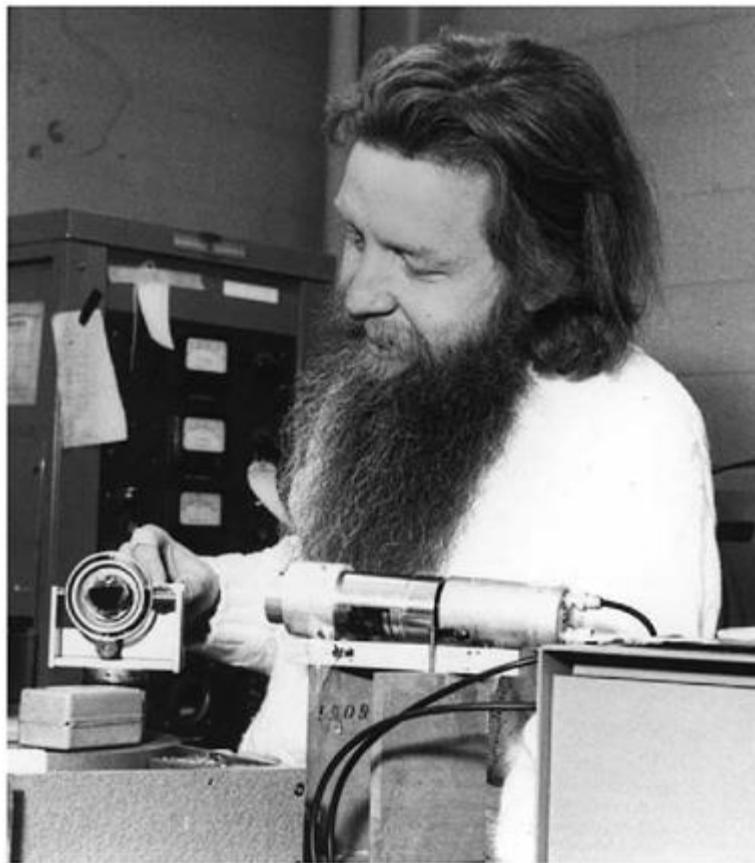


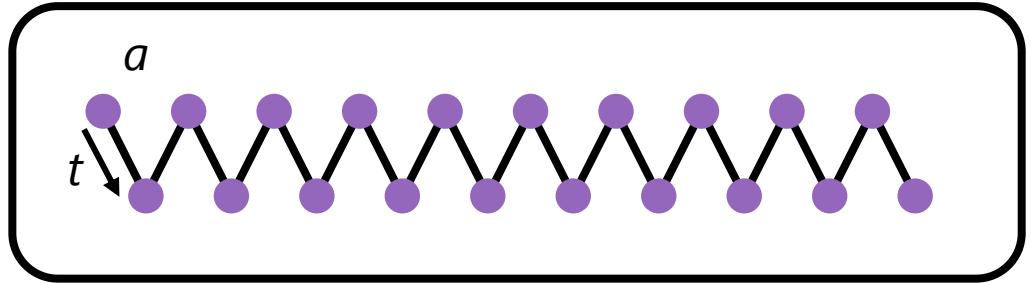
Making Carbon Magnetic

Aaron Sharpe
Bill McMillan Colloquium
October 2nd, 2024

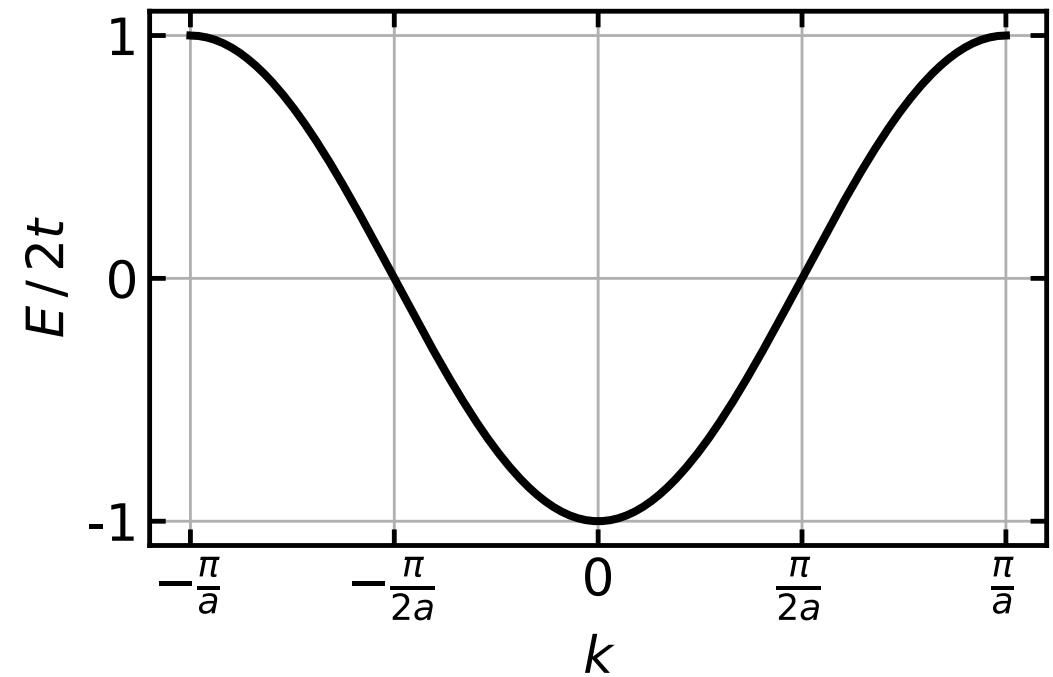
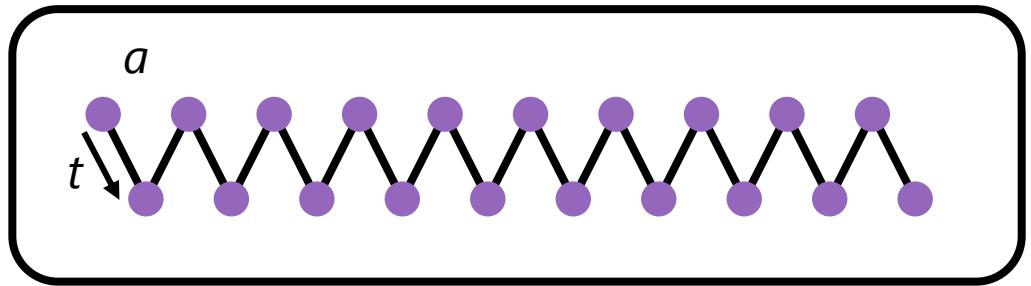
Slides available @ aaronsharpe.science



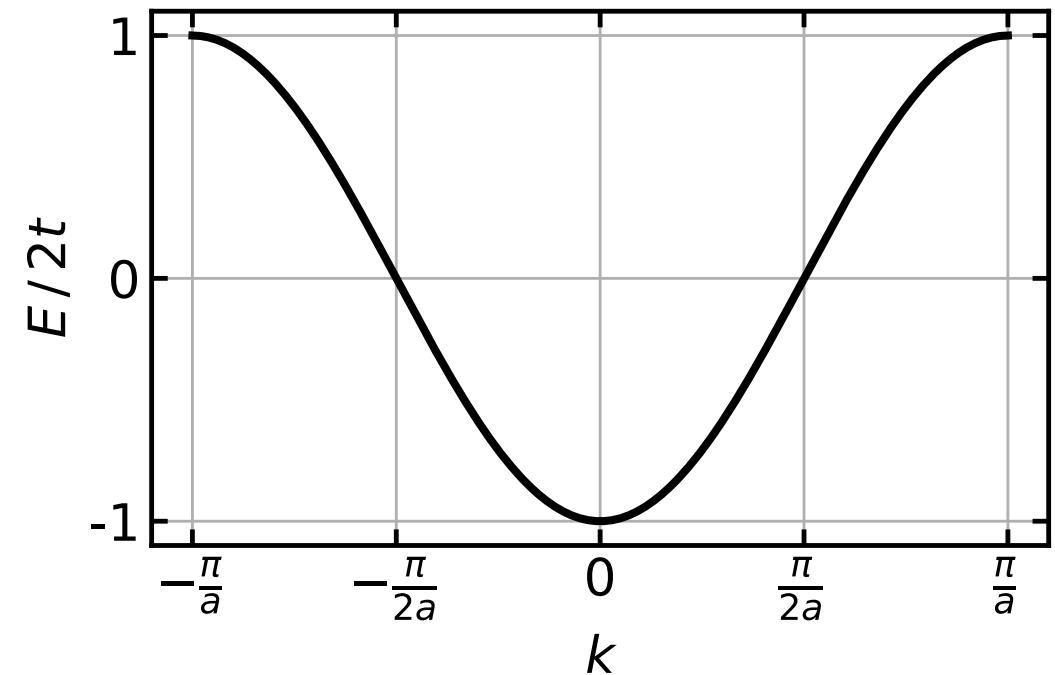
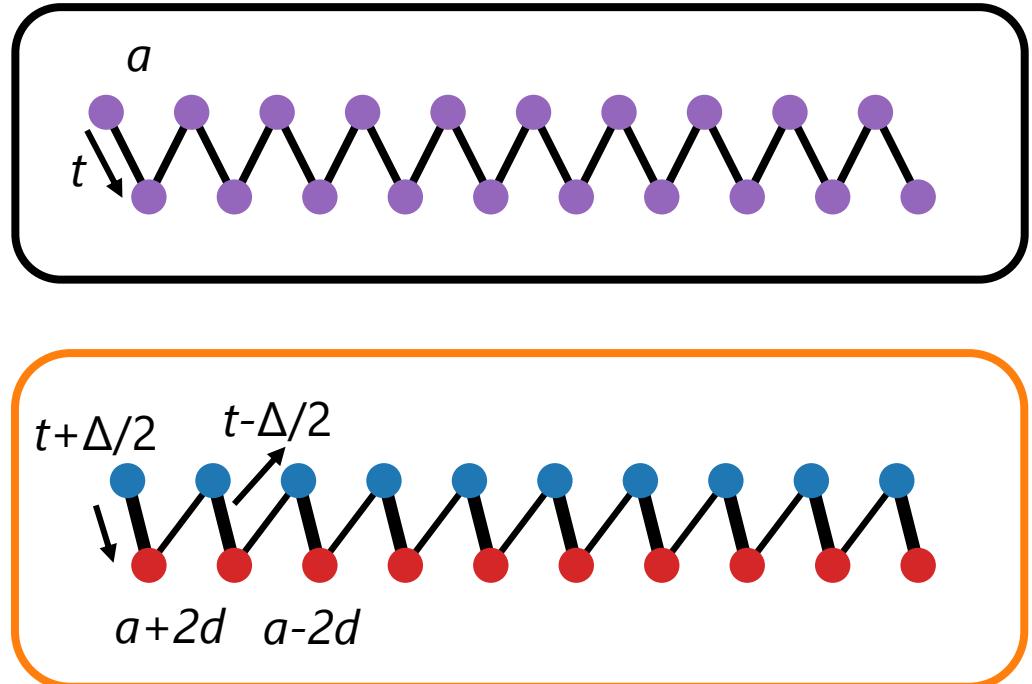
Zig-Zag Chain



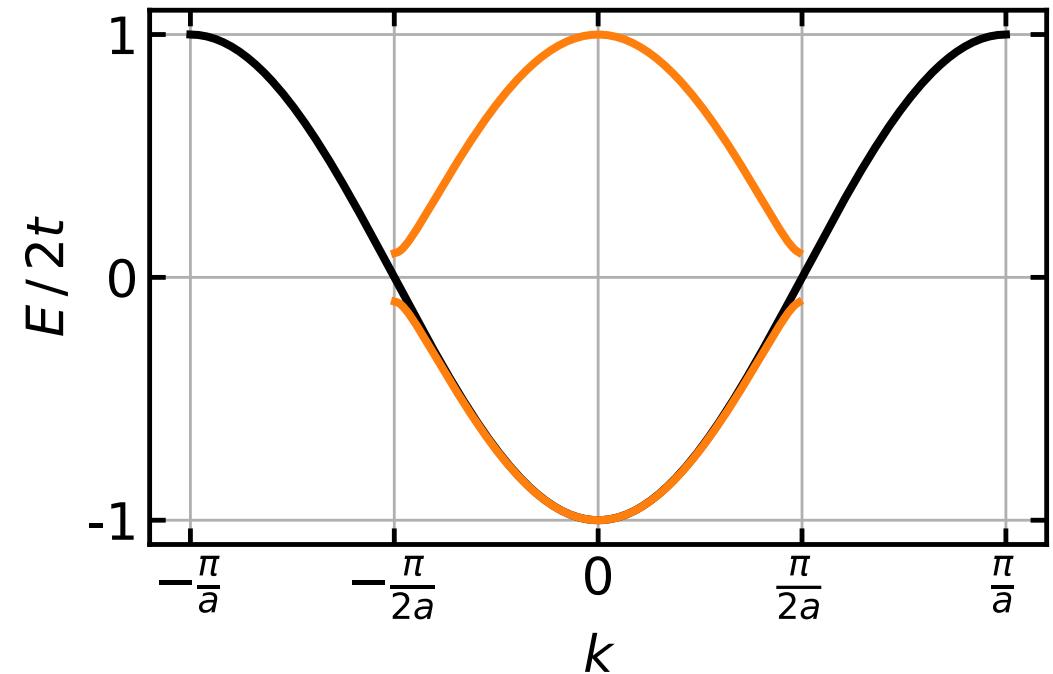
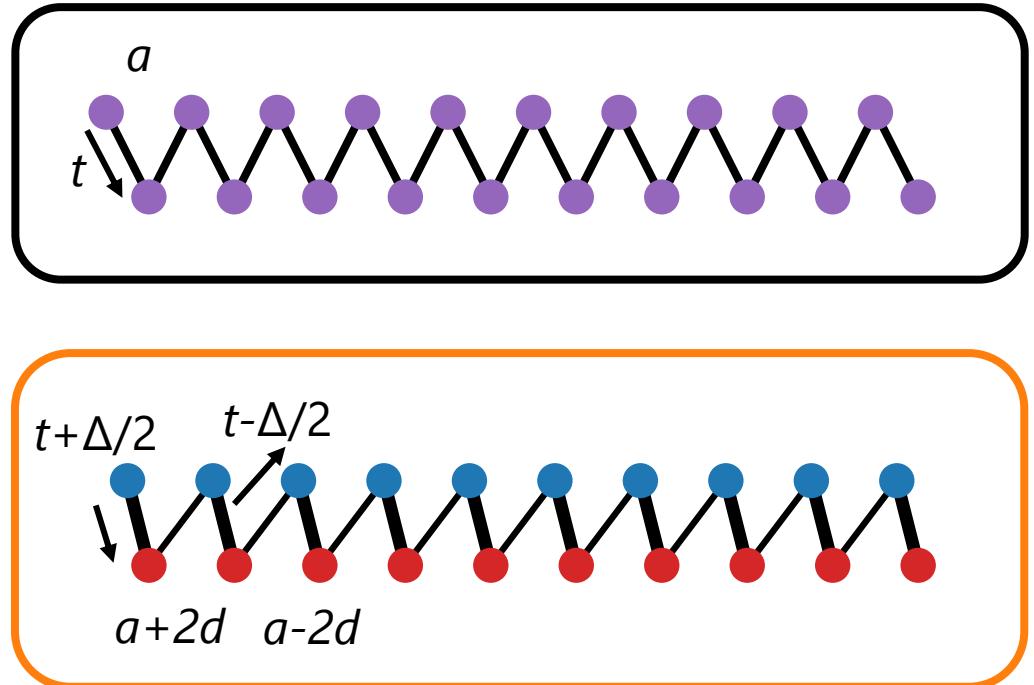
Zig-Zag Chain



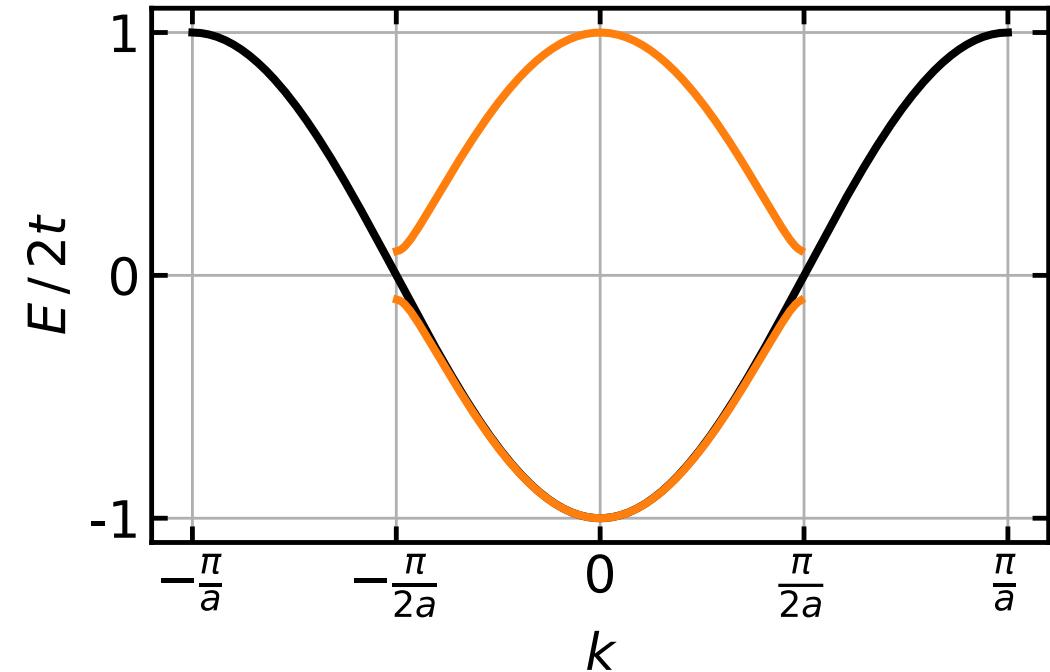
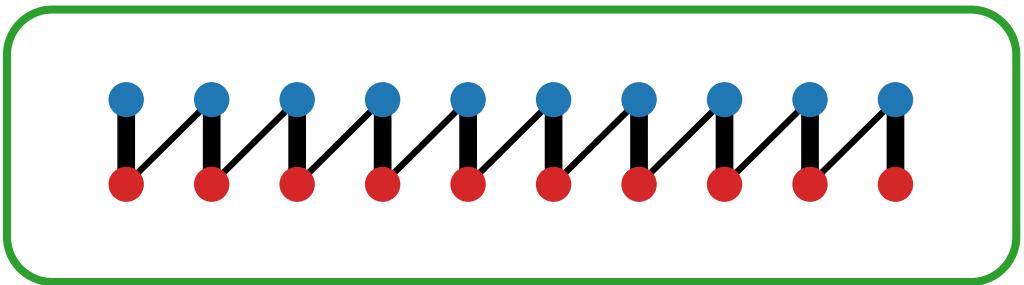
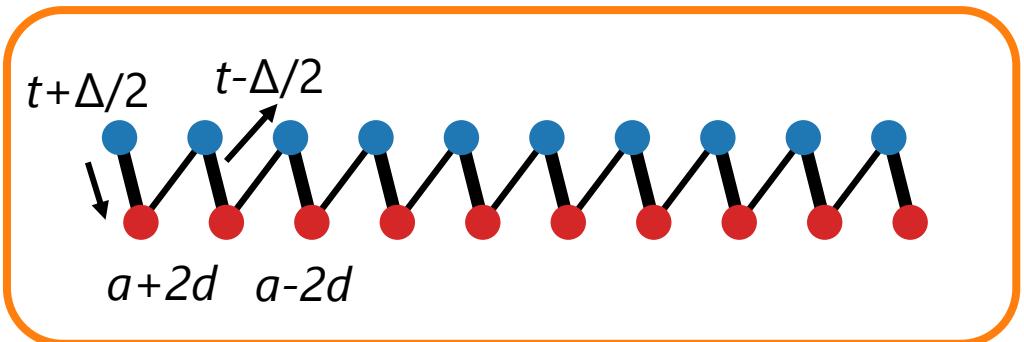
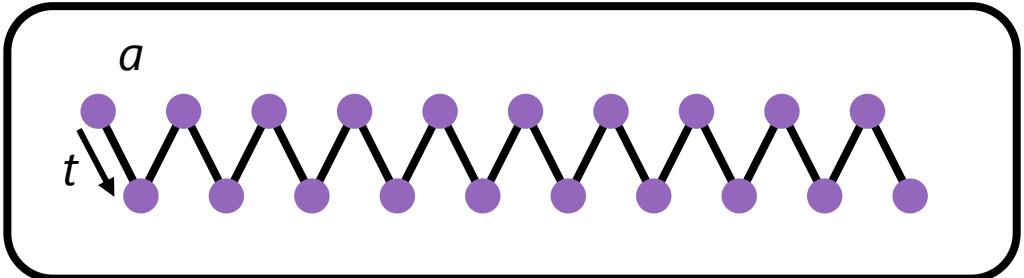
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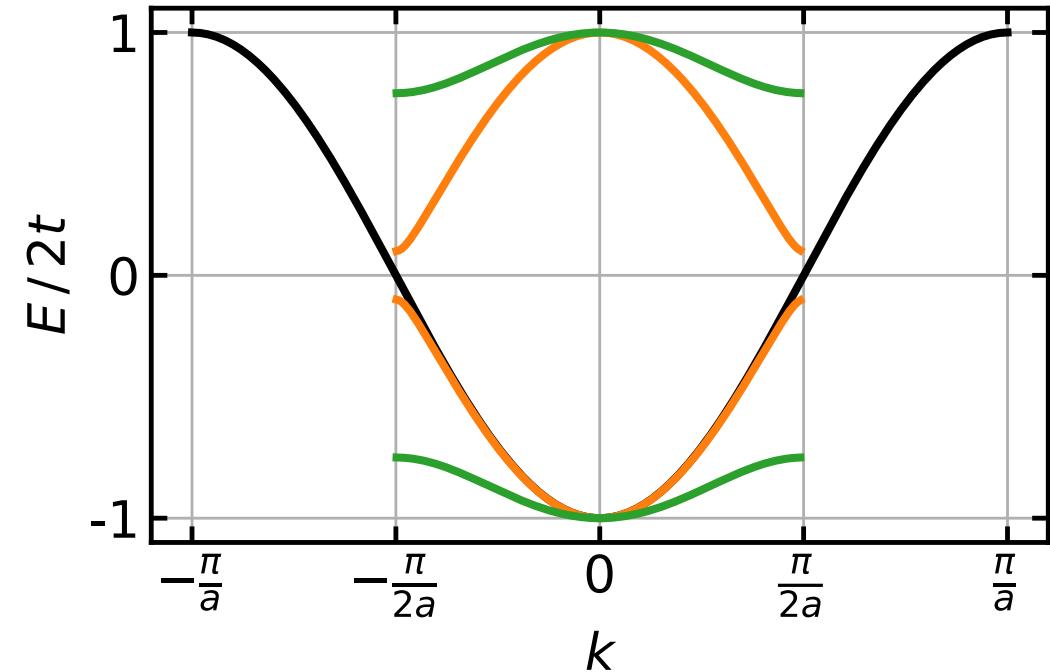
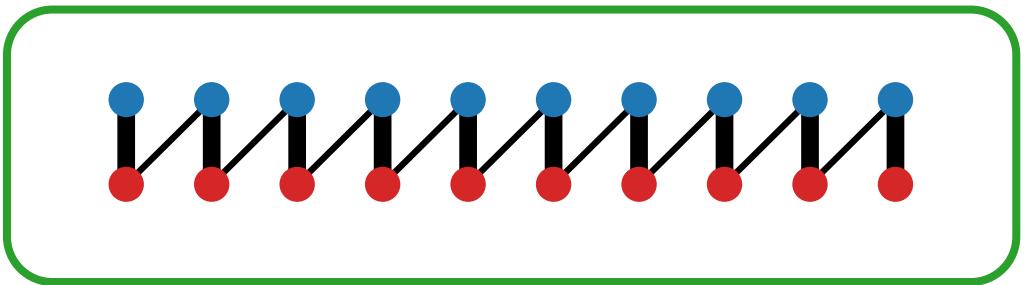
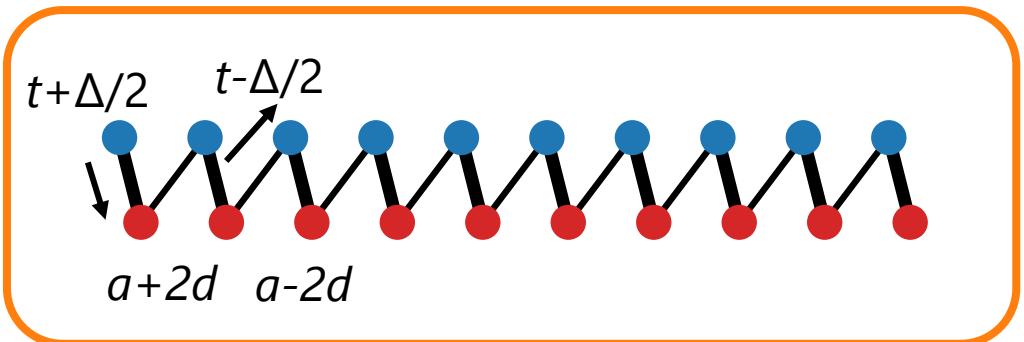
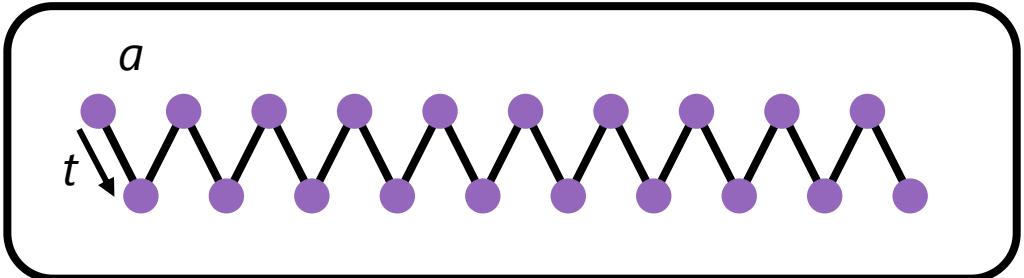
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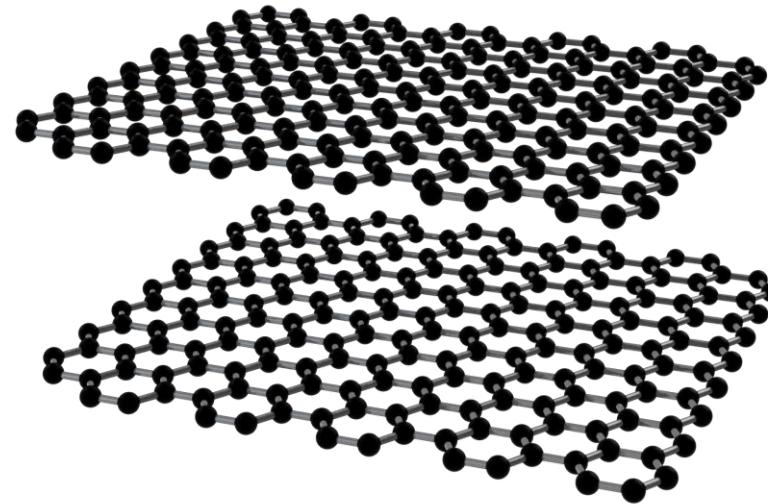
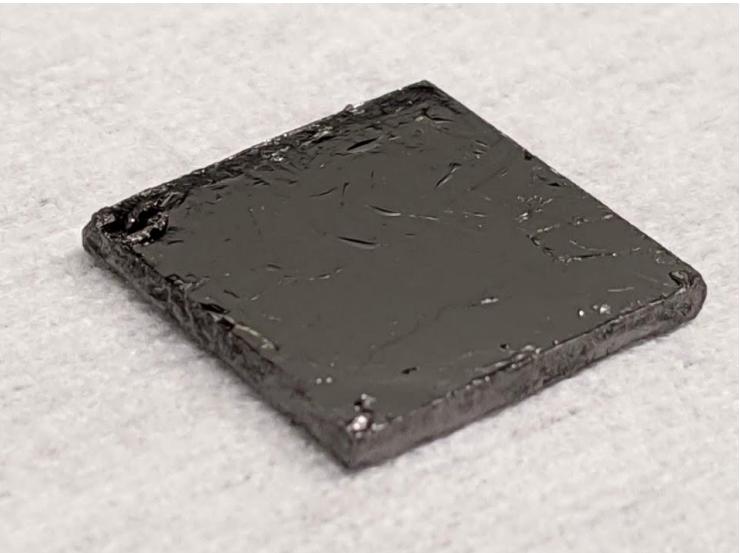
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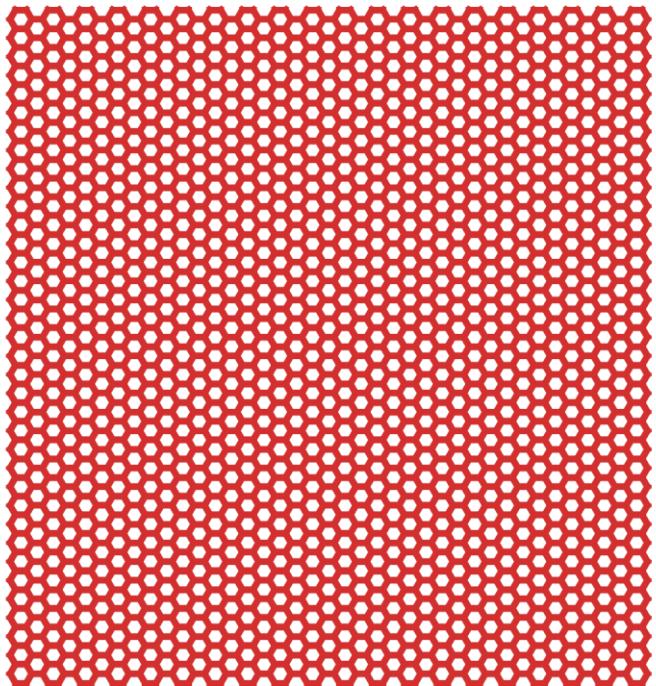
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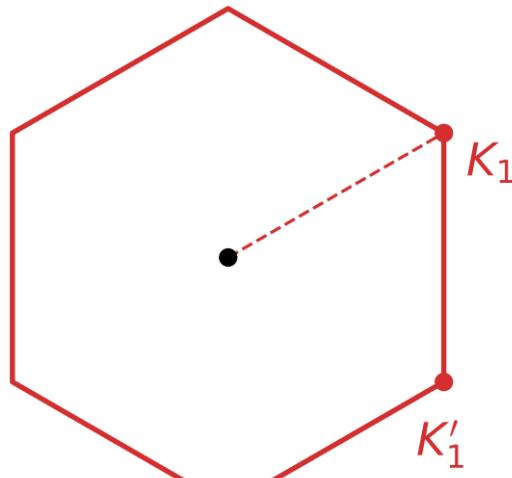
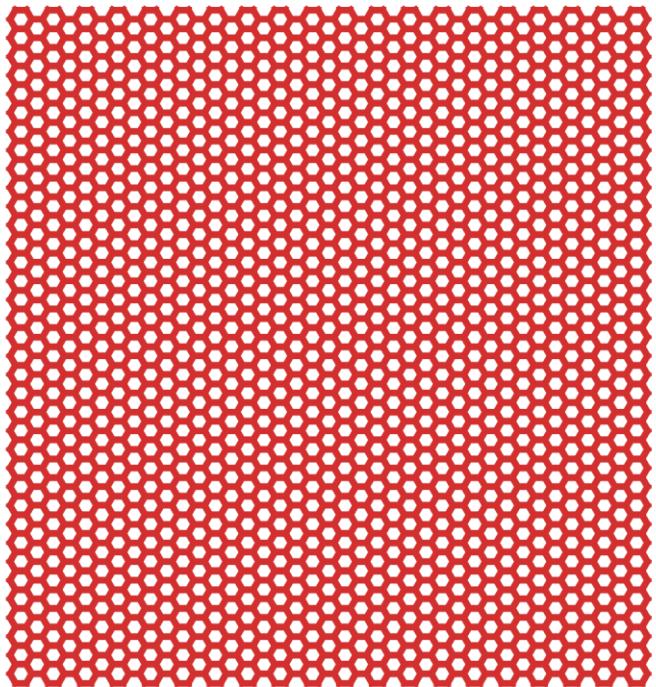
Graphene



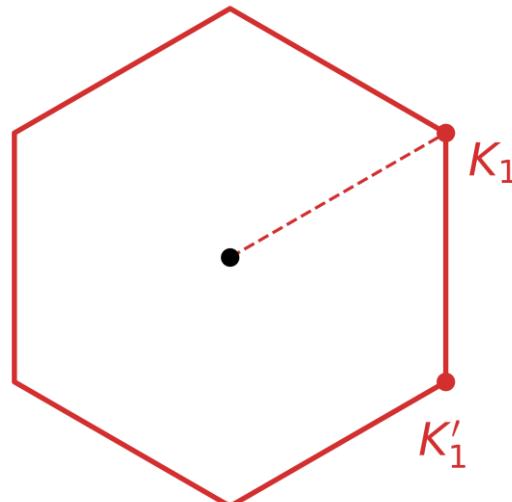
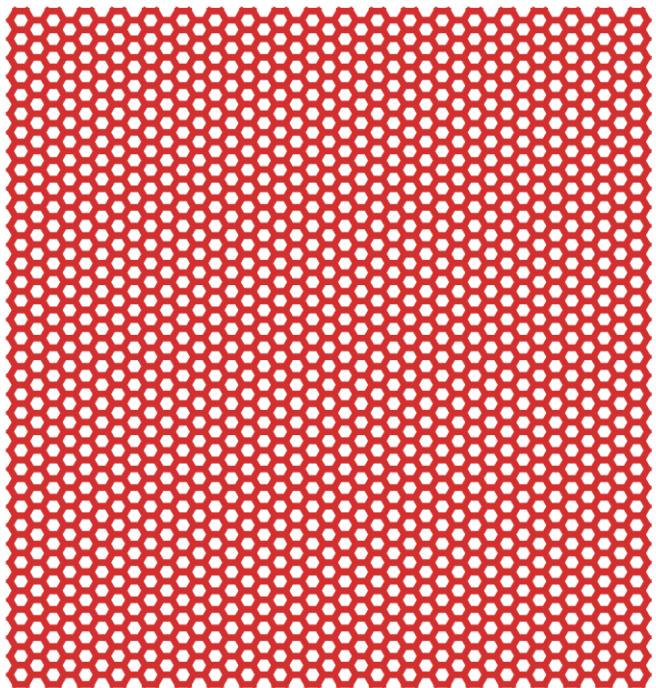
Bandstructure engineering with moirés



Bandstructure engineering with moirés

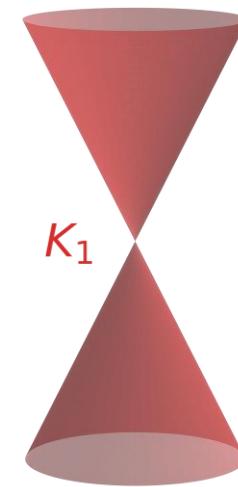
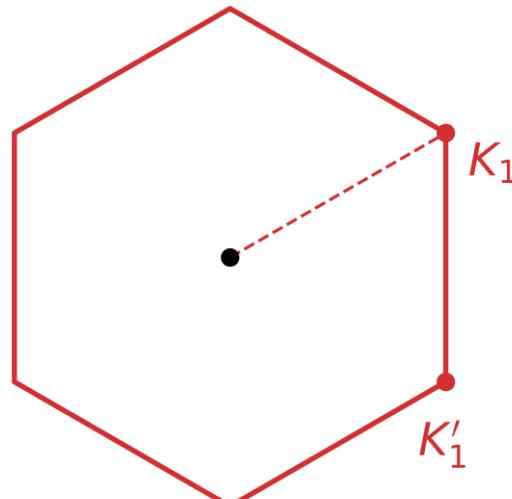
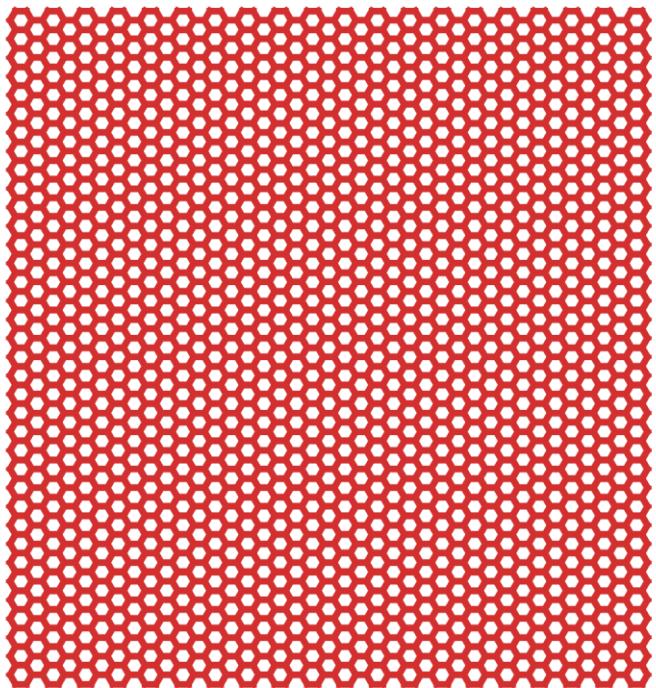


