Kekulé Spirals in Twisted Bilayer Graphene

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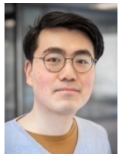
Tomohiro Soejima


Michael Zaletel

Berkeley


Erez Berg
Weizmann

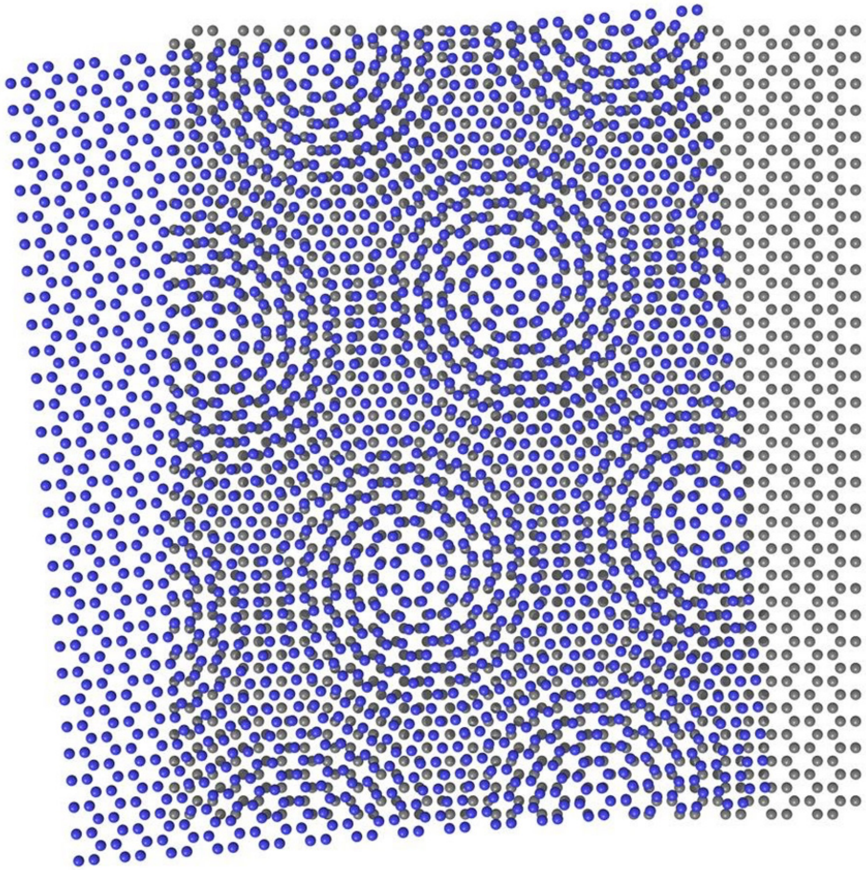

Yichen Hu
Florida


Nilotpal Chakraborty
Dresden

Kwan et al PRX **11,** 041063 (2021); PRX 12, 031020 (2022); PRB 110 (8), 085160 (2024);...
Wagner et al, PRL **128**, 156401 (2022)...; Wang et al PRB **109**, L201119 (2024)...

Do not capitalize moiré (unless it starts a sentence).

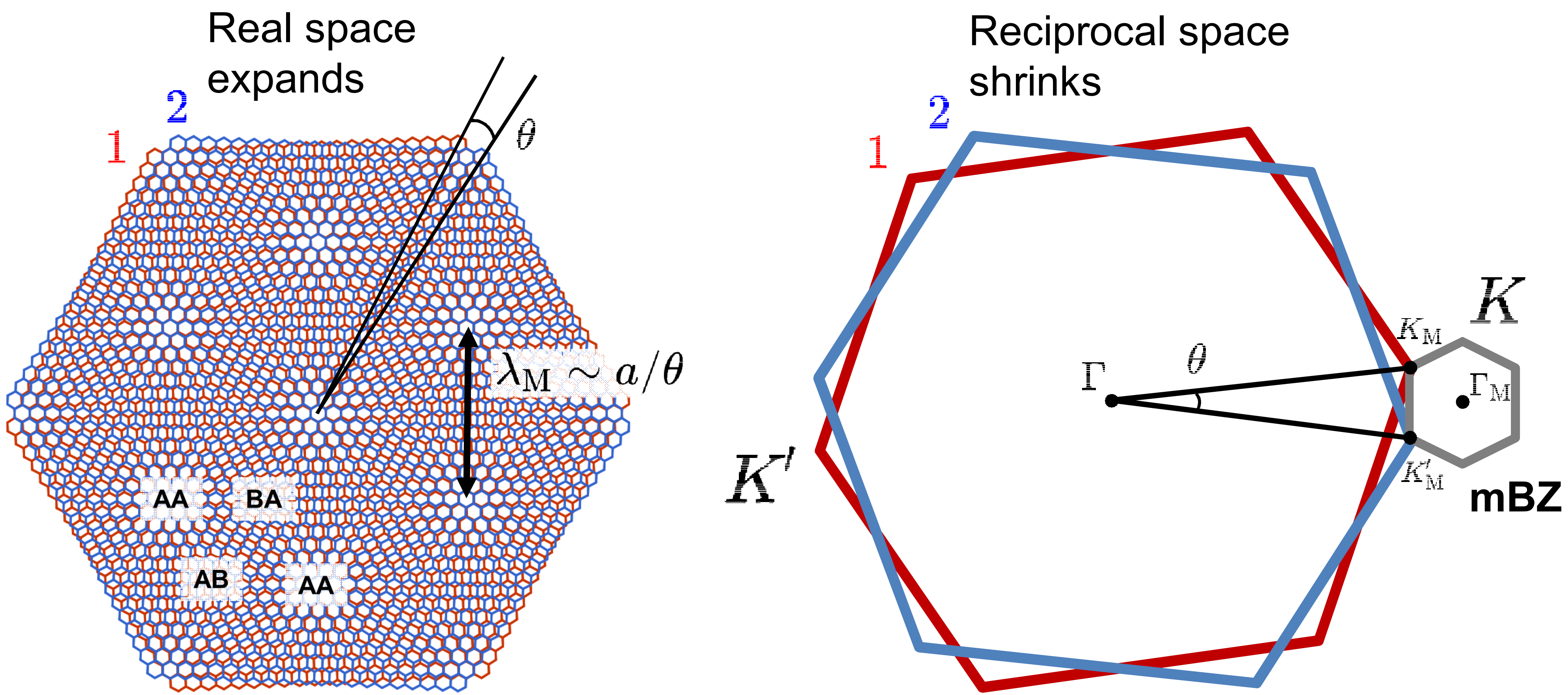
moiré: from *moirer*, from mohair, from *mukhayyar* or *khayyara* [chosen or best]



Picture: NIST

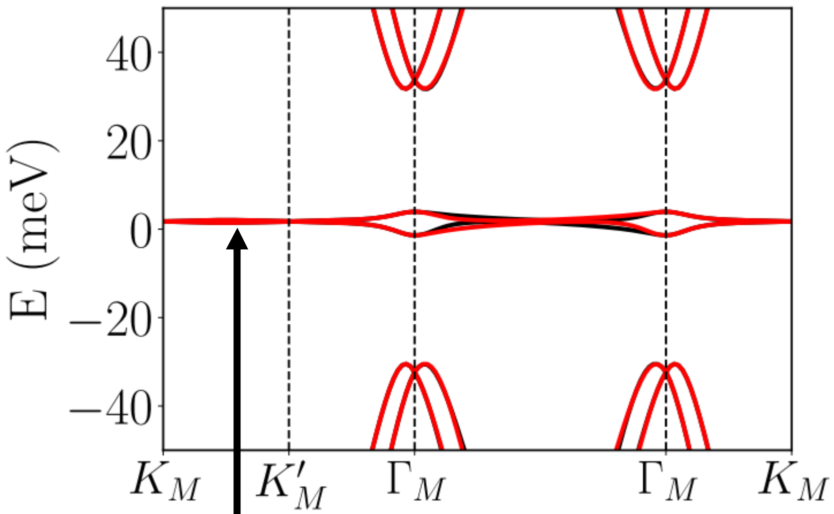
Moiré is the best!

moiré pattern



moiré pattern can generate flat bands:
Twisted Bilayer Graphene Bistritzer & MacDonald, *PNAS* (2011)

FLAT BAND → INTERACTIONS DOMINATE



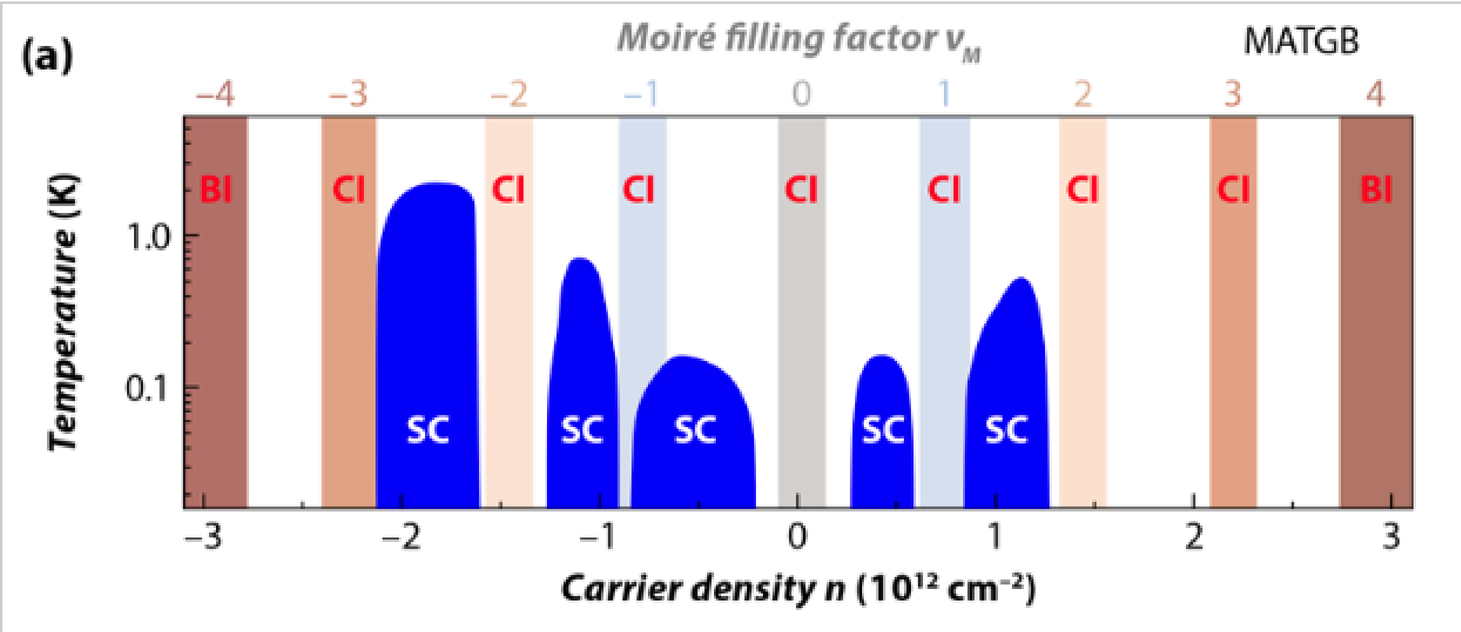
Magic Angle TBG

8 central bands
incl. spin and valley

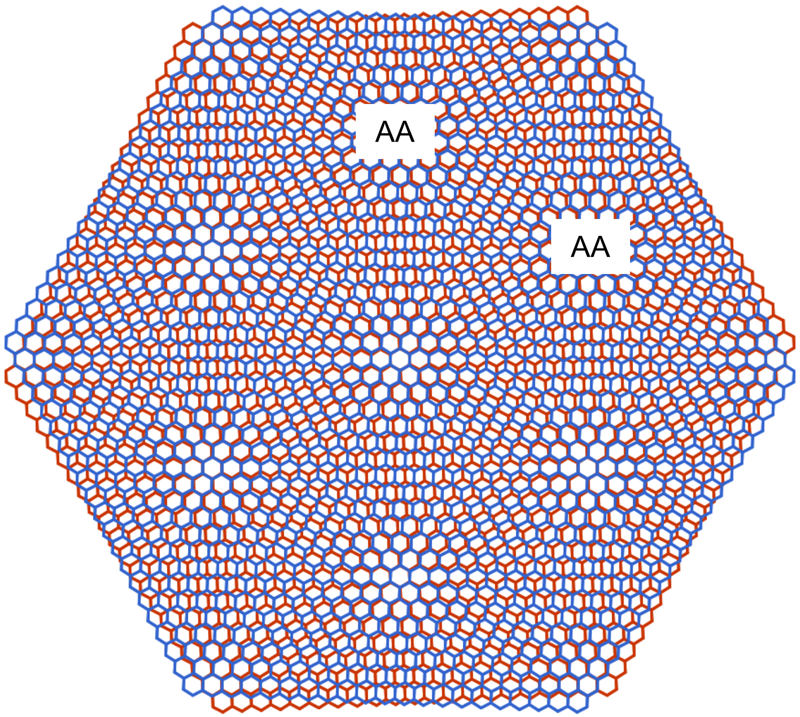
- focus on central bands

filling factor $\nu = -4, \ldots, 0, \ldots, 4$
(CNP)

Rough phase diagram (Stolen from MacDonald):



Q: What is the Correlated Insulator (Usually?)



