tunable Superconducting Domes** temperature. This is a common experimental and one that we made in experimental and one that we made in out in nent Field Tunable Superconducting Domes

**Temperature dependent domes** 2003. The black dashed in the black dashed in the black dashed from this procedure are  $\frac{1}{2}$ power-law in low current range and the power exponent range and the power exponent rapidly decreases with incr<br>In low current rapidly decreases with increasing the power exponent rapidly decreases with increasing the power

**HI-RESOLUTION FIGURE PROOF**



*n* binding to the anti-order due to the anti-order due to the anti-order due to the anti-order due to the anti-ر<br>11. 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<sup>1,2,3</sup>\*, Robert Polski<sup>1,2</sup>\*, Cyprian Lewandowski<sup>2,3</sup>, Alex Thomson<sup>2,3,4</sup>, Yang Peng<sup>5</sup>, Youngjoon Choi<sup>1,2,3</sup>, Hyunjin Kim<sup>1,2,3</sup>, Kenji Watanabe<sup>6</sup>, Takashi Taniguchi<sup>6</sup>, Jason Alicea<sup>2,3</sup>, Felix von Oppen<sup>7</sup>, Gil Refael<sup>2,3</sup>, and Stevan Nadj-Perge<sup>1,2†</sup>

#### arXiv:2112.10760 *ifornia 91125, USA* 2*Institute for Quantum Information and Matter, California Institute of Technology, Pasadena, Cal-*

Magic-Angle Multilayer Graphene: A Robust Family of Moir´e Superconductors <sup>4</sup>*Department of Physics, University of California, Davis, California 95616, USA* <sup>5</sup>*Department of Physics and Astronomy, California State University, Northridge, California 91330,*

Jeong Min Park,<sup>1,\*,†</sup> Yuan Cao,<sup>1,2,\*</sup> Liqiao Xia,<sup>1</sup> Shuwen Sun,<sup>1</sup> Kenji Watanabe,<sup>3</sup> Takashi Taniguchi,<sup>3</sup> and Pablo Jarillo-Herrero<sup>1,†</sup> <sup>7</sup>*Dahlem Center for Complex Quantum Systems and Fachbereich Physik, Freie Universitat Berlin, ¨*

# $arXiv:2201.01637$ arXiv:2112.10760v1 [cond-mat.supr-con] 20 Dec 2021  $l_{\text{SUSL}}$  insulation, and superconducting phases. While the original phases. While the original corrections of strong corrections o rX<br>rX

Emergence of Correlations at the Edge of the Magic Angle Regime in Alternating Twist Quadrilayer Graphene G. William Burg<sup>1</sup>, Eslam Khalaf<sup>2</sup>, Yimeng Wang<sup>1</sup>, Kenji Watanabe<sup>3</sup>, Takashi Taniguchi<sup>4</sup>,  $\mathcal{O}$  discovery of correlations and superconductivity in magic-angle twisted biternation twisted states states the trilayer and the trial to expatterns9–11. Here we demonstrate that magic-angle twisted tri-, quadri-, and pentalayers G. William Burg<sup>1</sup>, Eslam Khalaf<sup>2</sup>, Yimeng Wang<sup>1</sup>, Kenji Watanabe<sup>3</sup>, Takashi Taniguch

Emanuel Tutuc<sup>1</sup>  $E$ manuel Tutuc $\cdot$  $W = 1.75 \times 1$  $p$ lacement fields, despite bands international graphene of dispersive bands in  $p$ 



#### **Twisted Quadruple Layer Graphene: Dispersive versus Flat Bands**



Phinney et al., in preparation

## **Superconductivity with proximity induced SOC**



# **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



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## **Experiment**

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## **Theoretical**

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# **Mathematics**

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





**National Research Foundation of Korea** 



# **Graphene: Moire Superlattice**



# **Imaging AB/AC domain in Bernal stacked bilayer graphene**



# Strain solitons and topological defects in bilayer graphene

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

"School of Applied and Engineering Physics, "Department of Chemistry and Chemical Biology, "Kavli Institute at Cornell for Nanoscale Science, and "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



Conclus

Got crosses







Alden et al., PNAS (2013)

## **Lattice Shift Vector: Order Parameter for Relaxation Process**

#### **Unrelaxed**



#### **Define Lattice Shift Vector:**

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



#### **Order parameter maps of unrelaxed lattice**



**Intensity corresponds to distribution densities**



#### **2D Periodicity in configuration space**



We also define  $u = 0$  for untwisted sample and  $u = 0$  for a AA site as a rotational center.