Bandstructure engineering with moirés



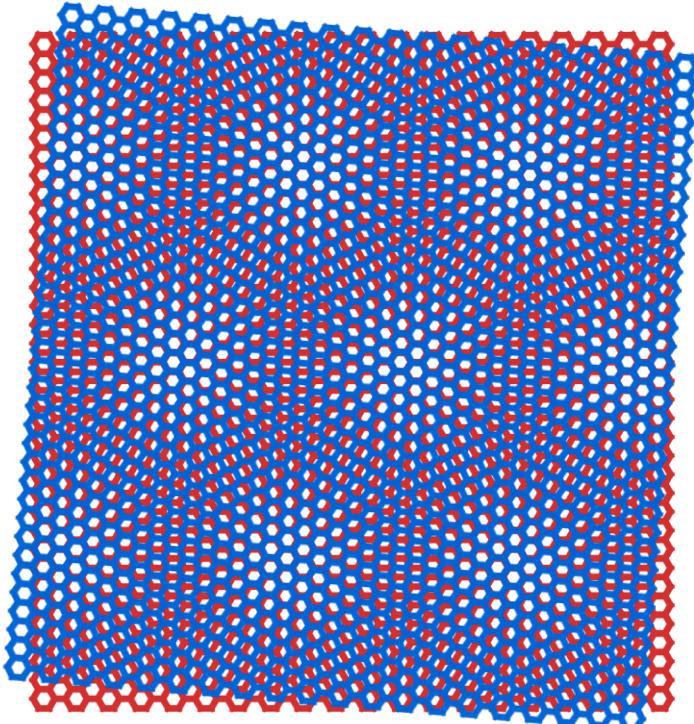
4-fold degenerate
spin x valley

Bandstructure engineering with moirés

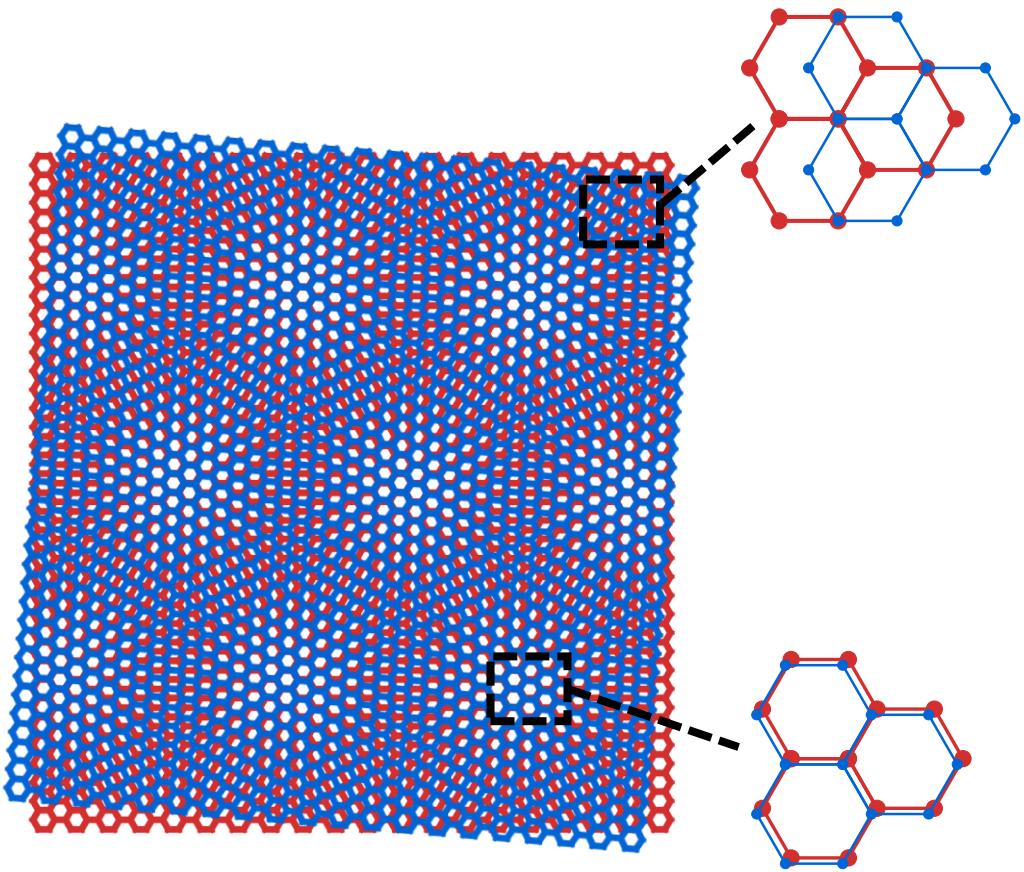


4-fold degenerate
spin x valley

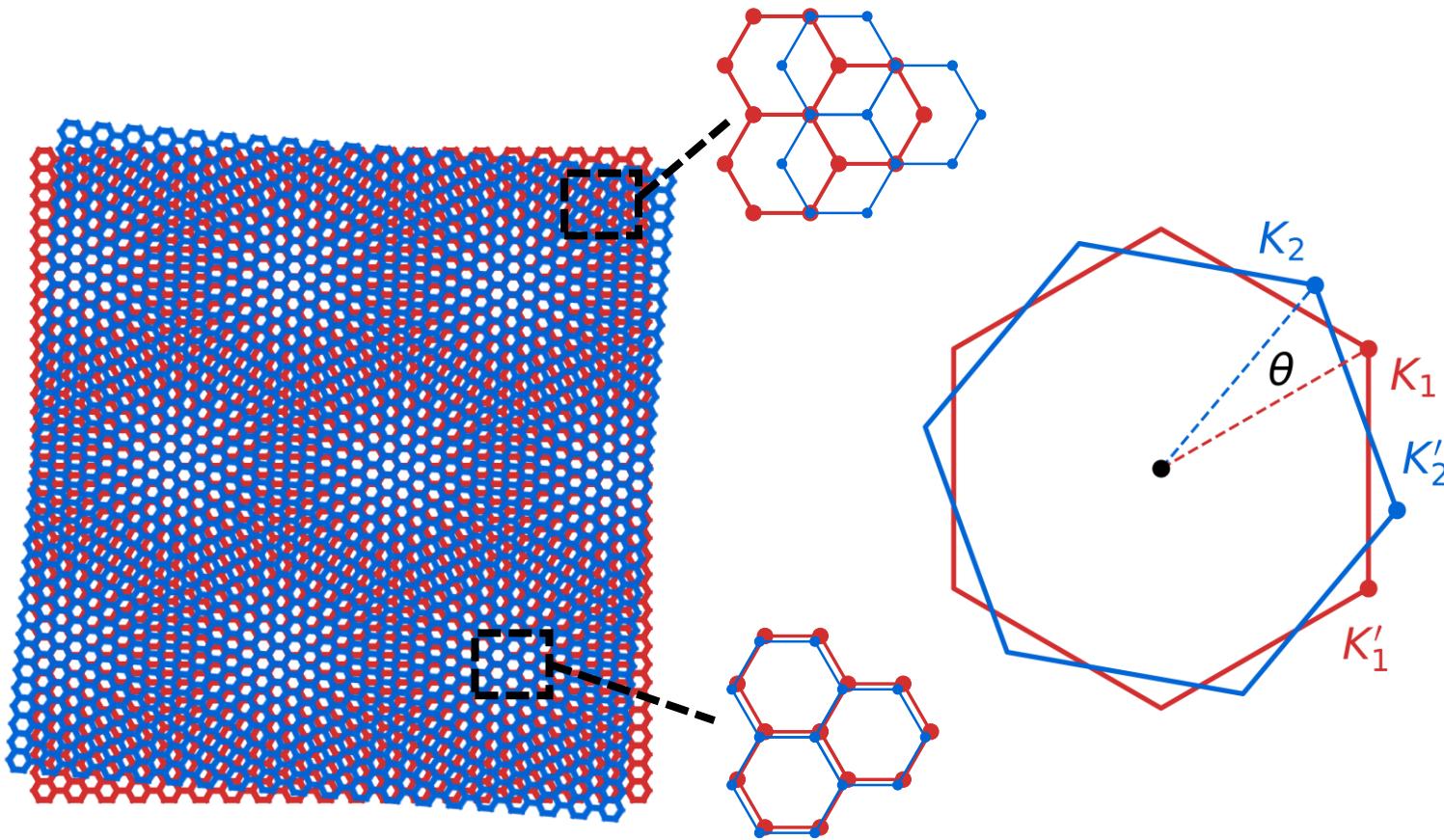
Bandstructure engineering with moirés



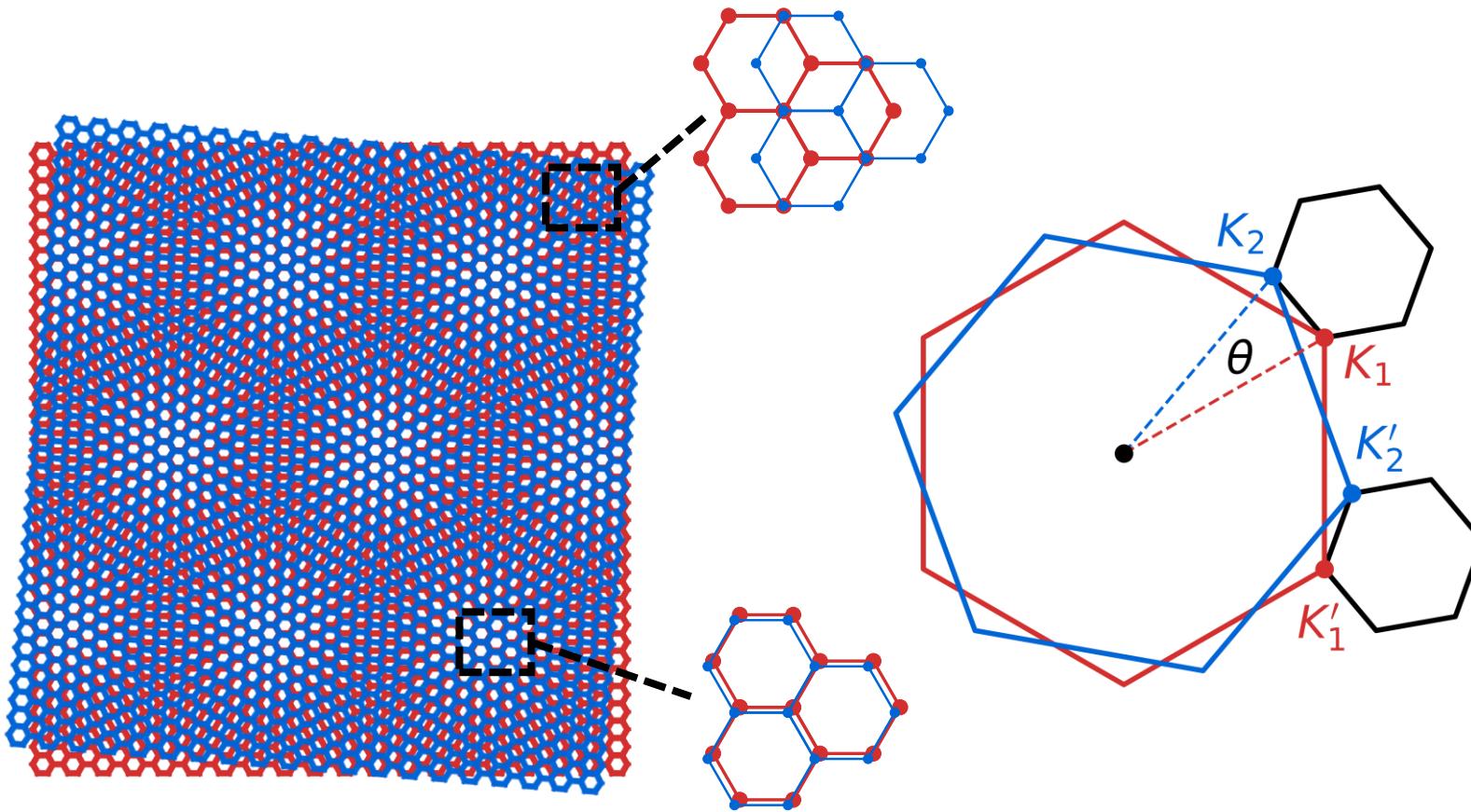
Bandstructure engineering with moirés



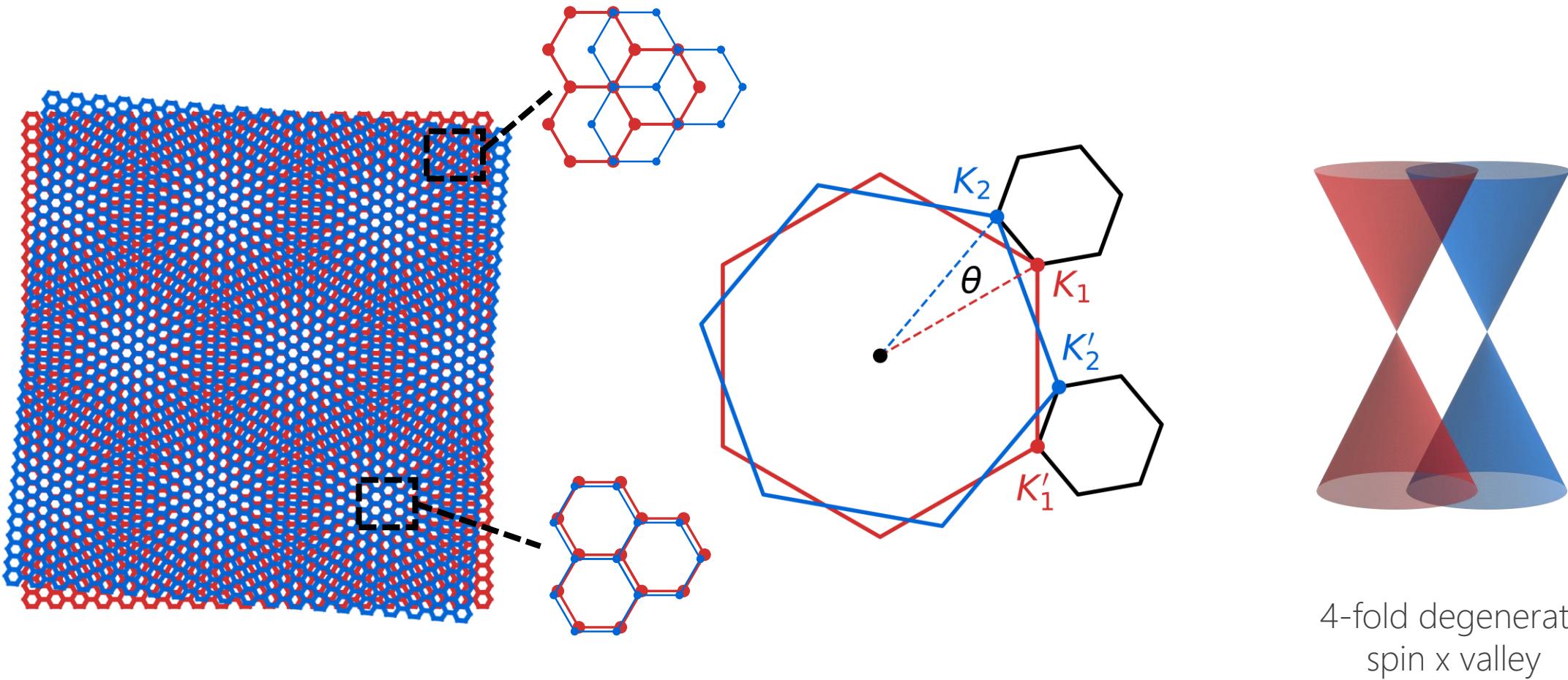
Bandstructure engineering with moirés



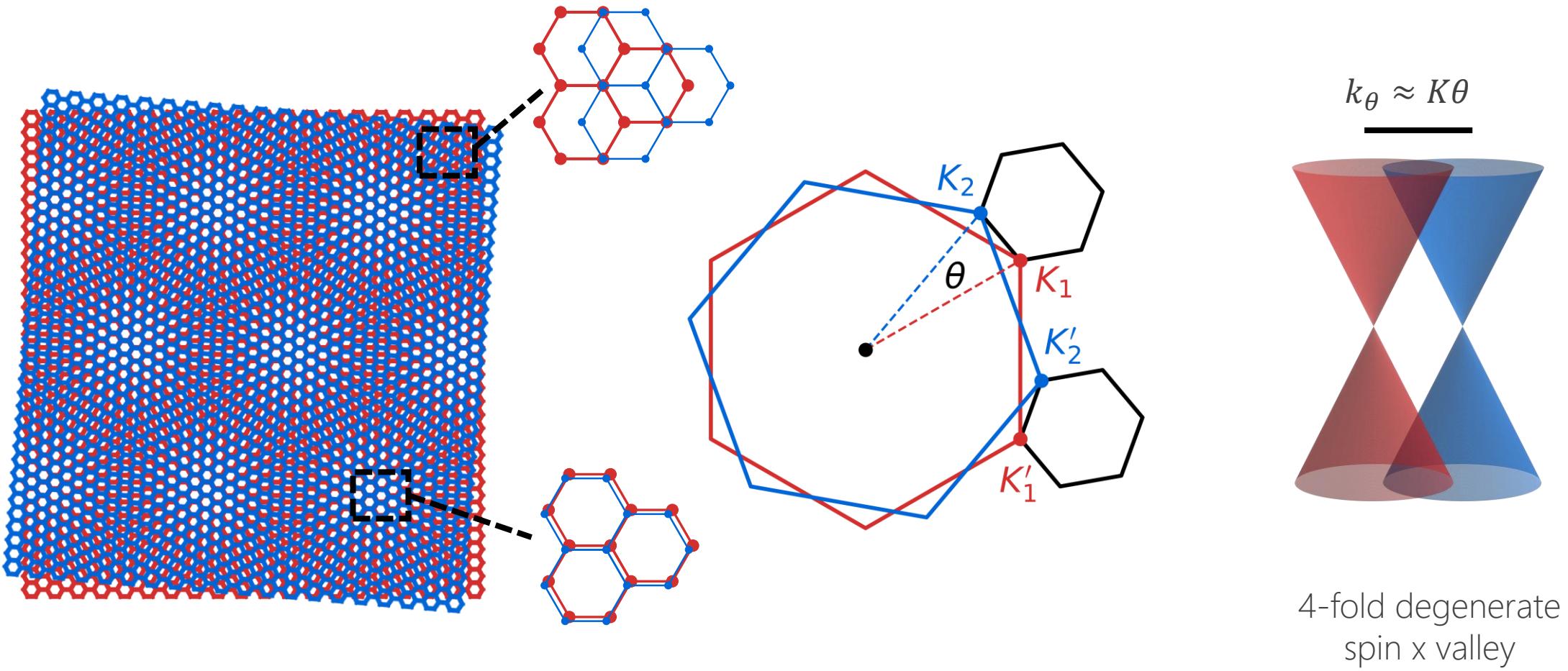
Bandstructure engineering with moirés



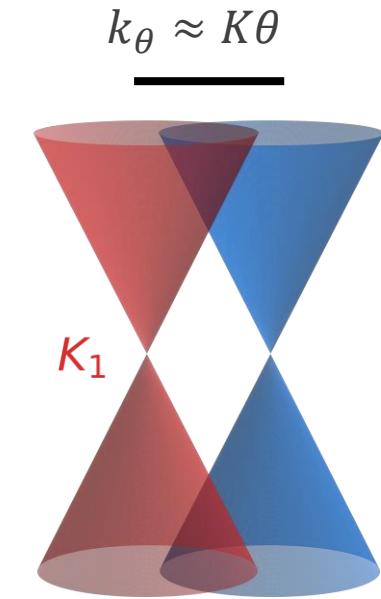
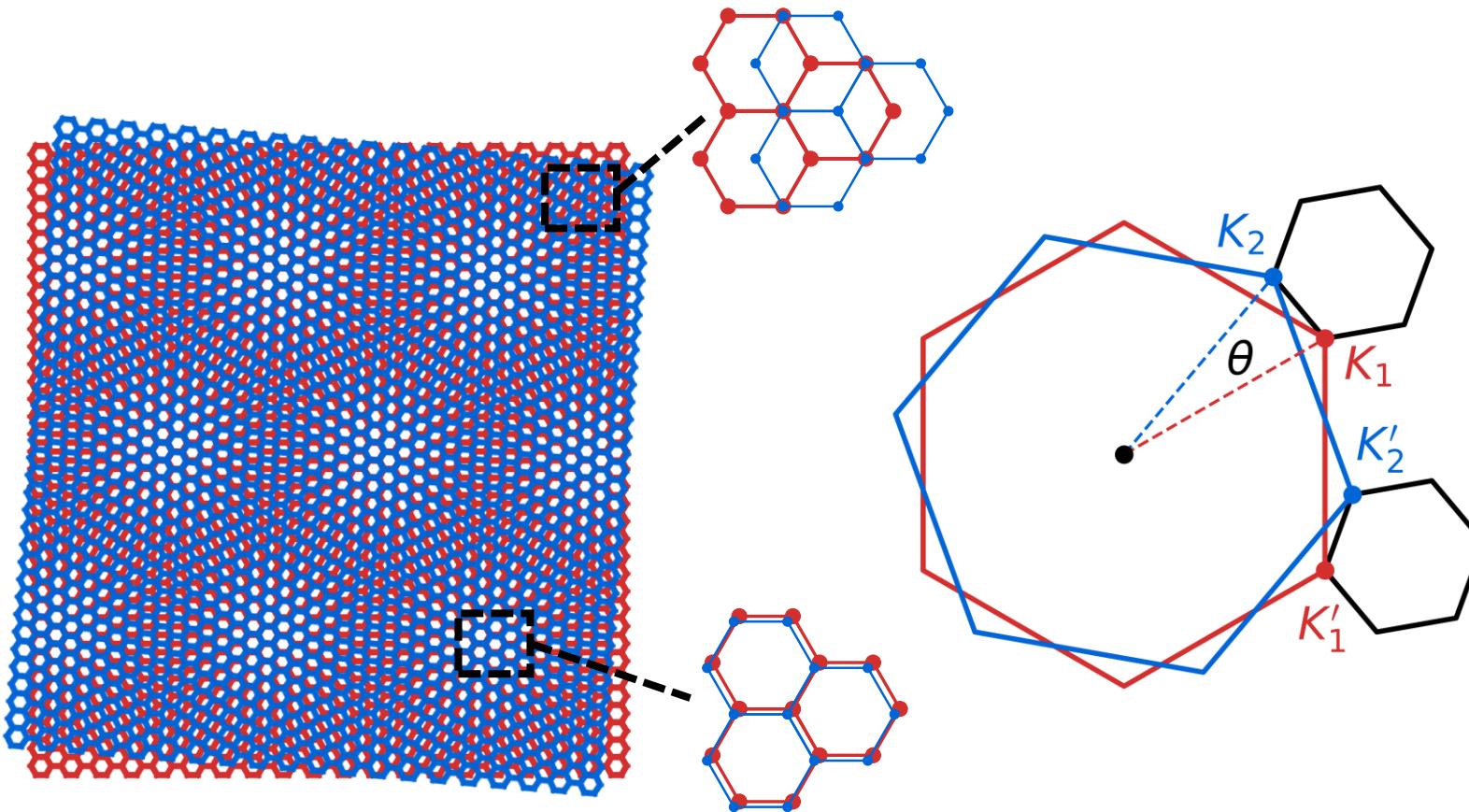
Bandstructure engineering with moirés



Bandstructure engineering with moirés



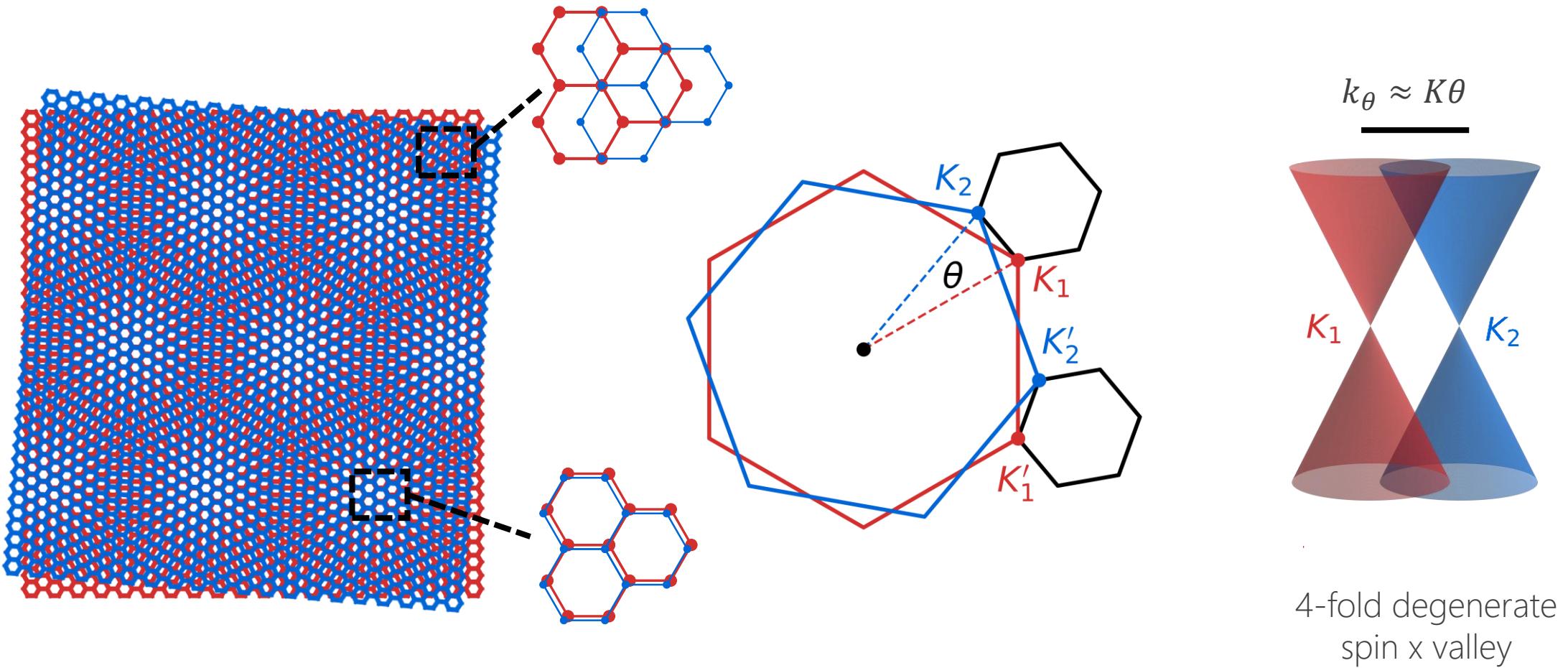
Bandstructure engineering with moirés



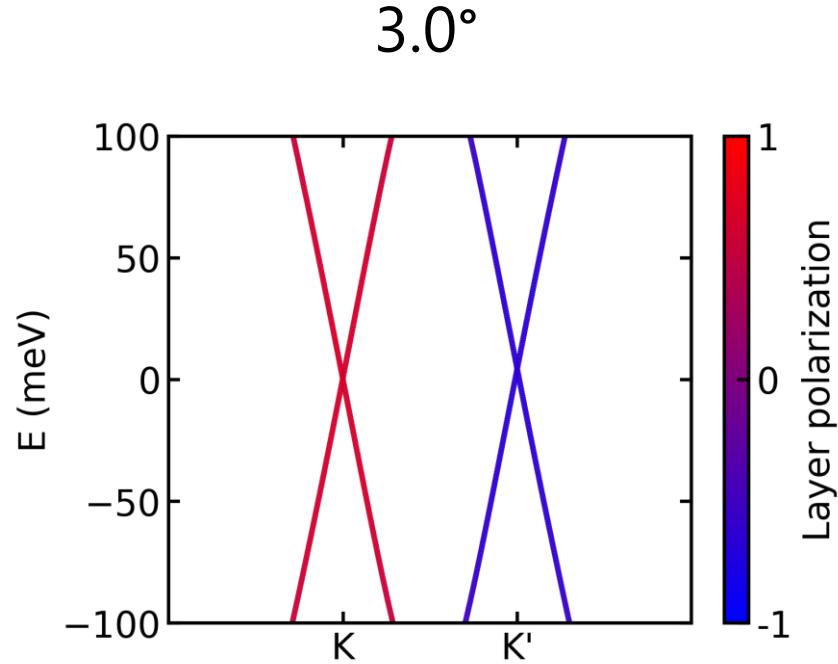
$k_\theta \approx K\theta$

4-fold degenerate
spin x valley

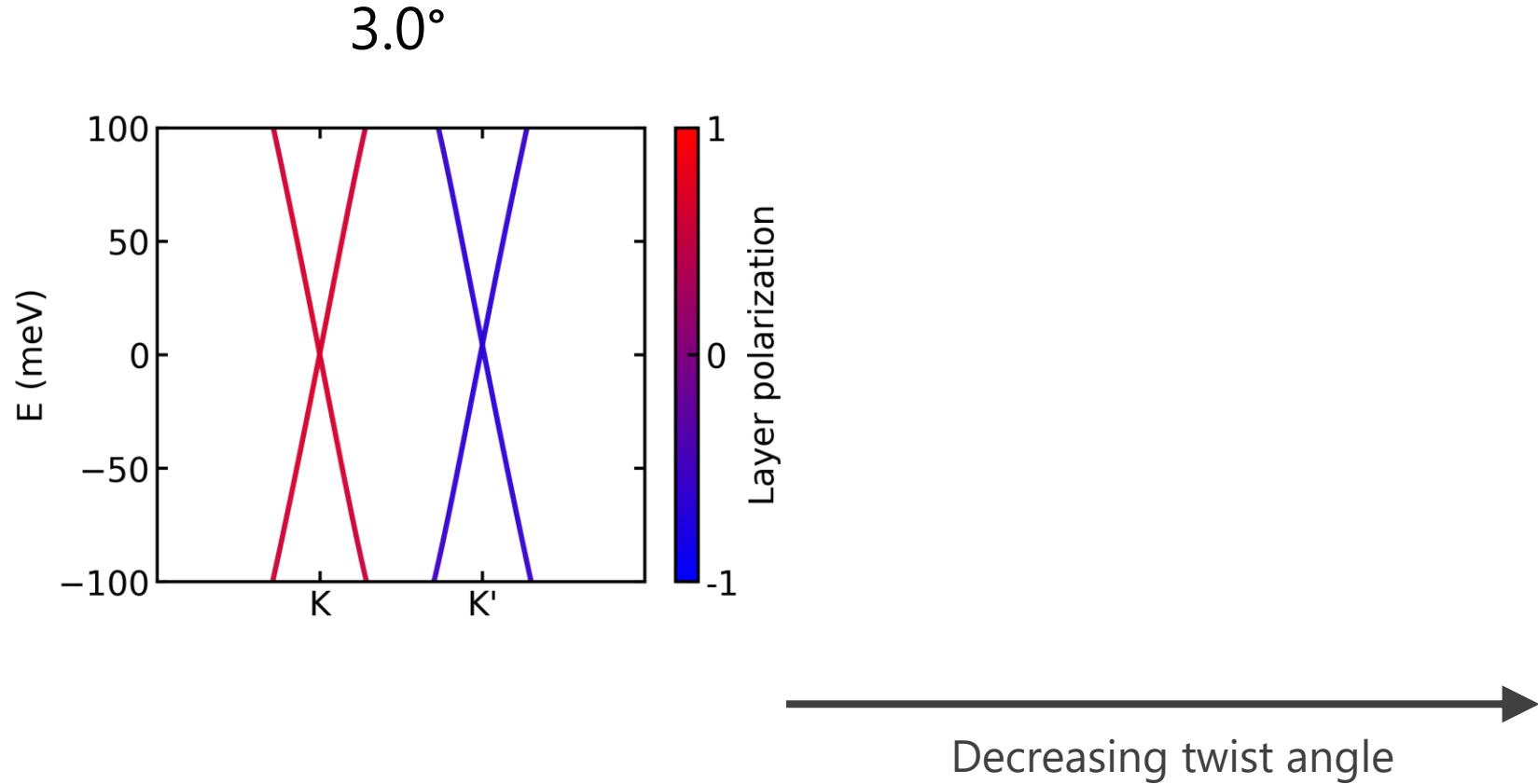
Bandstructure engineering with moirés



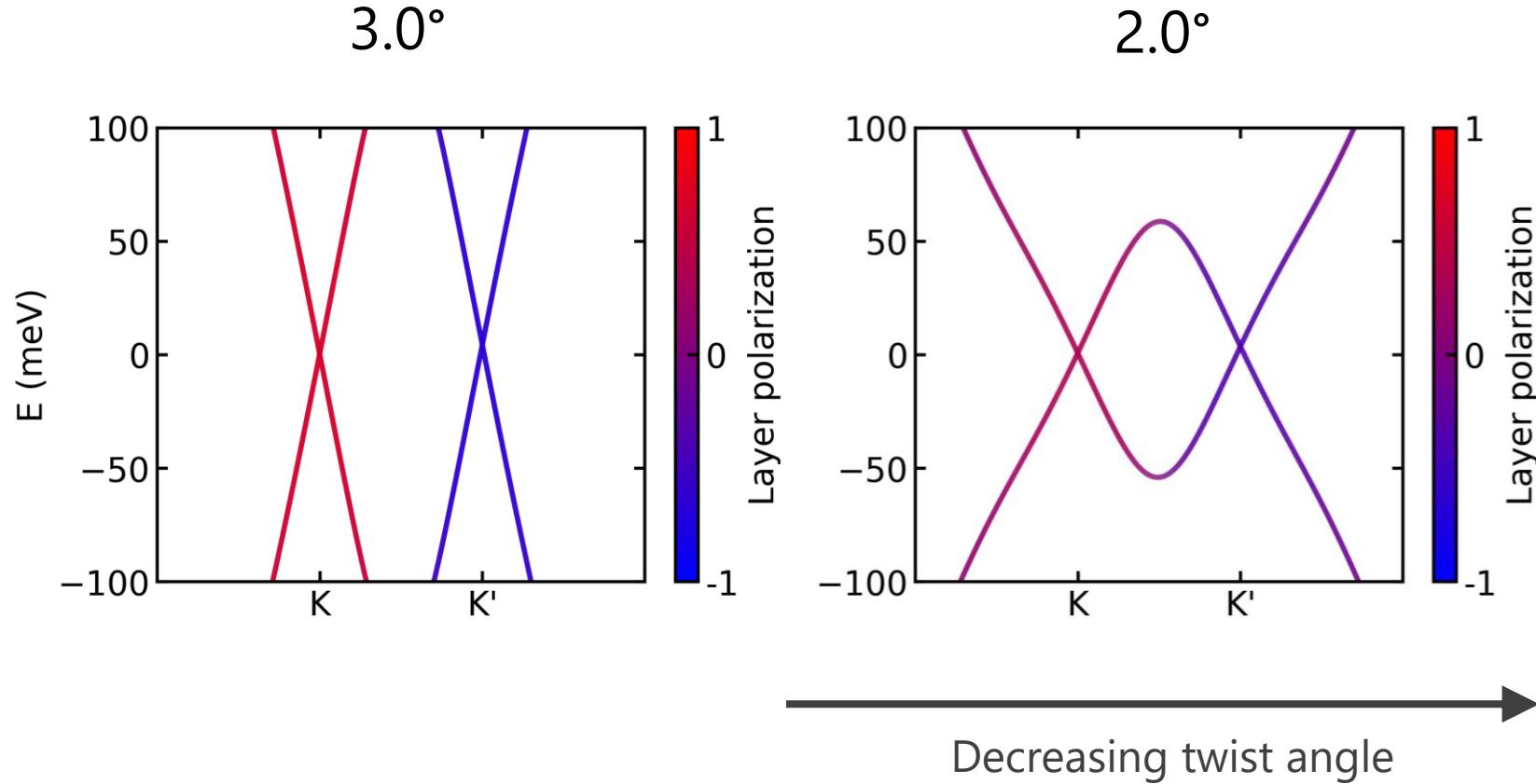
Bandstructure engineering with moirés



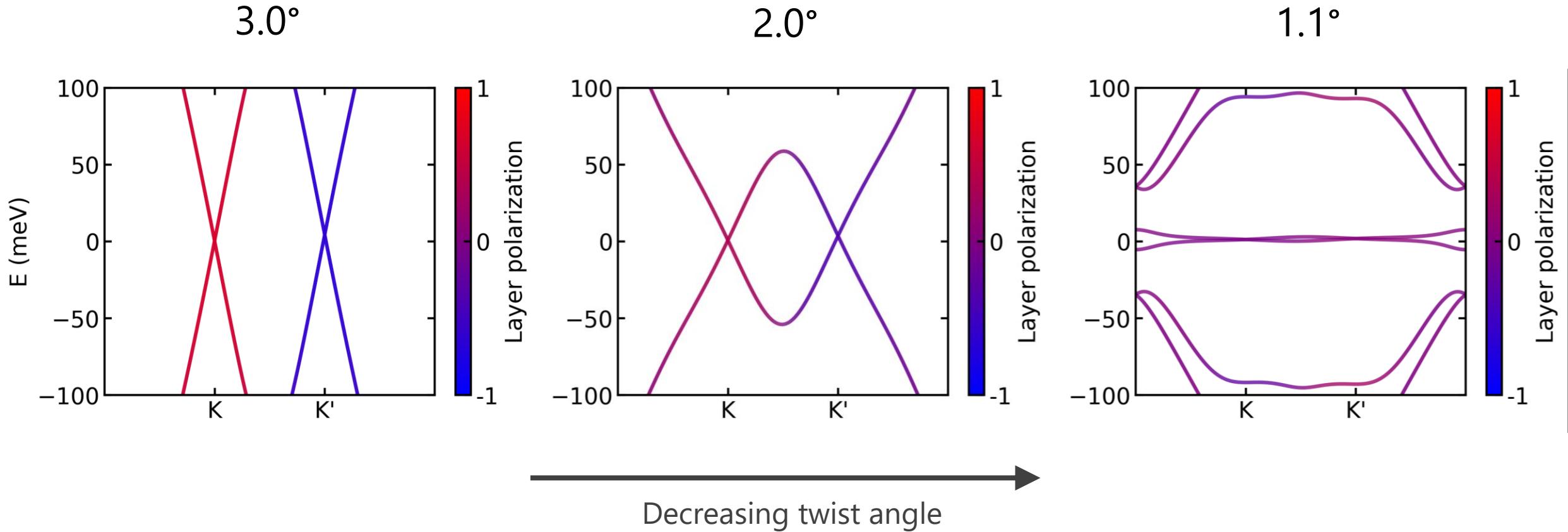
Bandstructure engineering with moirés



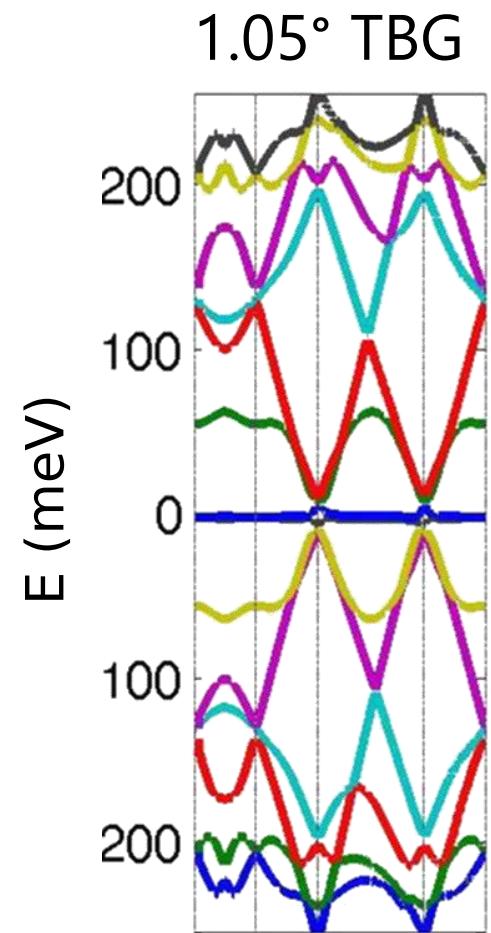
Bandstructure engineering with moirés



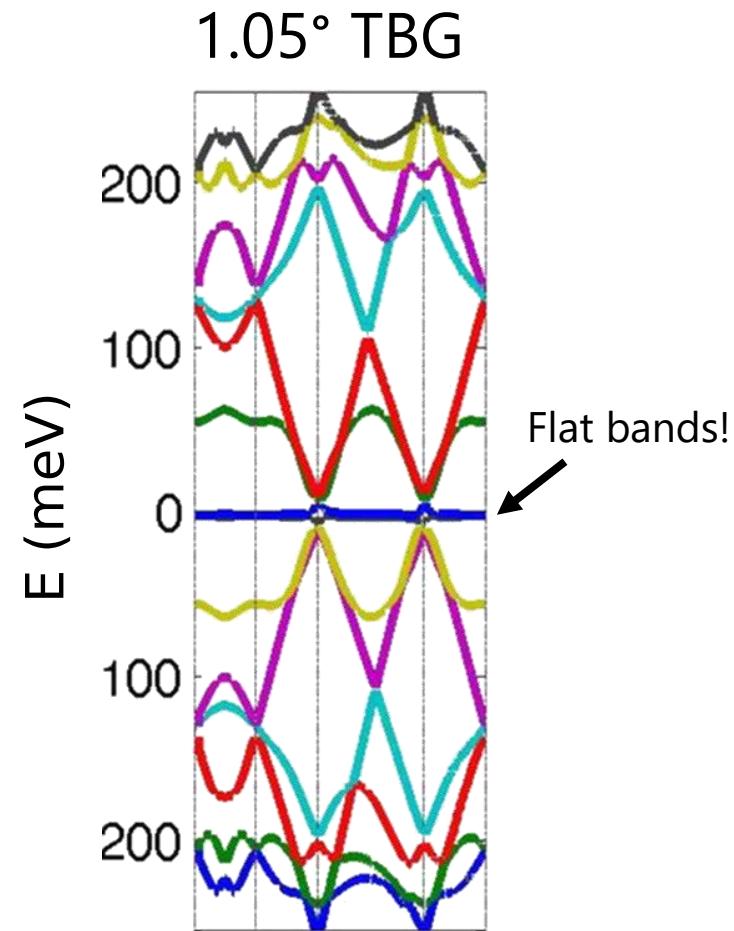
Bandstructure engineering with moirés



First experimental realization of TBG

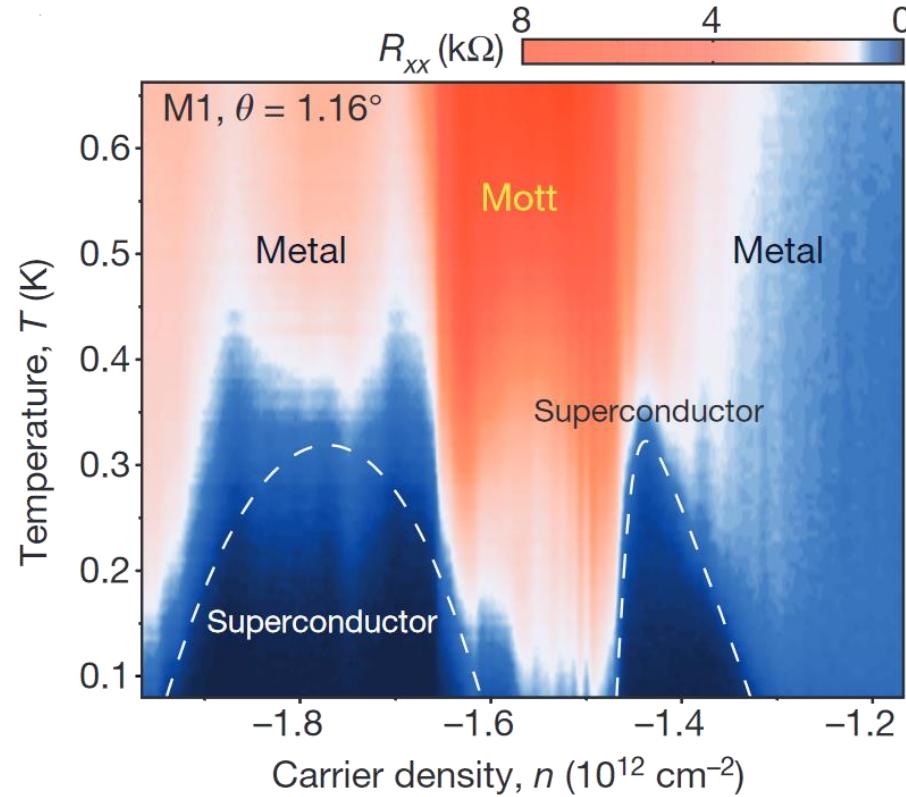
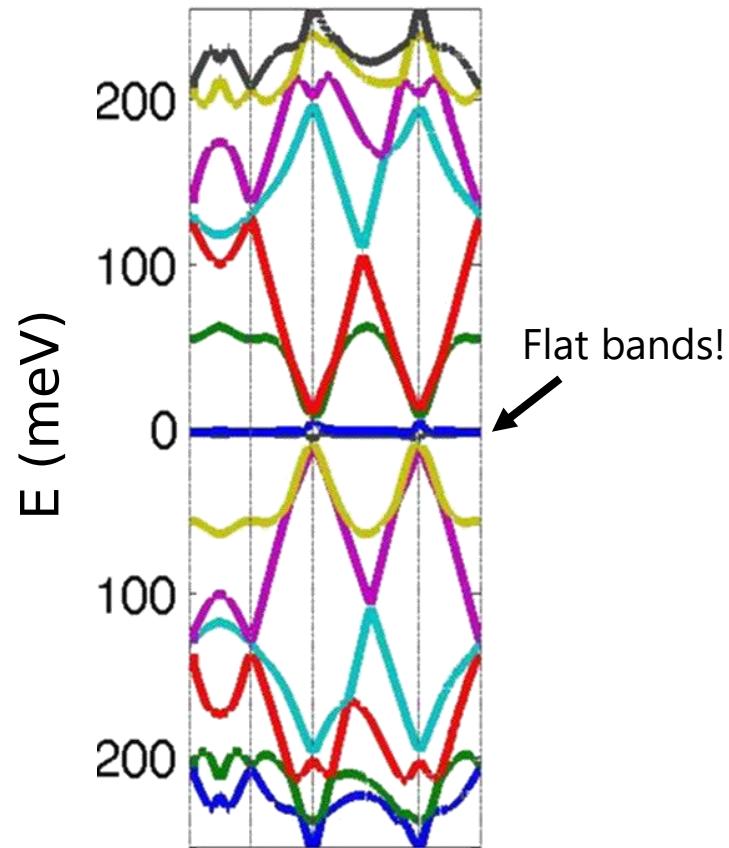