Q: What is the Correlated Insulator (Usually?)

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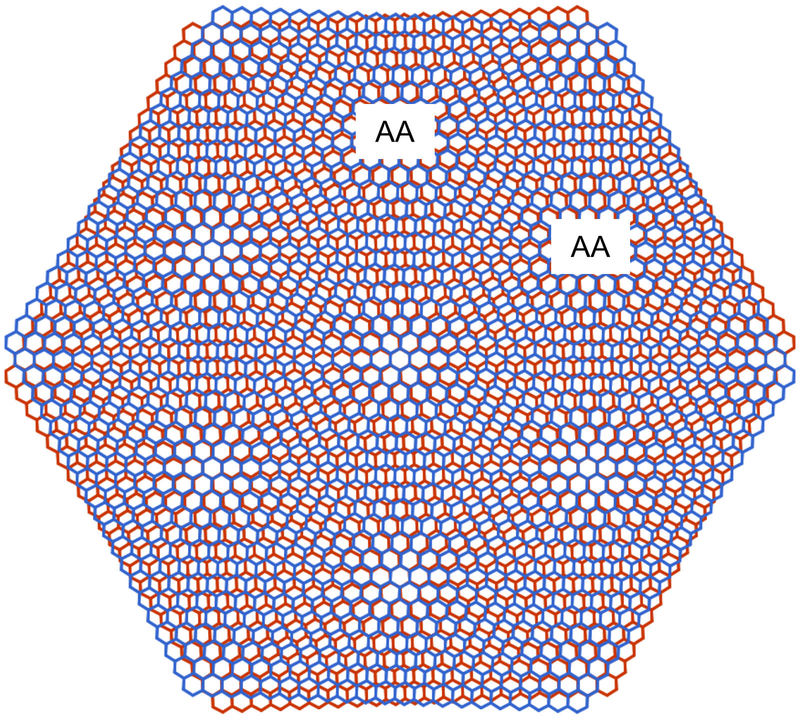

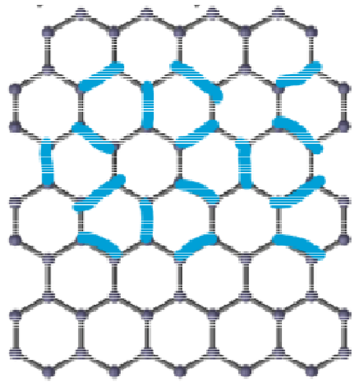

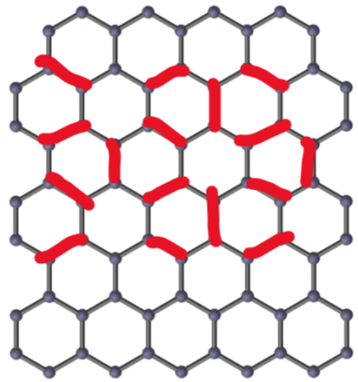

<u>Q: What is the Correlated Insulator (Usually?)</u>

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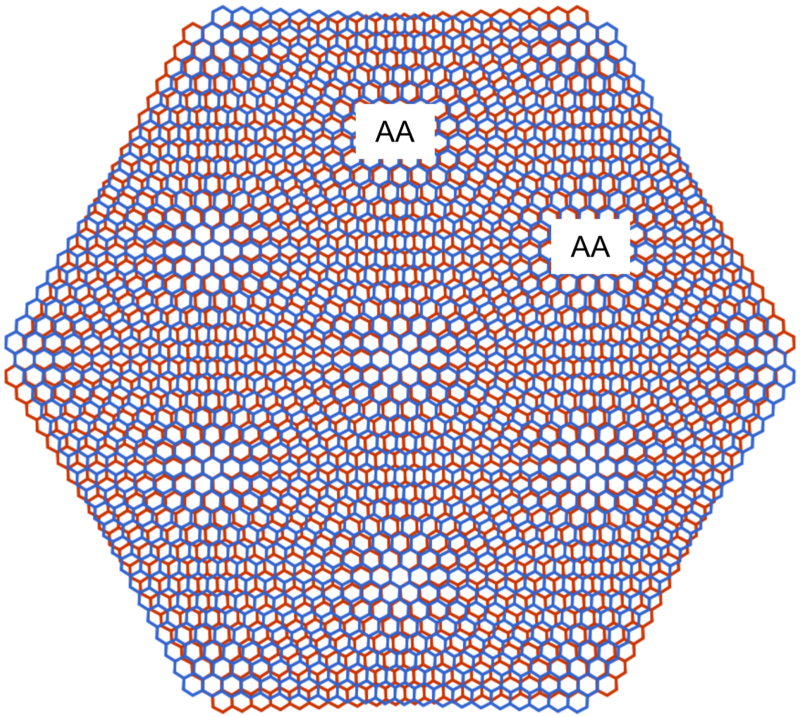

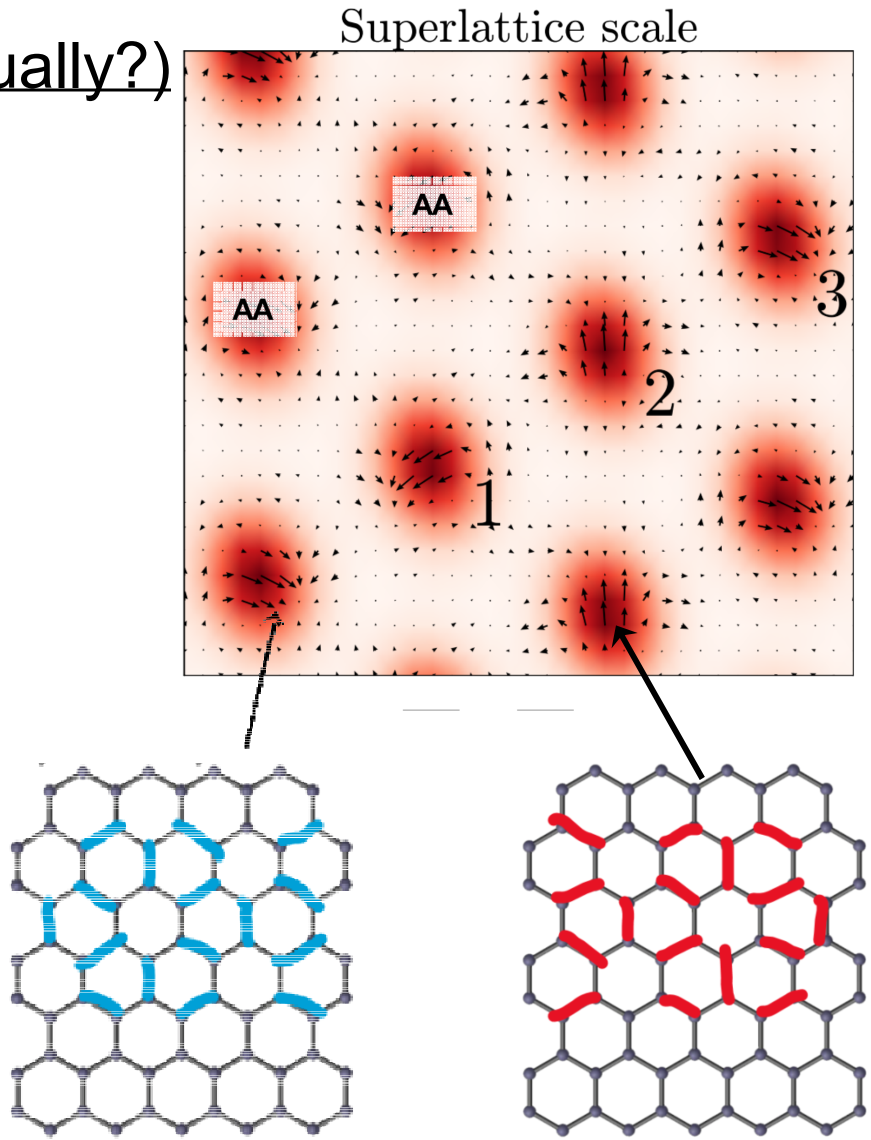

Q: What is the Correlated Insulator (Usually?)

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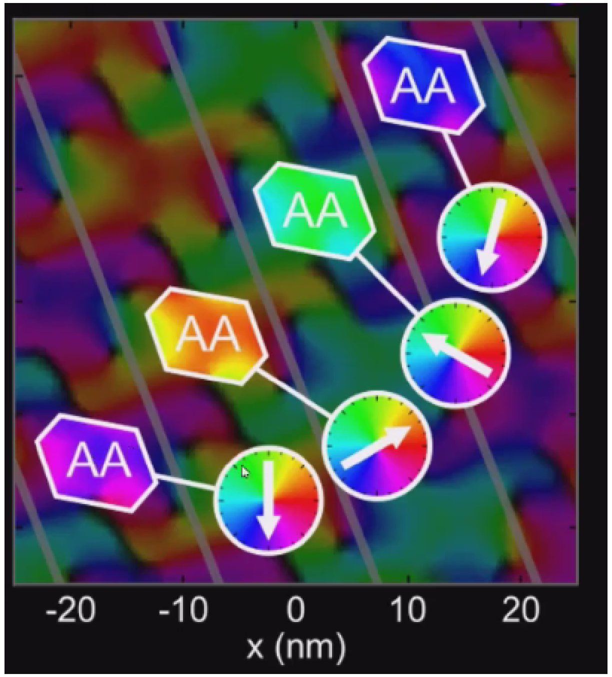
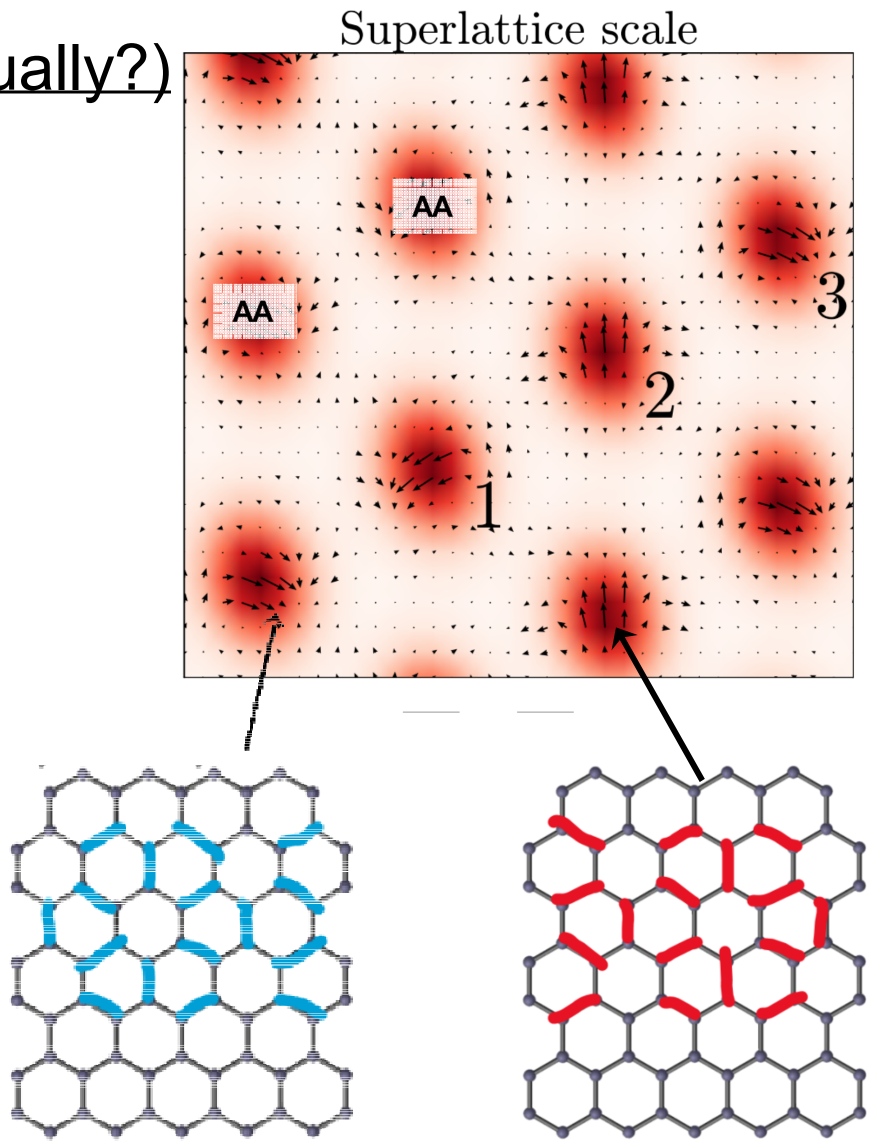
This is real:



(Screenshot from talk by Nuckolls/Yazdani)
Nature 620, 525 (2023) Nuckolls et al (Yazdani)



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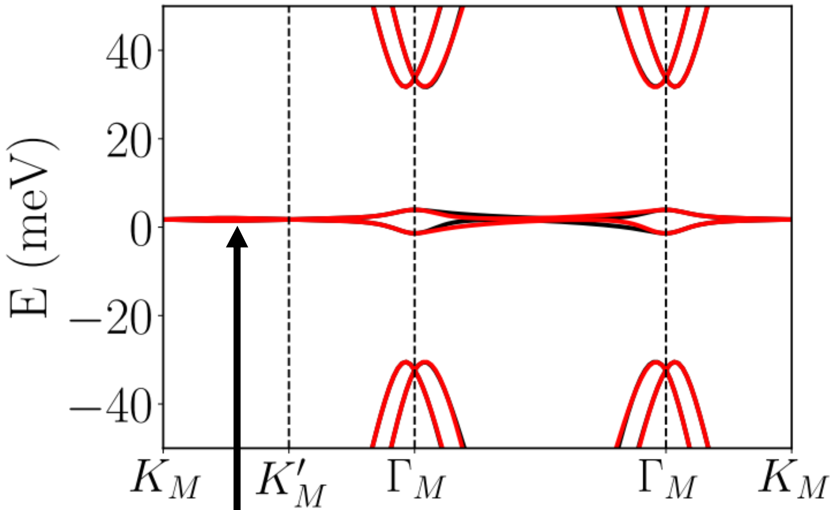


Magic Angle TBG

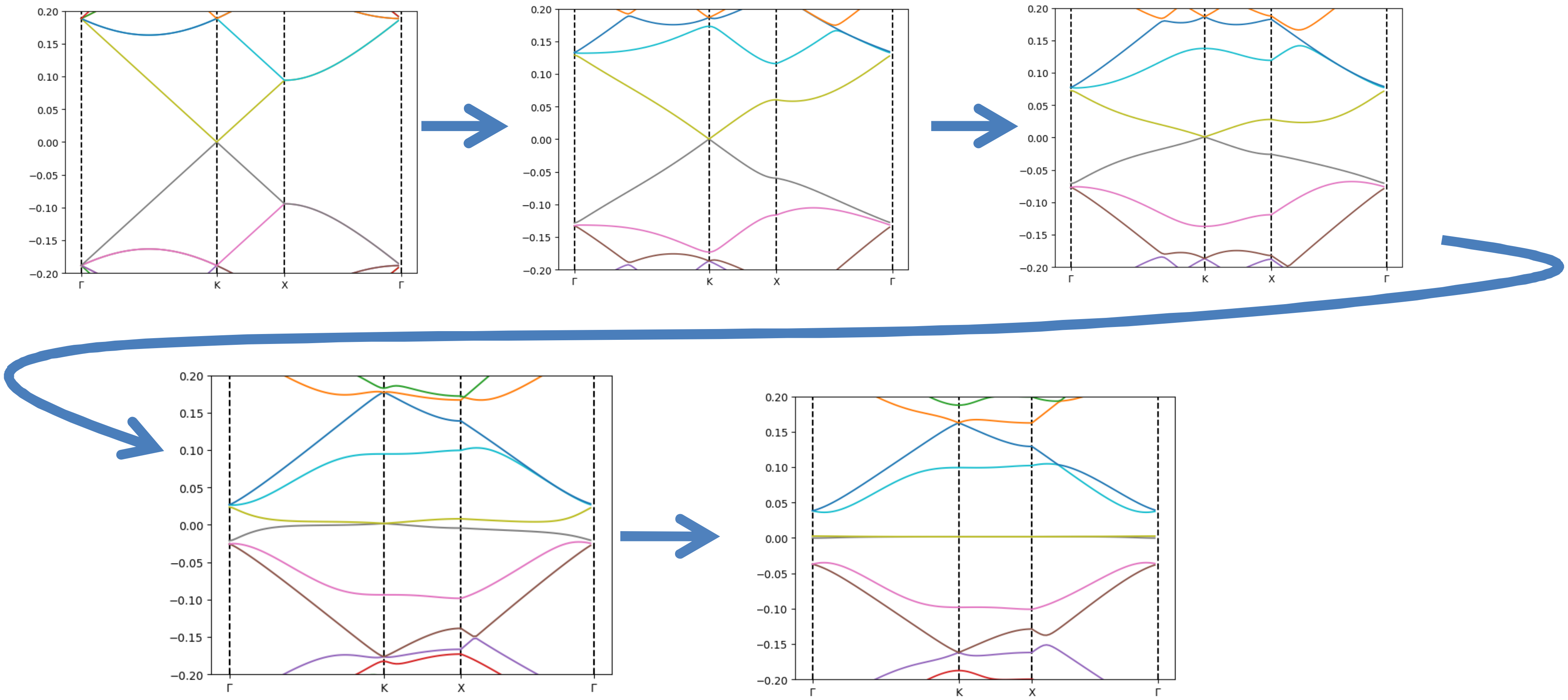
8 central bands incl. spin and valley

- focus on central bands

filling factor $\nu = -4, \ldots, 0, \ldots, 4$
(CNP)

Slowly turning on the layer coupling:

No coupling

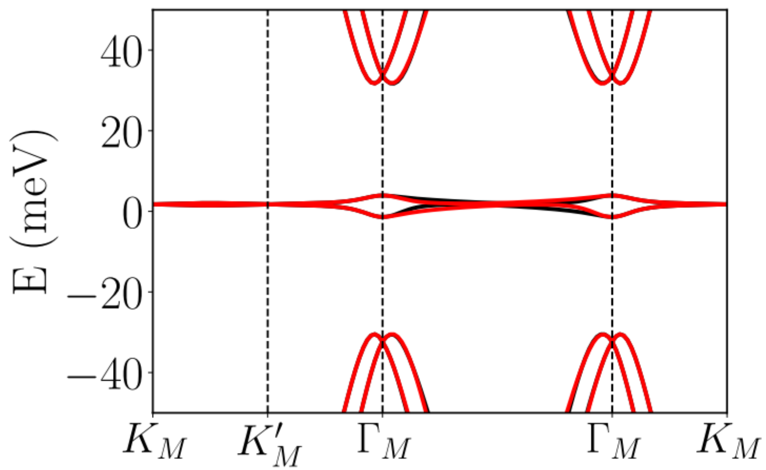


Full coupling

“Chiral” Limit of Bistritzer-MacDonald (BM) Model = TKV Model

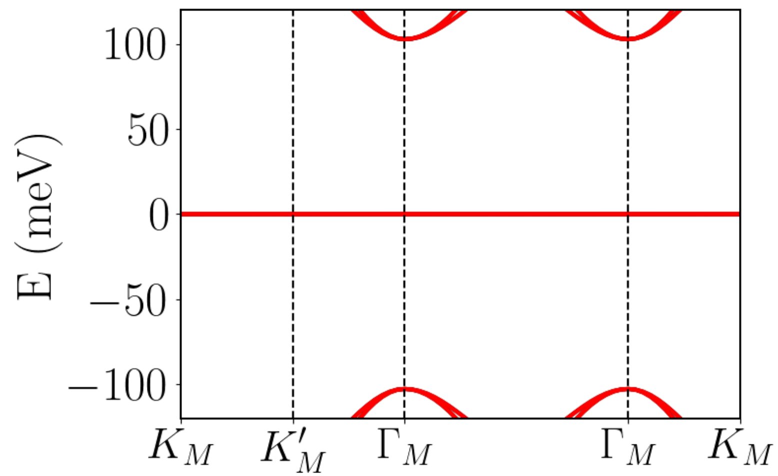
- **chiral ratio** $\eta = w_{AA}/w_{AB}$

$\eta = 0.8$



chiral limit

$\eta = 0$



Tarnopolsky, Kruchkov, Vishwanath, *PRL* (2019)

Chern Number:

Shiing-Shen Chern

Theorem:

A two-dimensional insulator has a quantized Hall conductivity

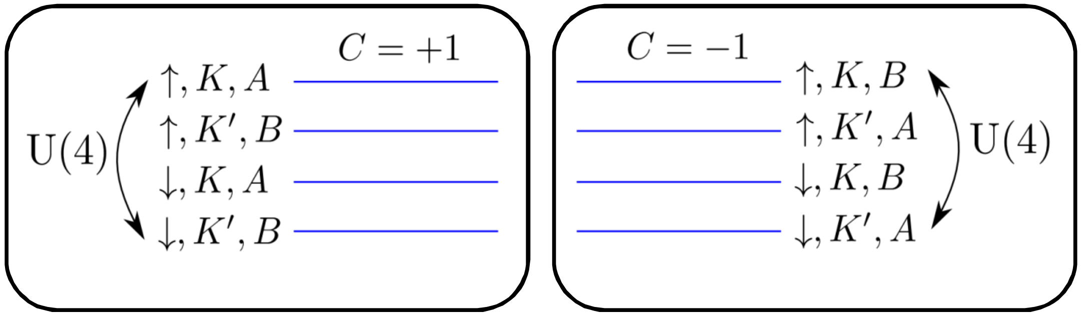
$$G_{xy} = C\frac{e^2}{h}$$

where C is the Chern number.

For non-interacting electrons:
(insulator=filled band and a gap)
C is integer.