# **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**





R

L

**AB**



 $L = gb^{-1}$  $R = br^{-1}$  $\rightarrow$   $RL \neq LR$ A loop around AA site corresponds to

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

R. Engelke et al., arXiv:2207.05276,

#### **Vortex and Anti-Vortex Pair** lortex and Anti Ve uration space that is isomorphic to *C*, the complex plane, with a physically motivated  $\mathcal{L}_{\mathcal{A}}$  motivated  $\mathcal{L}_{\mathcal{A}}$  and  $\mathcal{L}_{\mathcal{A}}$  $\mathbf{v}$ and study four independent forms of the *uij* matrix:



two-dimensional representation of the *uij* matrix.

under weak uniform strains.

numbers compared to a U(1), but still gives new insight to the U(1), but still gives new insight to the U(1), but still gives new insight to the U(1), but still give the U(1), but still give the U(1), but still give the U(

R. Engelke et al., arXiv:2207.05276, below the surface. By creating a surface  $\mathbf{e}$ 

# **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

# **Broken Mirror Symmetry and Spontaneous Dipoles**

#### MoSe<sub>2</sub>/MoSe<sub>2</sub> near 0 degree





**Electric Field Dependent Photoluminescence**



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

#### Interlayer dipole moment due to charge transfer!

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



#### **Ferroelectric and Anti-ferroelectric** below the Curie temperature, Fig. 4(d), the polarization  $\Gamma$ Fig. 4(e), the trace is an elhpse and when compensated icalcetric and Anti and potassium niobates are ferroelectric and are comparable with the more thoroughly investigated BaTi03. zone coexists with the 'normal value' of the corresponding sound velocity obtained from our Brillouin light scattering data, shown

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#### **Antiberromlastics Crystals**

C. Knrzr. Department of Physics, University of Californ&, Berkdey, CakiforrIia (Received January 10, 1951)

An antiferroelectric state is de6ned as one in which lines of ions in the crystal are spontaneously polarized, but with neighboring lines polarized in antiparallel directions. In simple cubic lattices the antiferroelectric state is likely to be more stable than the ferroelectric state. The dielectric constant above and below the antiferroelectric curie point is investigated for both erst- and second-order transitions. In either case the dielectric constant need not be very high; but if the transition is second order, e is continuous across the Curie point. The antiferroelectric state will not be piezoelectric. The thermal anomaly near the Curie point will be operature as in the ferroelectrical. A structure in as found in strontium titanate is not indicative of antiferroelectricity, unlike the corresponding situation in antiferromagnetism.





#### Antiferroelectric state of  $PbZrO<sub>3</sub>$  $\mathsf{Aut}(\mathsf{H})$  displacements in the cubic crystallographic crystallo

**Mode enti-ferroelectric**<br>ale anti-ferroelectric<br>Mode 3 le anti-ferroelectric c 3 **Atomic scale anti-ferroelectric versus** 2 **Moire scale anti-ferroelectric domains?**

**a** based on the contract of t

# **Ferroelectricity in vdW Hetero/homostructures**

#### **Atomically thin group-IV monochalcogenide**



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

#### **A few layer WTe<sub>2</sub> v** lew layer vv le $_2$



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

- $\epsilon \approx \theta$  the as a fitting as a function of the application of the app • Noncentrosymmetric
- pation of polar domaine respectively. He is estimated by the potential interest  $\sim$ • Formation of polar domains

RESEARCH | REPORT



RESEARCH | REPORT

 $\mathcal{L}$  by dots (blue dots)

 $\blacksquare$   $\blacksquare$ compare all possible configurations at all possible configurations at all  $\alpha$  $i_{\text{dense}}$  270, 1459 (2021). K. Yasuda et al., Science 372, 1458 (2021)

# **Dynamics of Domain Polarization Switching**



# **Hysteretic domain dynamics**



H. Yu et al., submitted

# **Summary and Outlook**

![](_page_32_Figure_1.jpeg)

Commensurate domains and domain boundary formation

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