First experimental realization of TBG



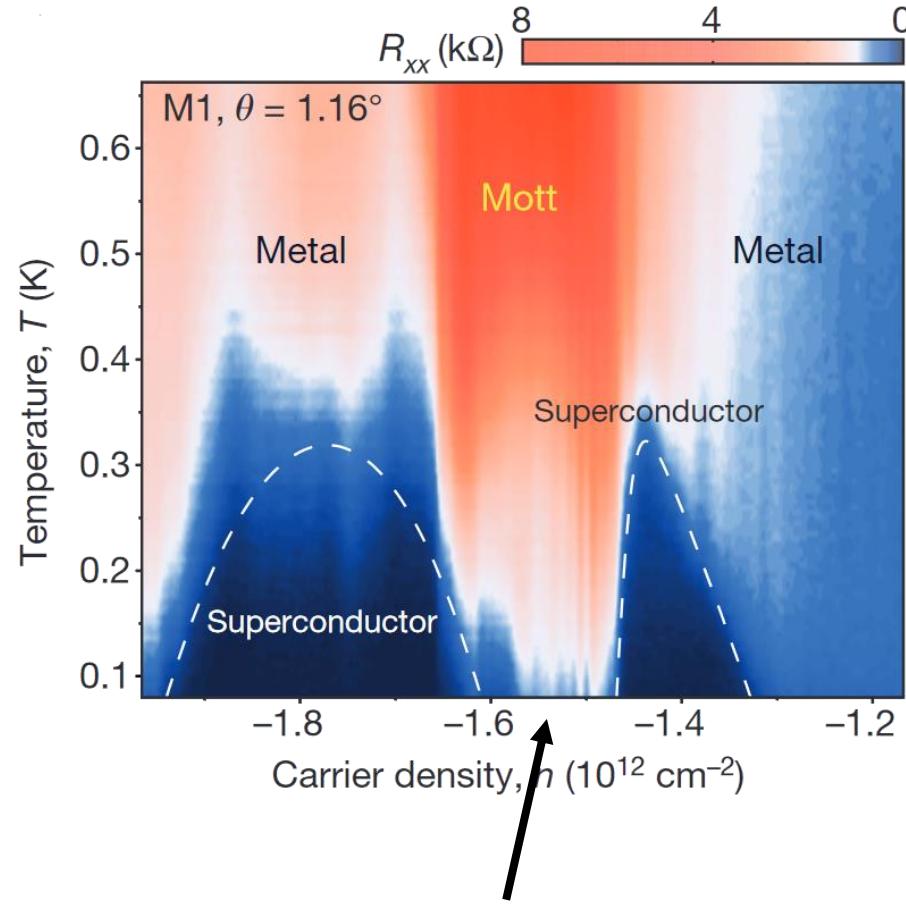
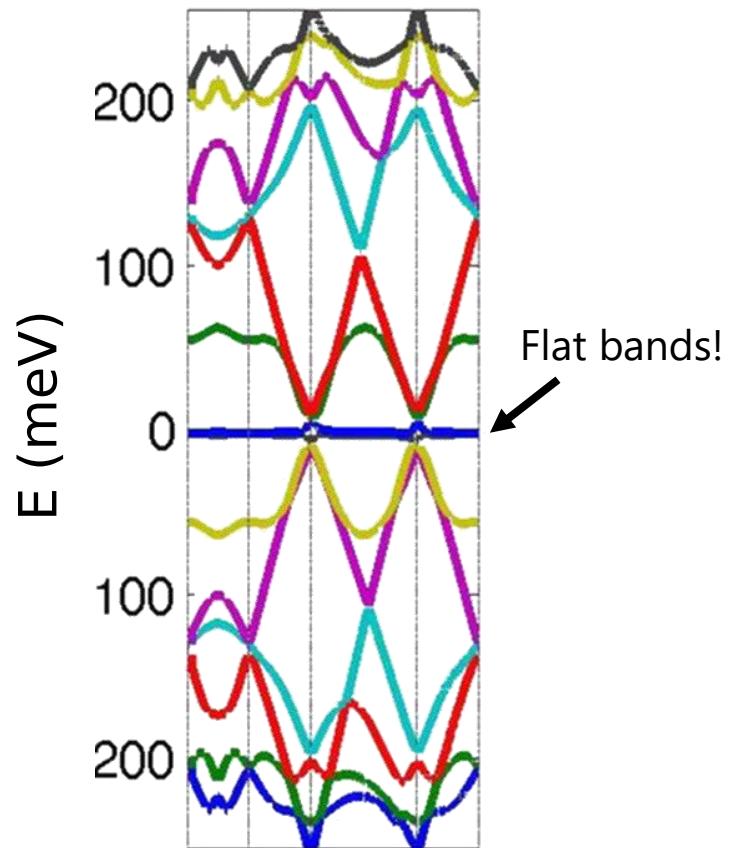
First experimental realization of TBG

1.05° TBG



First experimental realization of TBG

1.05° TBG

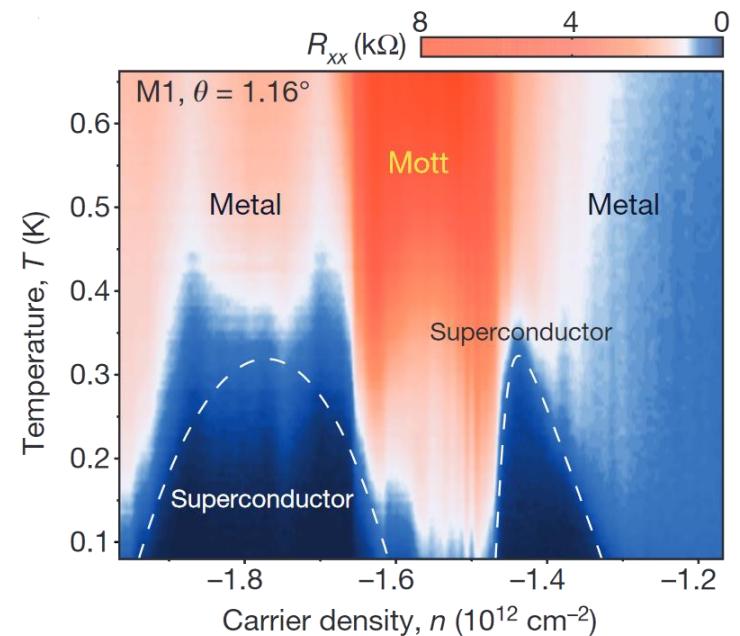
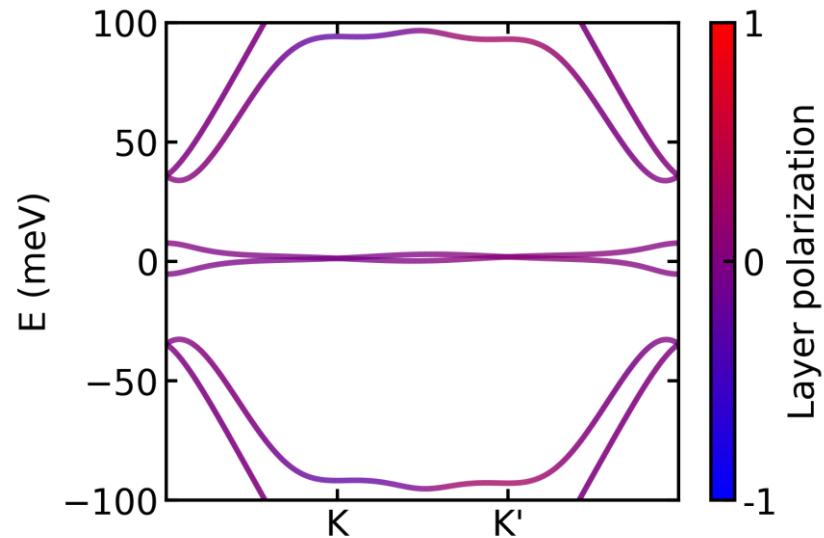


Half filled band – symmetry broken phase
Think Hund's rules

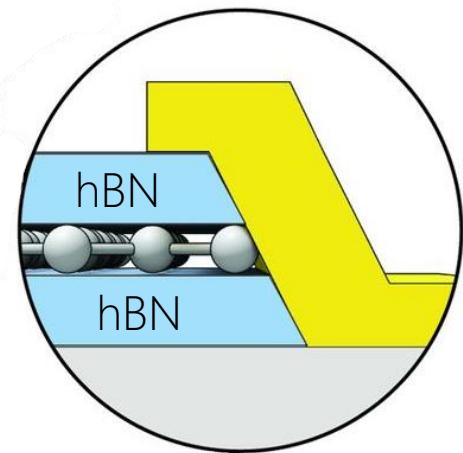
Summary

By controlling twist angle, graphene multilayers can become strongly correlated!

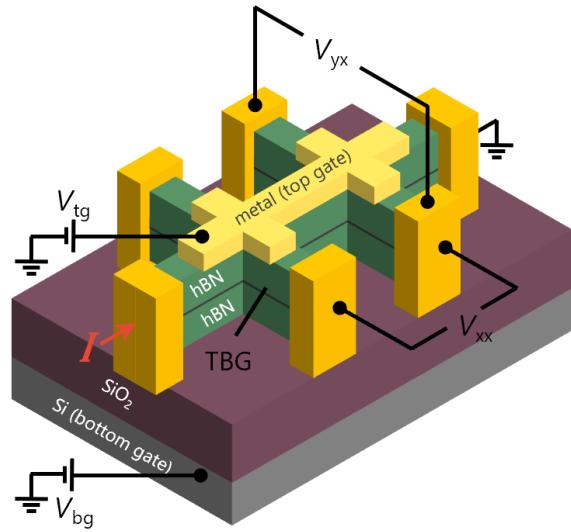
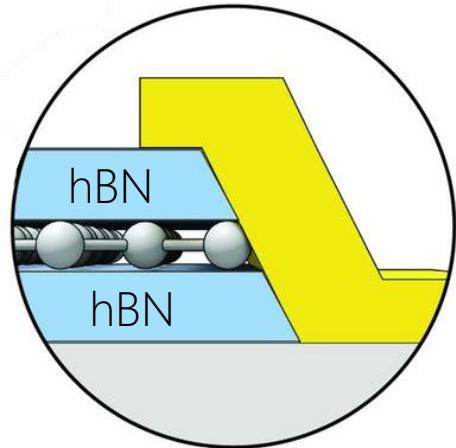
Let's make some samples!



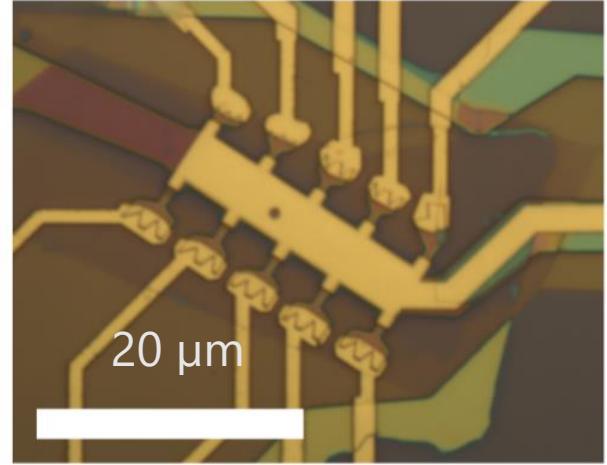
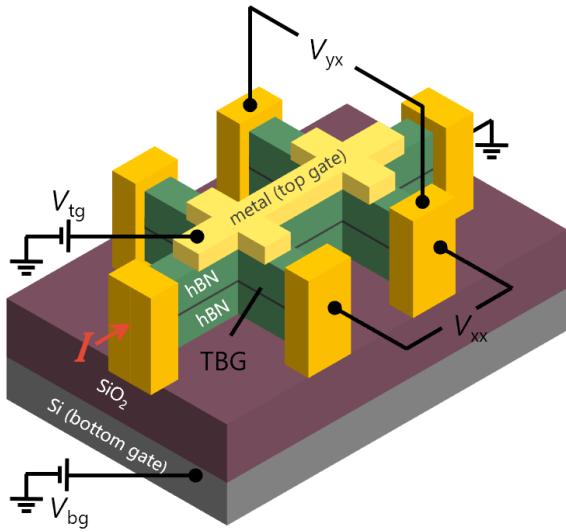
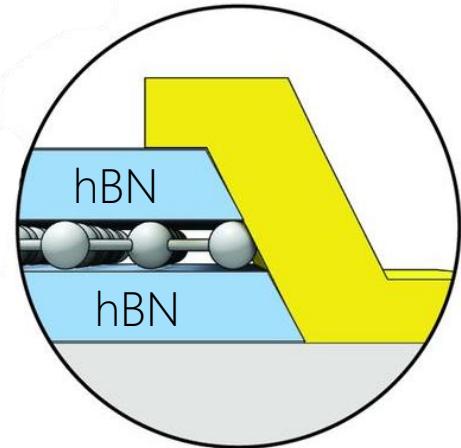
Typical twisted bilayer graphene (TBG)



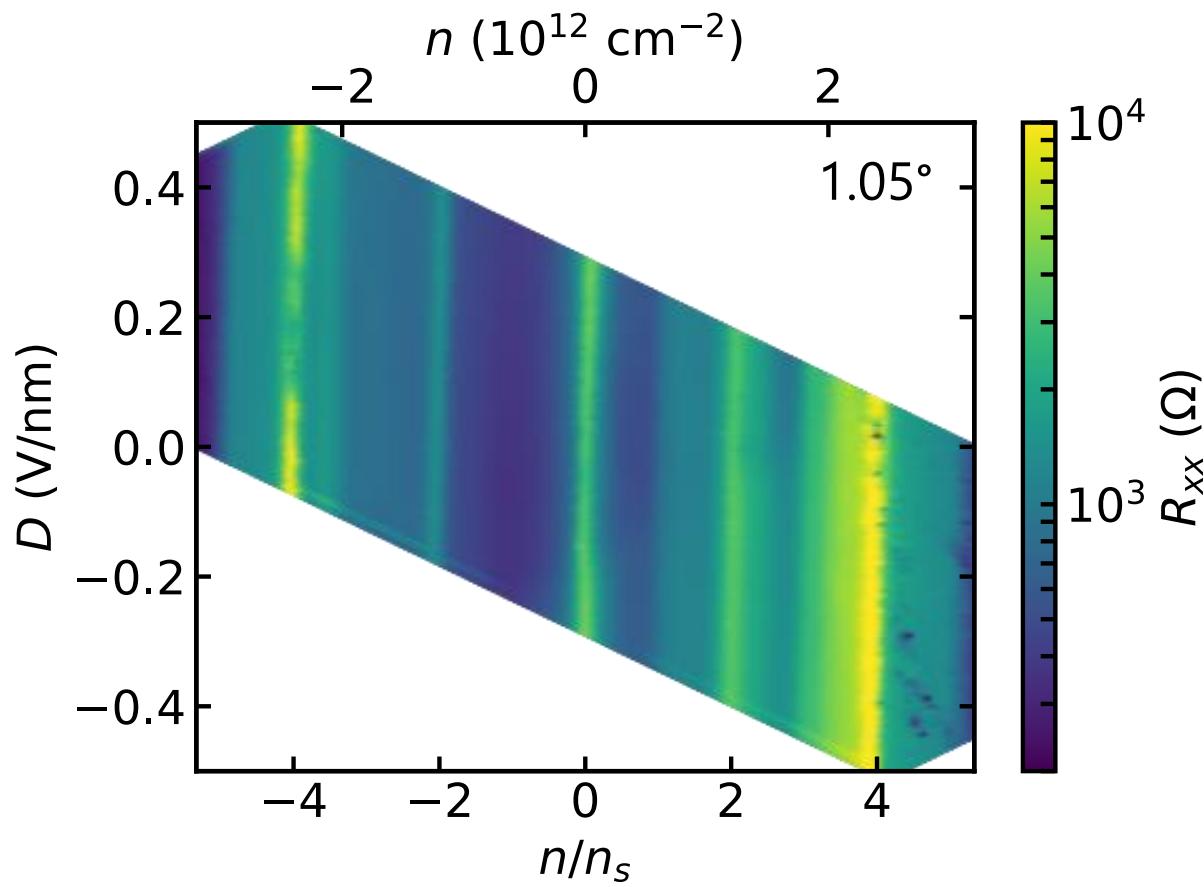
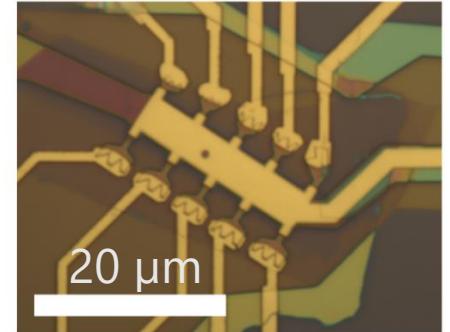
Typical twisted bilayer graphene (TBG)