Chiral Limit (Strong Coupling)

Bultinck et al., *PRX* '20
Bernevig et al., *PRB* '21
Lian et al., *PRB* '21

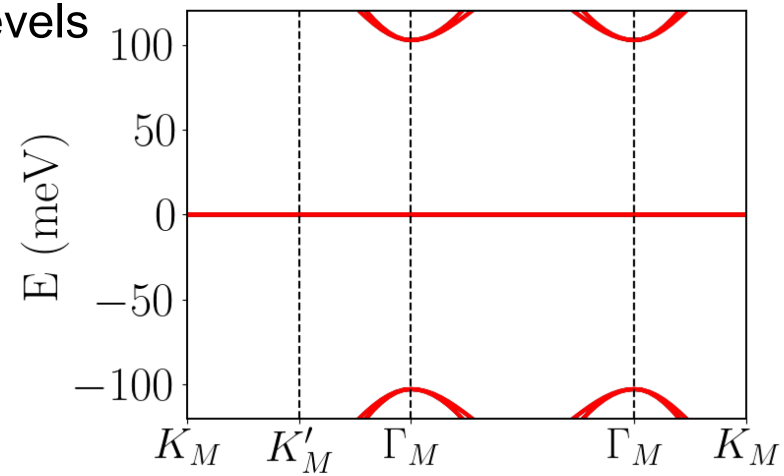


Two 'Chern quartets' : like multicomponent Landau levels

Expect flavor ferromagnetism

chiral limit

$\eta = 0$



Tarnopolsky, Kruchkov, Vishwanath, *PRL* (2019)

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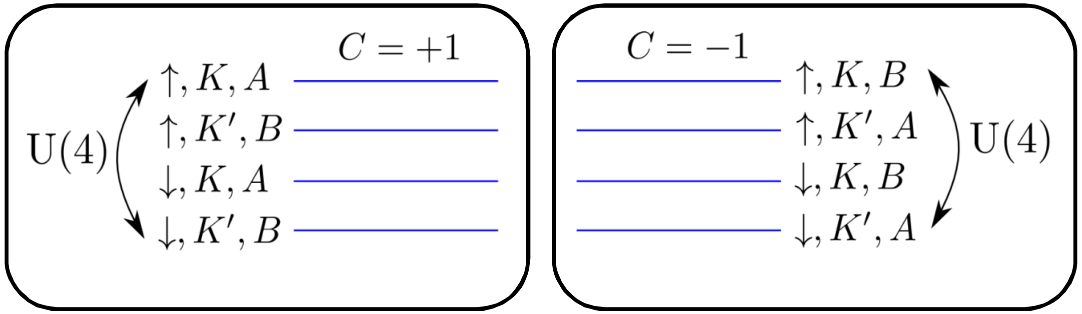


Two ‘Chern quartets’ : like multicomponent Landau levels

Expect flavor ferromagnetism = Slater determinant ground states

Fill any $4+\nu$ out of 8 central Chern bands (generalized ferromagnets – can break T)

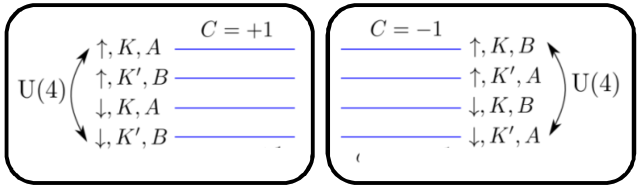
But can rotate within a Chern-Band, ex, fill $|K A \uparrow\rangle + e^{i\phi}|K' B \downarrow\rangle$

tilting the pseudospin before filling the band

Superposing K and K’ introduces wavevector |K – K’| which triples graphene unit cell

Chiral Limit (Strong Coupling)

Bultinck et al., *PRX* ‘20
Bernevig et al., *PRB* ‘21
Lian et al., *PRB* ‘21

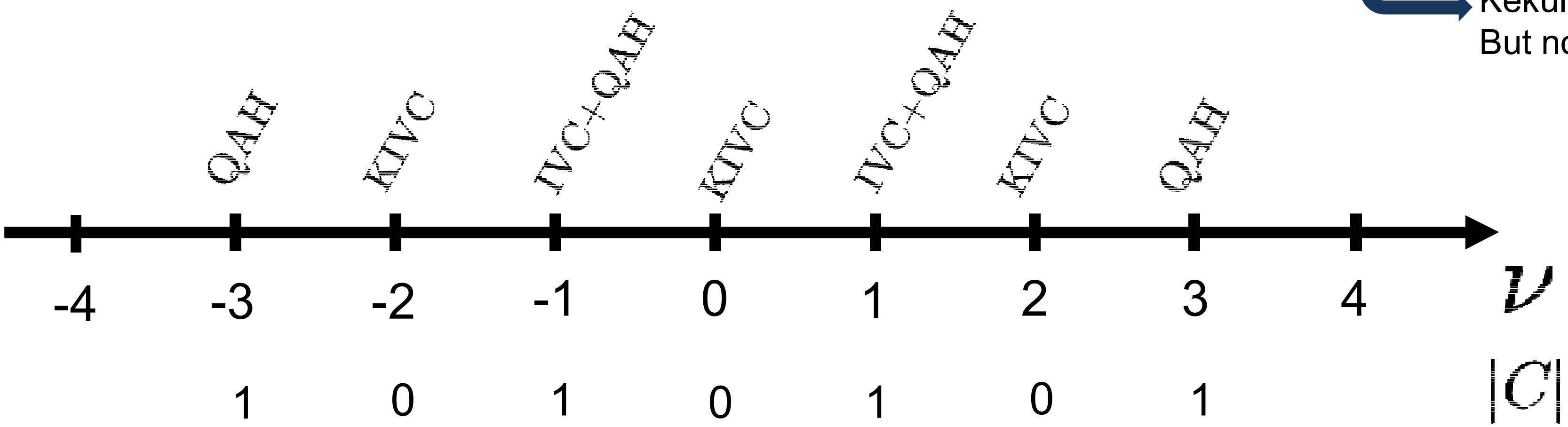


- single-particle terms and deviations from chiral limit treated perturbatively

select out subset of generalized ferromagnets
i.e. **‘strong-coupling’ insulators**

QAH = Quantum Anomalous Hall
IVC = Intervalley Coherent
KIVC = Kramers IVC

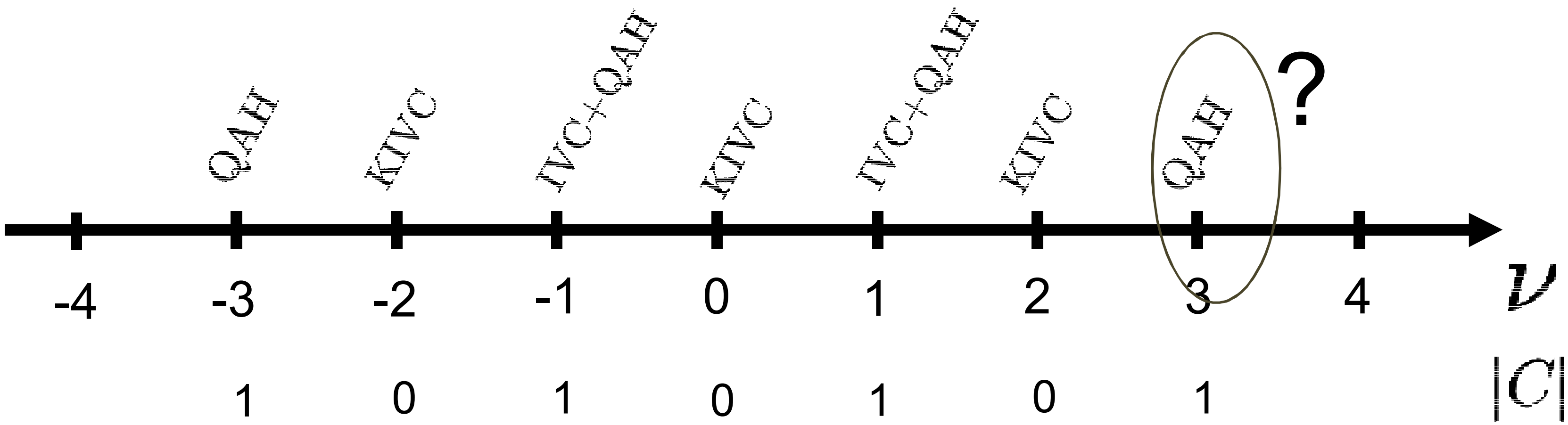
Kekule currents
But not density



Bultinck et al., *PRX* ‘20
Xie & MacDonald, *PRL* ’20
Cea & Guinea, *PRB* ’20
Zhang et al., *PRB ’20*
Lian et al., *PRB* ‘21
Xie et al., *PRB* ‘21
Potasz et al., *PRL* ‘21
Parker et al., *PRL* ‘21
Kwan et al., *PRX* ‘21
…

Strong Coupling & Experiments

- ✓ correlated insulators at integer ν as flavour ferromagnetism
- ✗ experimentally CNP is often semimetallic, but large-gap robust insulators in strong-coupling
- ✗ experimentally metallic/weak anomalies at $\nu = \pm 1$, but gapped insulators in strong-coupling
- ✗ observation of ‘Chern-odd’ insulators, e.g. C=0 at $\nu = 3$ Pierce et al., Nat Phys, ’21



Beyond Strong Coupling

"Perturbations"

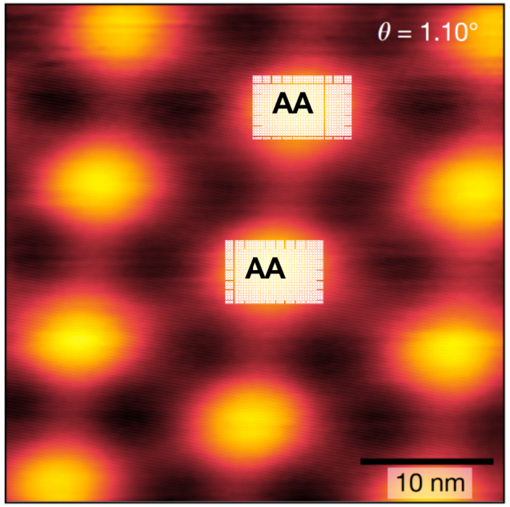
- Not exactly flat band
- p-h sym breaking terms
- substrate potential
- Strain

Treat these more seriously from the start?

Strain

- uniaxial heterostrain ϵ

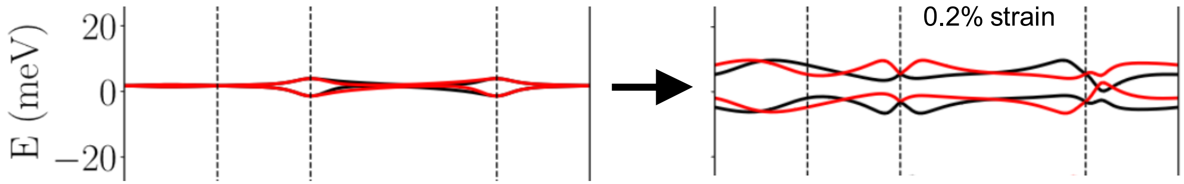
Bi, Yuan, and Fu PRB 2019



strain is ubiquitous in experiment

typically $\epsilon \simeq 0.1 - 0.7\%$

Xie et al., *Nature* '19
Choi et al., *Nat. Phys.* '19
Kerelsky et al., *Nature* '19

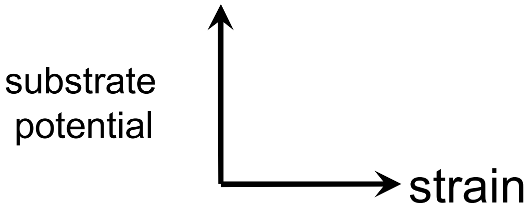


- theoretically known to degrade strong-coupling KIVC gap at charge neutrality (many expts see no gap at ν=0)