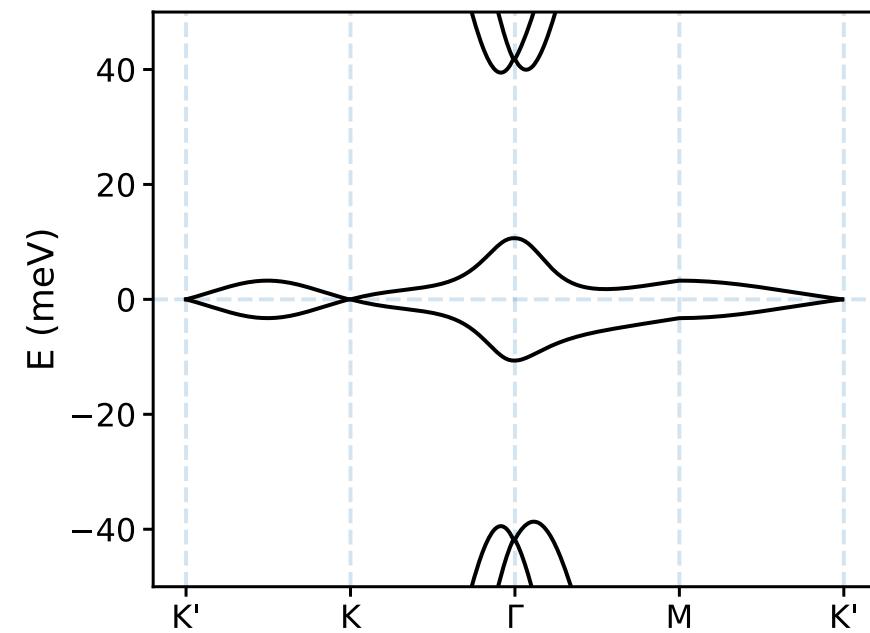
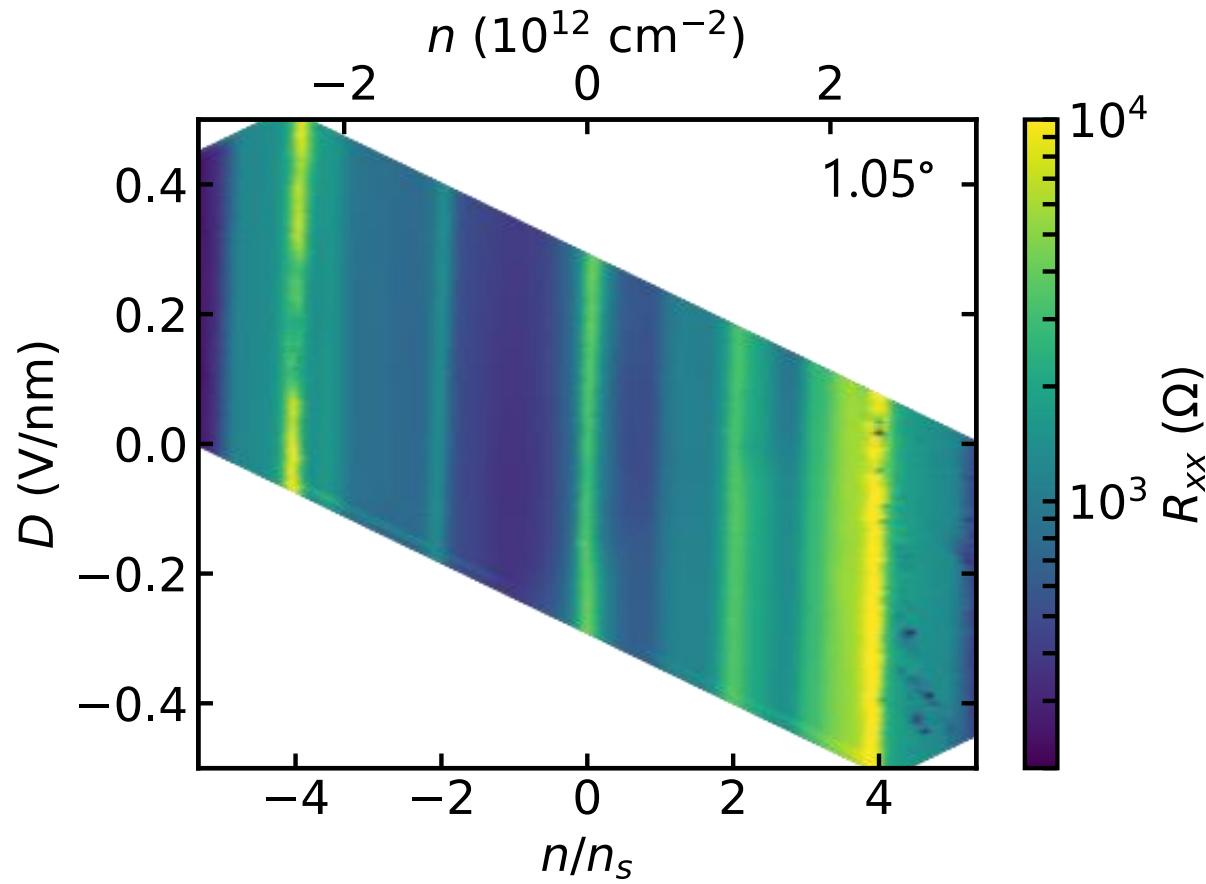
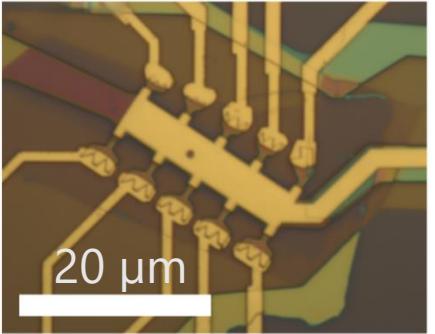
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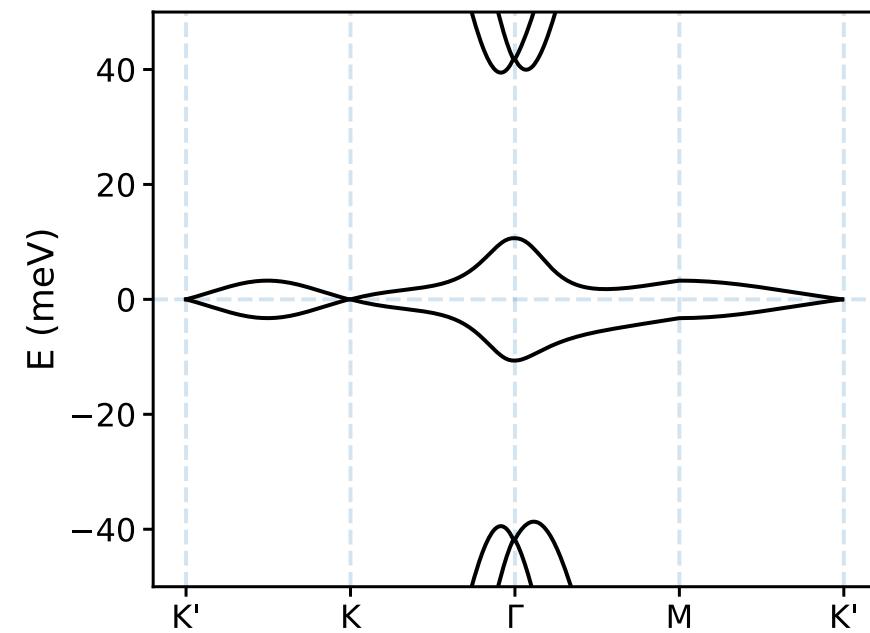
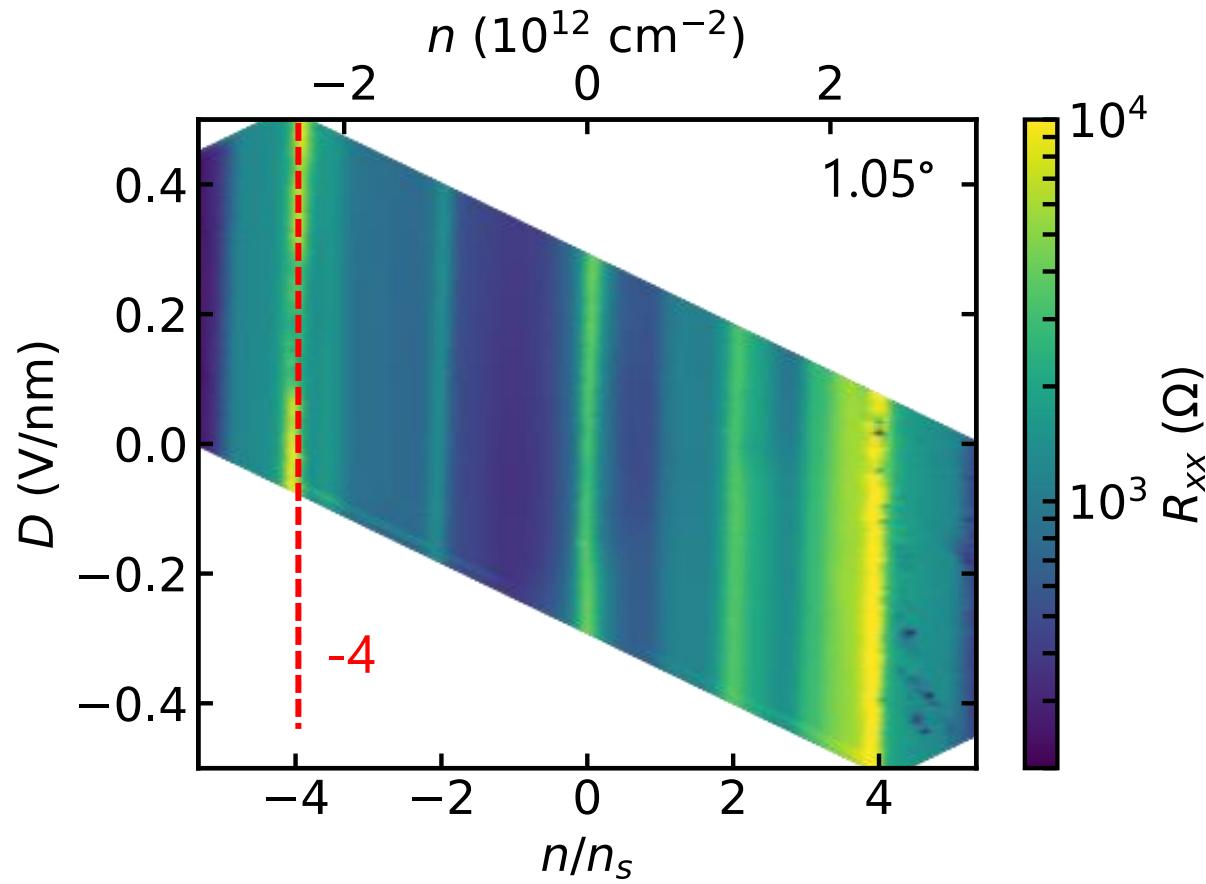
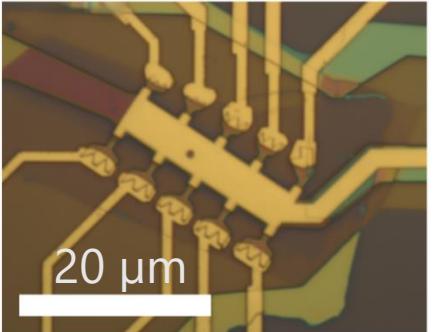
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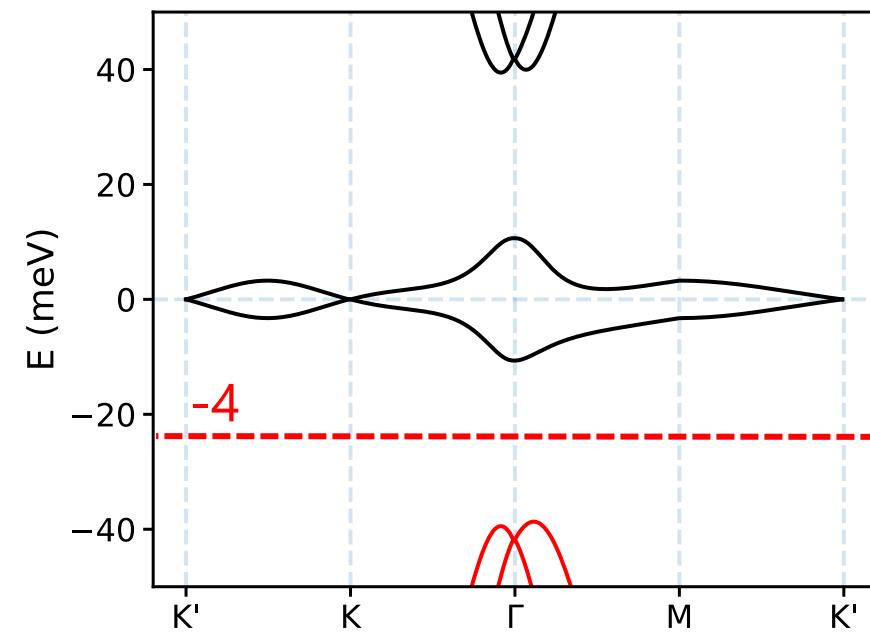
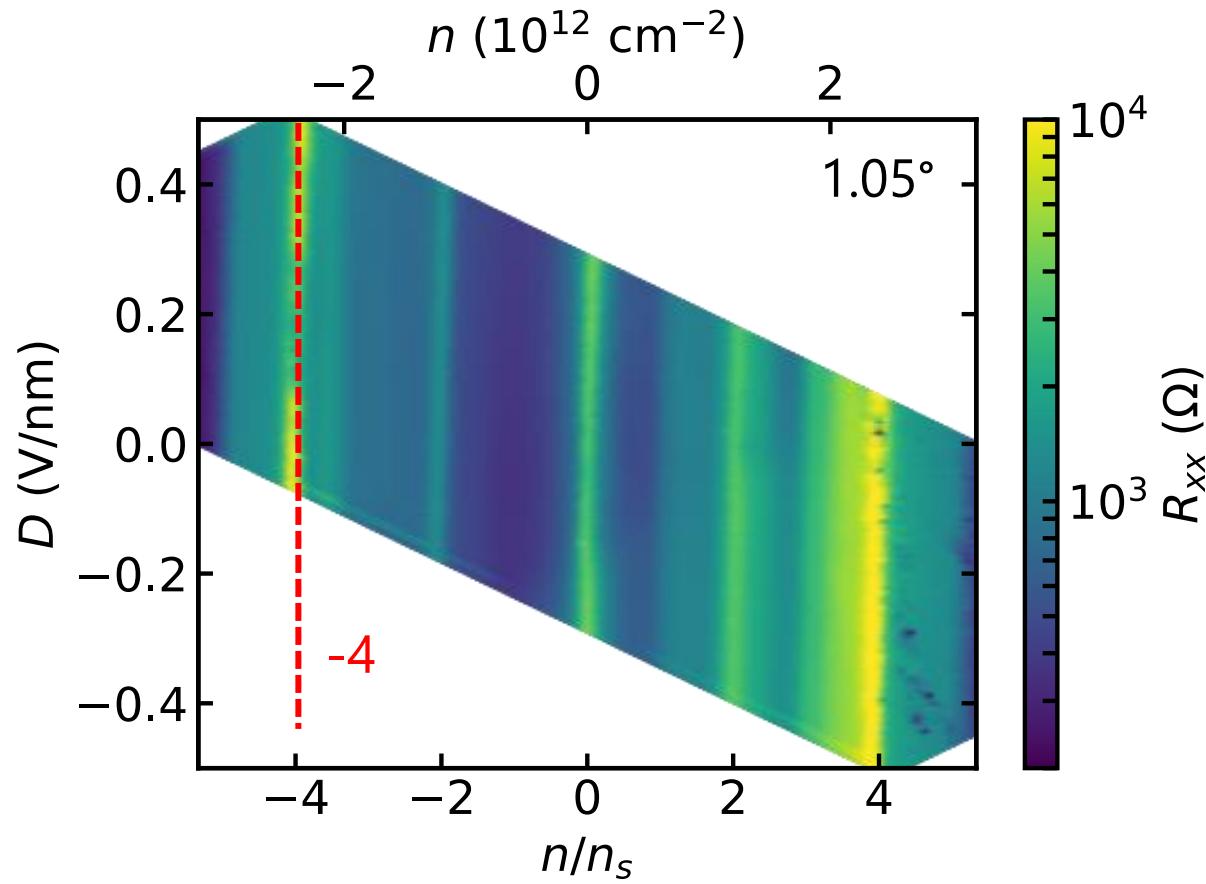
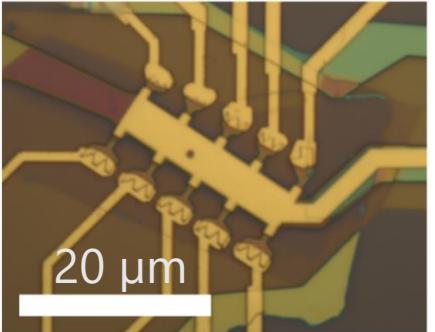
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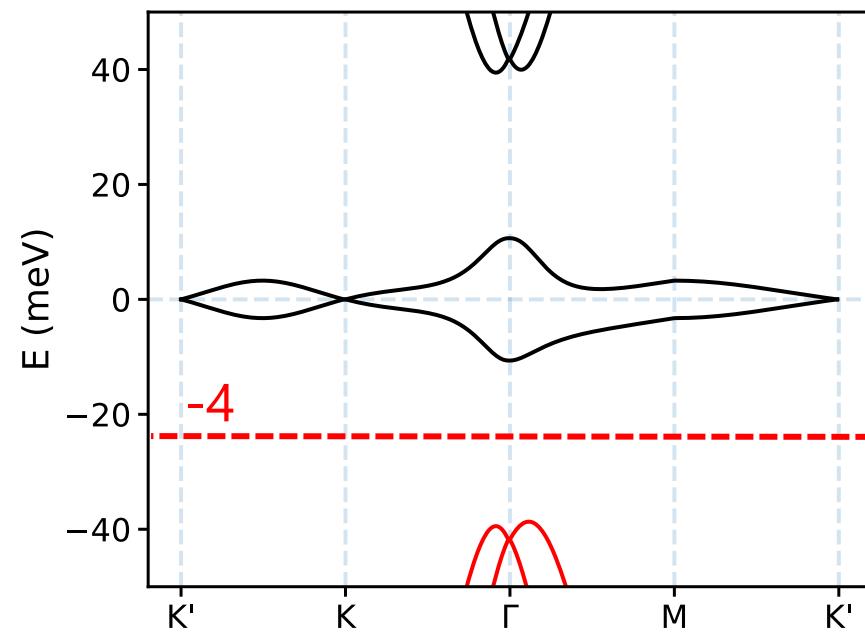
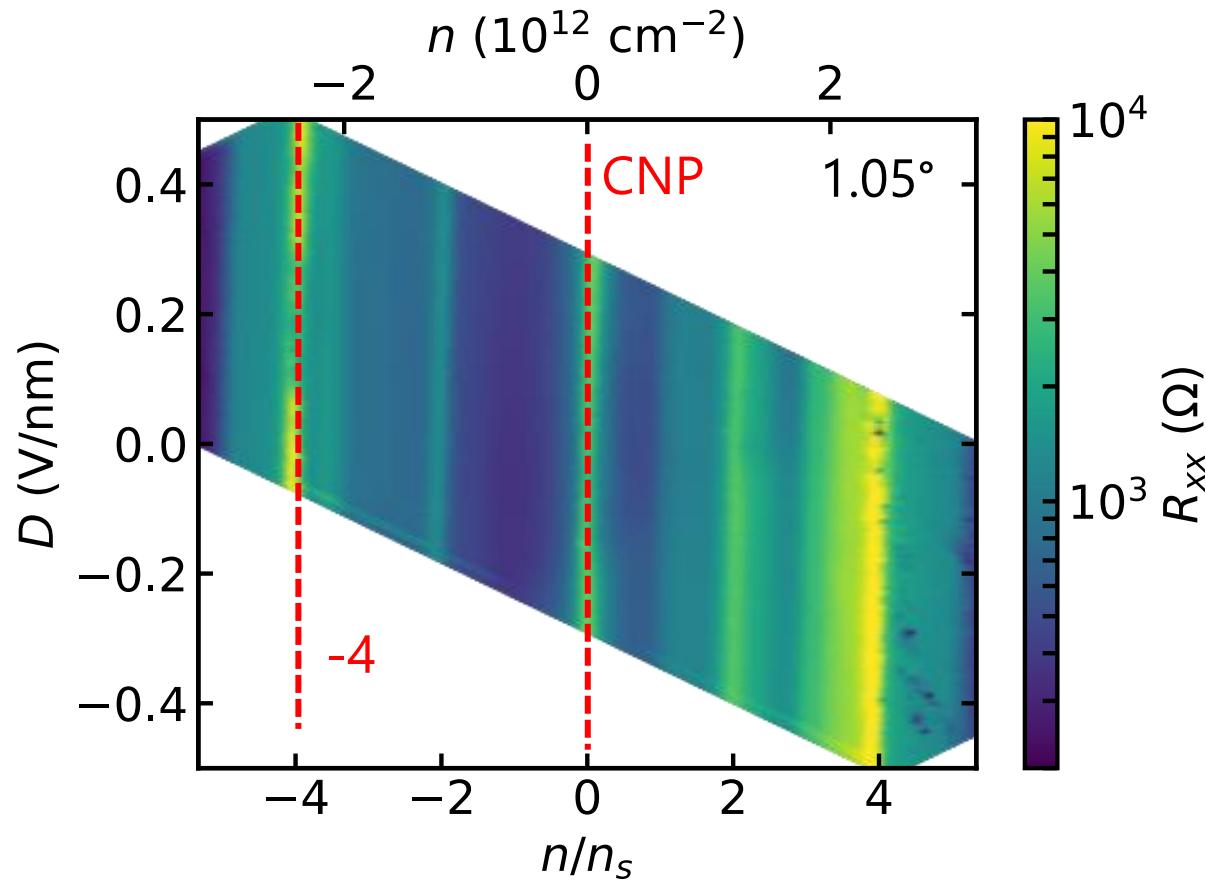
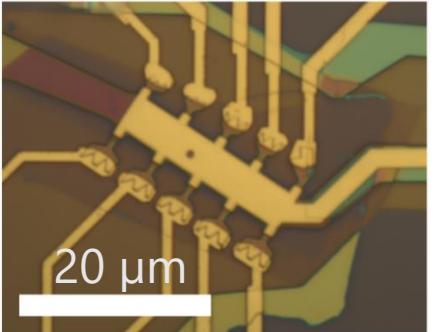
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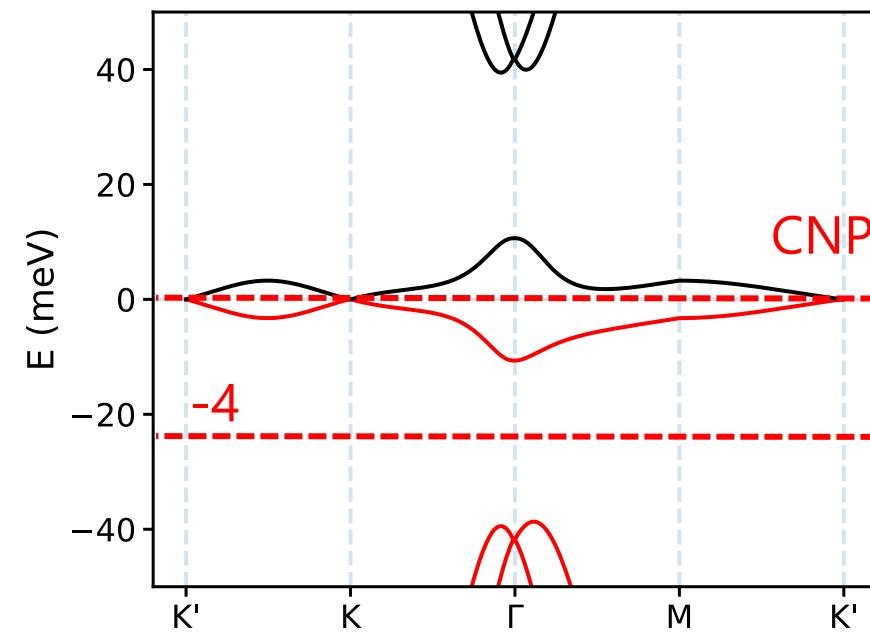
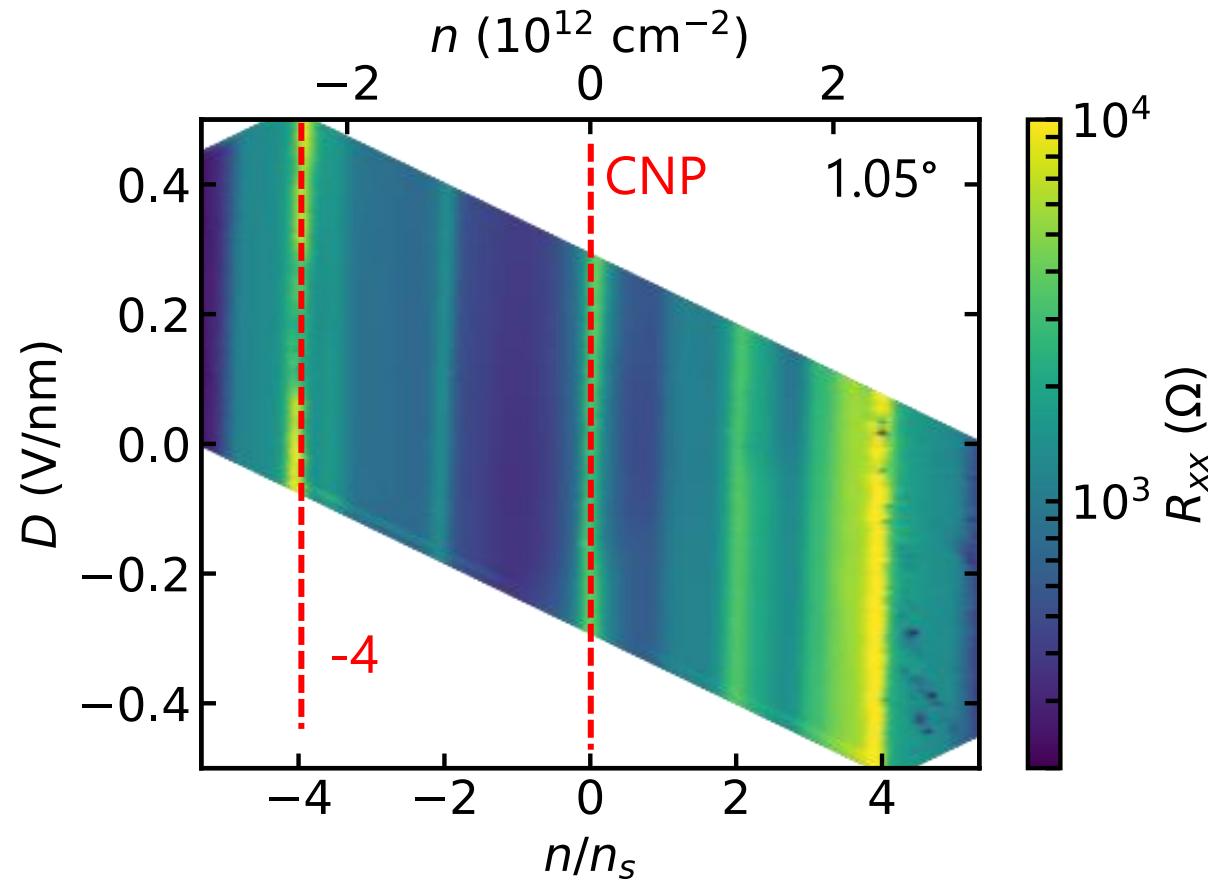
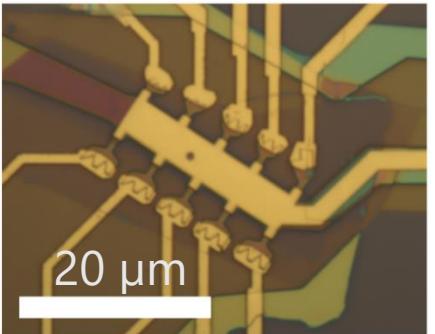
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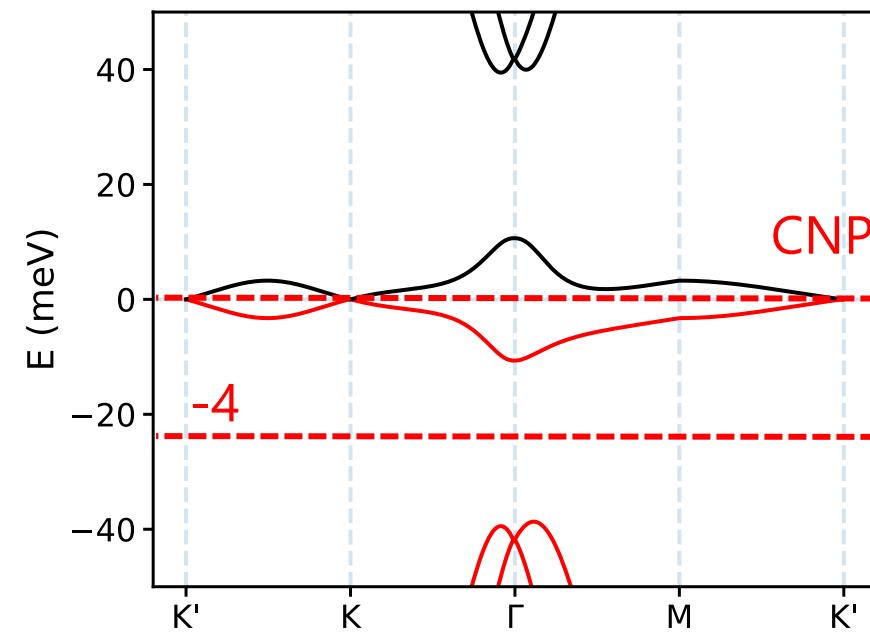
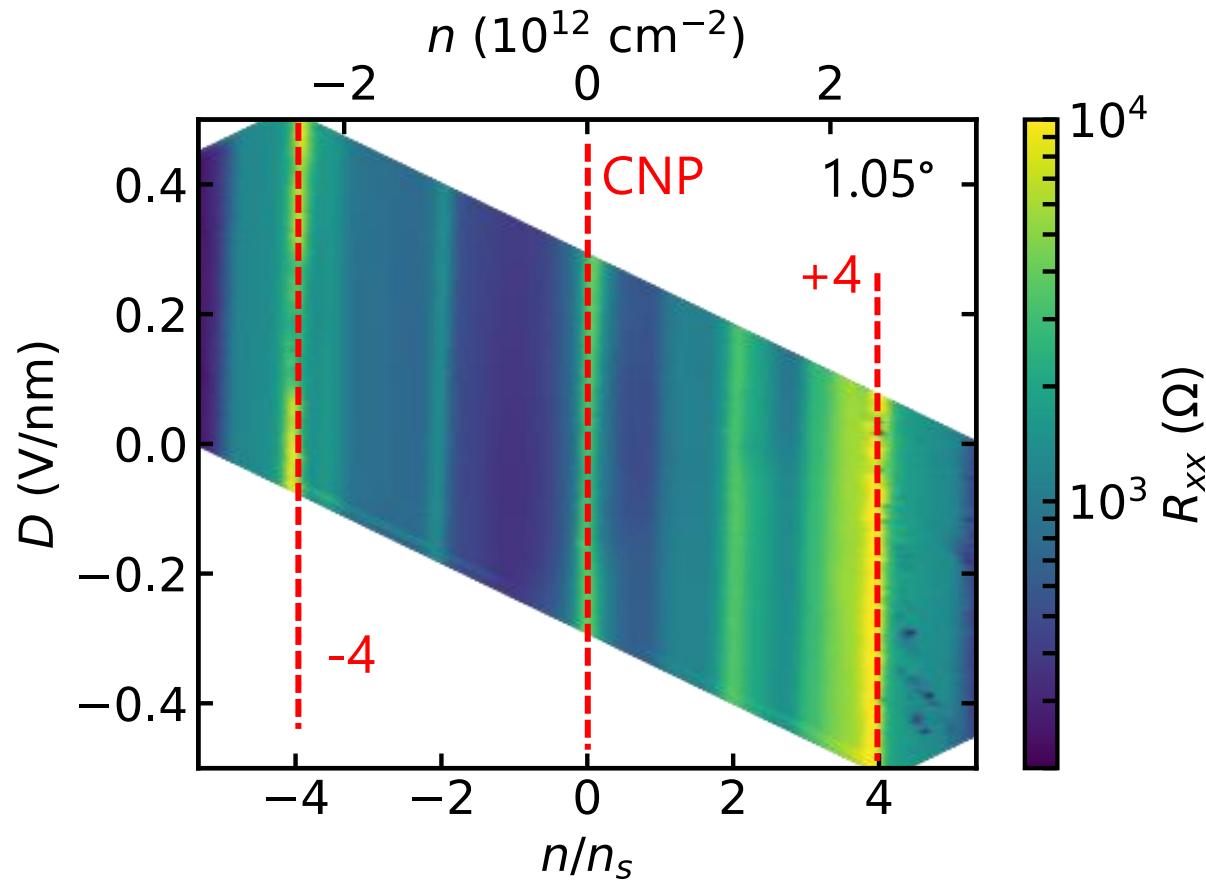
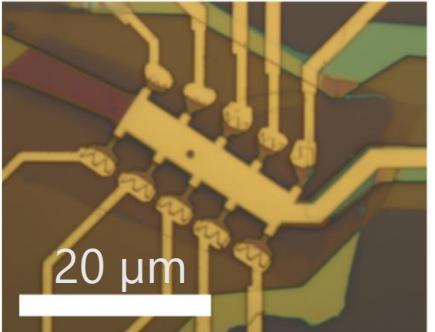
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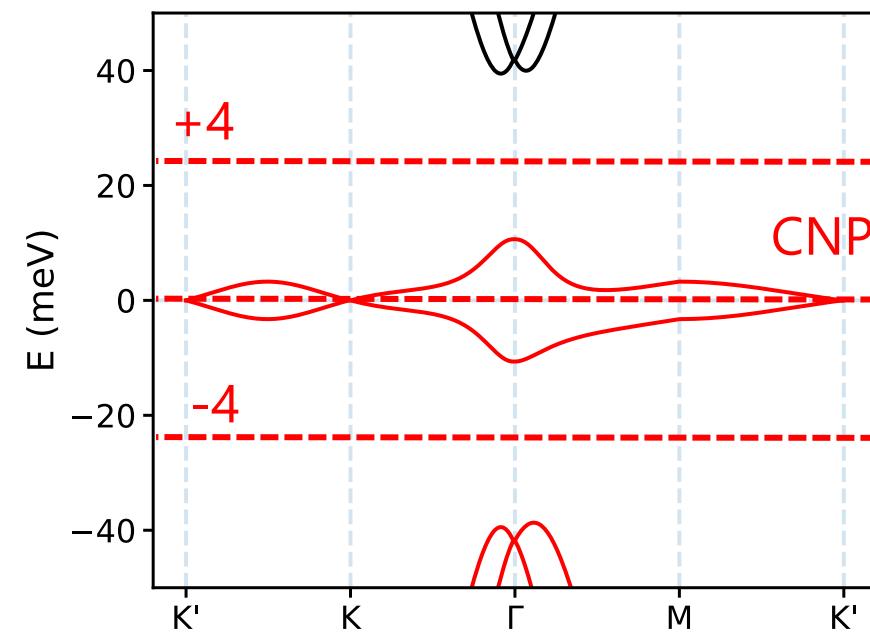
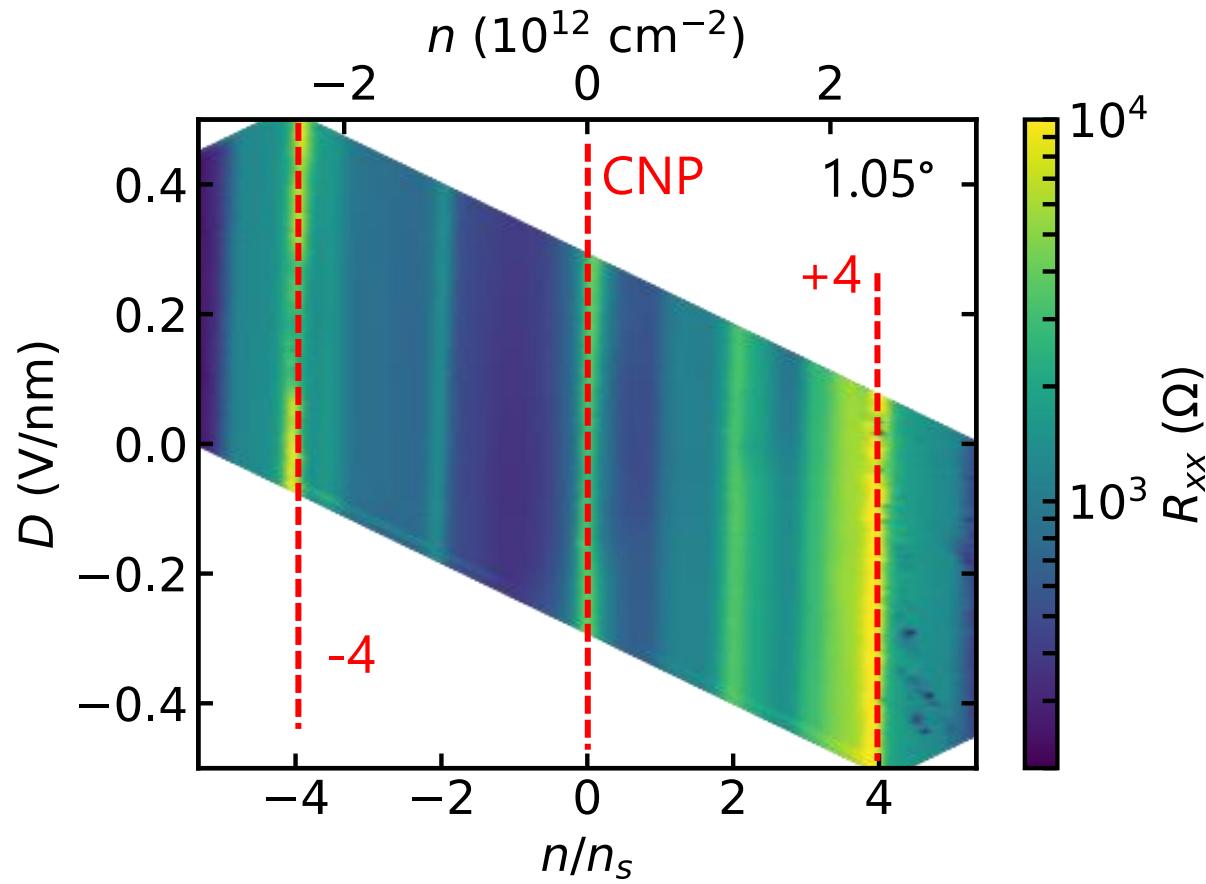
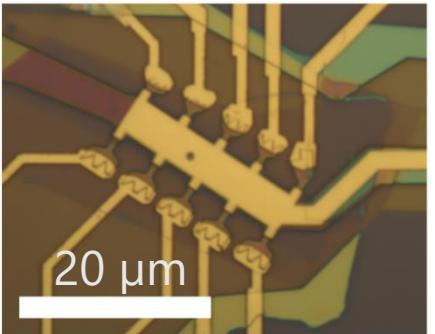
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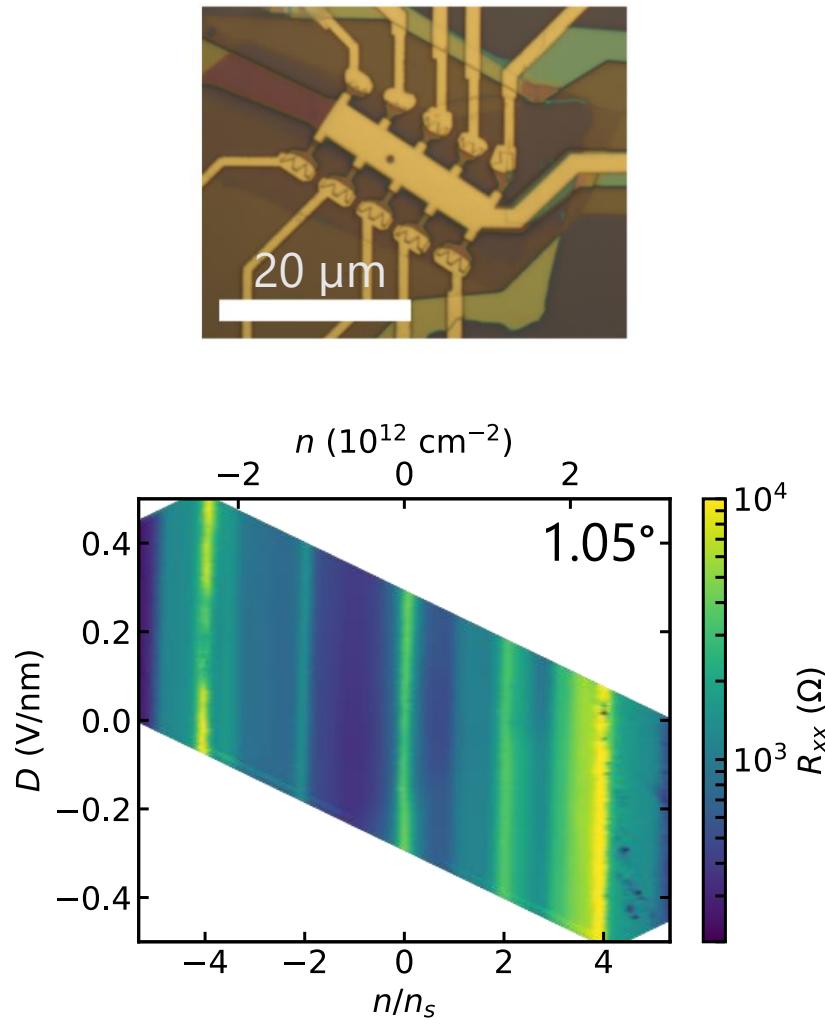
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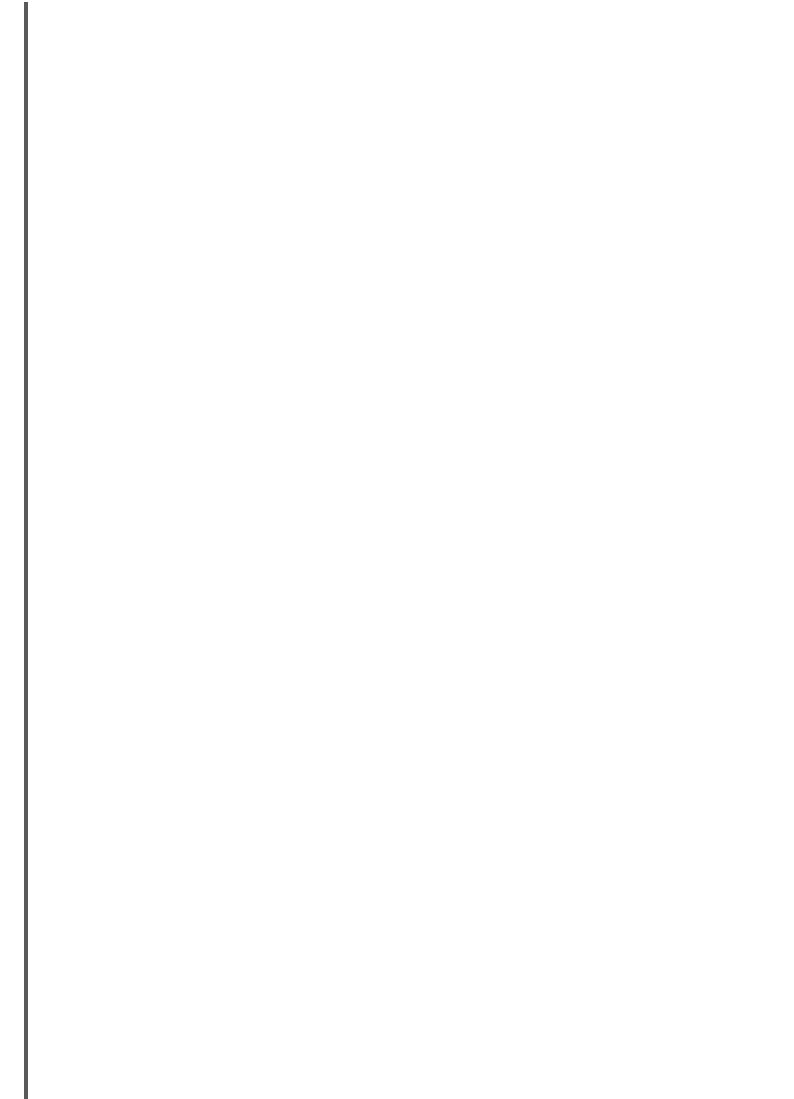
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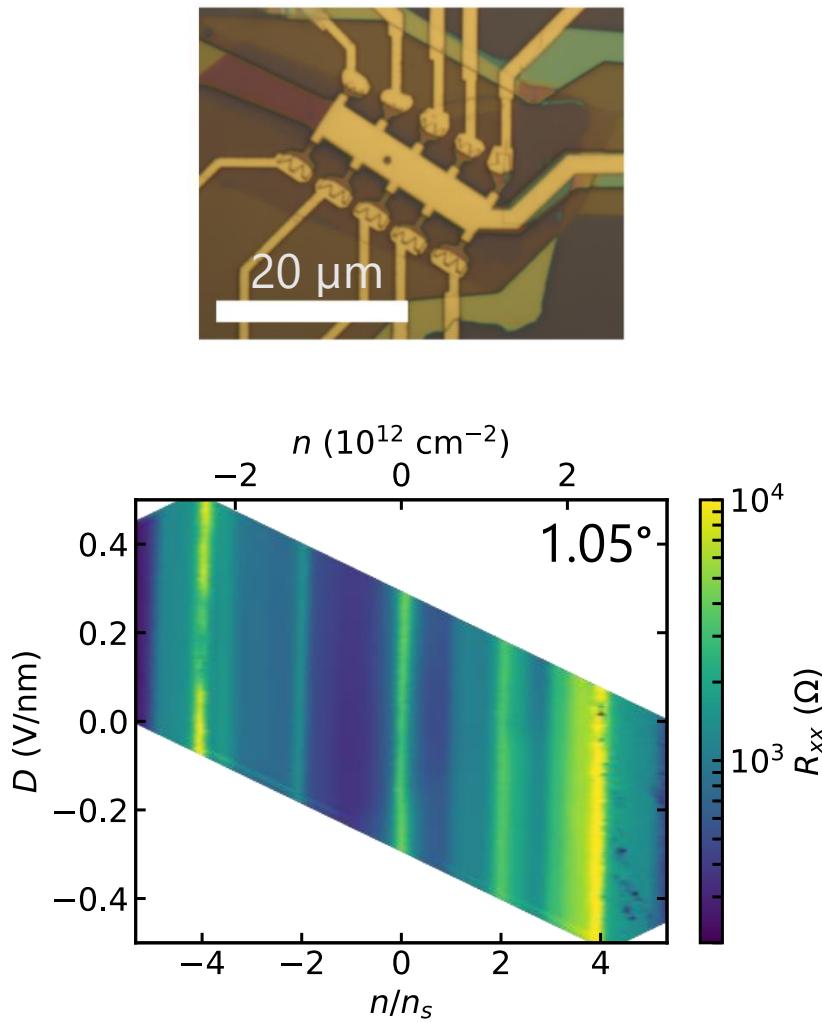
Typical TBG



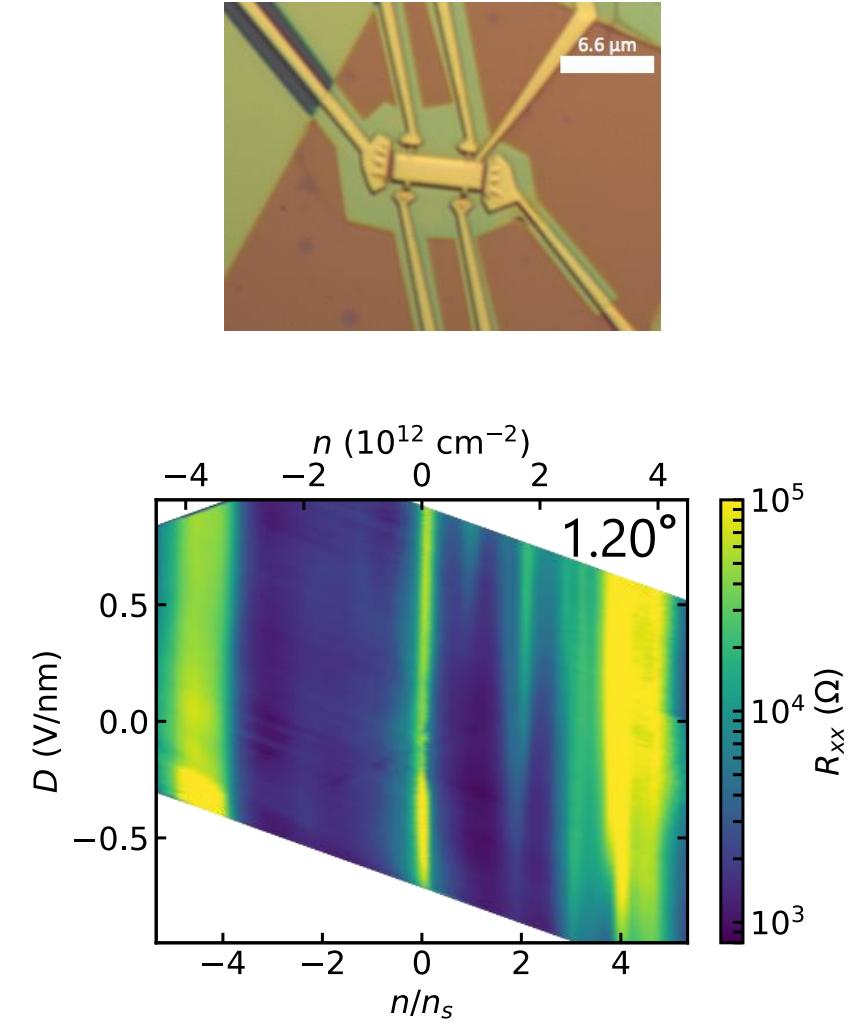
Atypical TBG



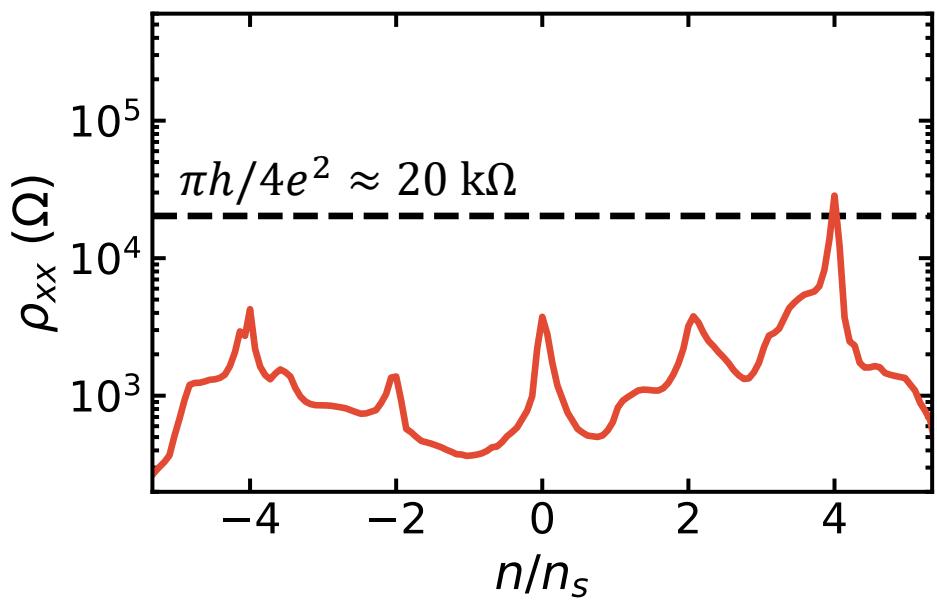
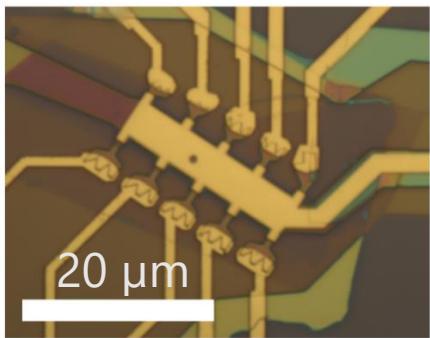
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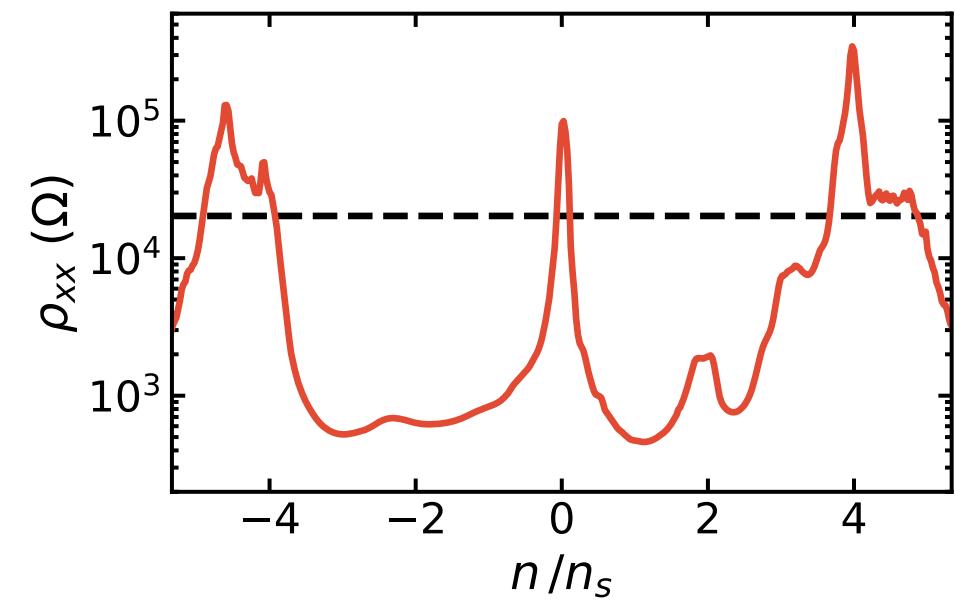
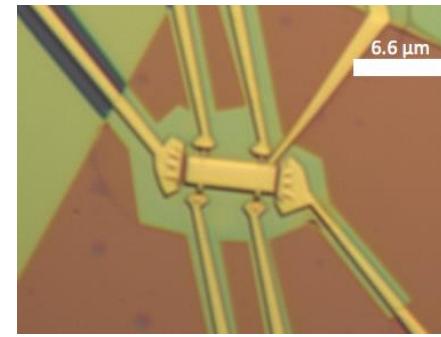
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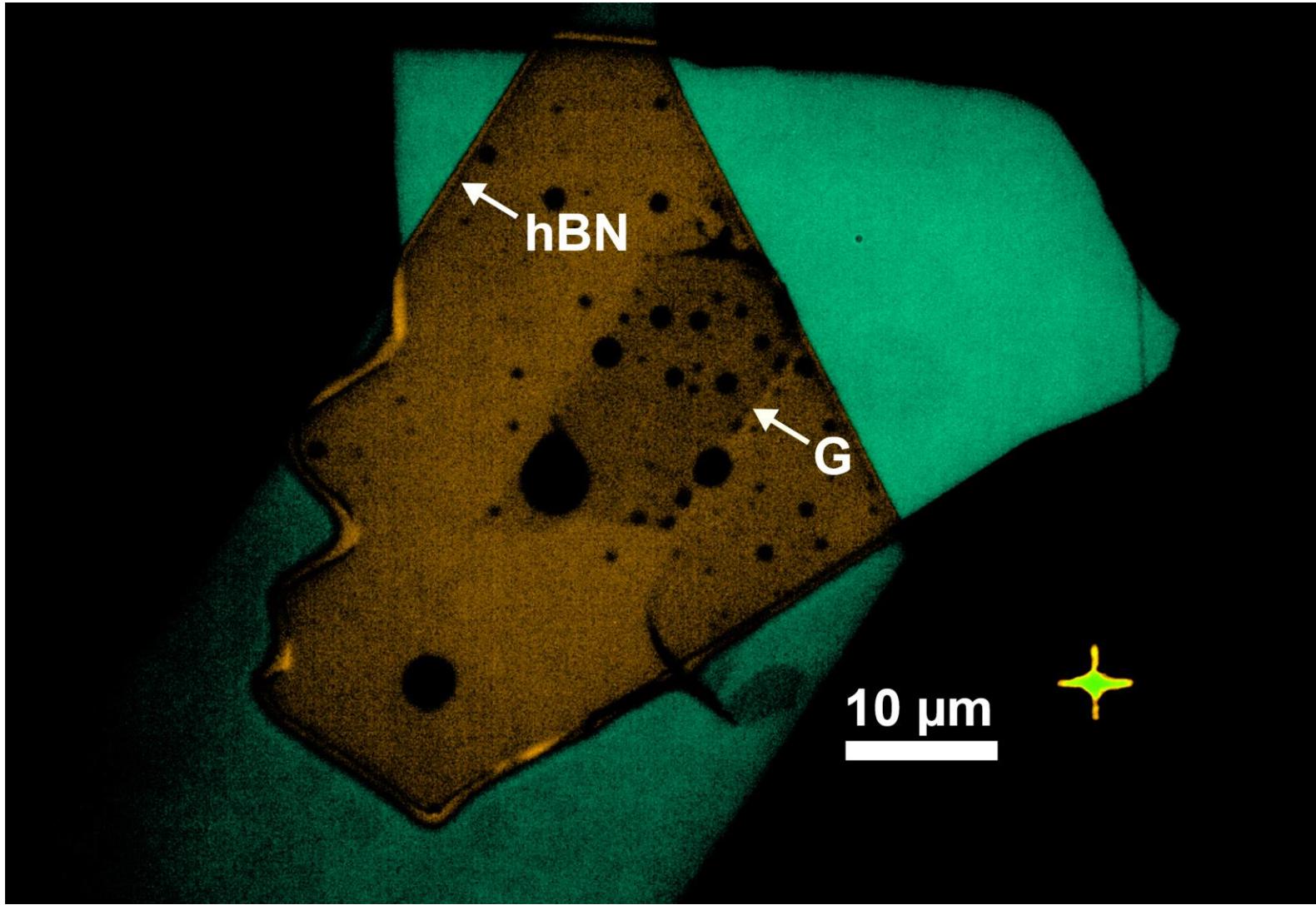
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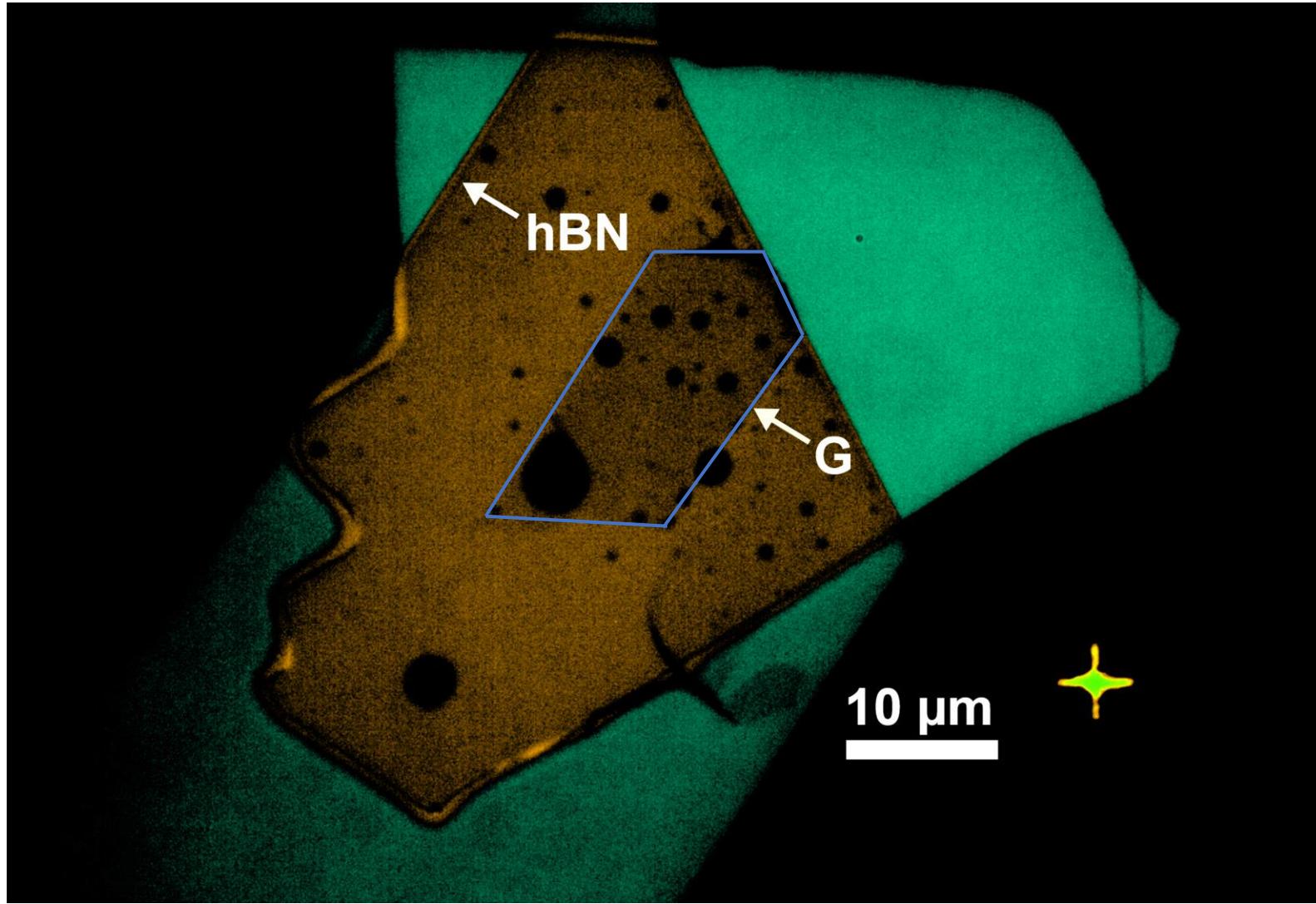
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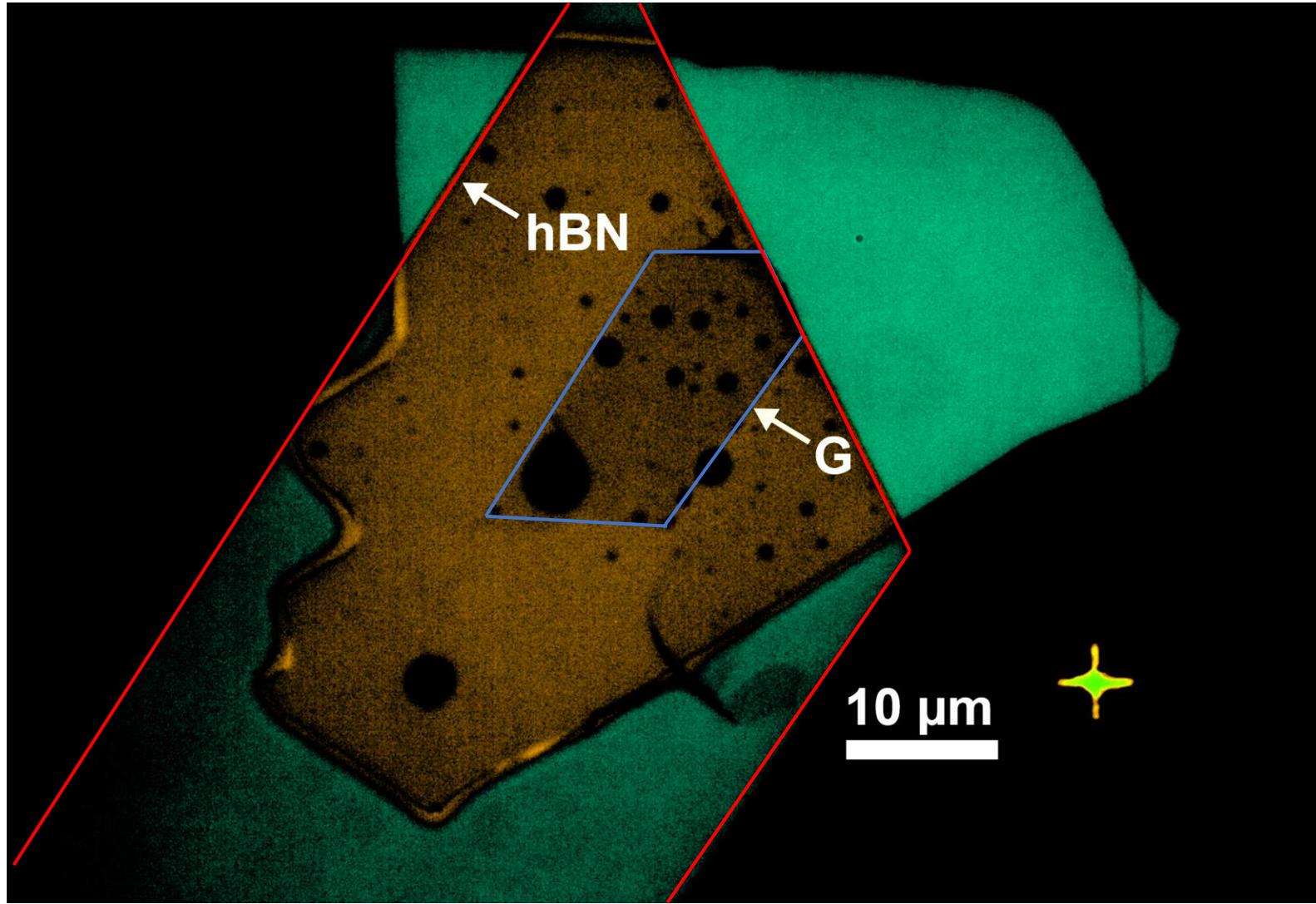
Visual hBN alignment



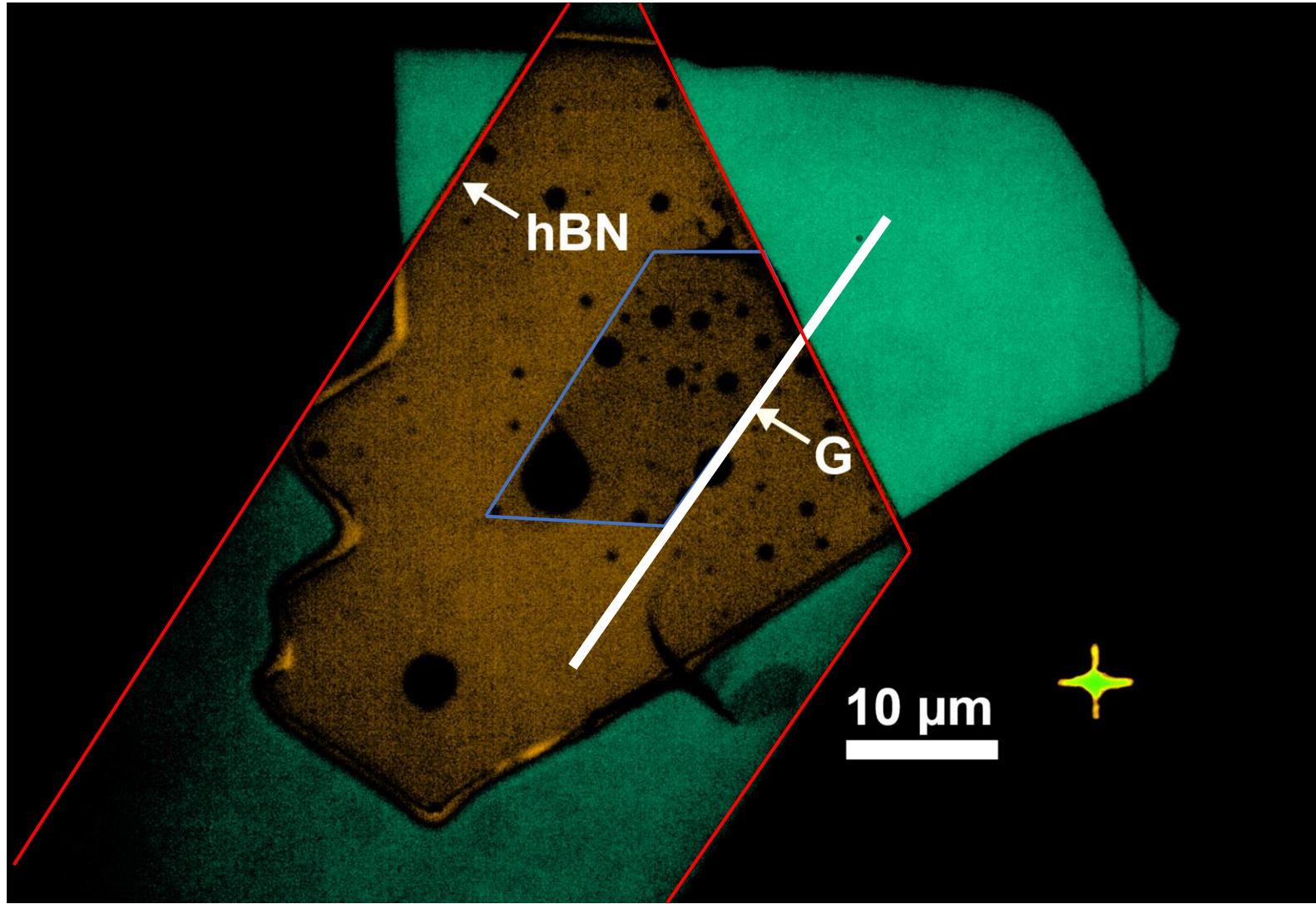
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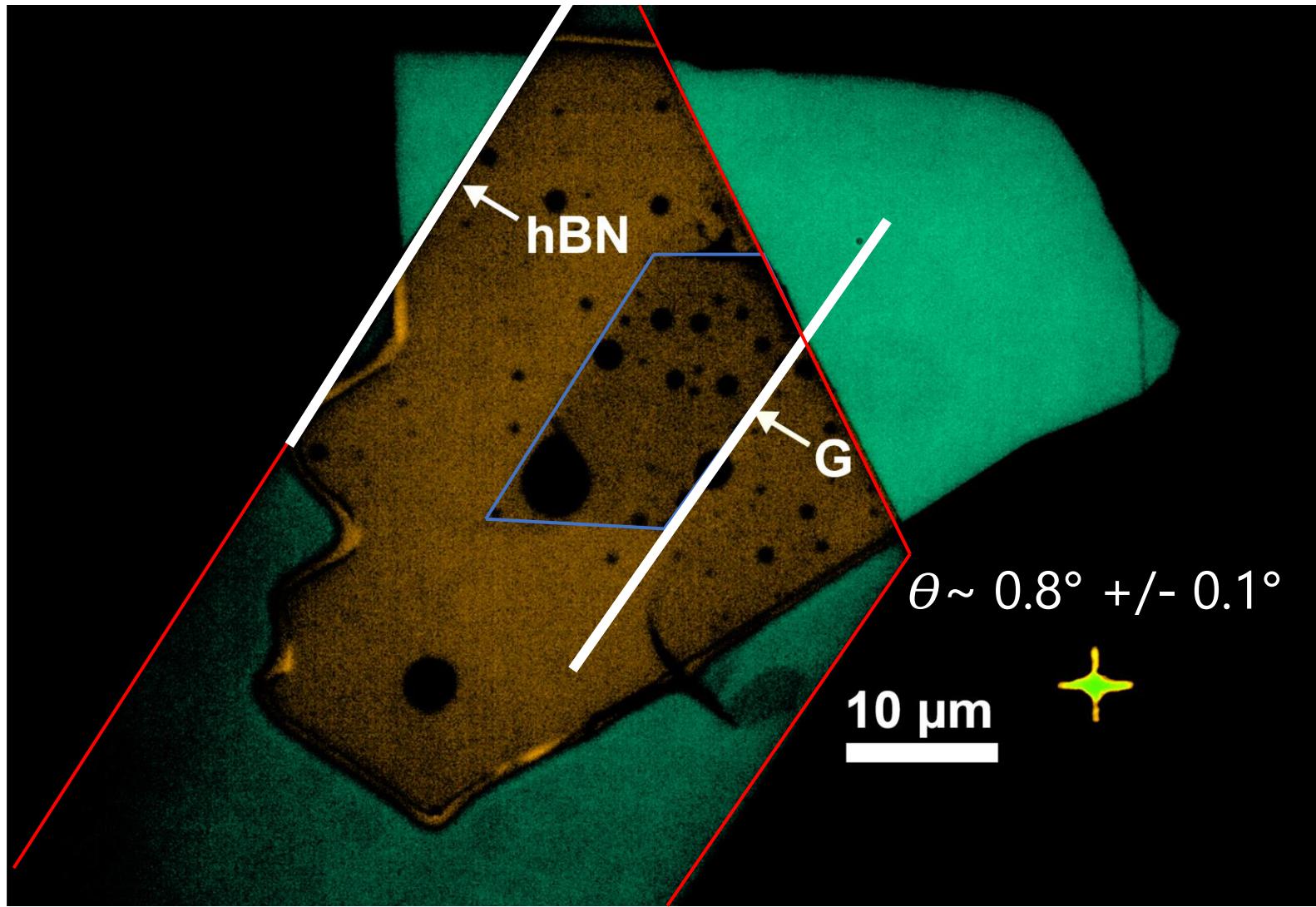
Visual hBN alignment



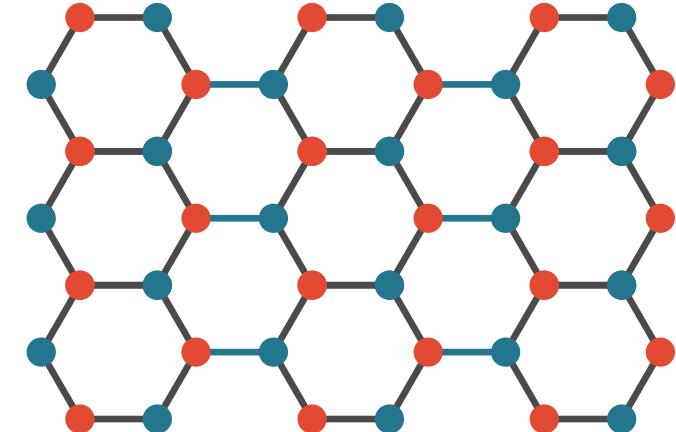
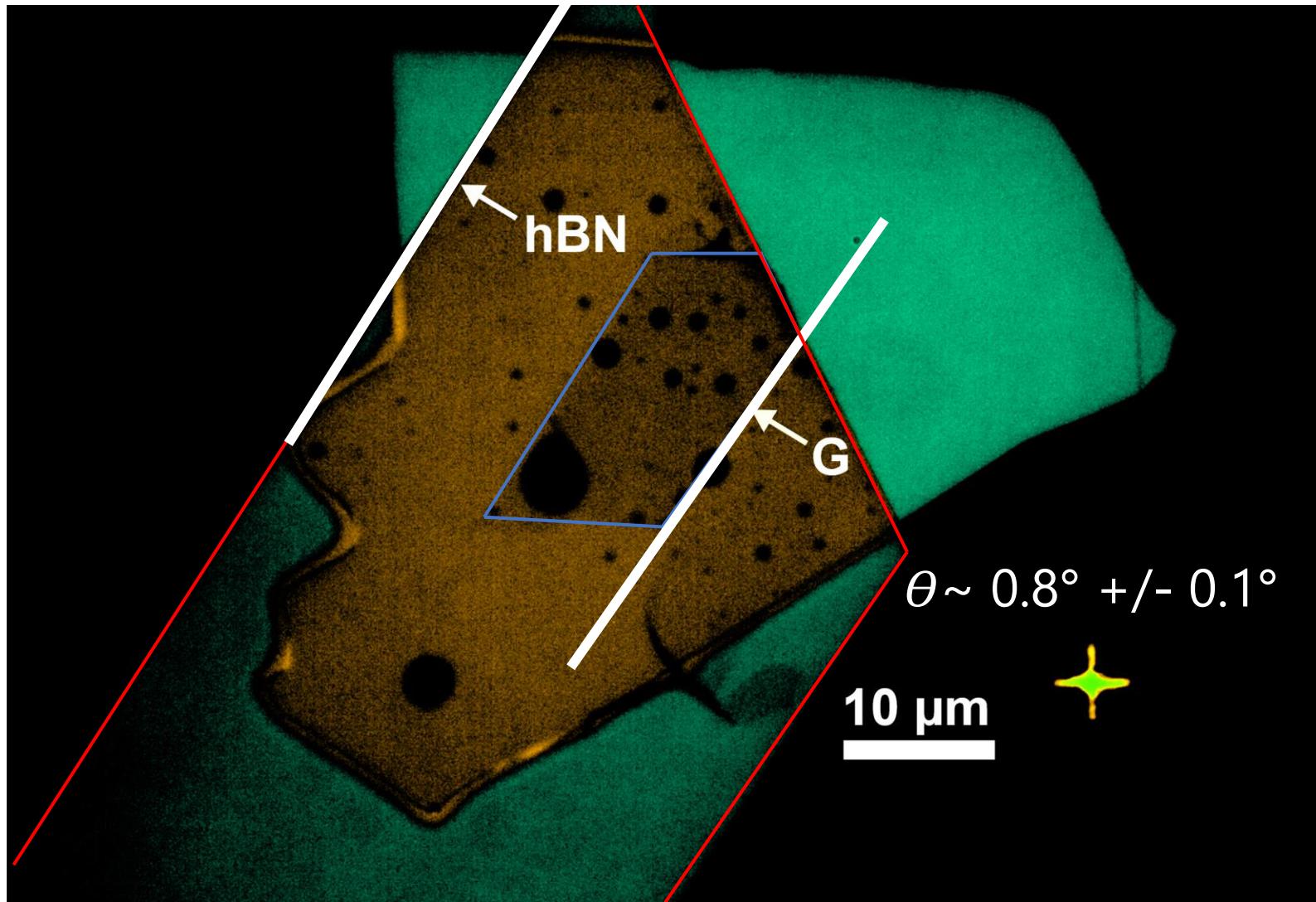
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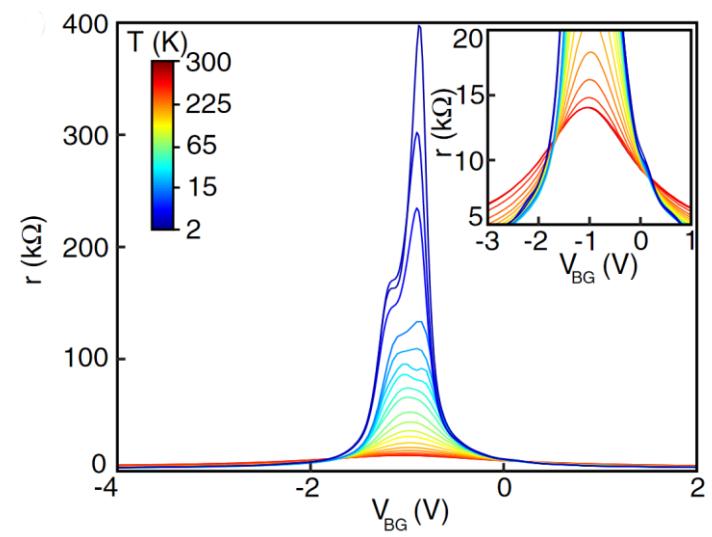
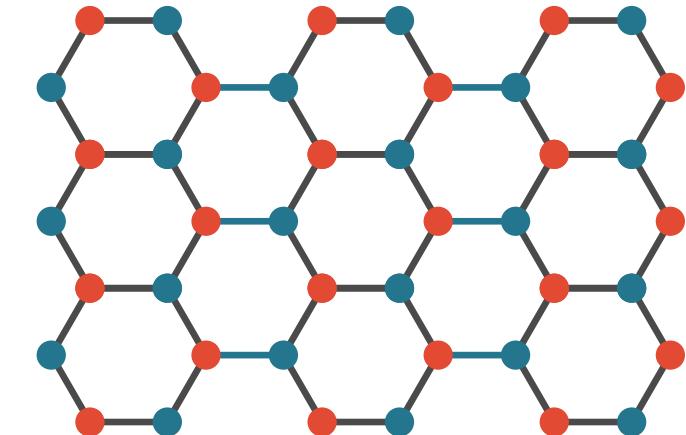
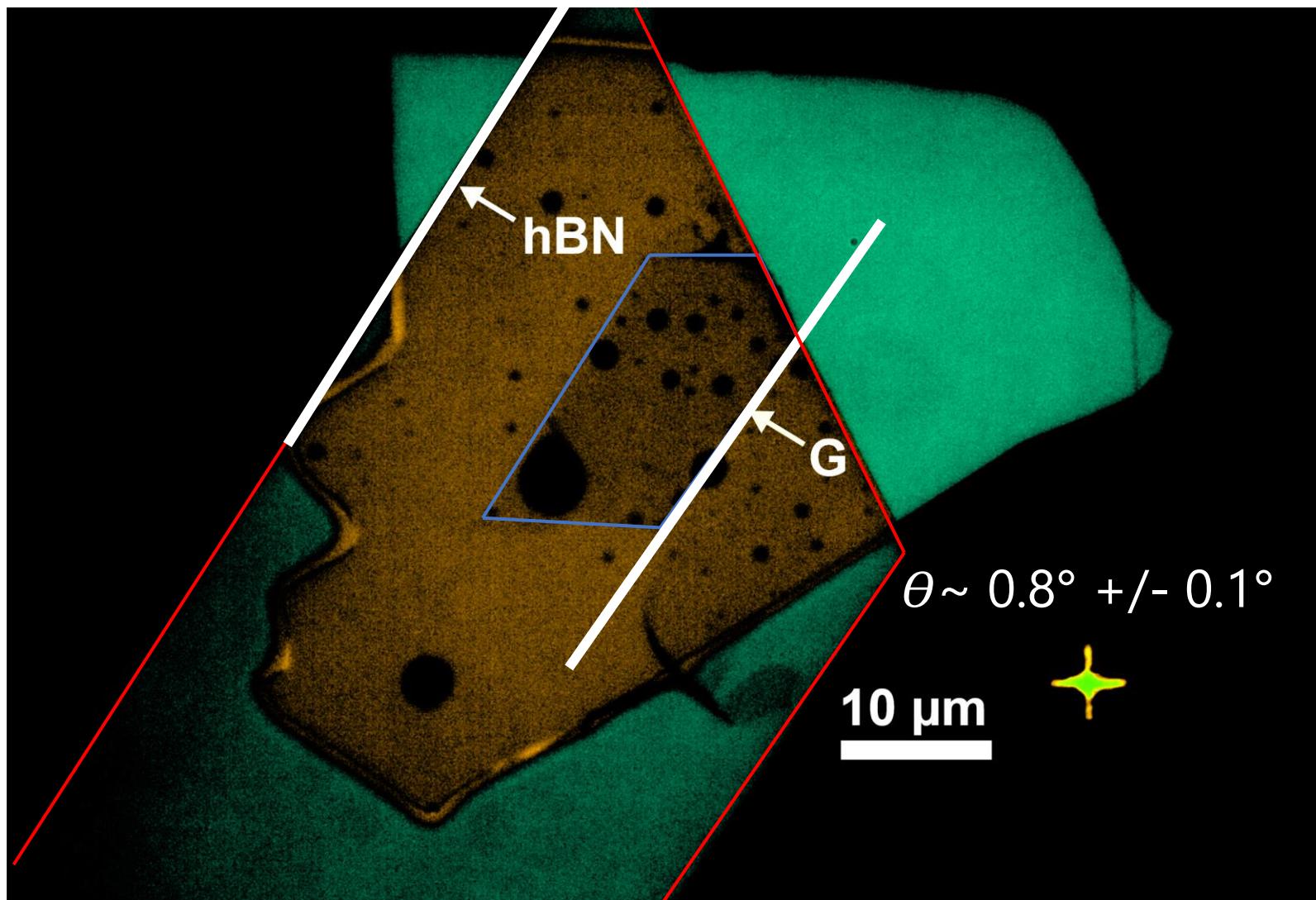
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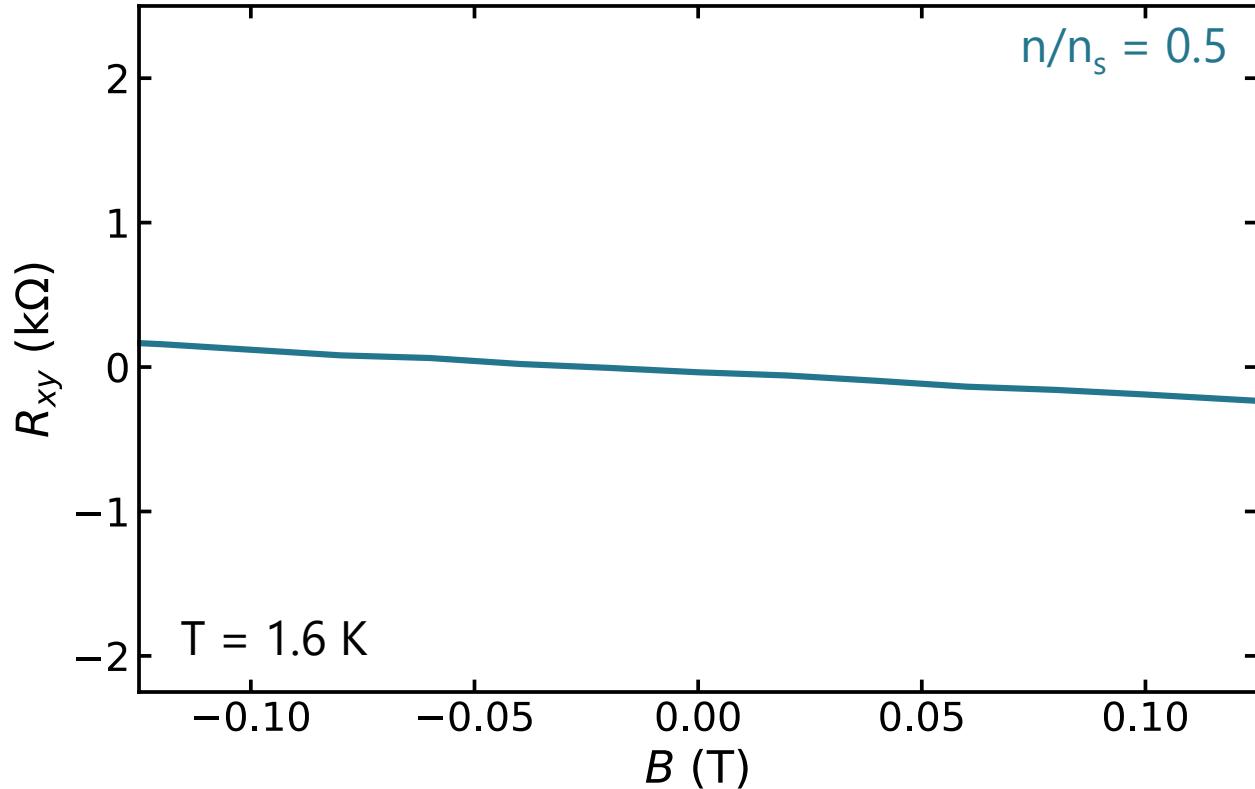
Visual hBN alignment



Visual hBN alignment

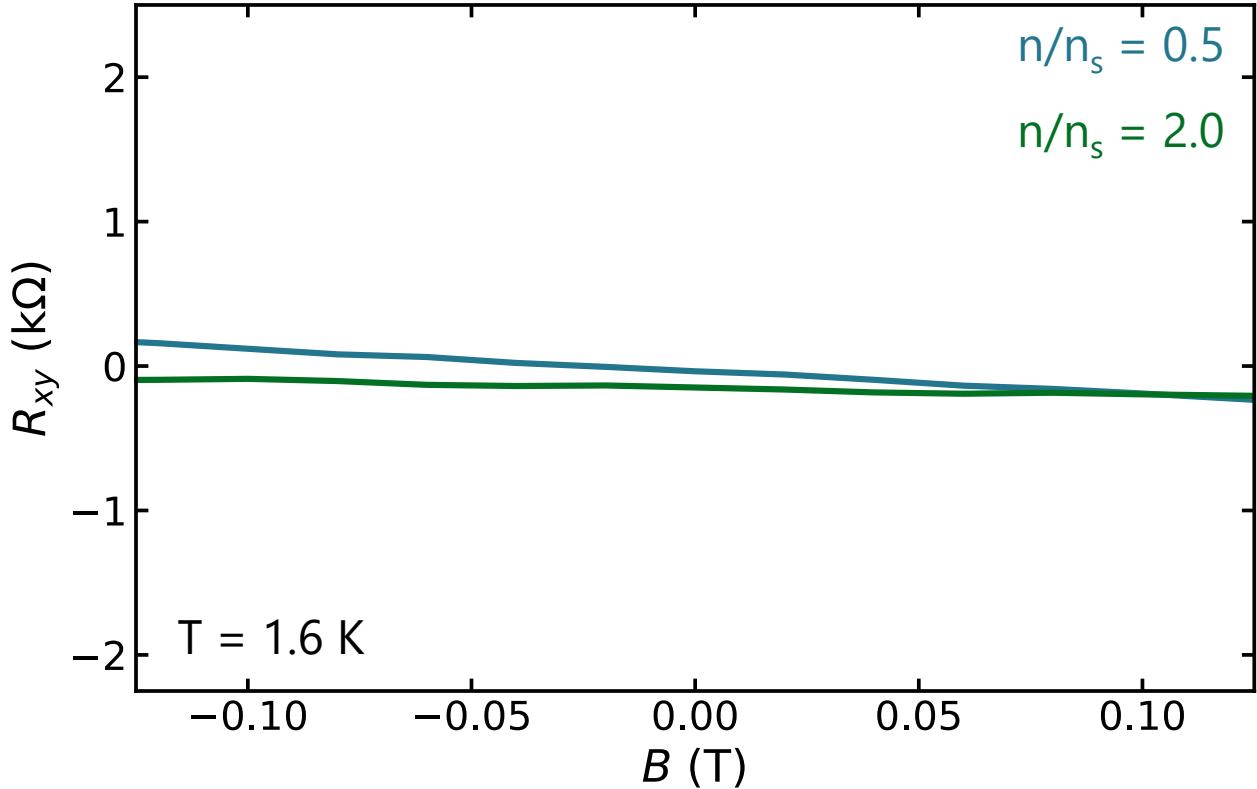


Hall slope density dependence



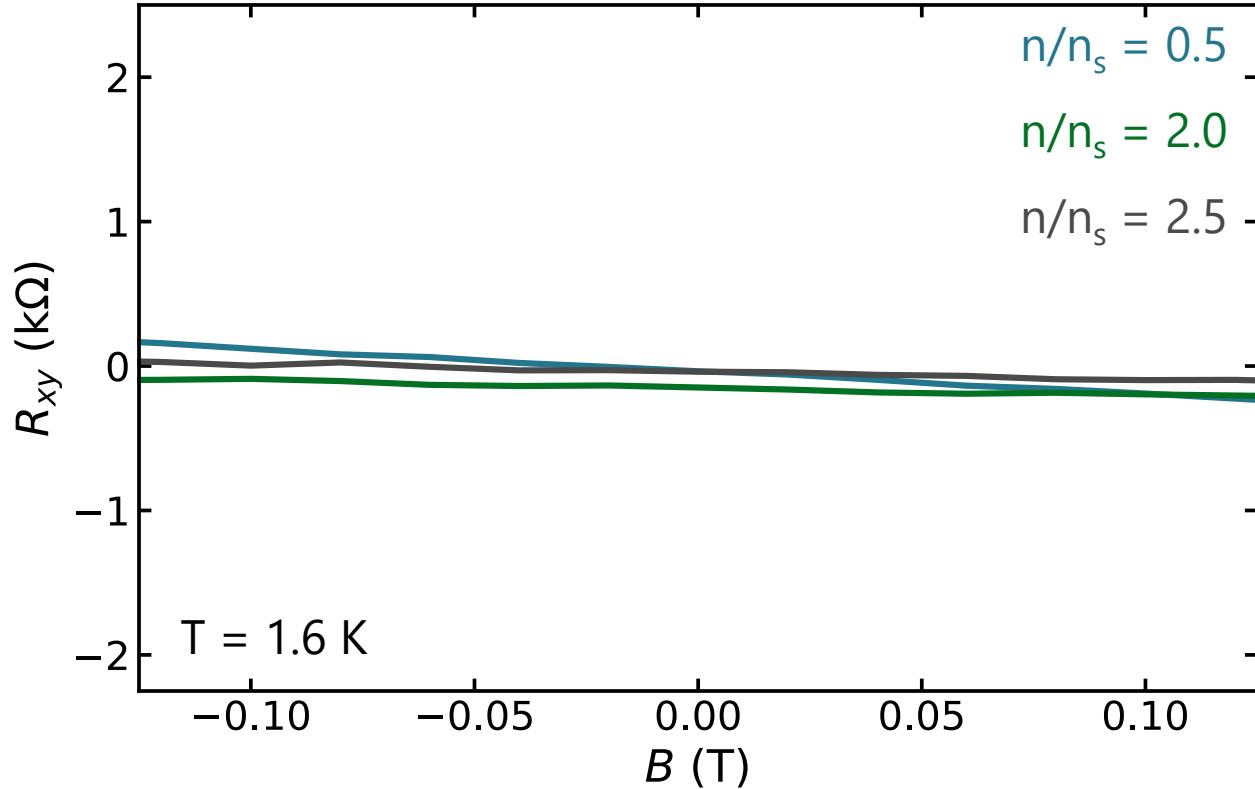
$$\text{Classical Hall: } R_{xy} = \frac{V_H}{I} = -\frac{B}{ne}$$

Hall slope density dependence



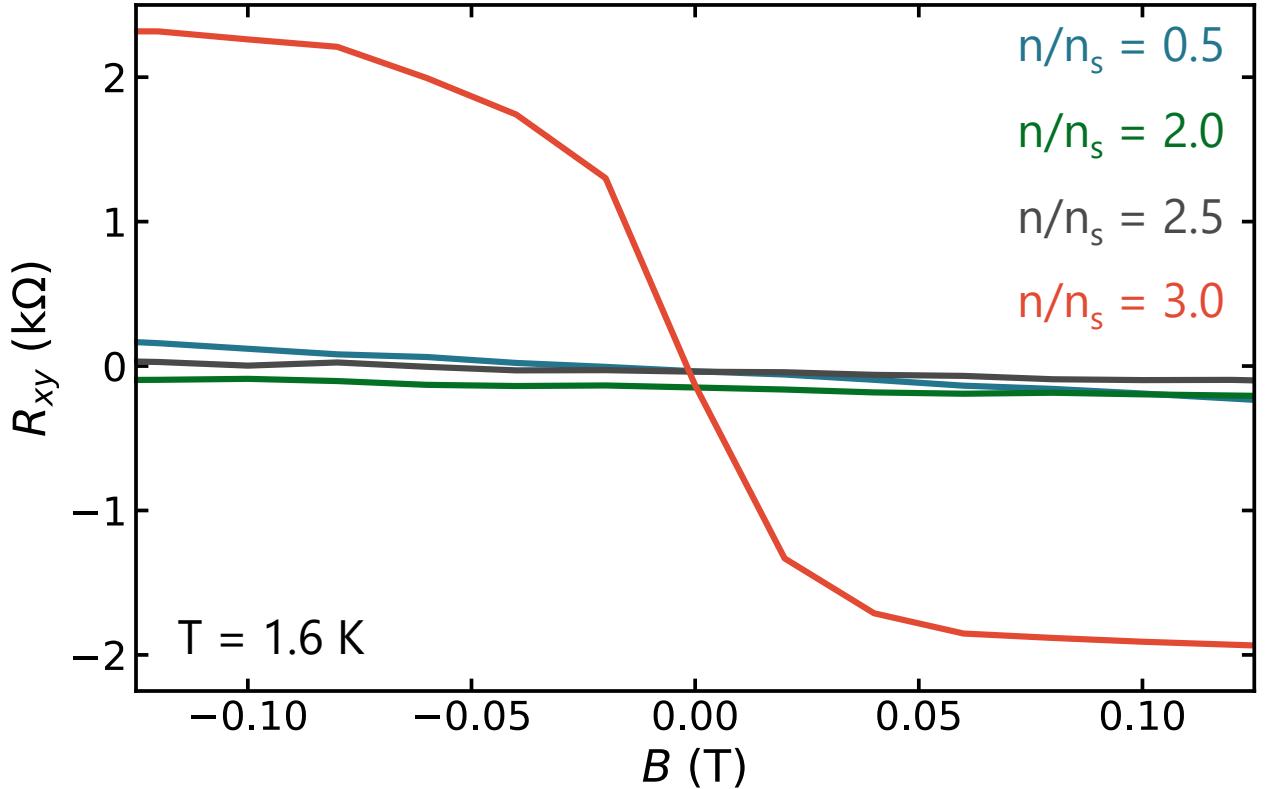
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Hall slope density dependence