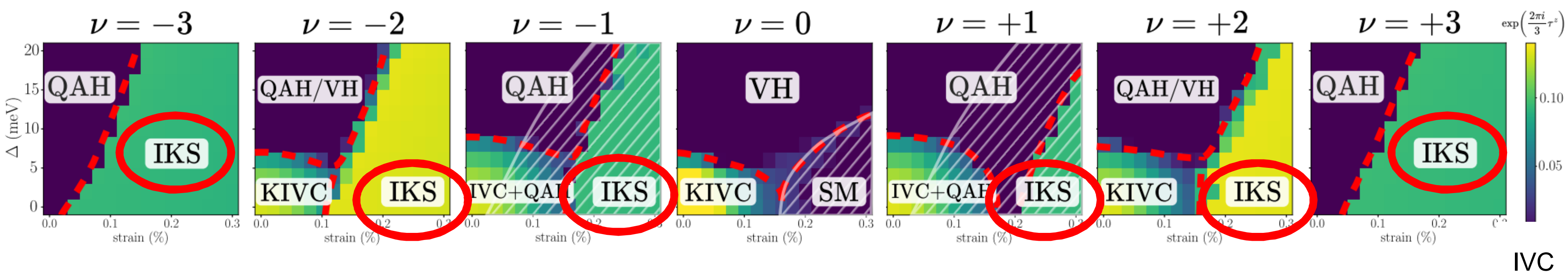
Parker et al., *PRL* '21

Results

Kwan et al., *PRX* 2021

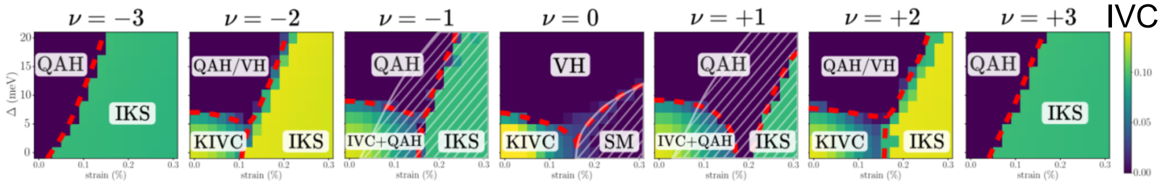


<u>IKS: Incommensurate Kekulé Spiral</u>
QAH: quantized anomalous Hall state
KIVC: Kramers intervalley coherent state
VH: valley Hall state
IVC: intervalley coherence
SM: semimetal
Hatched: no HF gap



- self-consistent Hartree-Fock with realistic parameters
- check for completely general translational symmetry-breaking
- **IKS dominates all non-zero integers with strain**

What is IKS? Symmetries



For any integer ν :

- Superposes K and K' (i.e., Kekule distortion, breaks $U(1)_v$)
- breaks translation symmetry $\hat{T}_{\mathbf{a}_i}$

q is wavevector of the spiral

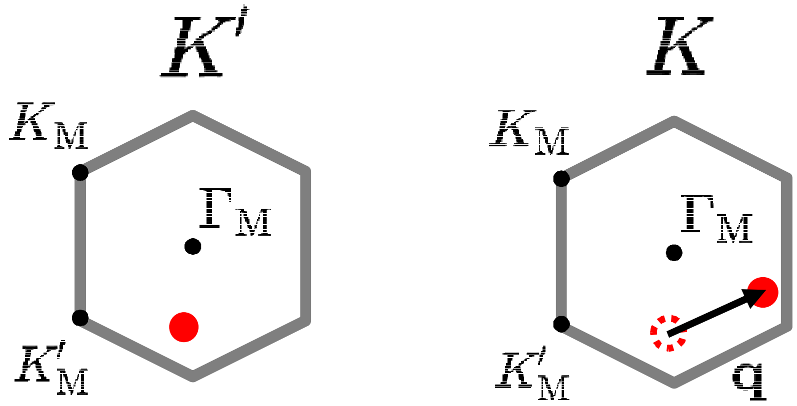
- preserves modified translation symmetry $\hat{T}'_{\mathbf{a}_i} = \hat{T}_{\mathbf{a}_i} e^{i\mathbf{a}_i \cdot \mathbf{q}\tau_z/2}$
  - combination of moiré translation and valley rotation

Kwan et al., 2105.05857

What is IKS? **q**

- valleys hybridize at finite wavevector **q**:

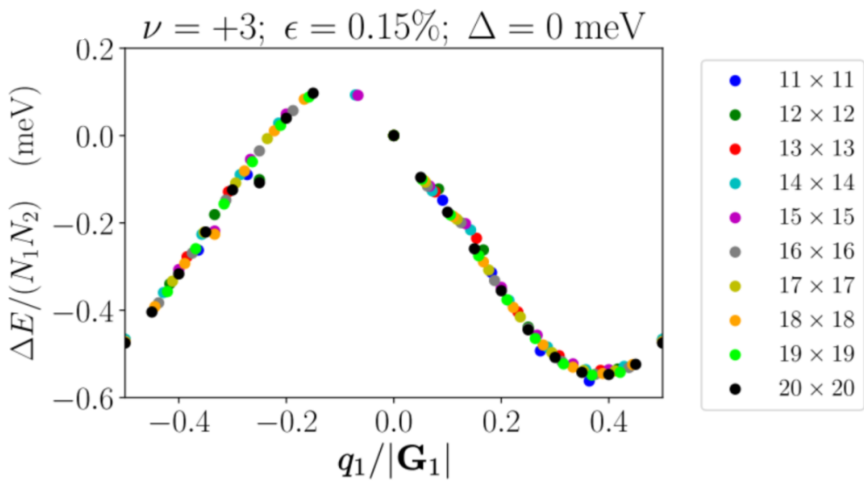
$$u_{\mathbf{k}}|K, \mathbf{k}-\mathbf{q}/\mathbf{2}\rangle + v_{\mathbf{k}}|K', \mathbf{k}+\mathbf{q}/\mathbf{2}\rangle$$



labelled by $\tilde{\mathbf{k}}$

$\tilde{\mathbf{k}} = \mathbf{k} + \tau_z \mathbf{q}/2$

- **q** is very soft, and generally incommensurate

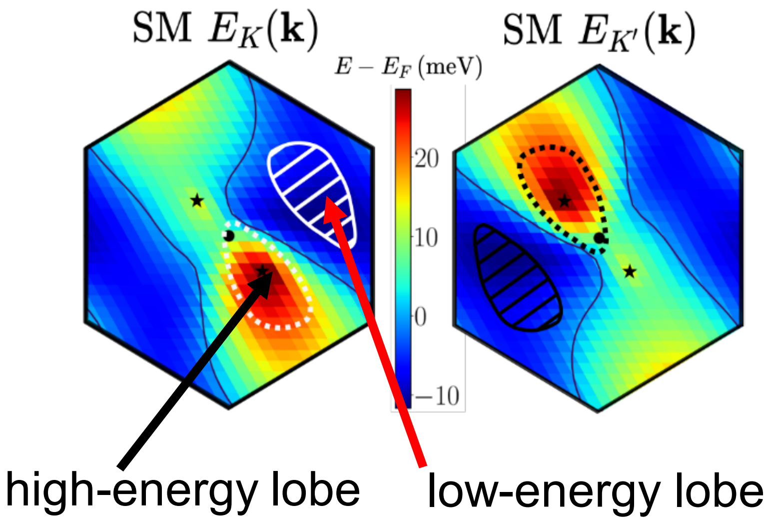


IKS: Incommensurate Kekulé Spiral

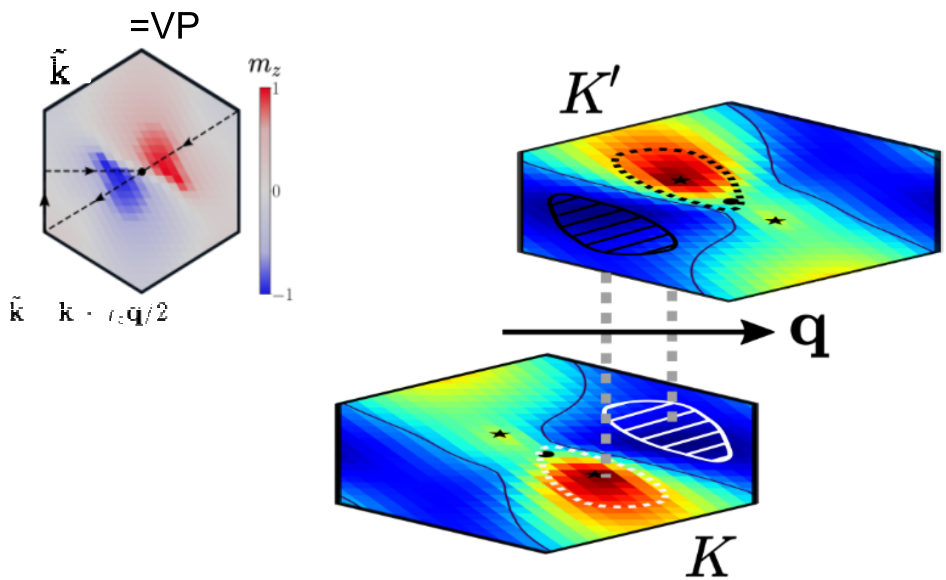
Kwan et al., *PRX* '21

Physical mechanism for IKS

consider HF bands of symmetric metal



- Align high-energy lobe in one valley with low-energy lobe in other valley to find **q**. IVC everywhere else.



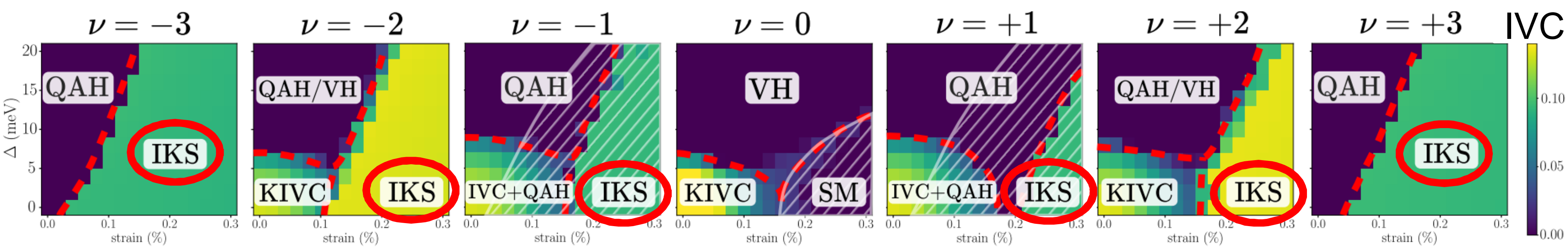
Strain increases dispersion → Strong Coupling Ferromagnetism Fails!