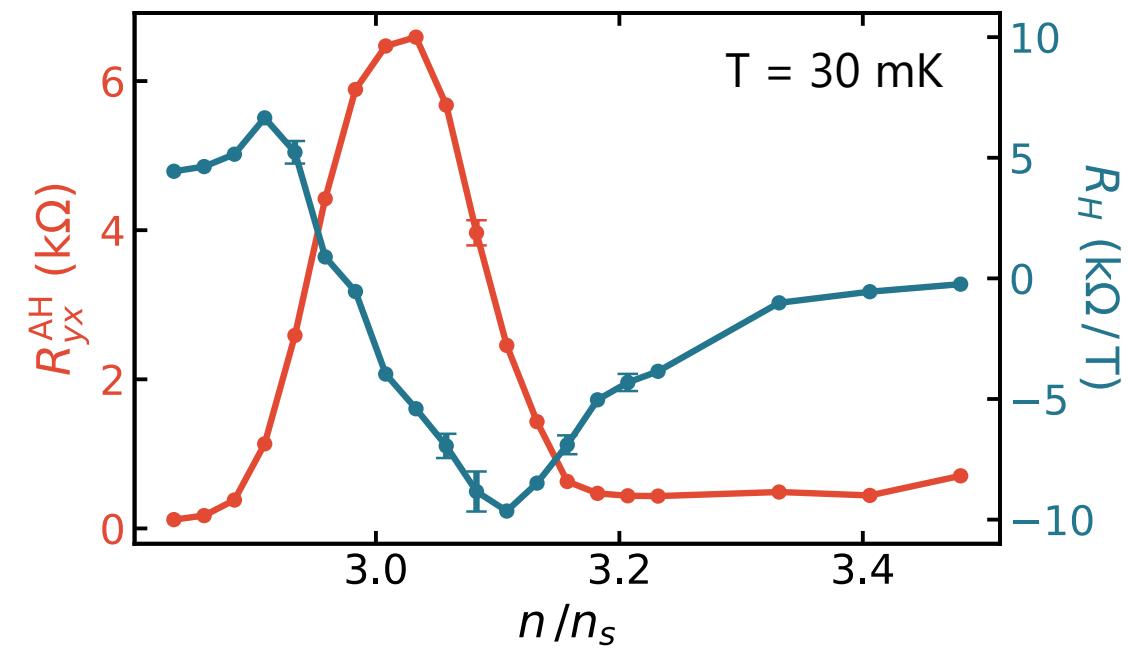
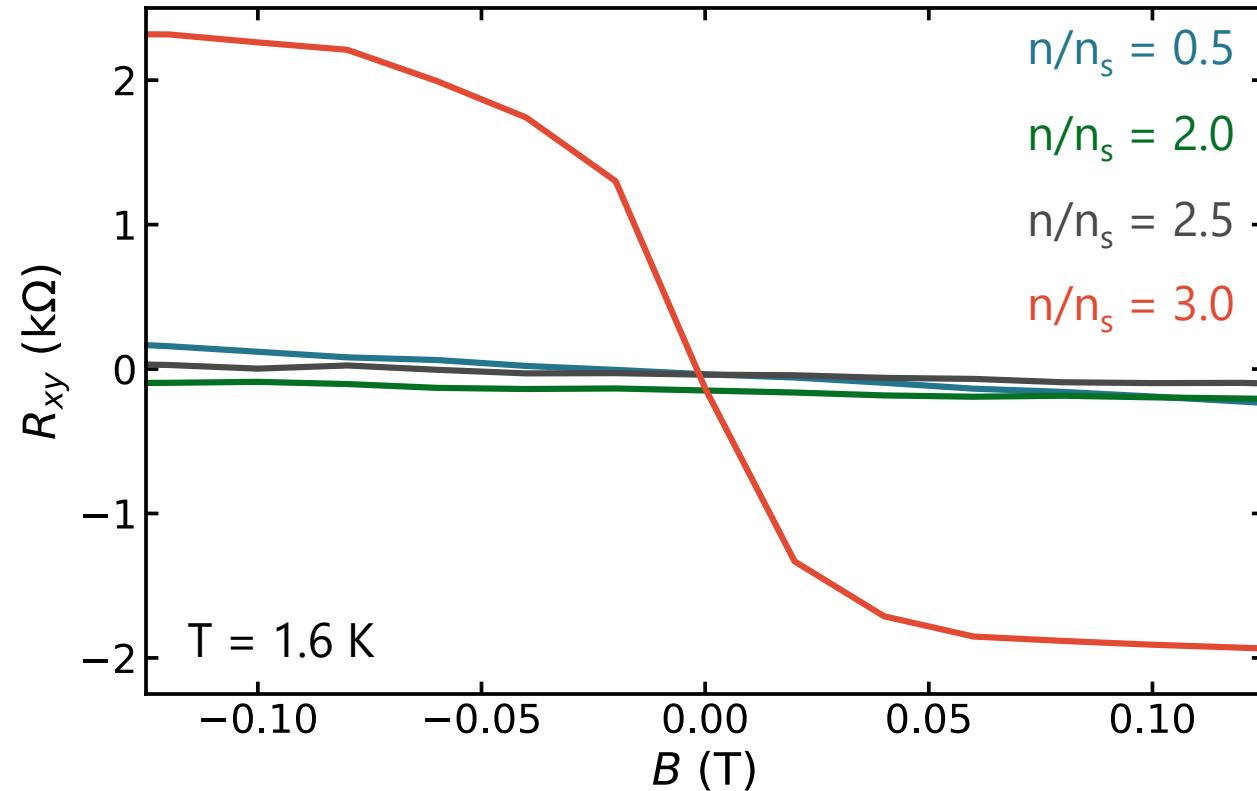
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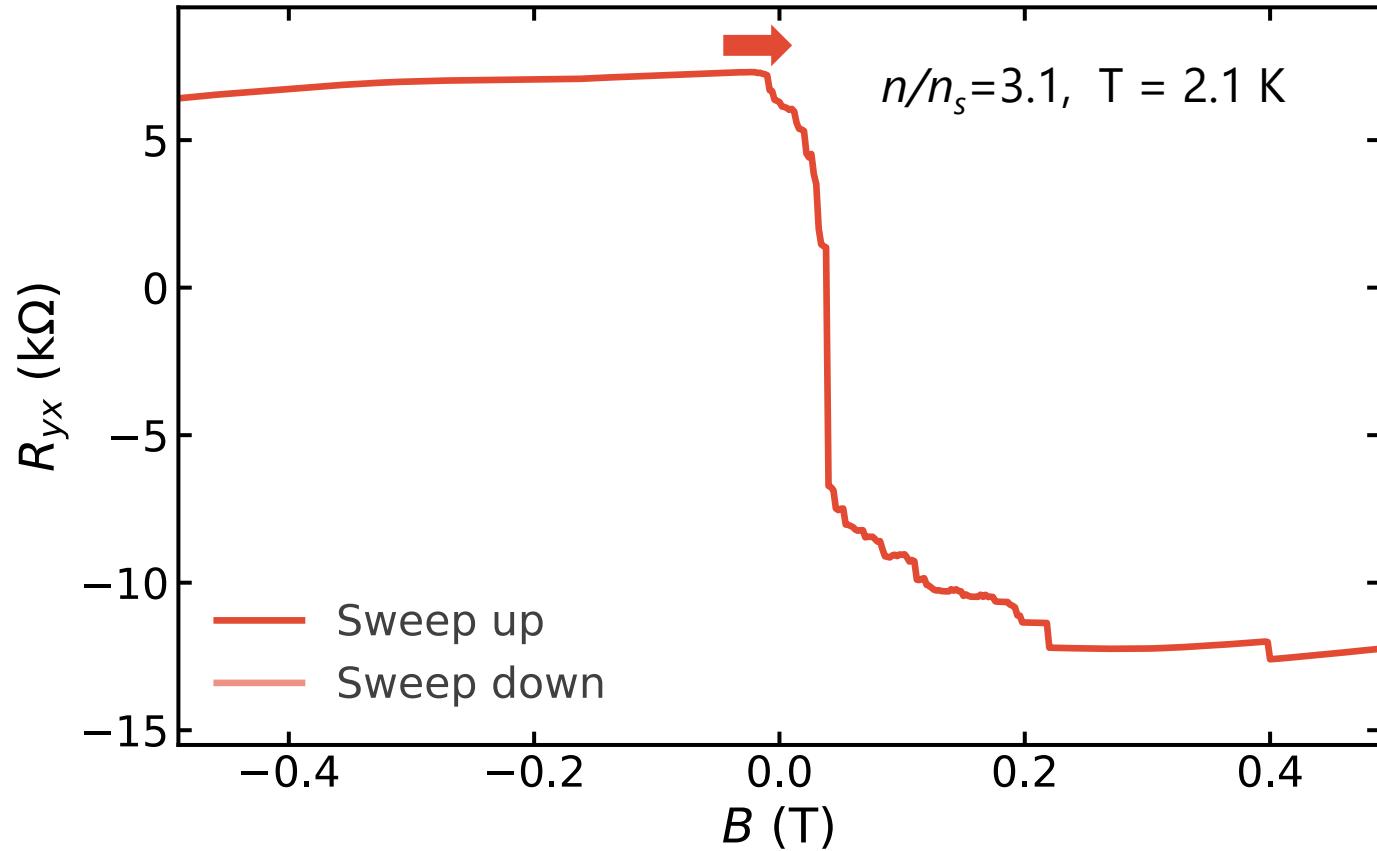
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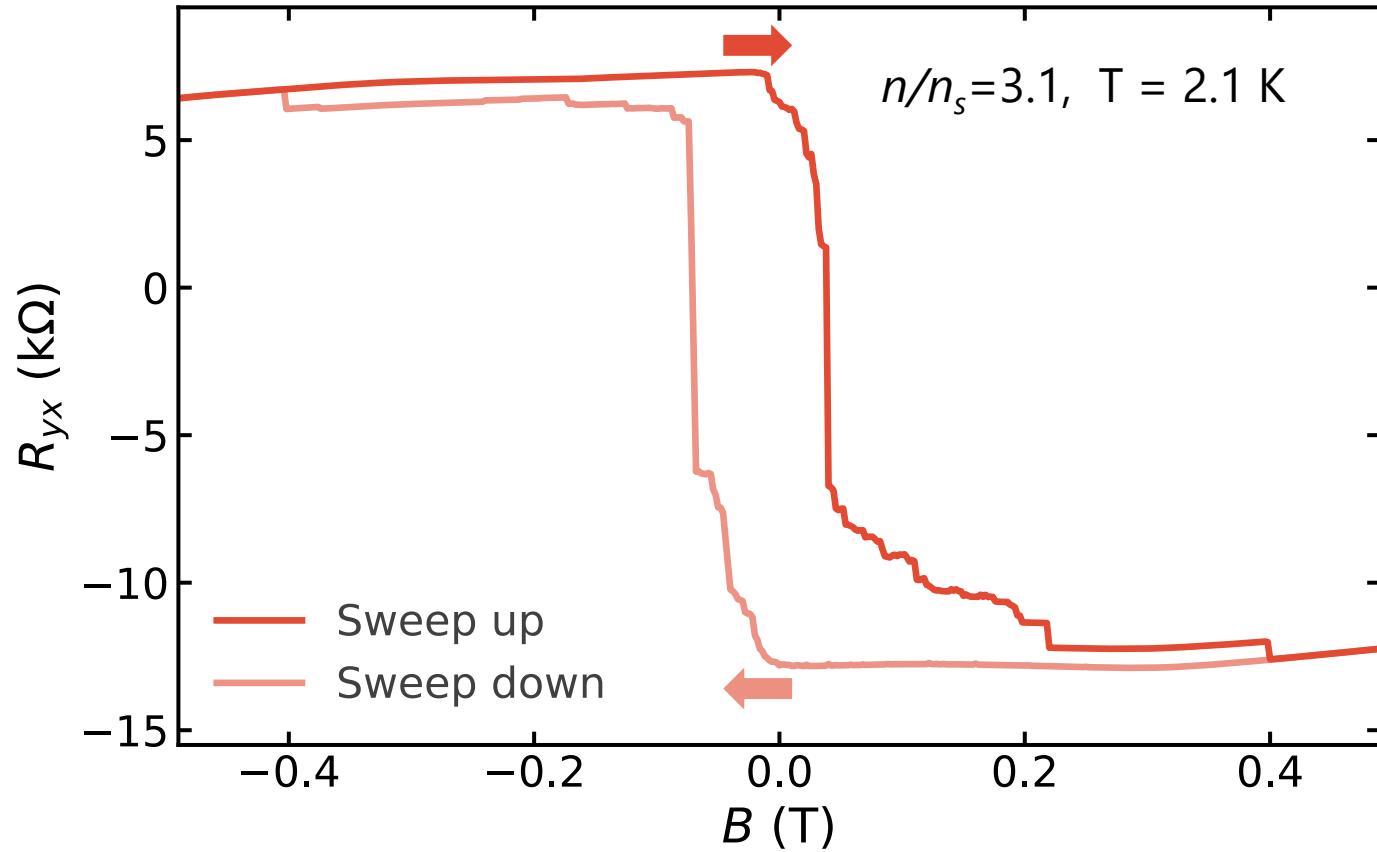
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Anomalous Hall signal can be really large!



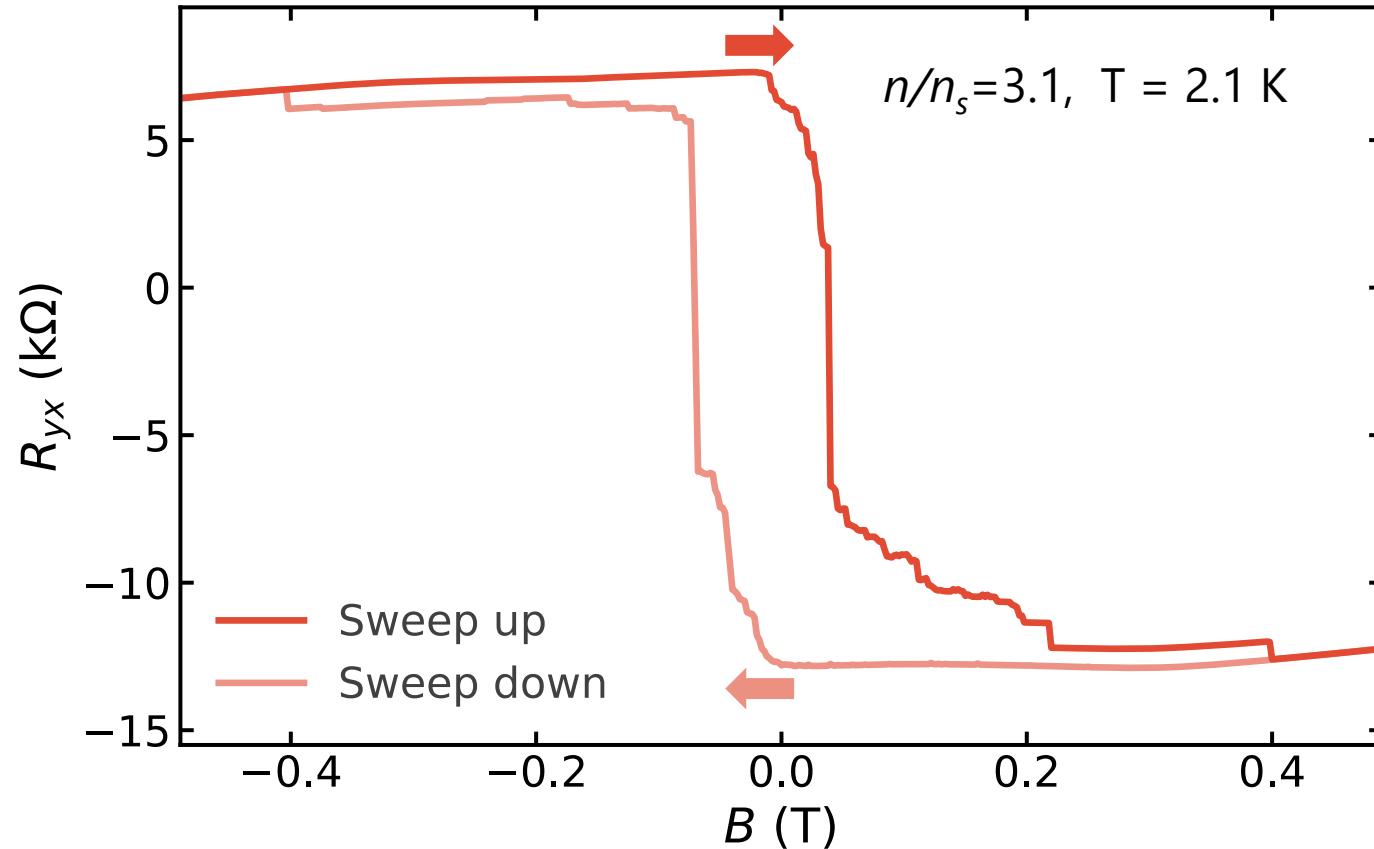
$$R_q = h/e^2 \approx 26 \text{ k}\Omega$$

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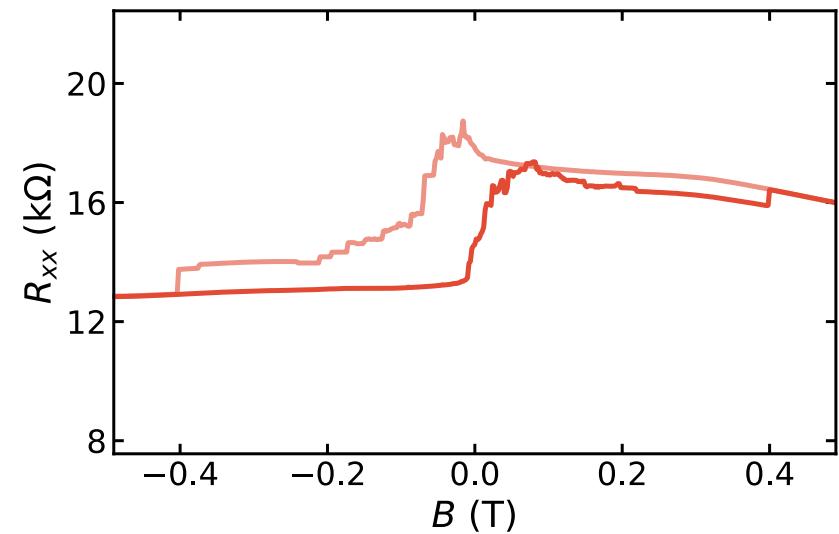


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Summary

By controlling twist angle, graphene multilayers can become strongly correlated!

Large anomalous Hall (and R_{xx})

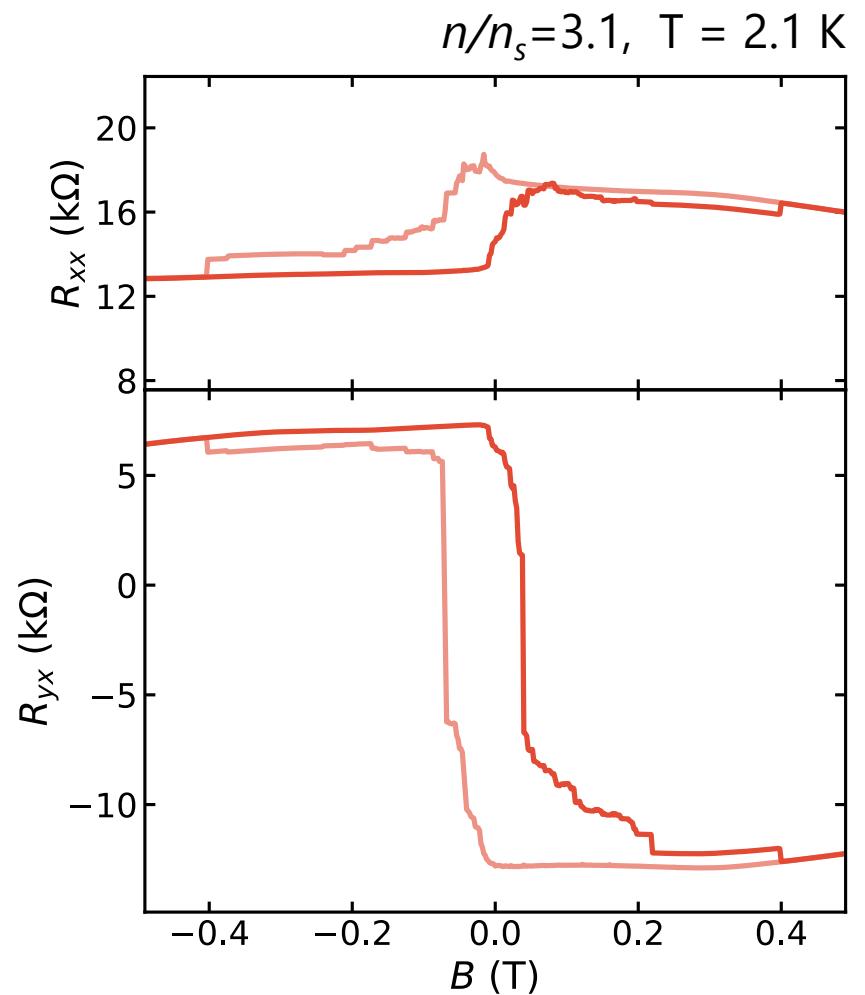
Apparent insulating state

Non-local transport

Survives up to $\sim 5\text{K}$

Evidence of domains

Reminiscent of early Magnetic Tis \rightarrow Chern insulator?



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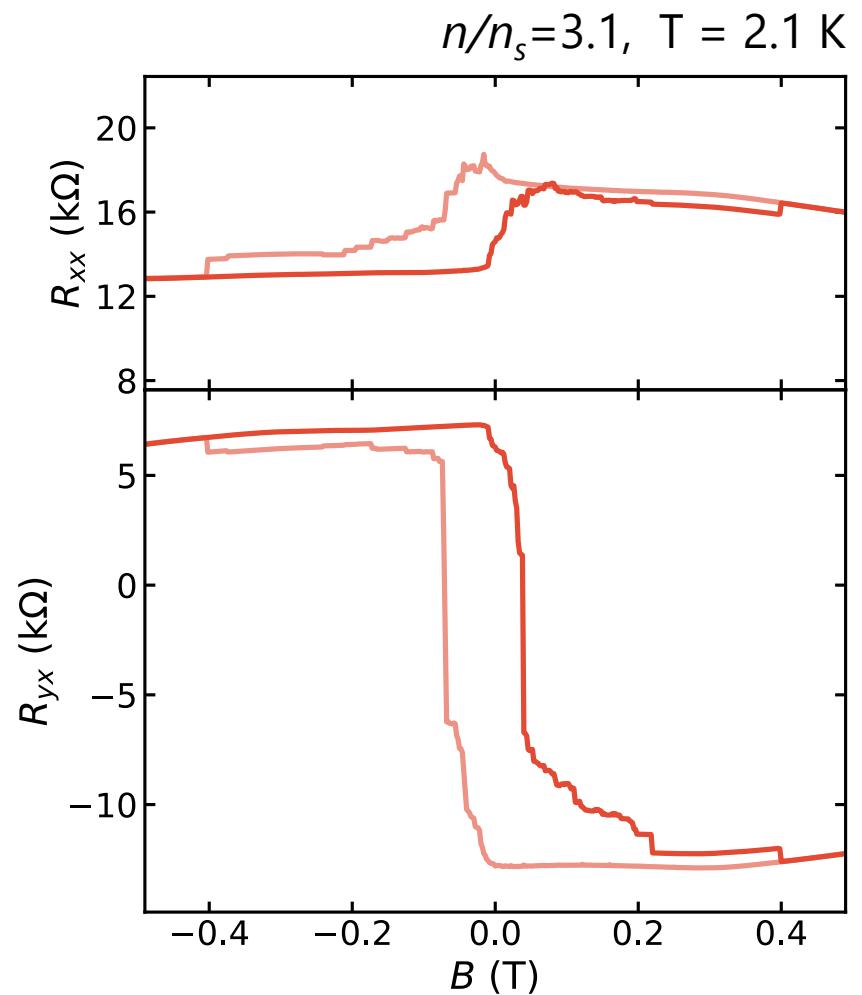
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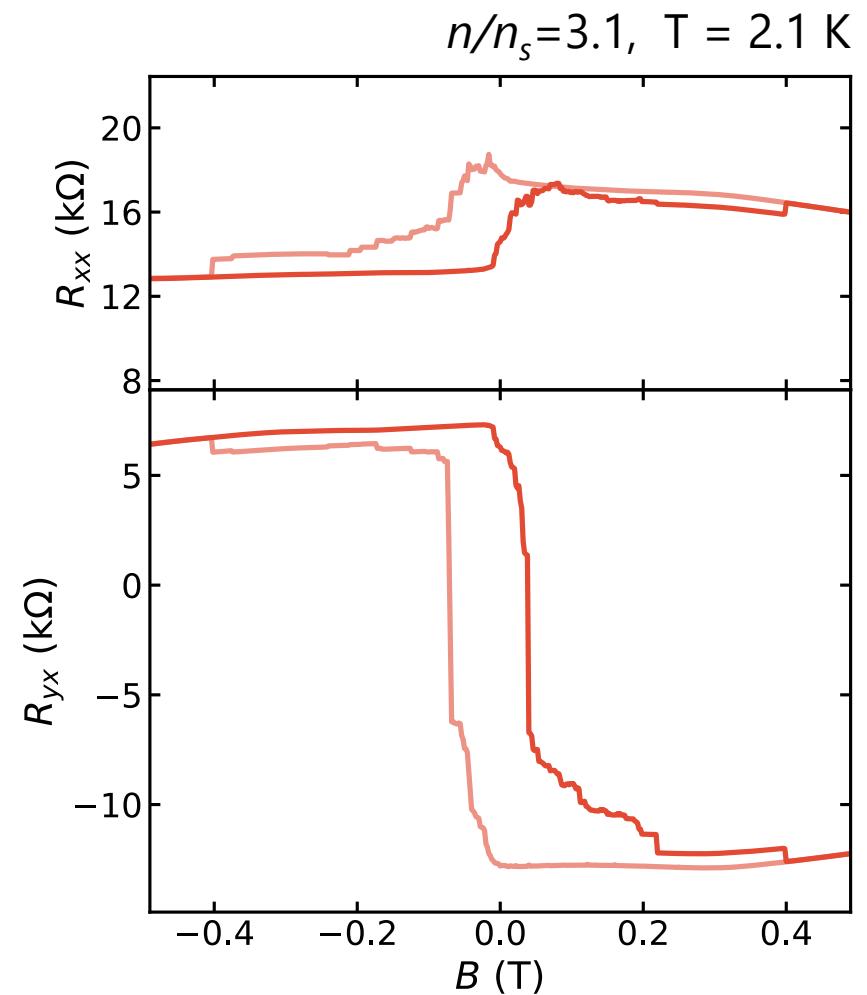
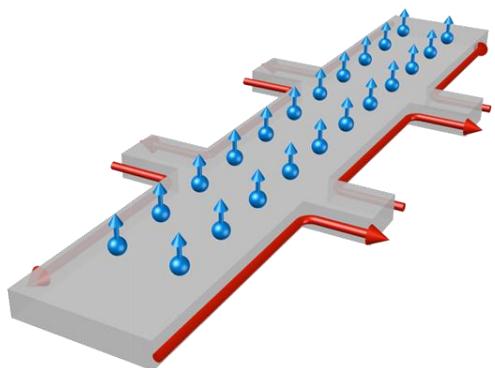
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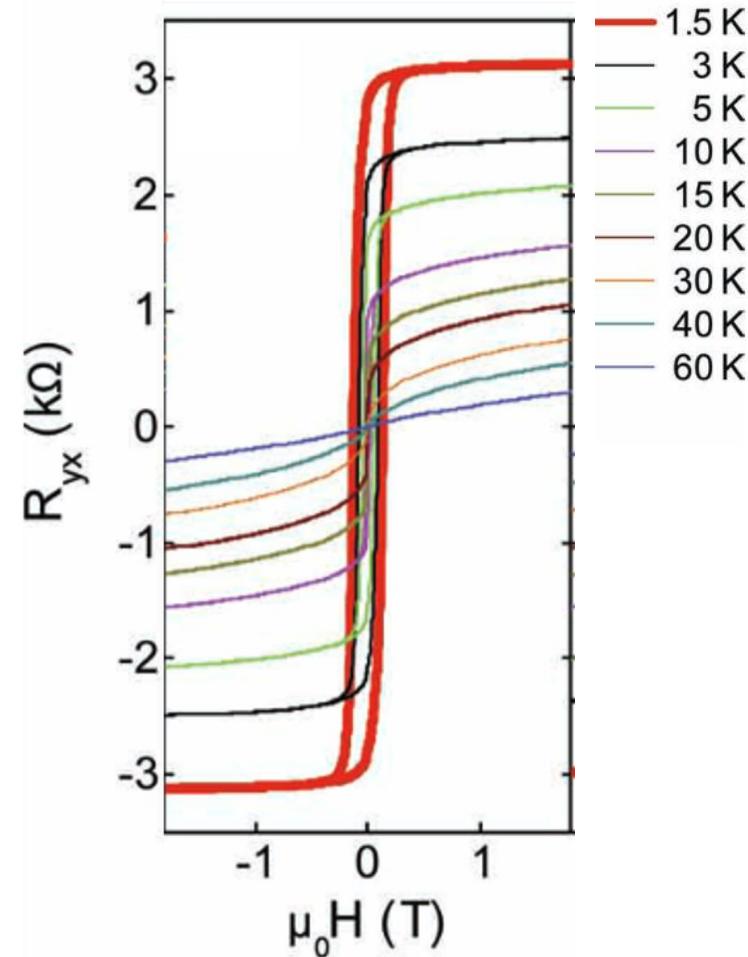
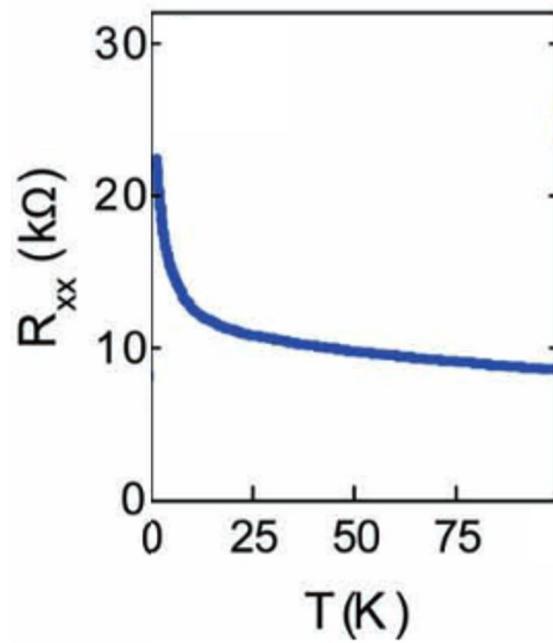
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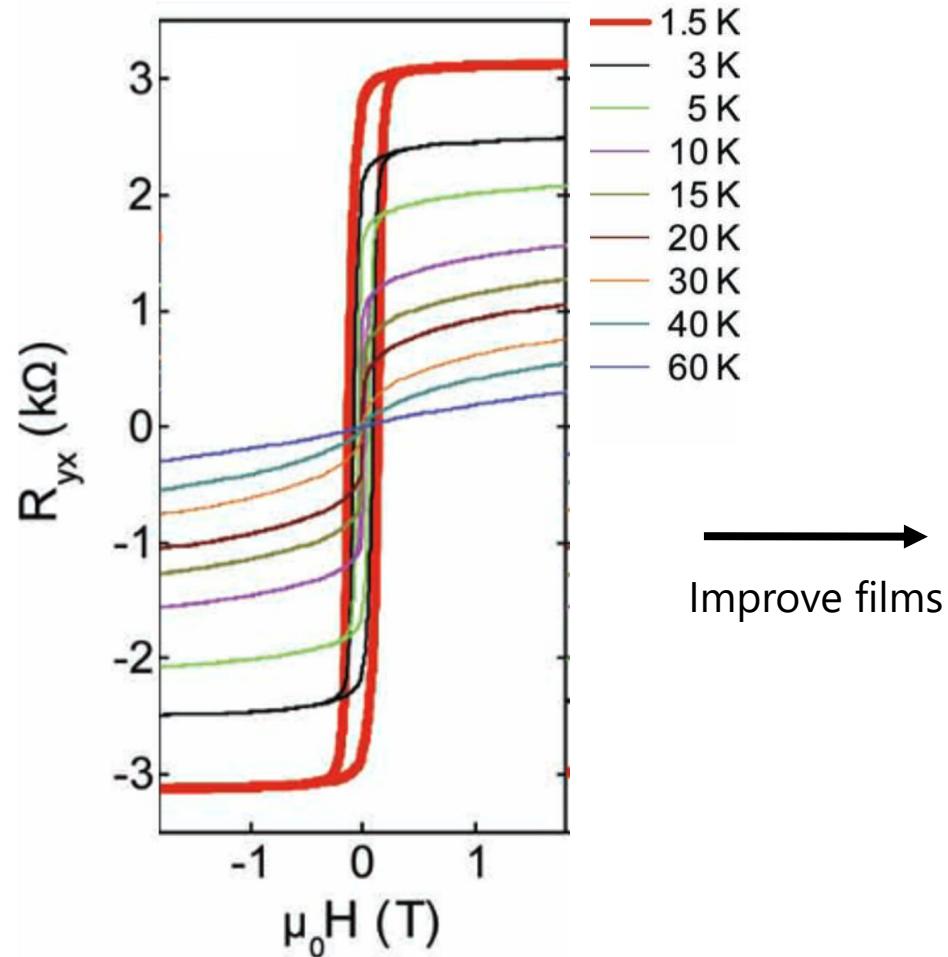
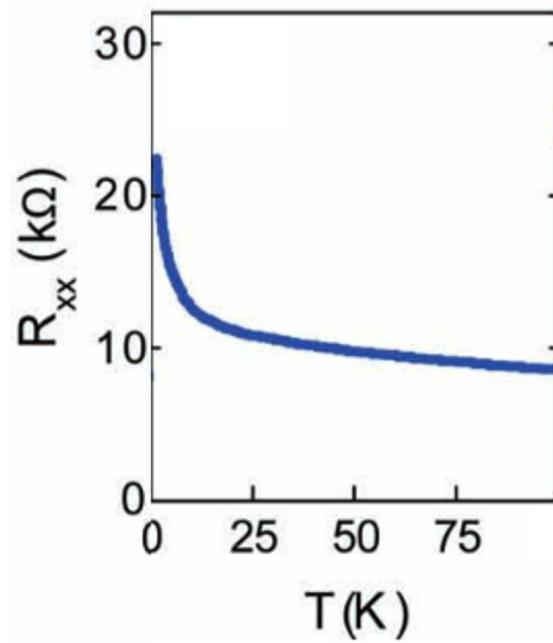
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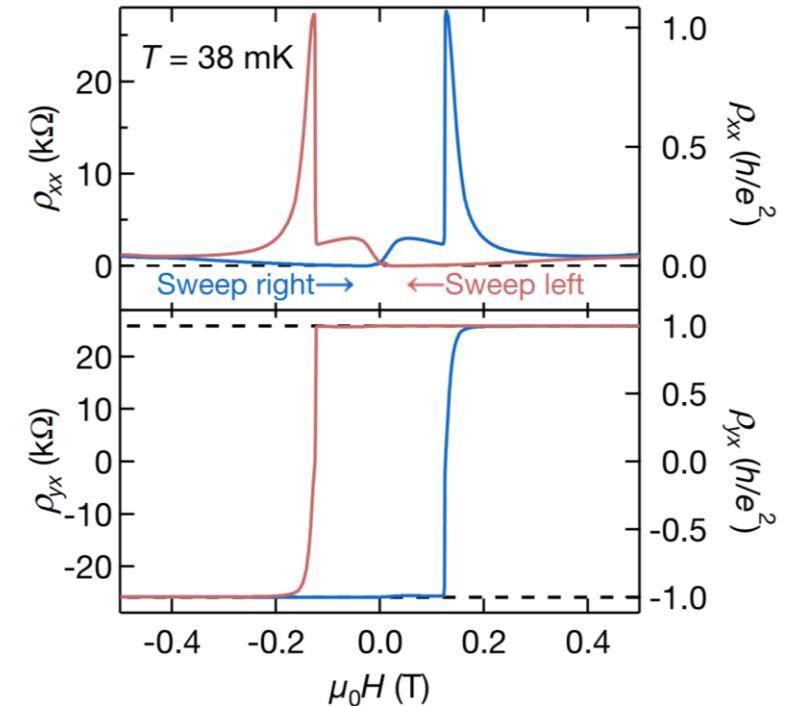
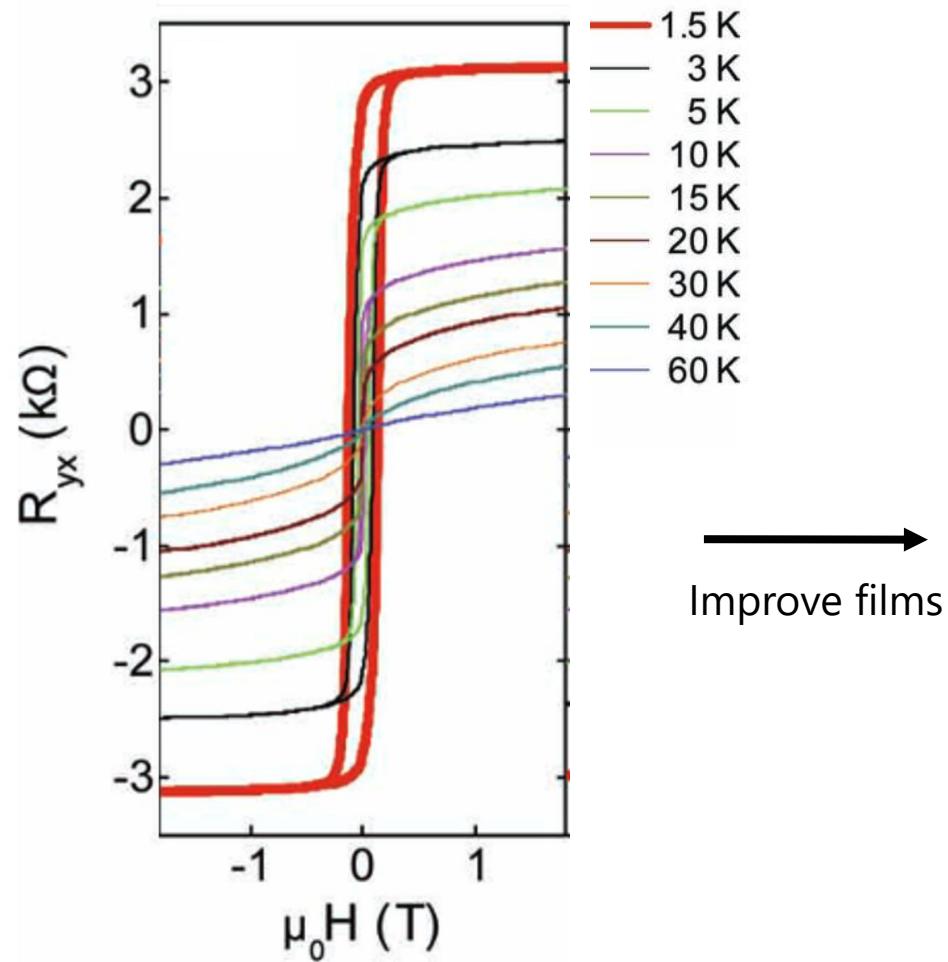
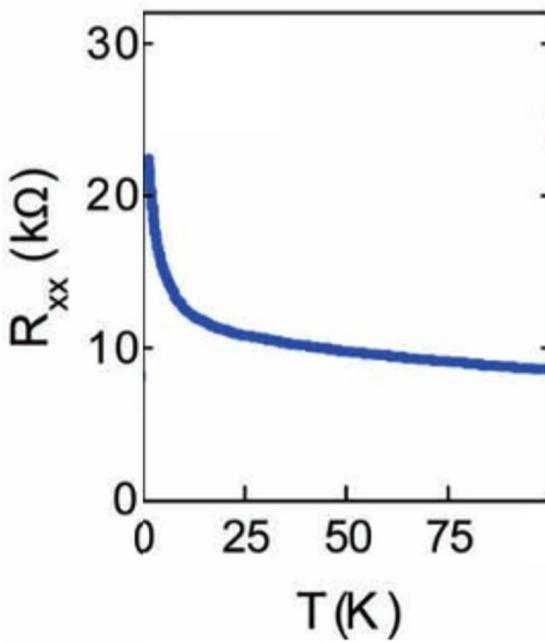
Early magnetically doped topological insulators



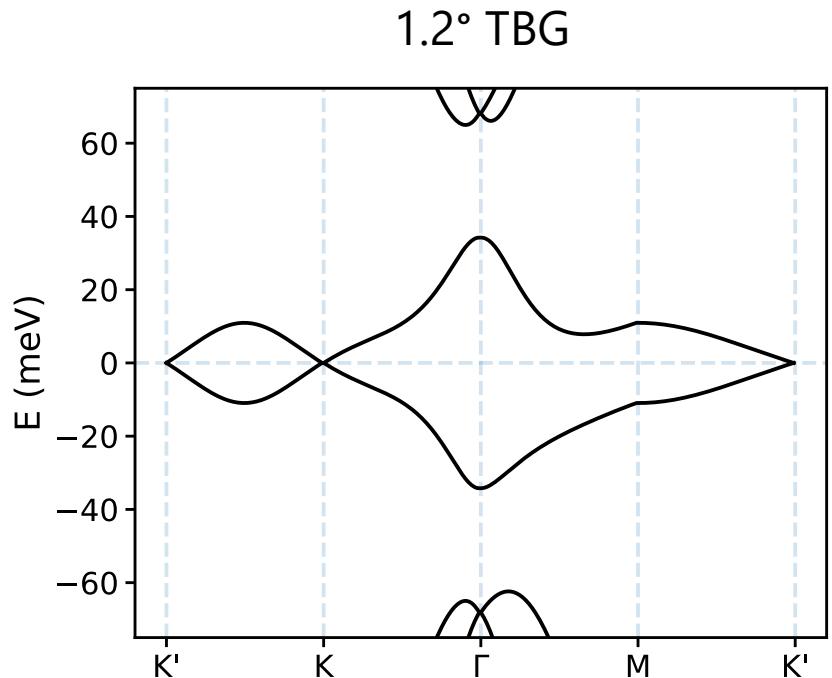
Early magnetically doped topological insulators



Early magnetically doped topological insulators



hBN alignment is the key for AH?



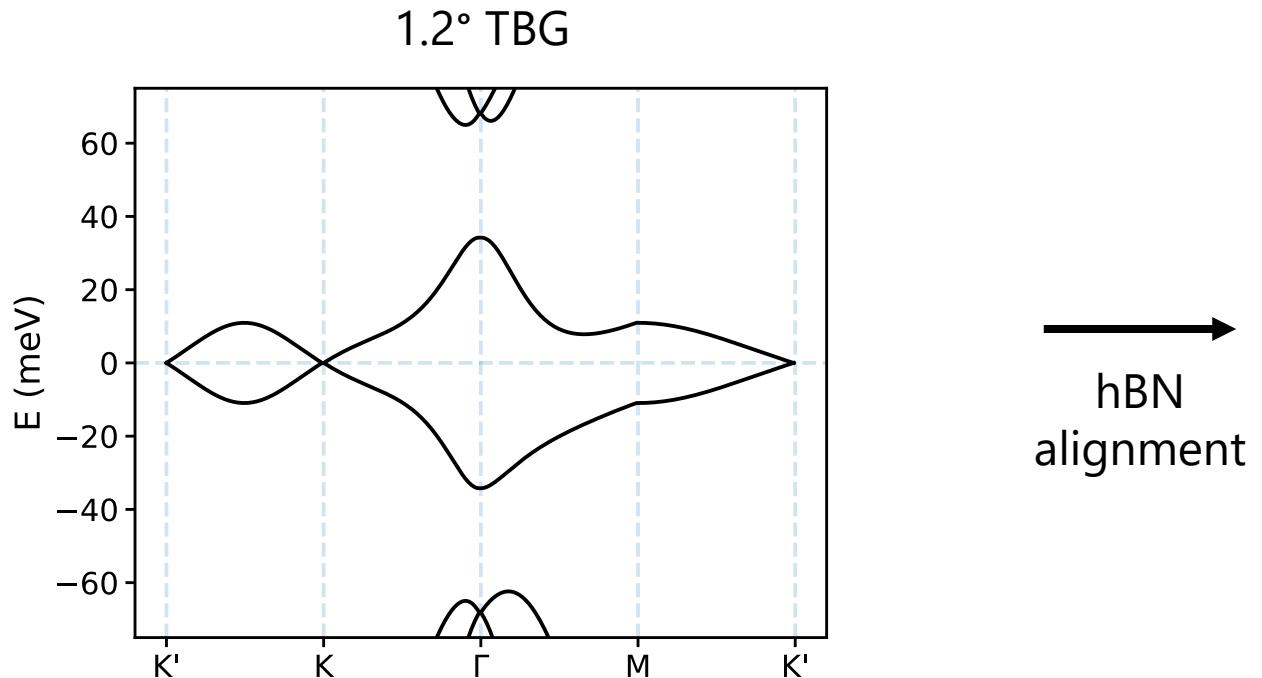
Theory:

Zhang, et al. PRB (2019)

Bultinck, et al. PRL (2020)

Xie and MacDonald, PRL (2020) – Gap open spontaneously?