Kwan et al., *PRX* ‘21

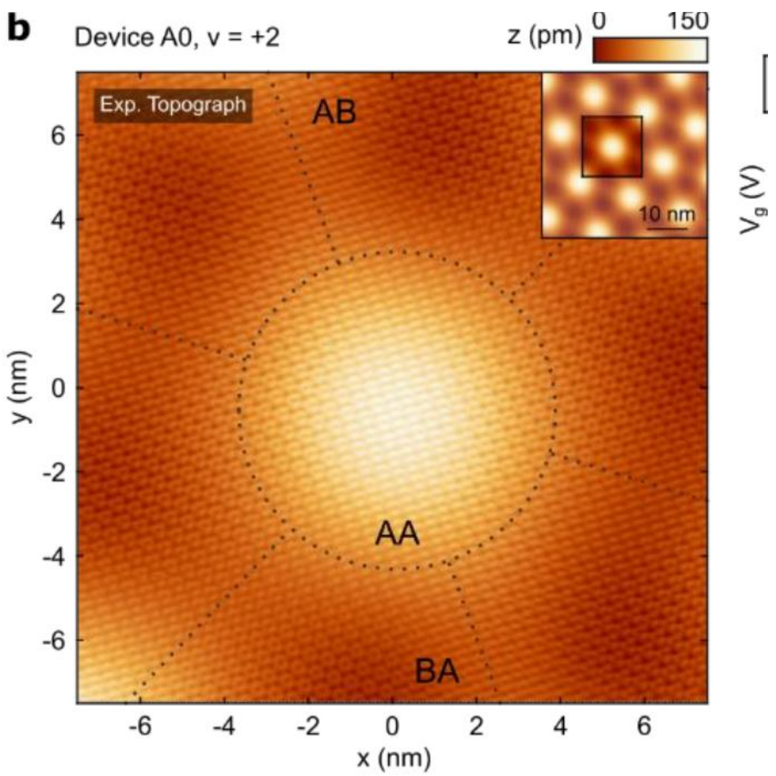
Connections of IKS/HF to experiment

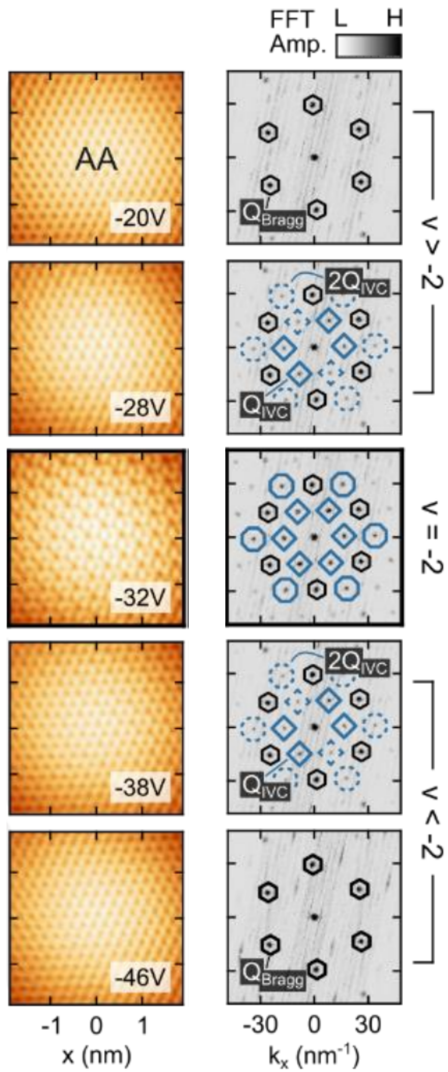
- ✓ strain significantly degrades gap and induces semimetallic behaviour at CNP
- ✓ IKS at $\nu = \pm 1$ is a 'near-insulator'
- ✓ gapped IKS at $\nu = \pm 2$ is spin-unpolarized Yankowitz et al., *Science* '19
- ✓ gapped IKS at $\nu = \pm 3$ has C=0 Pierce et al., Nat Phys, '21
- ✓ IKS emerges at strain ratios well within experimental limits

✓ KEKULÉ SPIRAL OBSERVED IN STM! Nature 620, 525 (2023) Nuckolls et al (Yazdani)



Kwan et al., *PRX* '21





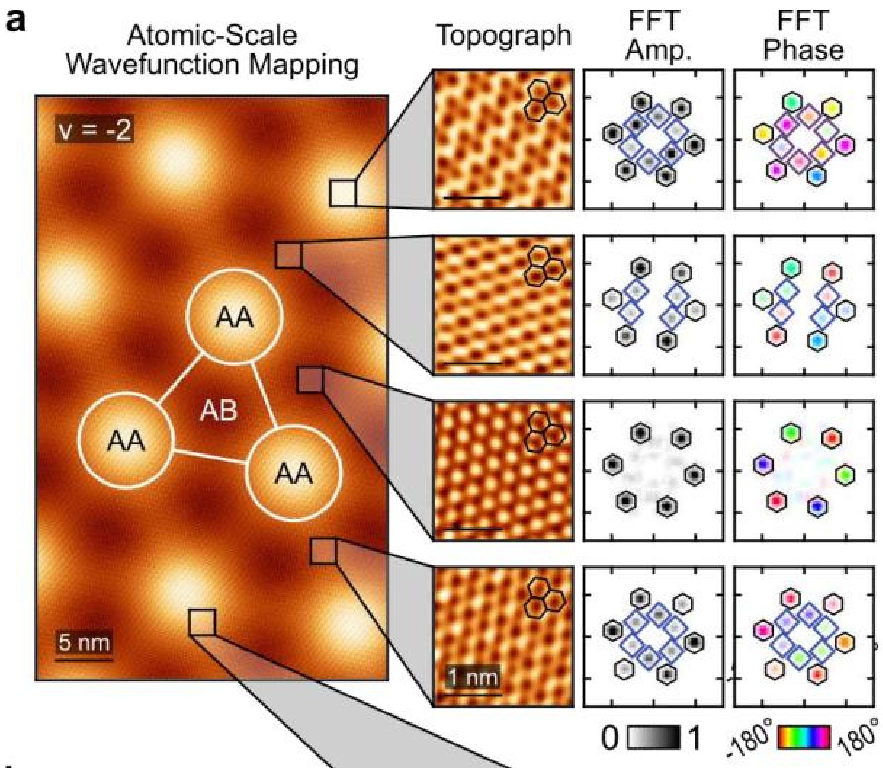
Graphene Bragg Peaks

Also Kekulé distortion

Kekulé distortion = Intervalley Coherence,
.....BUT NOT KIVC!
Hong et al *PRL* 22
Calugaru et al *PRL* 22

"Quantum textures of the many-body wavefunctions in magic-angle graphene"

Nature 620, 525 (2023) Nuckolls et al (Yazdani)

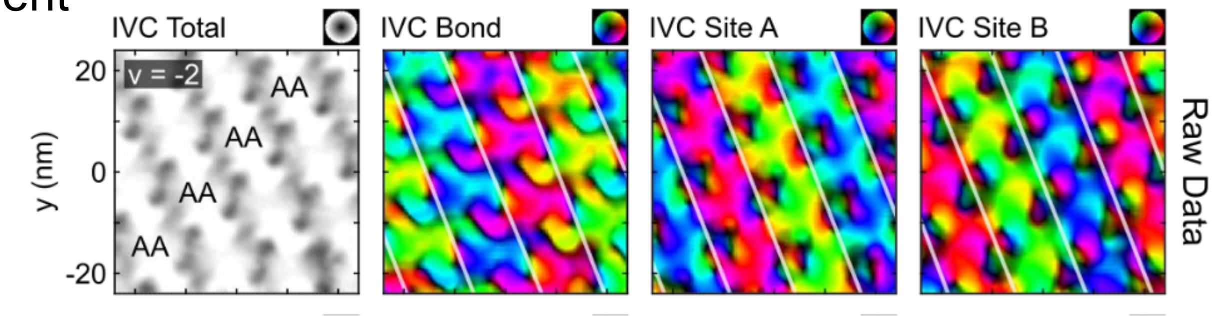


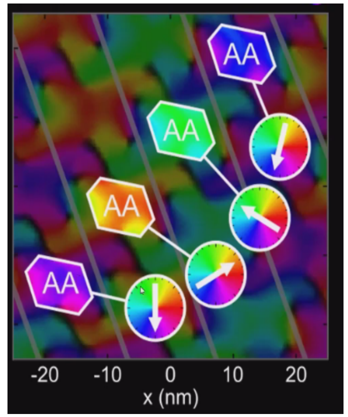
Kekule Bragg Peaks seen almost everywhere, but pattern changes

Analysis by phases of FFT peaks

Nature 620, 525 (2023) Nuckolls et al (Yazdani)

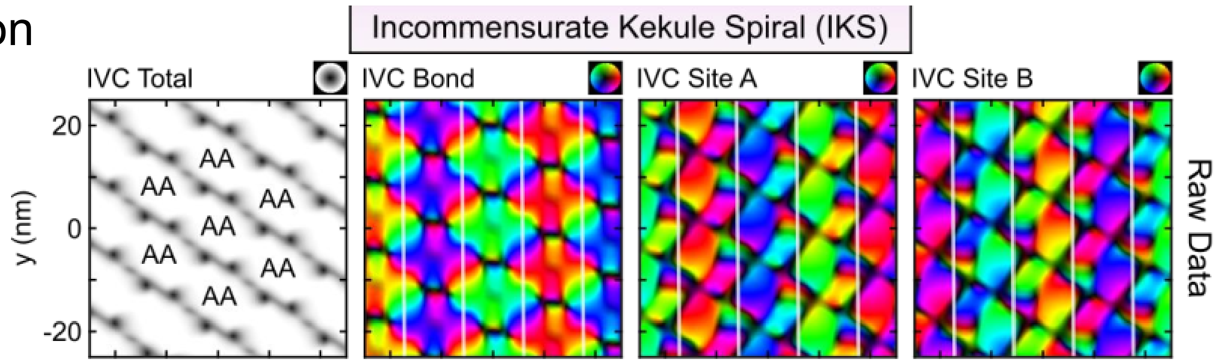
Experiment





(Screenshot from talk by Nuckolls/Yazdani)

Simulation



IKS is seen in all “typical” samples at $\nu = \pm 2$.

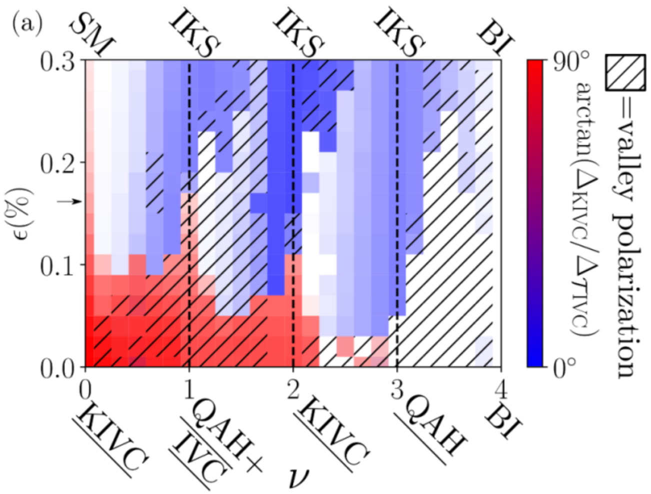
In a few samples with ultra-low strain (< 0.1%) they see something different (but not KIVC!)

Nature 620, 525 (2023) Nuckolls et al (Yazdani)

Non-integer fillings

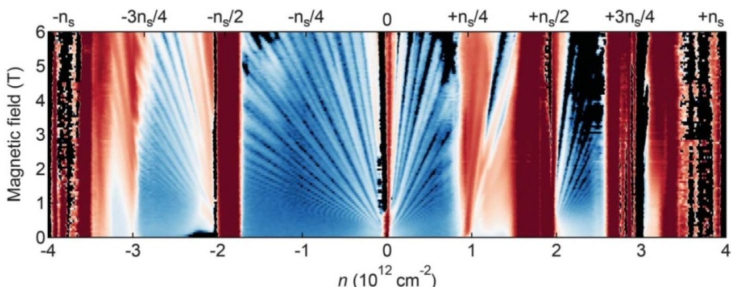
Wagner et al., *PRL* '22

- IKS persists for a large range of dopings at finite strain

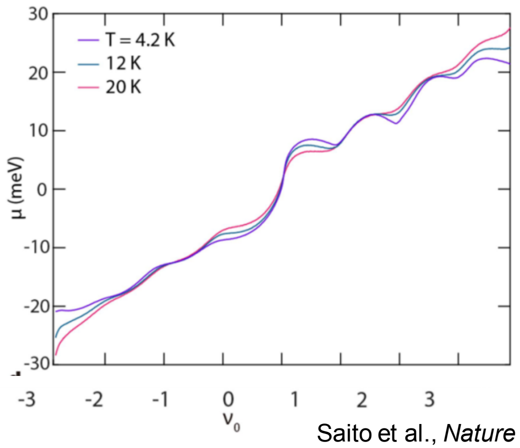


✔ Landau fan degeneracy at finite strain is consistent with experiments
4,-,2,1 degeneracy at fillings 0,1,2,3

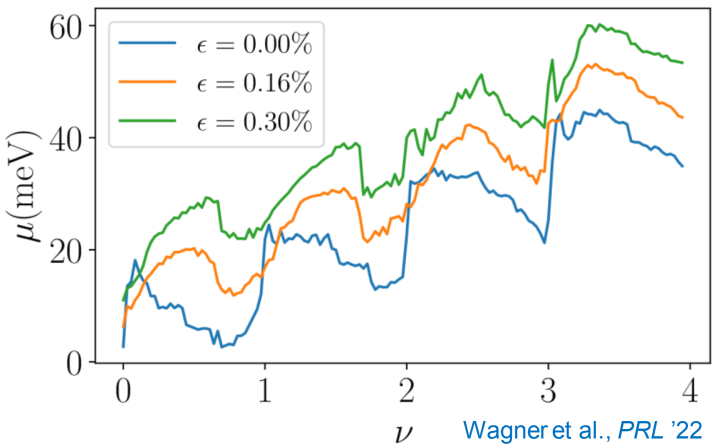
✔ 'cascade' transitions consistent with experiment Zondiner et al., *Nature* '20



Yankowitz et al., *Science* '19



Saito et al., *Nature* '21



Wagner et al., *PRL* '22

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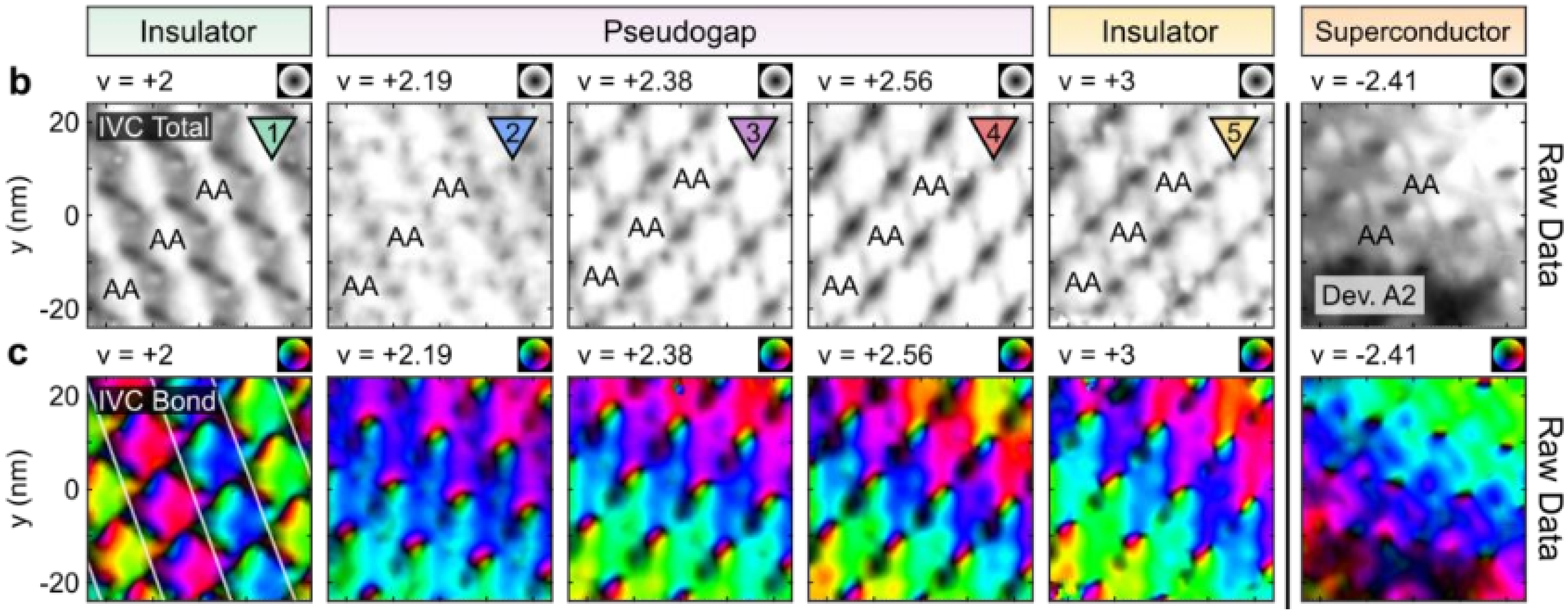
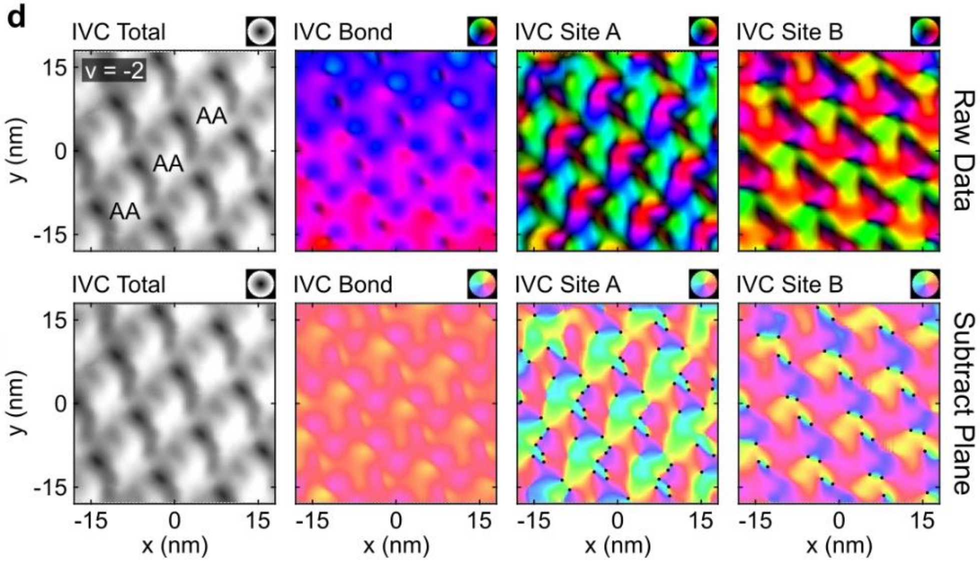


IKS-ish?

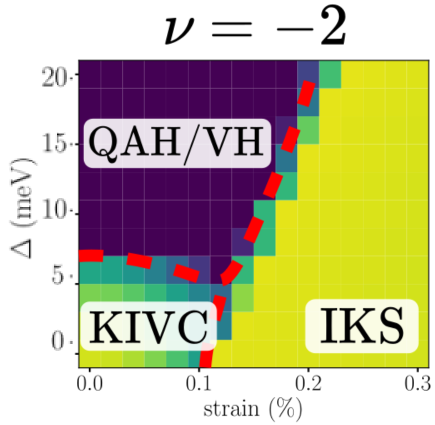
What about highly unstrained samples?

Hartree-Fock says KIVC

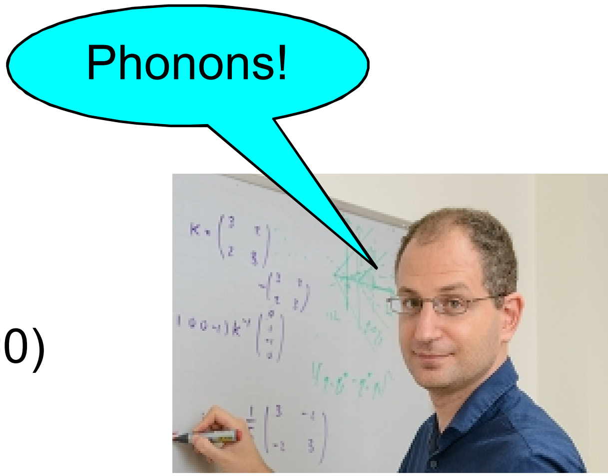
--- should show no Kekulé density pattern



Nature 620, 525 (2023) Nuckolls et al (Yazdani)



But data sees Kekulé density pattern (q=0)



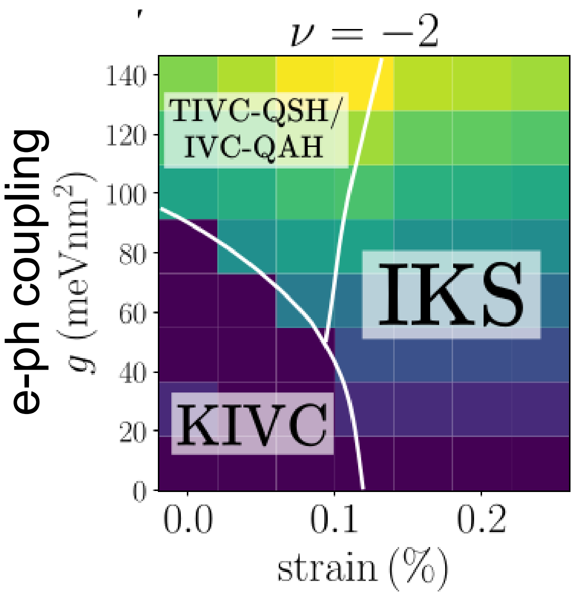
Electron-phonon coupling and competing Kekulé orders in twisted bilayer graphene

Yves H. Kwan,[1] Glenn Wagner,[2] Nick Bultinck,[3,4] Steven H. Simon,[3] Erez Berg,[5] and S.A. Parameswaran[3]

Phys. Rev. B **110**, 085160 (2024)

$$\hat{H} = \epsilon a^\dagger a + \gamma a + \gamma^* a^\dagger \xrightarrow[b^\dagger = a^\dagger + \gamma/\epsilon]{} \hat{H} = \epsilon b^\dagger b - \frac{|\gamma|^2}{\epsilon}$$

An electron state with density modulation lowers total phonon energy.

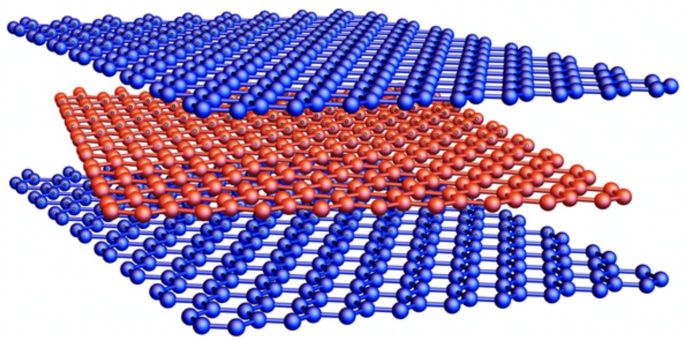


TIVC-QSH $\quad \big(|KA\uparrow\rangle + |\bar{K}B\uparrow\rangle\big)\big(|KB\downarrow\rangle + |\bar{K}A\downarrow\rangle\big)$

IVC-QAH $\quad \big(|KA\uparrow\rangle + |\bar{K}B\uparrow\rangle\big)\big(|KA\downarrow\rangle + |\bar{K}B\downarrow\rangle\big)$

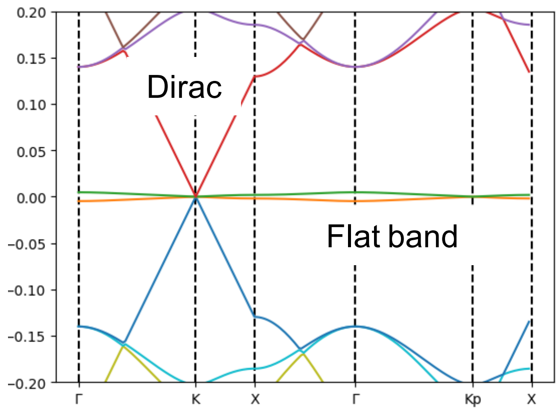
“Imaging inter-valley coherent order in magic-angle twisted *trilayer* graphene”

Kim et al (Nadj-Perge) Nature 693 942, 2023

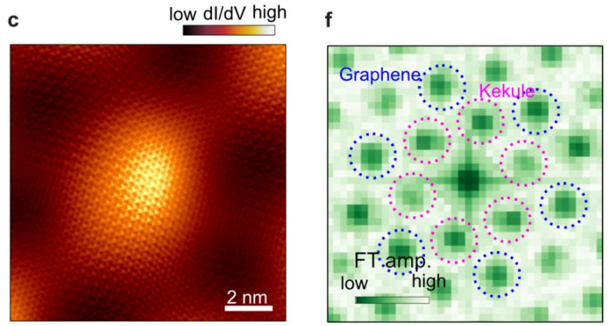


Twisted trilayer graphene: 1st and 3rd layers aligned

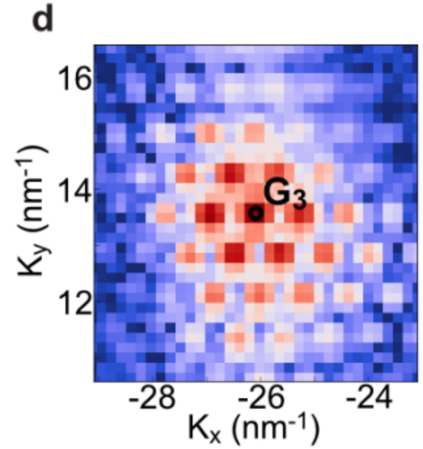
Noninteracting spectrum = $\underbrace{\text{Twisted Bi-Layer graphene}}_{\text{symmetric combination of layers 1 and 3 couples to layer 2.}}$ + $\underbrace{\text{One Dirac Cone left over}}_{\text{antisymmetric combination of layers 1 and 3 decouples}}$