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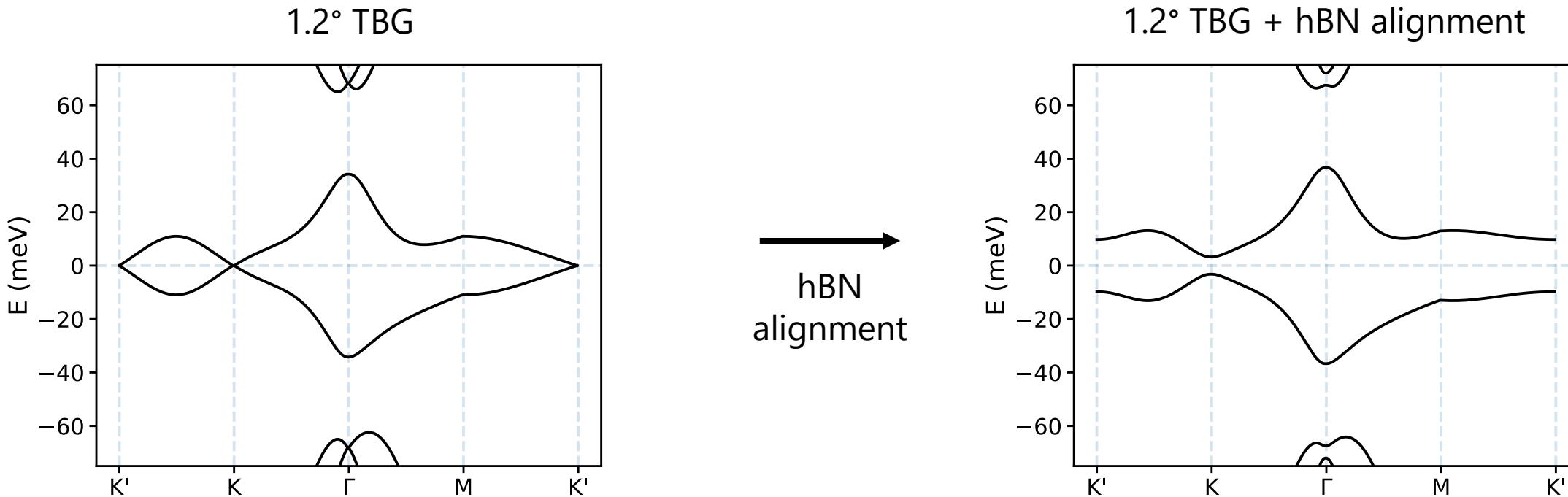
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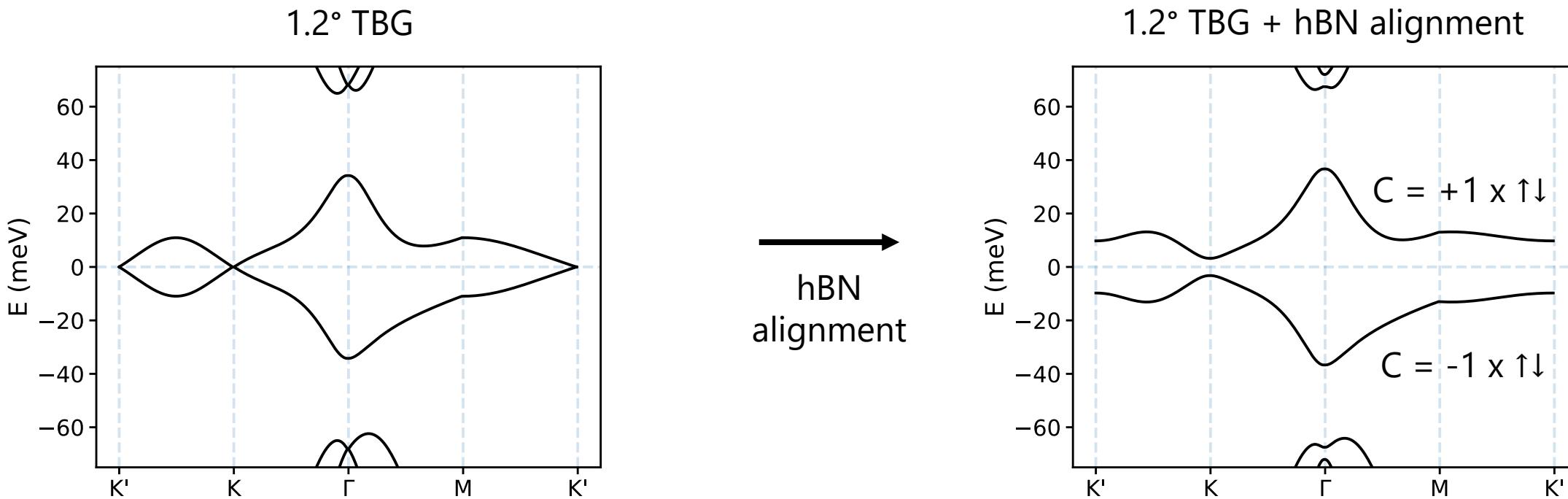
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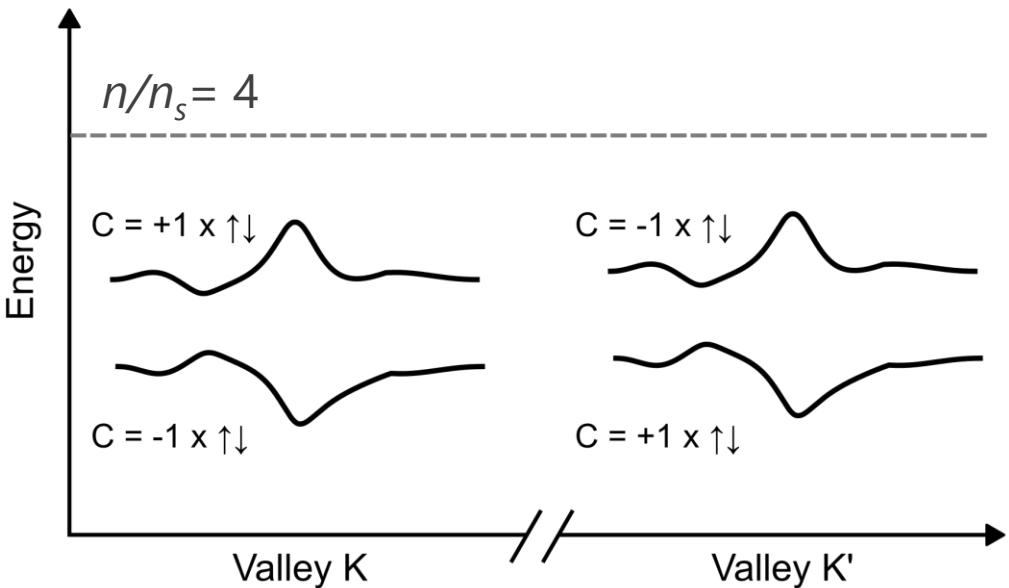
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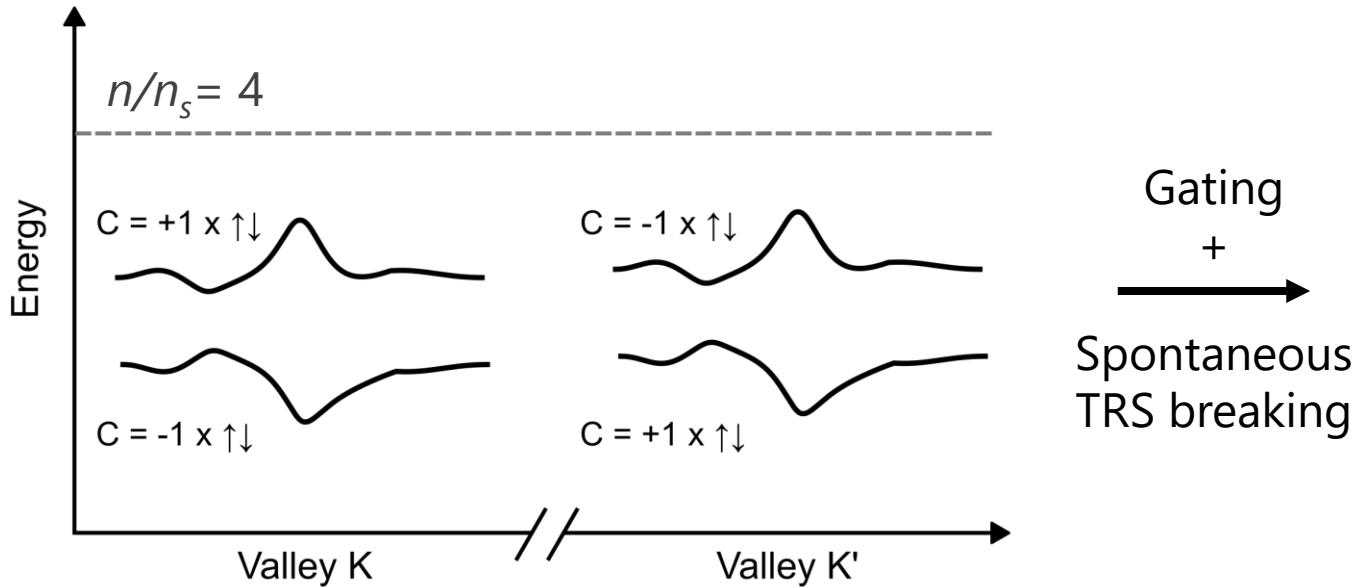
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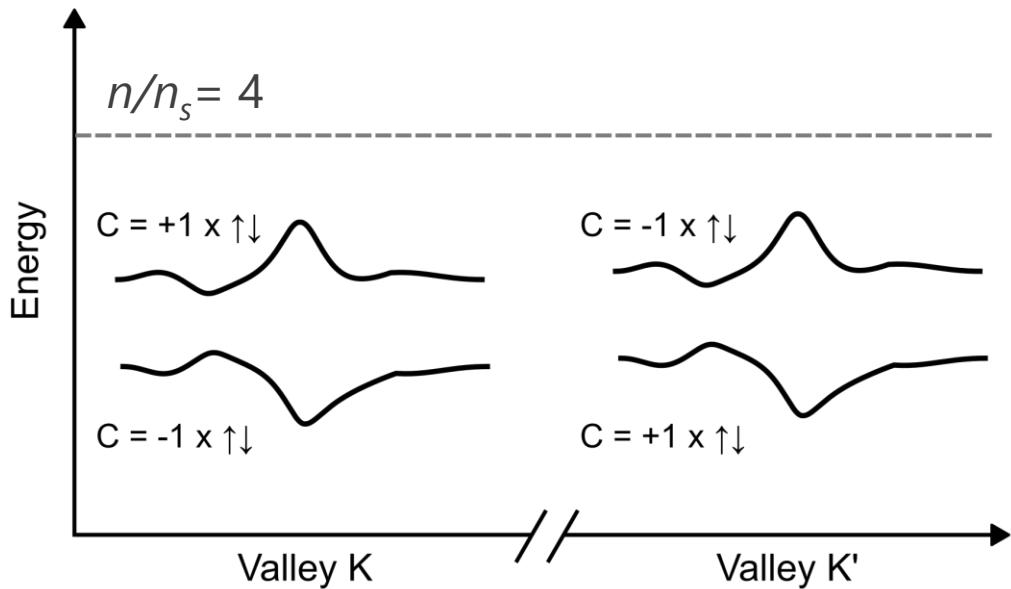
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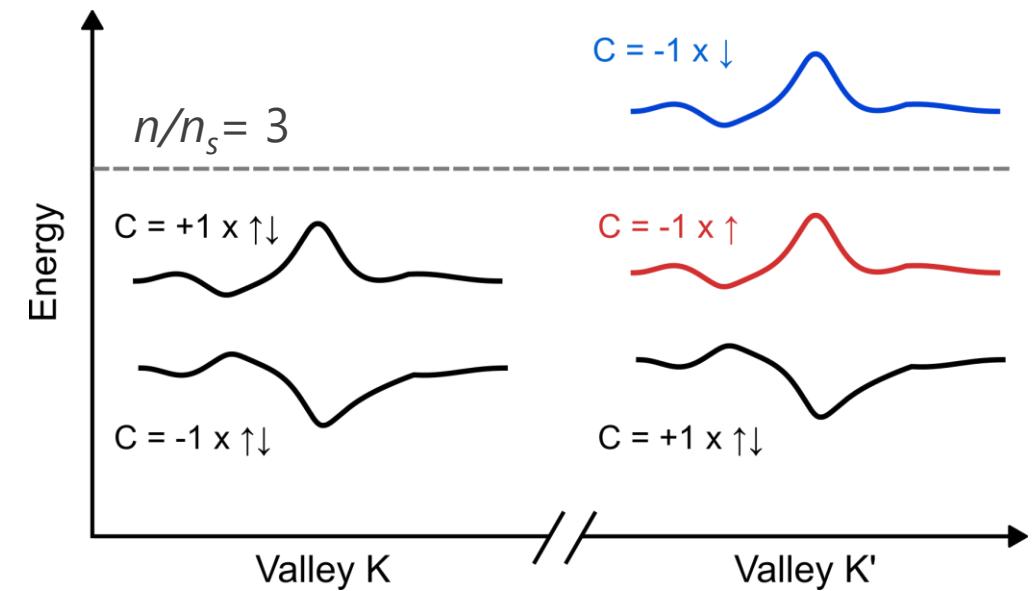
Bultinck, et al. PRL (2020)

Xie and MacDonald, PRL (2020) – Gap open spontaneously?

hBN alignment is the key for AH?



Gating
+
Spontaneous TRS breaking

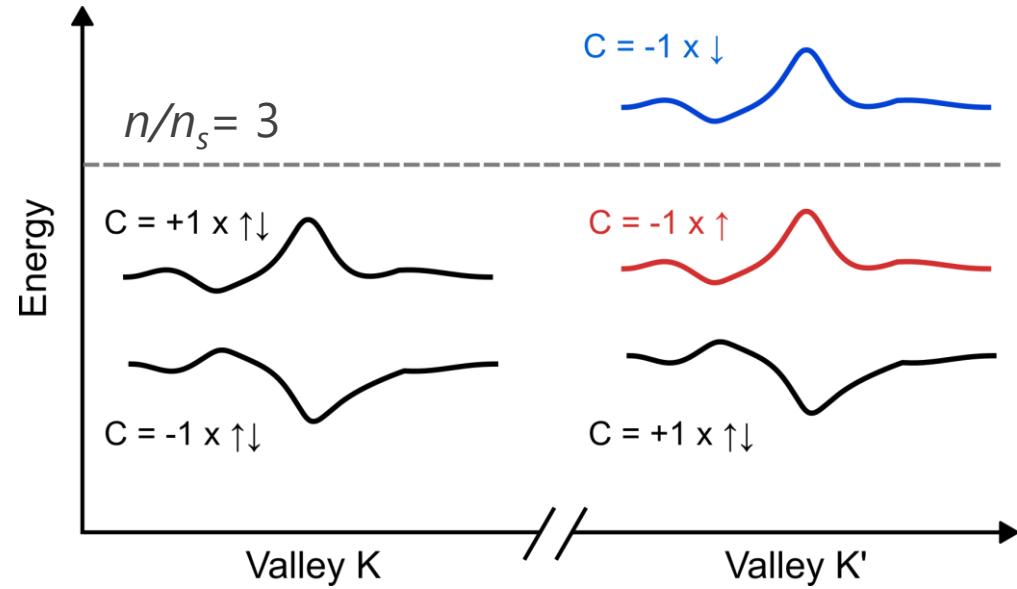


Theory:

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Possible Scenarios and in-plane response

Interaction driven spin/valley polarization



Xie et al. PRL (2020)

Zhang et al. PRR (2019)

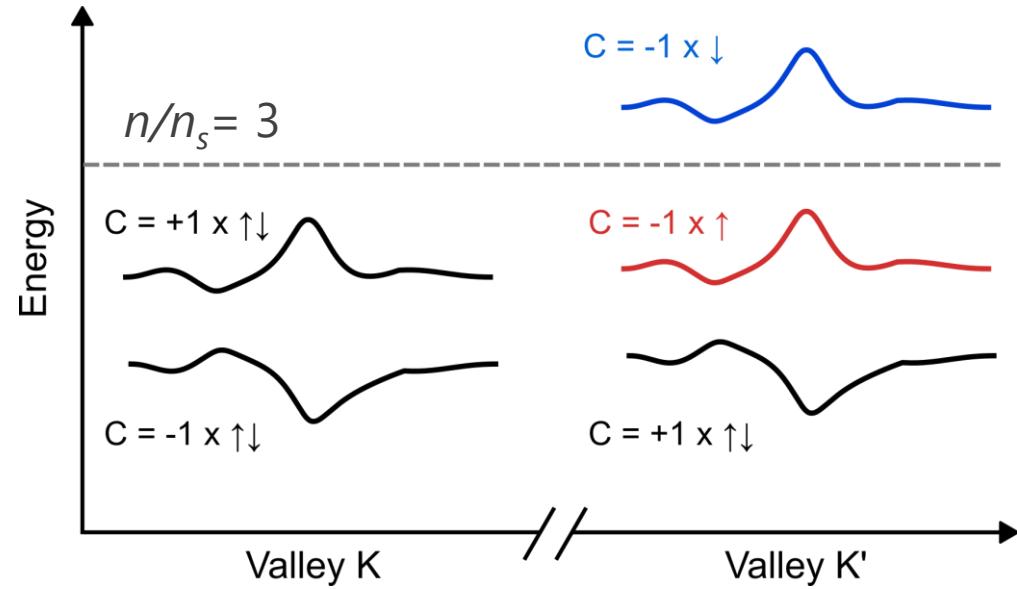
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Spin?



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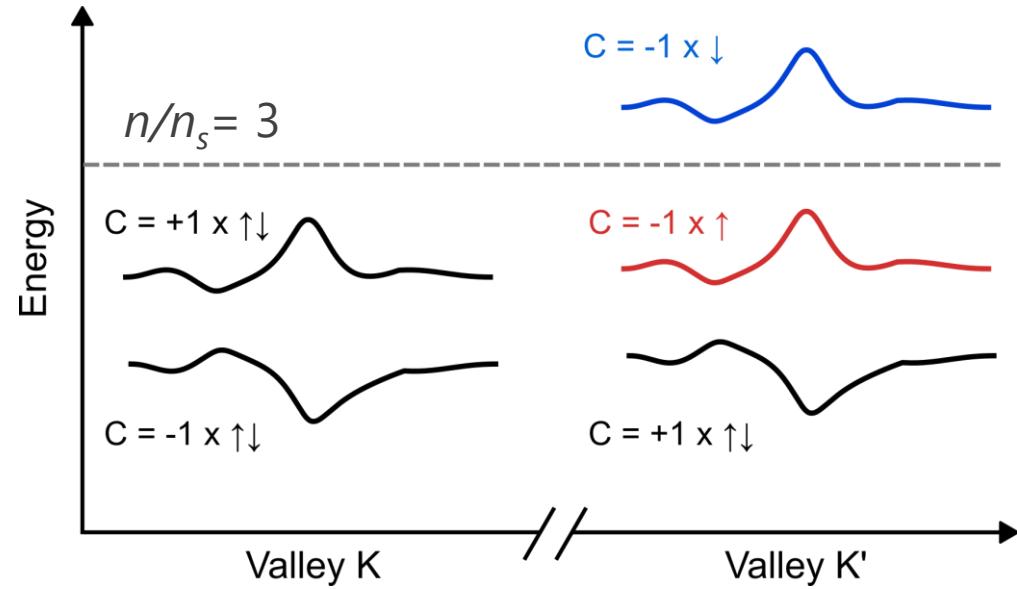
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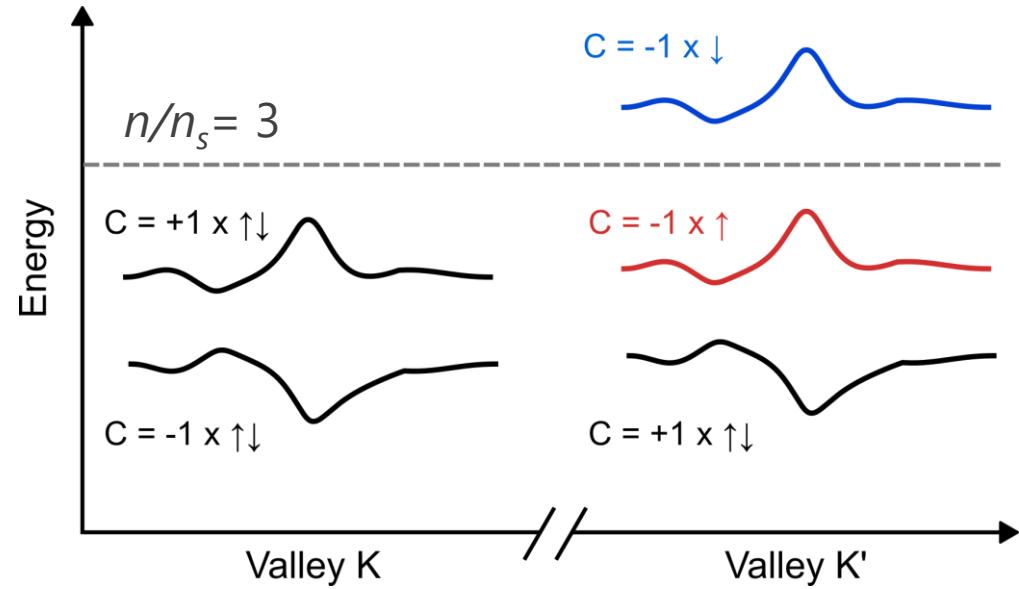


Orbital?

- Xie et al. PRL (2020)
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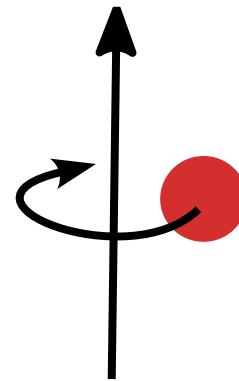
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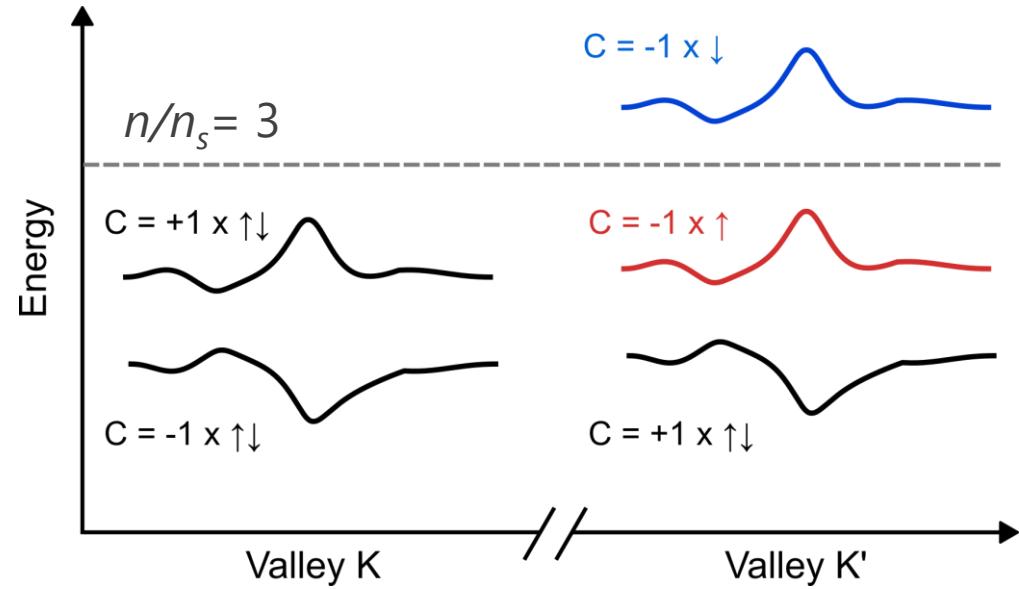
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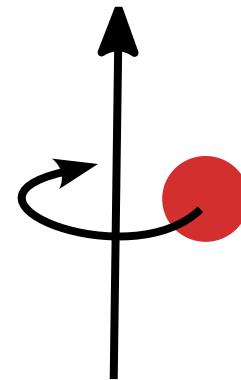
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Orbital?



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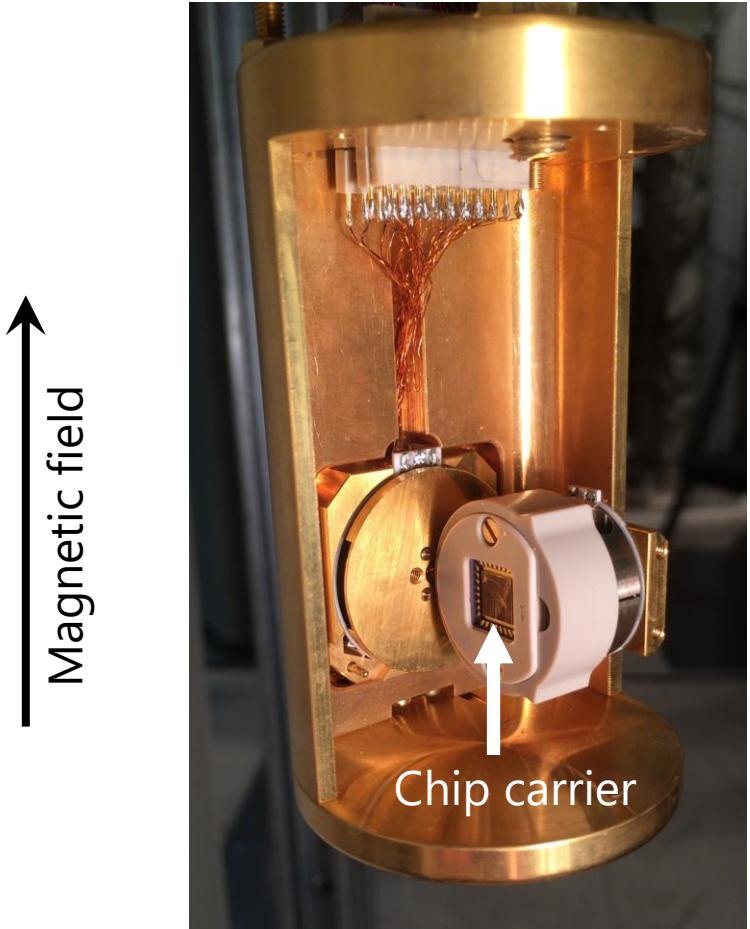
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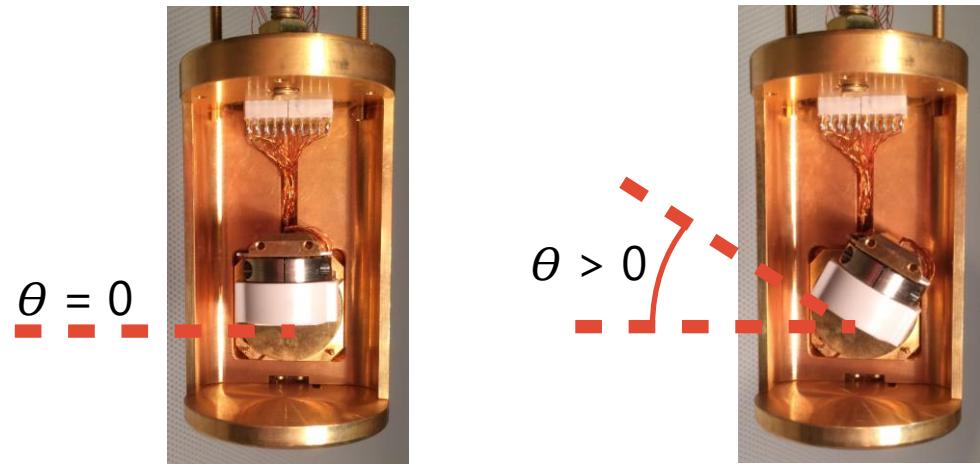
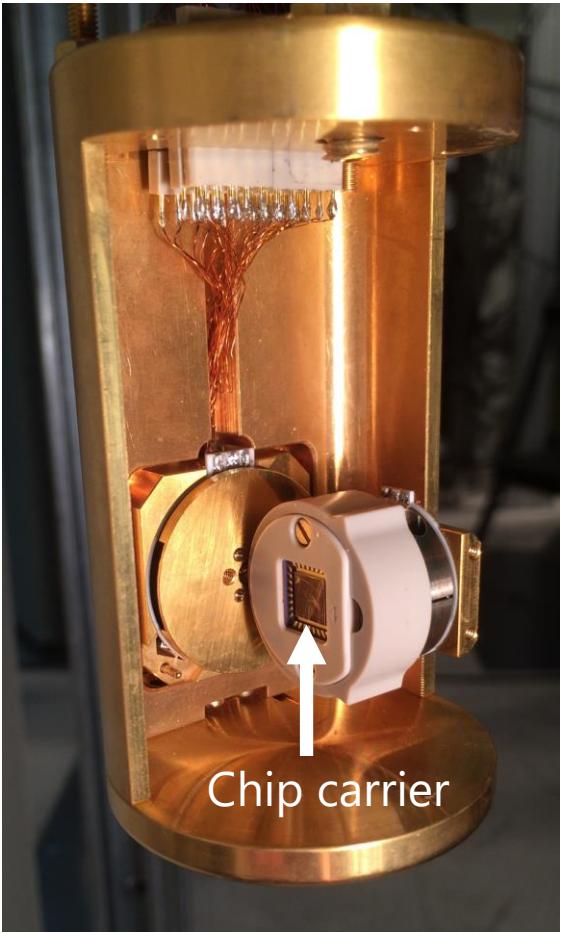
In-plane field can couple to valley!

Probing Nature of Magnetism



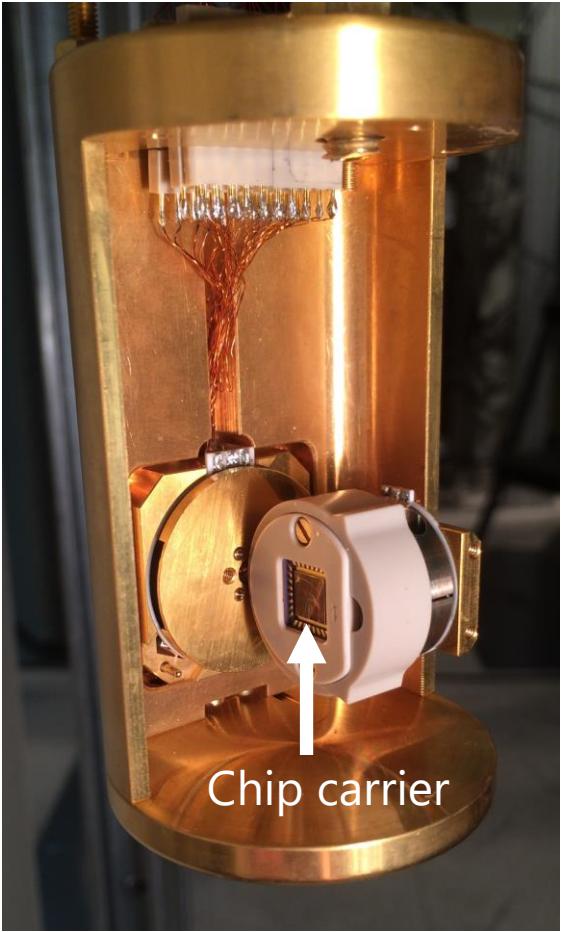
Probing Nature of Magnetism

Magnetic field
↑



Probing Nature of Magnetism

Magnetic field
↑



$$\theta = 0$$



$$\theta > 0$$



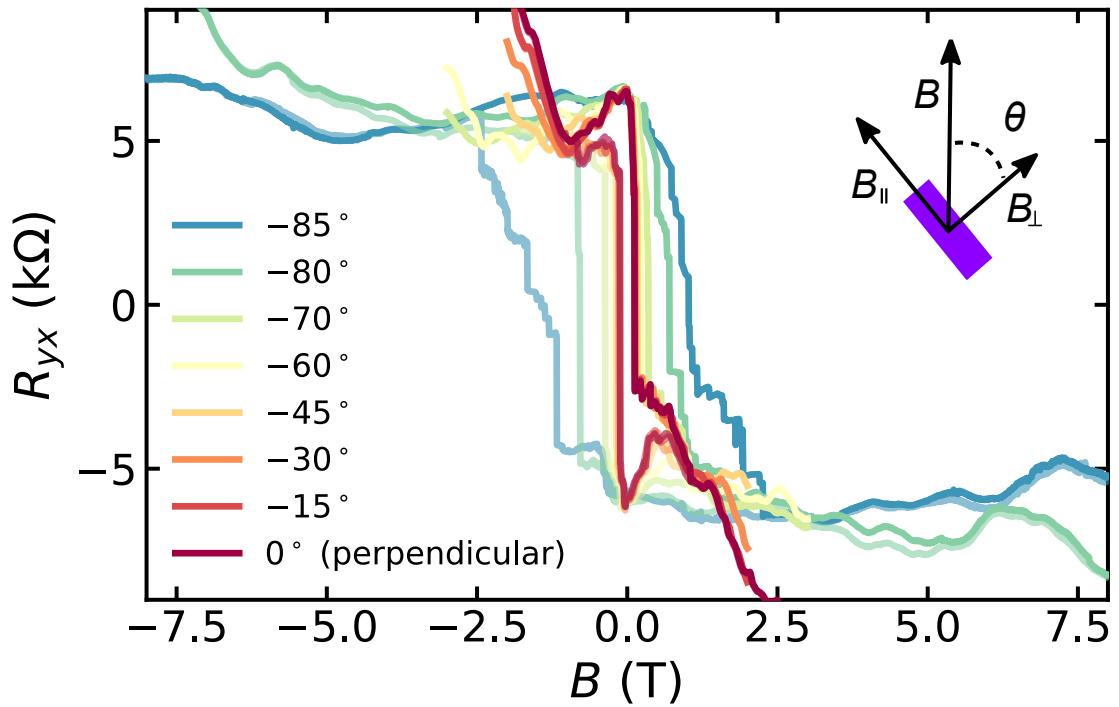
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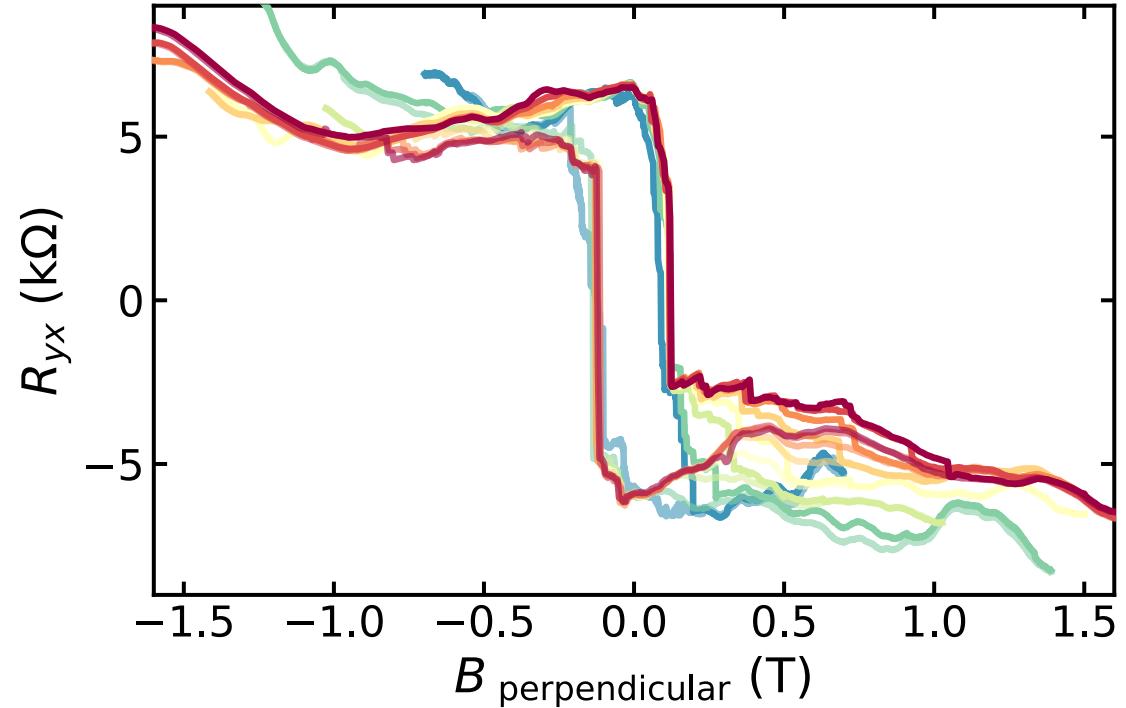
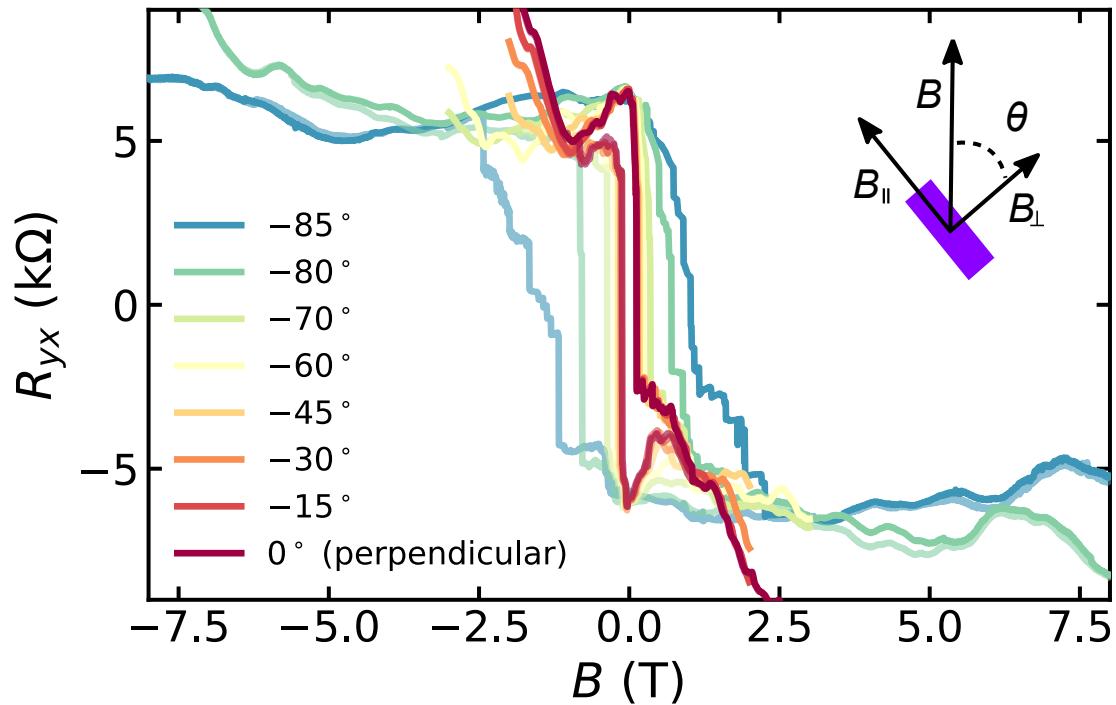


Hysteresis loops in tilted field



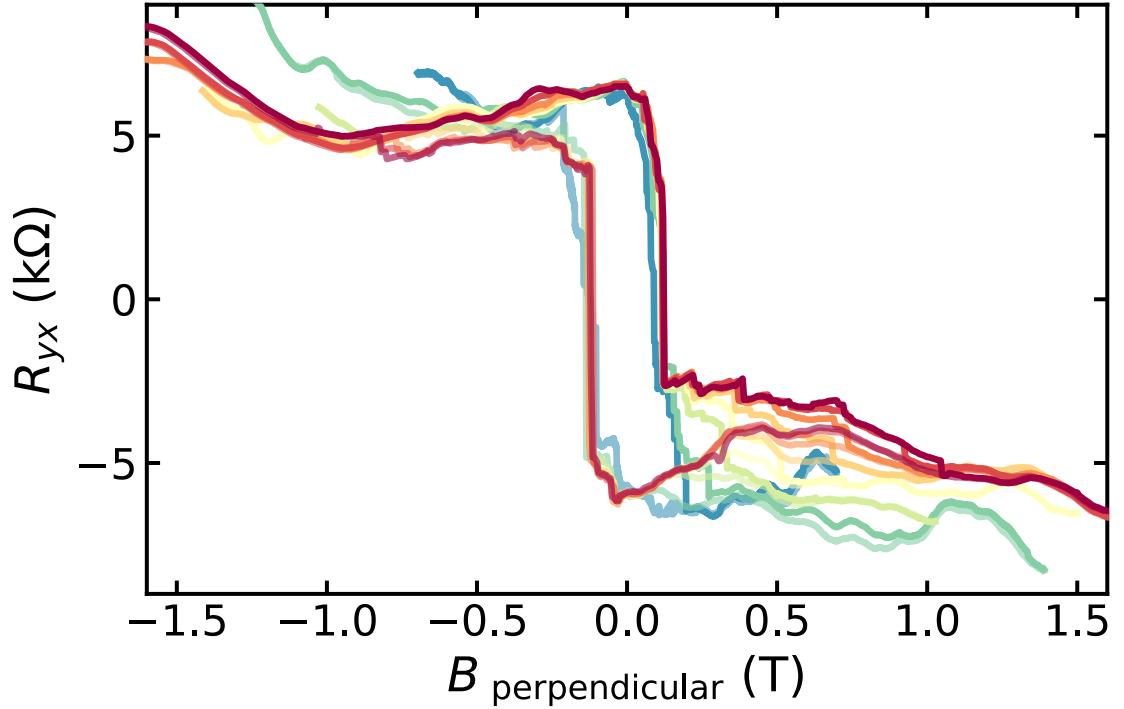
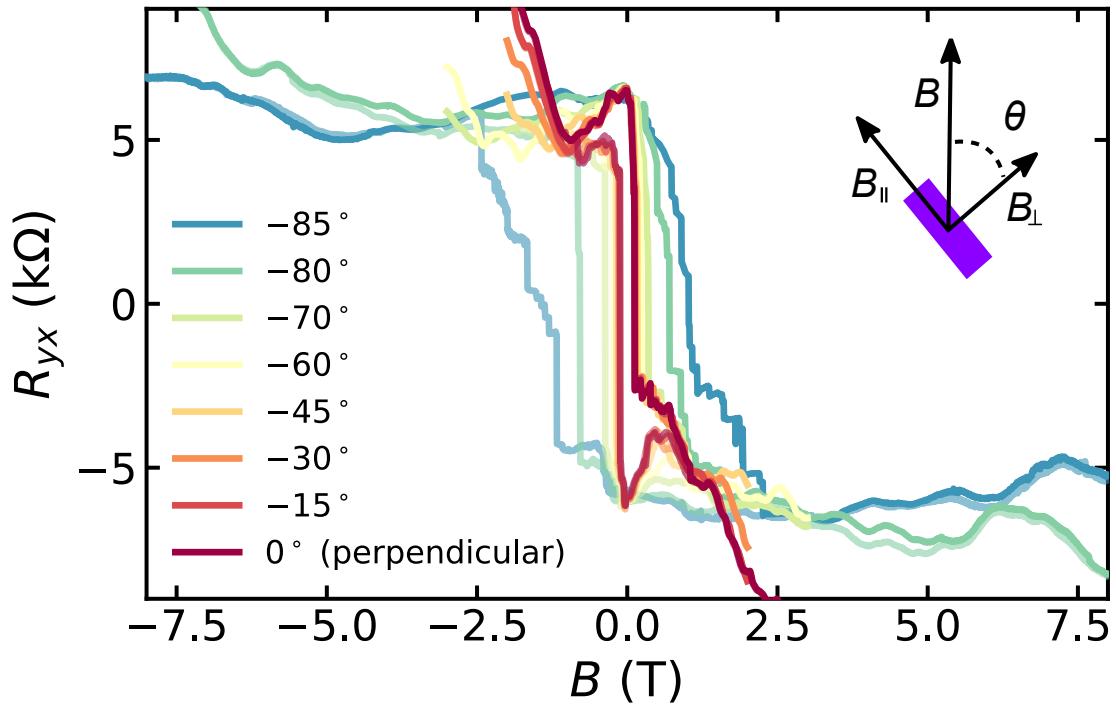
Similar conclusion: Tschirhart, et al. Science (2021)
Sharpe, et al. Nano Lett. (2021)