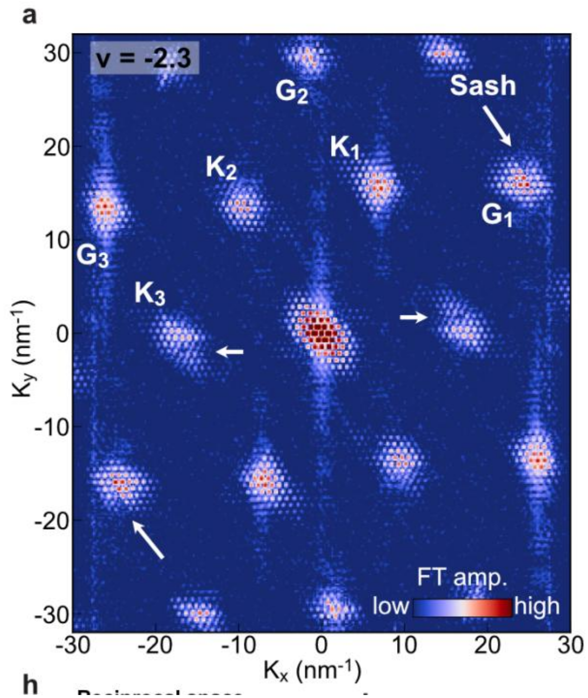


How different is this from bilayer?



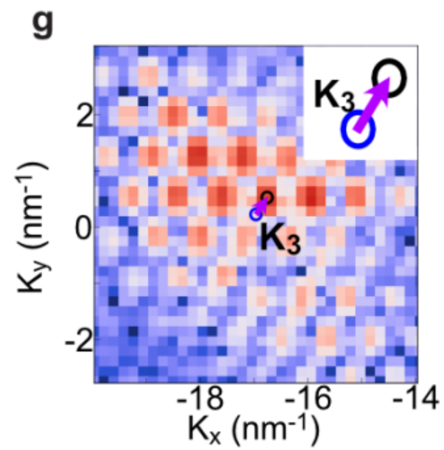
Looking for an additional wavevector q_{IKS}

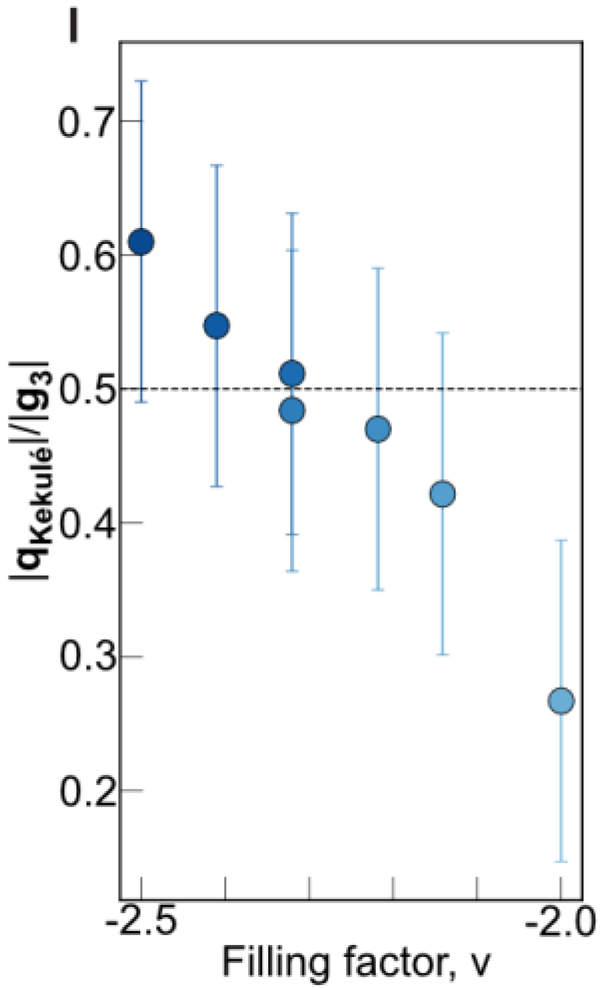


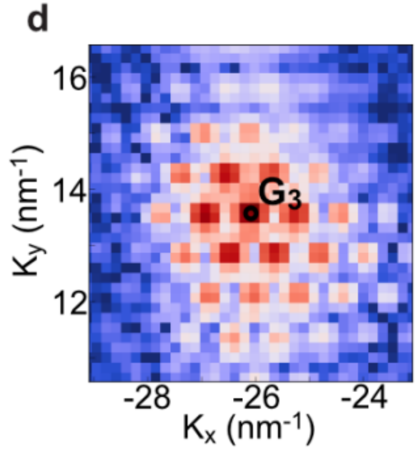


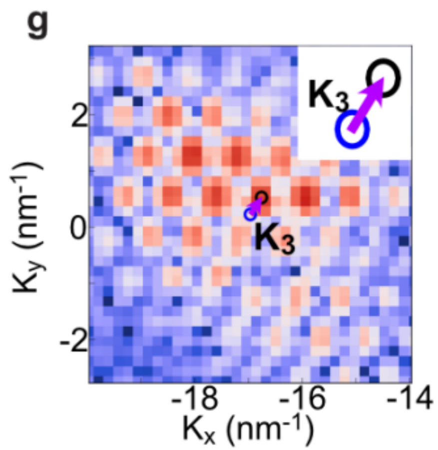
Additional periodicity
From Moire

Displacement of pattern at G compared to pattern at K gives another wavevector!




l
0.7
0.6
0.5
0.4
0.3
0.2
$|q_{Kekulé}|/|g_3|$
-2.5
-2.0
Filling factor, ν


d
16
14
12
K_y (nm^{-1})
G_3
-28
-26
-24
K_x (nm^{-1})


g
2
0
-2
K_y (nm^{-1})
K_3
K_3
-18
-16
-14
K_x (nm^{-1})

Z. Wang et al: Arxiv 2310.16094:

Kekule spirals and charge transfer cascades in twisted symmetric trilayer graphene

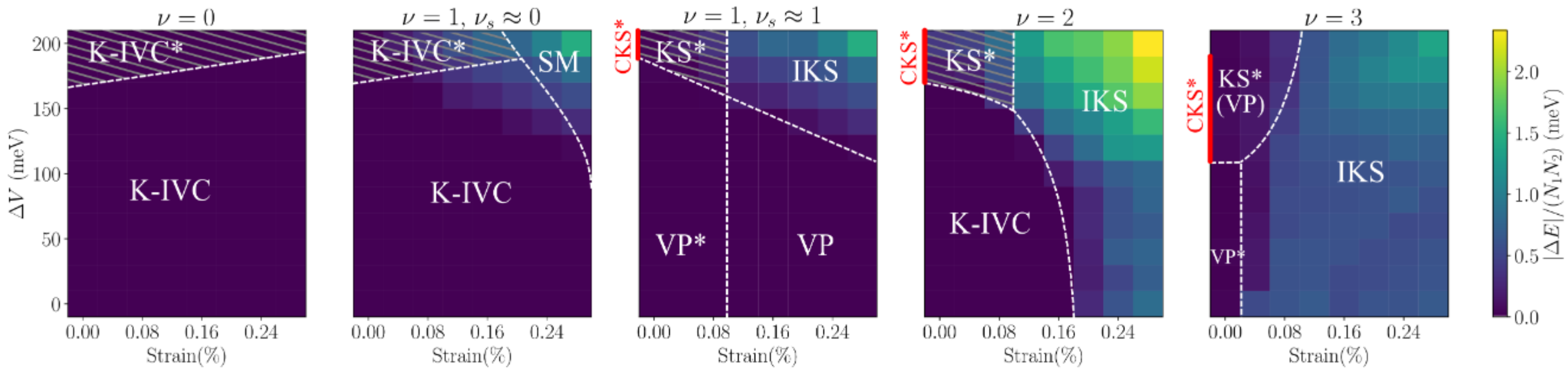


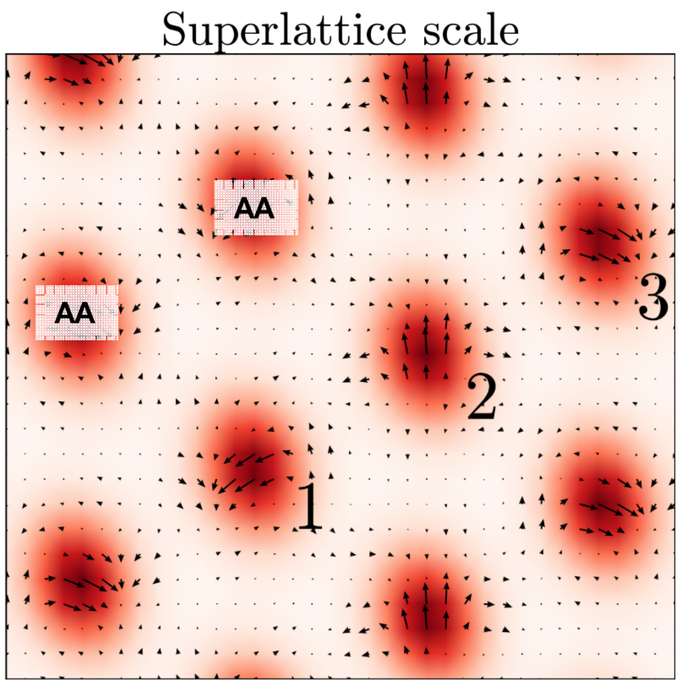
FIG. 2. Phase diagrams as a function of strain and interlayer potential at integer fillings ν. We have performed 10×10 self-consistent Hartree-Fock calculations, scanning over all possible Kekulé spiral vectors $\boldsymbol{q}$. ΔE is the energy difference between the $\boldsymbol{q} = 0$ state with the overall energy minimum. The acronyms are: K-IVC for Kramers-intervalley coherence, VP for valley polarization, (I)KS for (incommensurate) Kekulé spiral, SM for (compensated) semi-metal. Asterisk (*) denotes states that break C_2T-symmetry. The KS*(VP) region in $\nu = 3$ consists of Kekulé spiral states with possible finite valley polarization. The red lines on the zero-strain axis indicate *commensurate* Kekulé spirals. The shaded regions have a finite charge gap. For $\nu = 1$, we have shown the phases and charge gaps of the two spin sectors separately, where ν_s denotes the filling of each spin sector. As ΔE cannot be separately defined for each spin sector, the data are simply duplicated in the two plots of $\nu = 1$.

Summary:

IKS: Incommensurate **K**ekulé **S**piral

It is there in “typical” samples

Explains a lot.... how much more will it explain?



Kekulé Spirals in Twisted Bilayer Graphene

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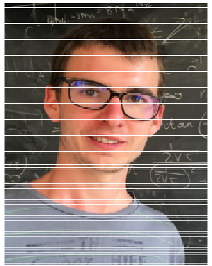
Thank you for listening!

Yves Kwan

→ Princeton

Glenn Wagner



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Kwan et al PRX **11,** 041063 (2021); arXiv:2303.13602; PRX 12, 031020 (2022); ...
Wagner et al, PRL **128**, 156401 (2022). Wang et al, arxiv 2310.16094