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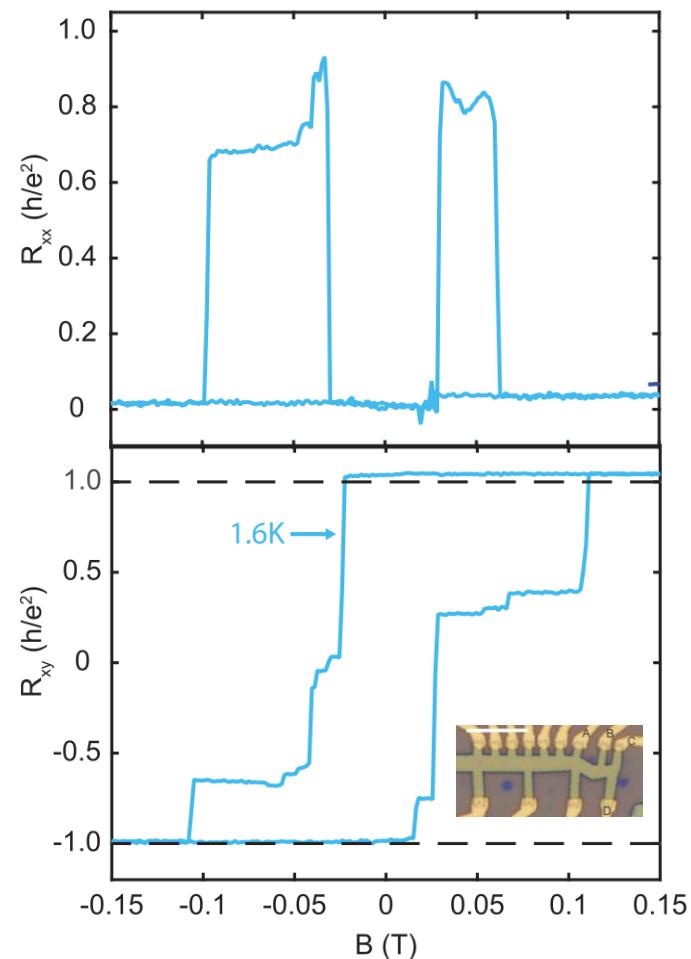
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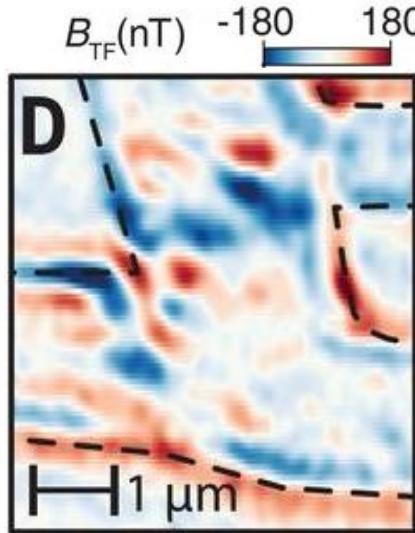
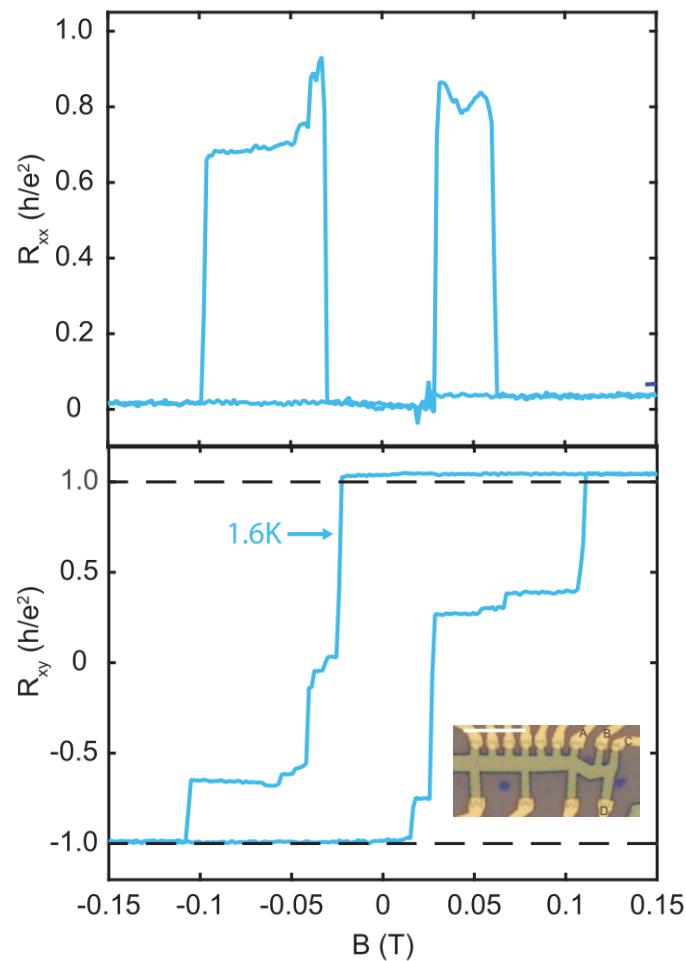
Data collapses when plotted vs B perpendicular!
Orbital Ferromagnet!

Quantum anomalous Hall in TBG

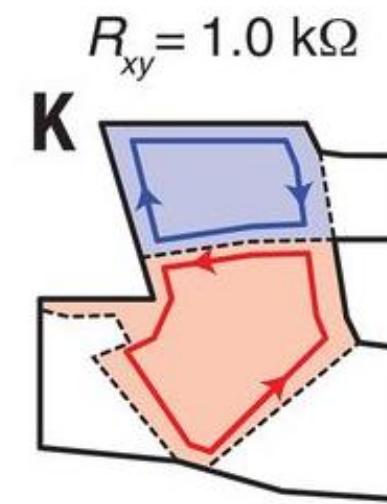
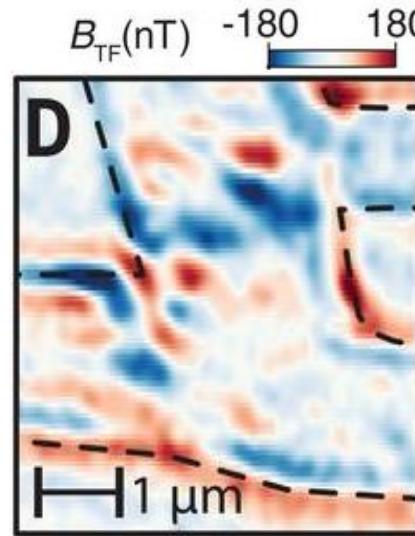
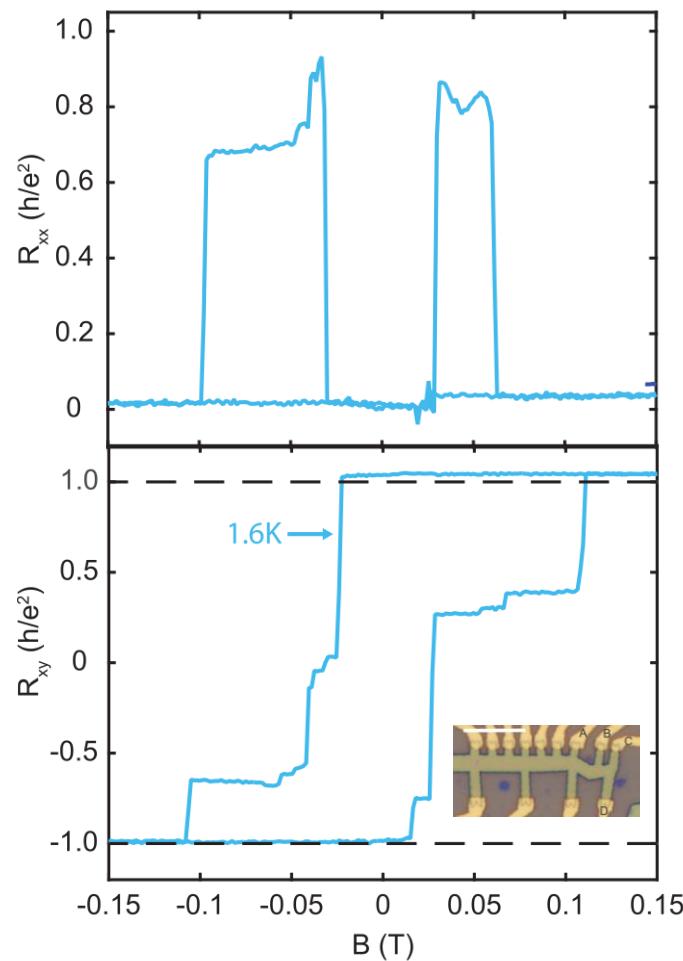
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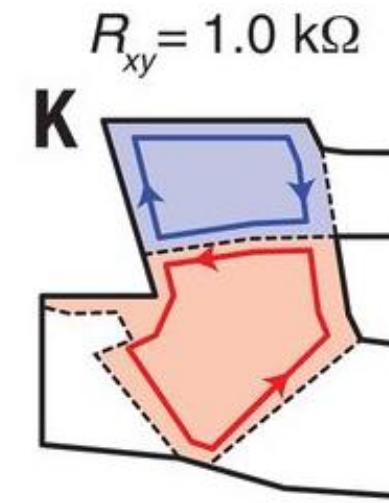
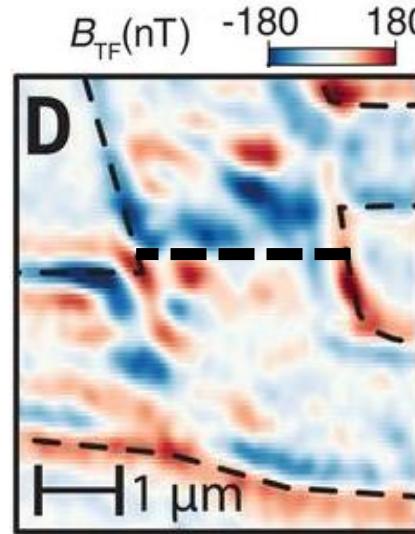
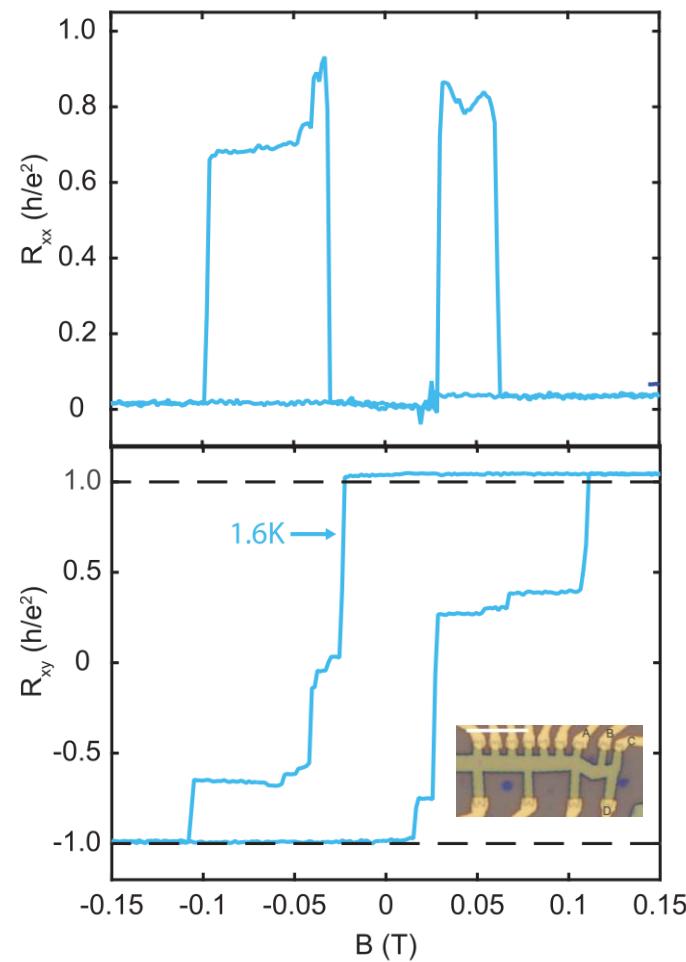
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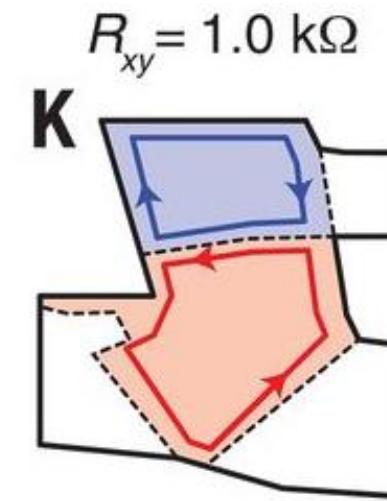
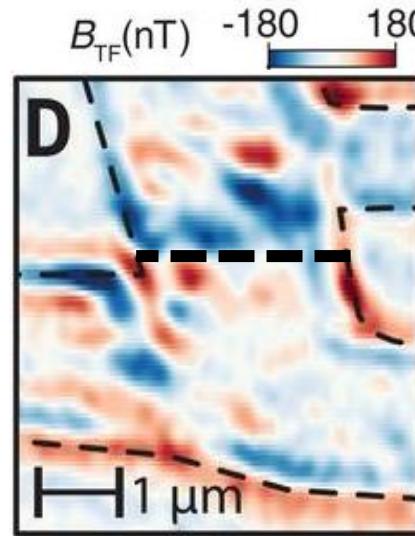
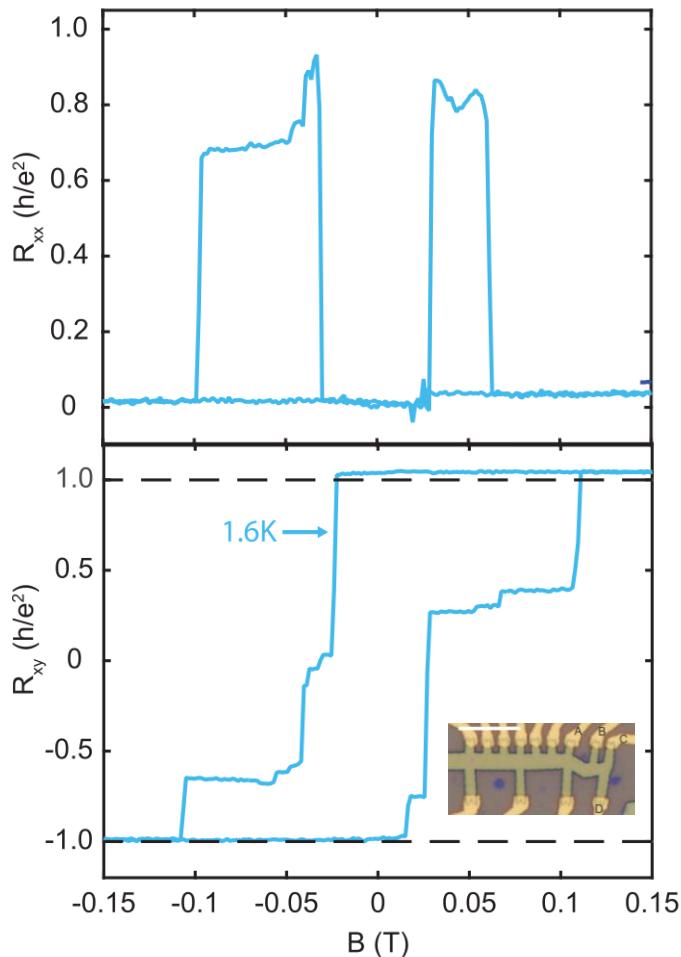
Quantum anomalous Hall in TBG



Quantum anomalous Hall in TBG



Quantum anomalous Hall in TBG



Splotchy magnet yet quantized?

(Q)AH is quite common in moirés!

MATBG aligned/commensurate with hBN:

- A. Sharpe, et al. Science (2019)
M. Serlin, et al. Science (2020)
Tschirhart, et al. Science (2021)
A. Sharpe, et al. Nano Lett. (2021)
P. Stepanov, et al. PRL (2021)
C-C. Tseng, et al. Nat. Phys (2022)
S. Grover, et al. Nat. Phys (2022)
Z. Zhang, et al. arXiv:2408.12509

MATBG + WSe2:

- J-X. Lin, et al. Science (2022)

Rhombohedral graphene

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G. Chen, A. Sharpe, et al. Nature (2020)
G. Chen, A. Sharpe, et al. Nano Lett. (2022)

Quadlayer + WSe2

- Y. Sha et al. Science (2024)
Pentalayer (FCI when aligned to hBN)

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Twisted bilayer MoTe2 (FCI)

- E. Anderson, et al. Science (2023)
J. Cai, et al. Nature (2023)
Y. Zeng, et al. Nature (2023)
H. Park, et al. Nature (2023)
F. Xu, et al. PRX (2023)

AB-stacked MoTe2/WSe2:

- T. Li, et al. Nature (2021)

Twisted mono-bilayer graphene:

- S. Chen, et al. Nat. Phys (2020)
H. Polshyn, et al. Nature (2020)
M. He, et al. Nat. Comms (2021)

Twisted double bilayer graphene:

- [AB-AB] M. Kuiji, et al. Nat. Comms (2022)
[AB-BA] M. He, et al. Nano Lett. (2023)

Helical trilayer graphene

- L-Q Xia, et al. Nat. Phys (2024)

M+N graphene:

- D. Waters, et al. arXiv:2405.05913

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- [AB-AB] M. Kuiiri, et al. Nat. Comms (2022)
- [AB-BA] M. He, et al. Nano Lett. (2023)

Helical trilayer graphene

- L-Q Xia, et al. Nat. Phys (2024)

M+N graphene:

- D. Waters, et al. arXiv:2405.05913

Summary

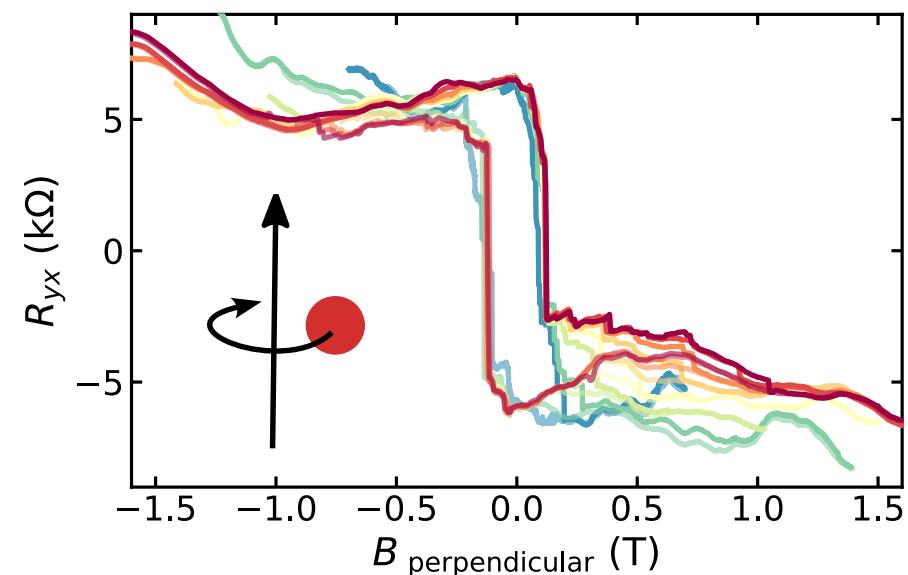
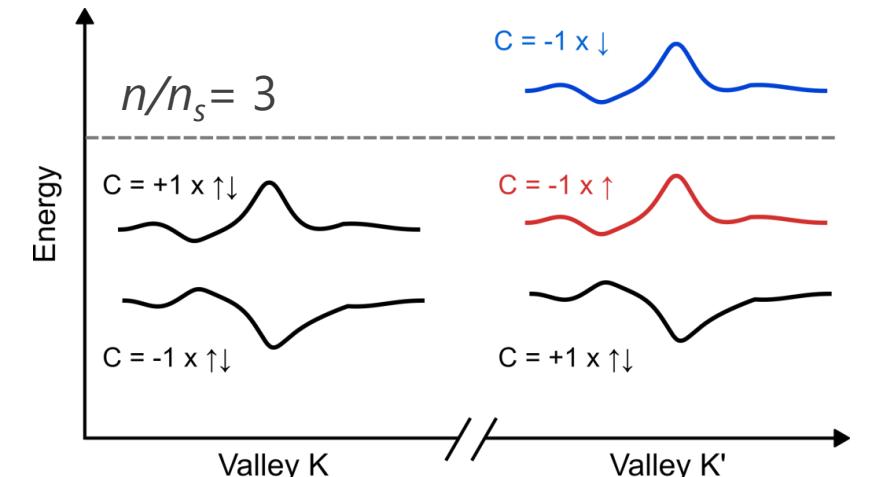
By controlling twist angle, graphene multilayers can become strongly correlated!

TBG is a Chern insulator at filling factor 3!

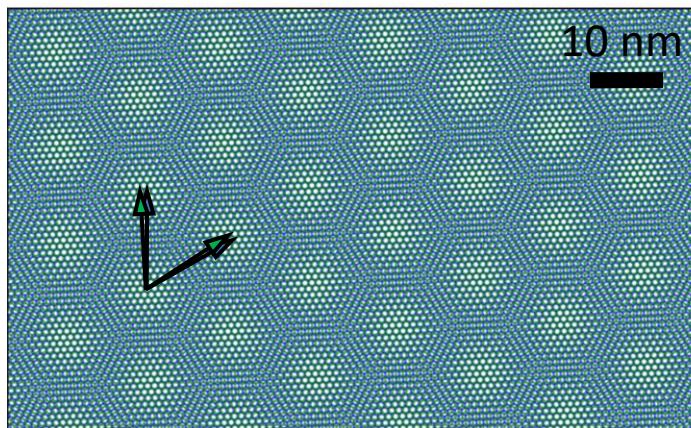
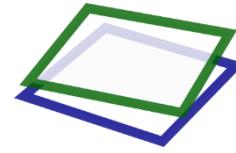
Magnetism is orbital in character
hBN alignment is likely crucial

Why is this orbital magnet so hard to reproduce in TBG?

Magnetism appears splotchy
(Q)AH appears to be quite ubiquitous in moirés
Shouldn't the hBN/graphene form a moiré?
How should we model this?



From moiré to multimoiré



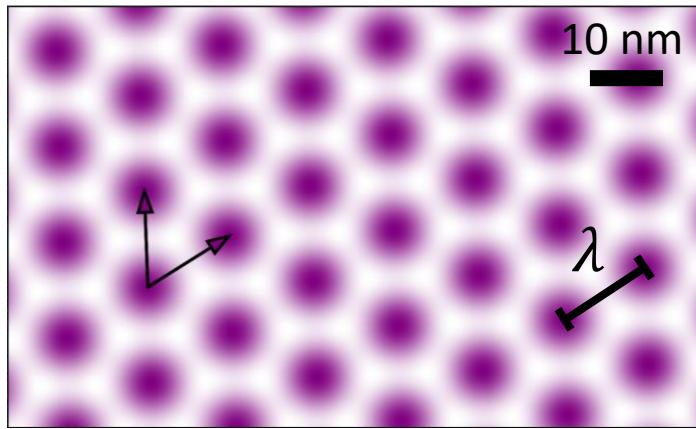
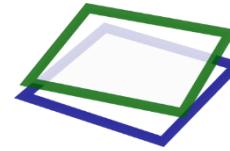
Yang, et al. arXiv:2310.12961

Nakatsuji, et al. PRX (2023)

Foo, et al. PRR (2024)

Bistritzer and MacDonald, PNAS (2011)

From moiré to multimoiré



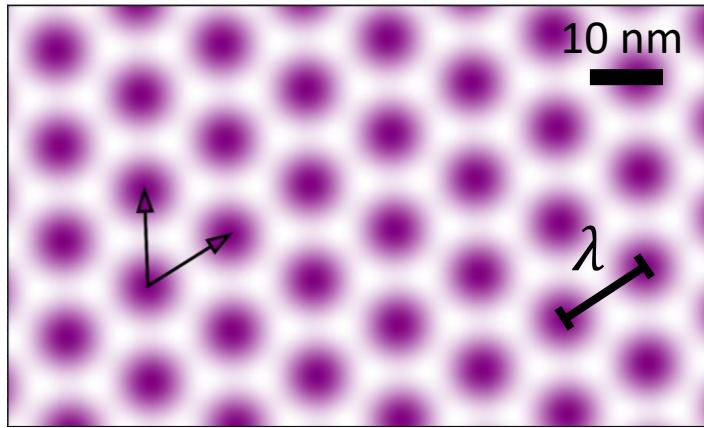
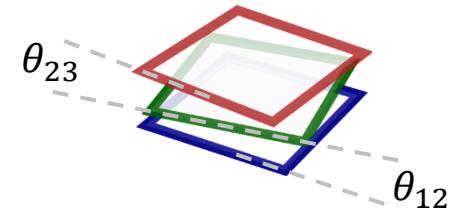
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From moiré to multimoiré



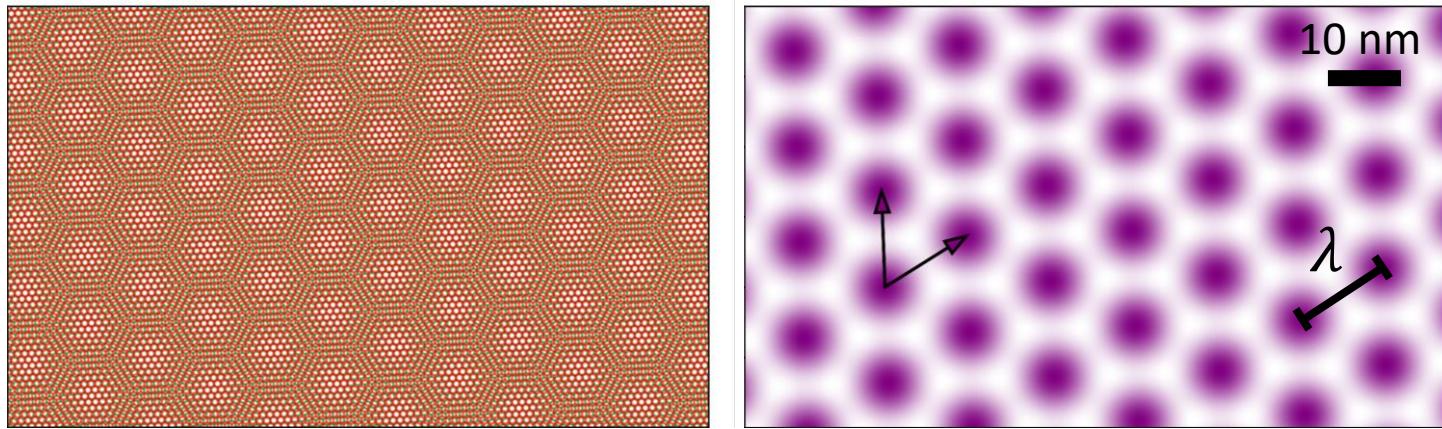
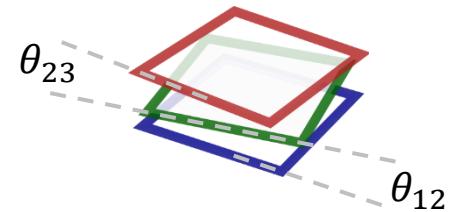
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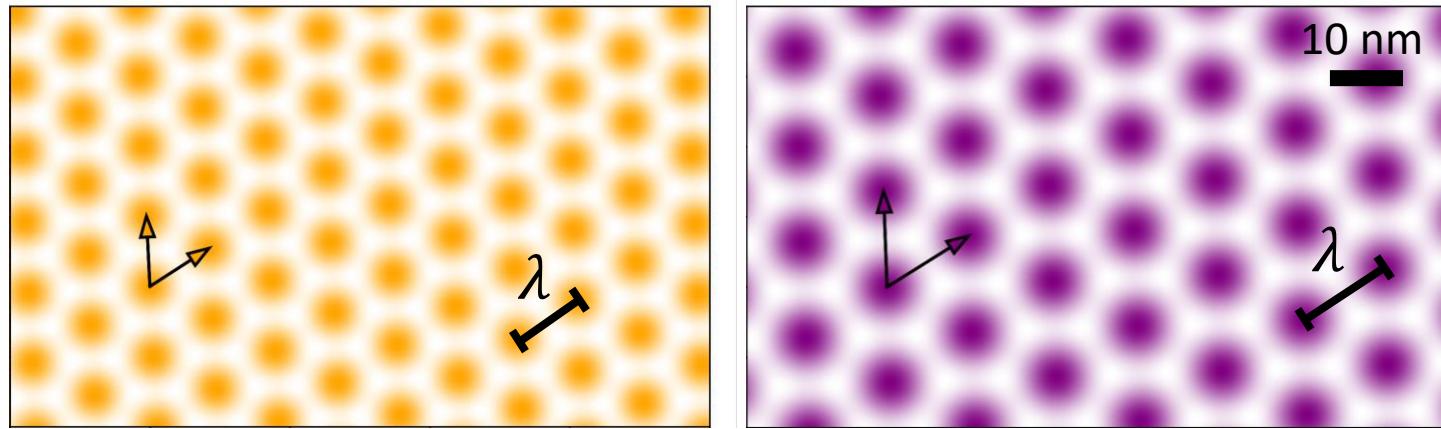
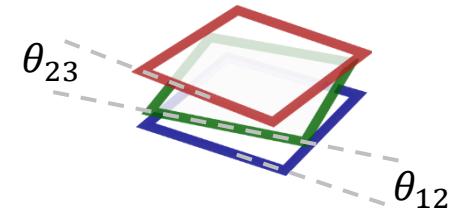
Bistritzer and MacDonald, PNAS (2011)

From moiré to multimoiré



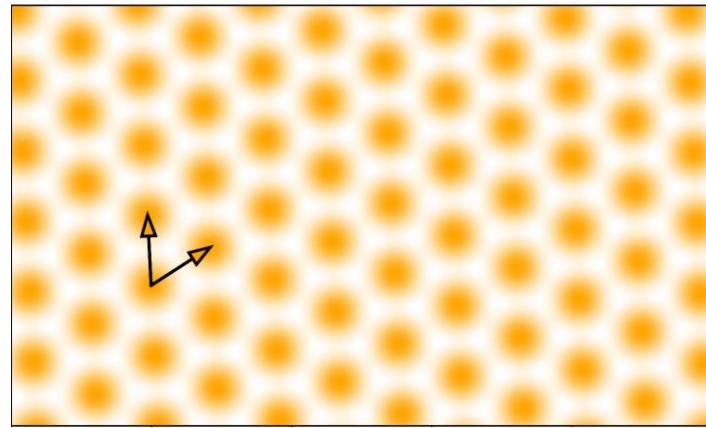
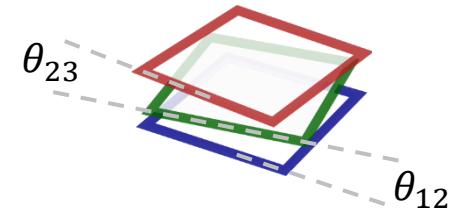
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From moiré to multimoiré



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From moiré to multimoiré



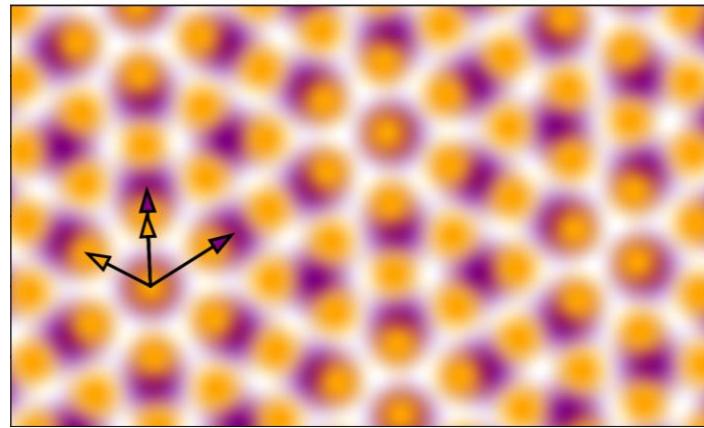
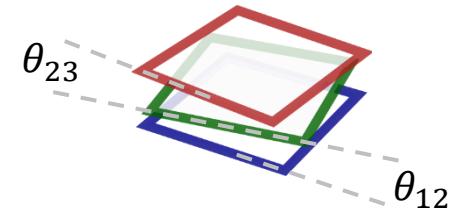
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From moiré to multimoiré



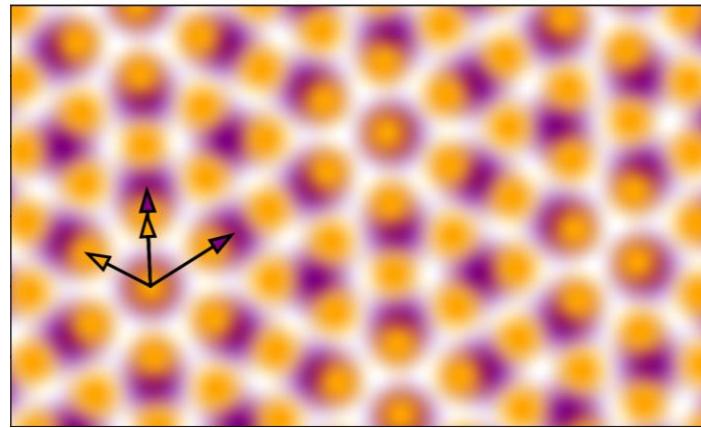
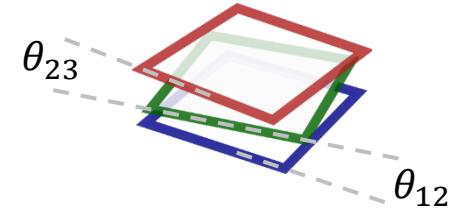
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Bistritzer and MacDonald, PNAS (2011)

From moiré to multimoiré



Moiré quasicrystal

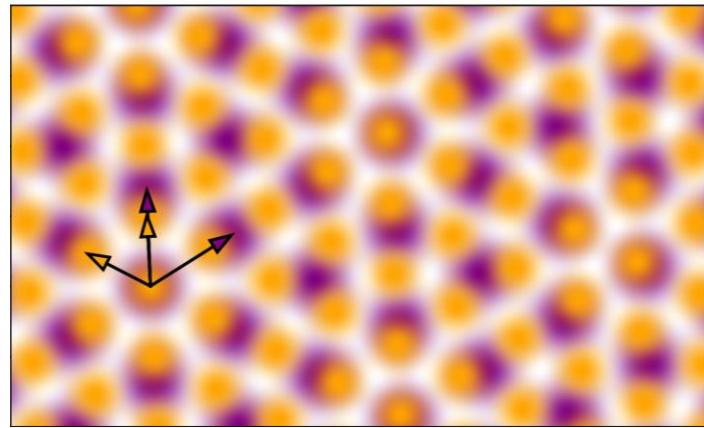
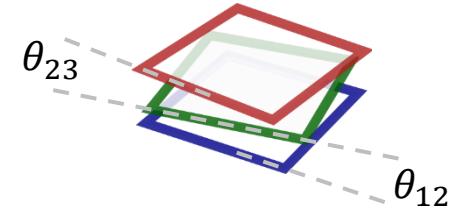
Yang, et al. arXiv:2310.12961

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From moiré to multimoiré



Moiré quasicrystal

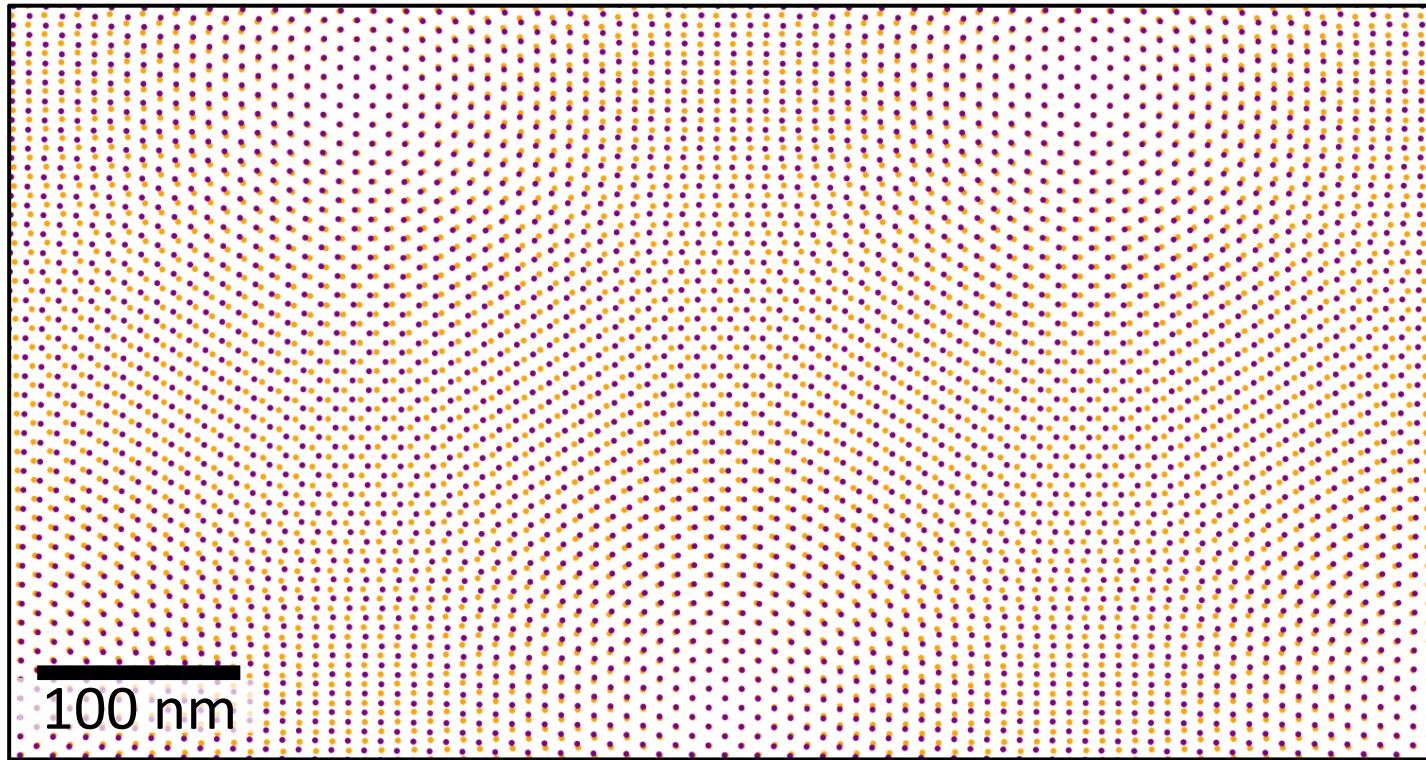
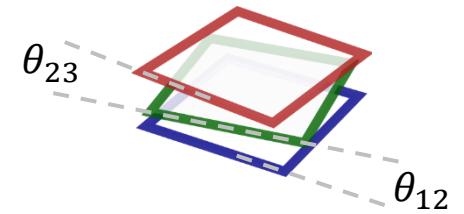
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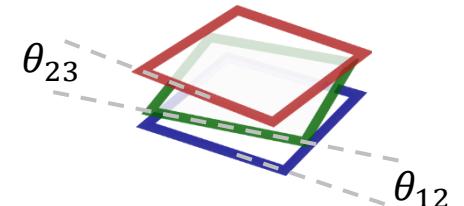
From moiré to multimoiré



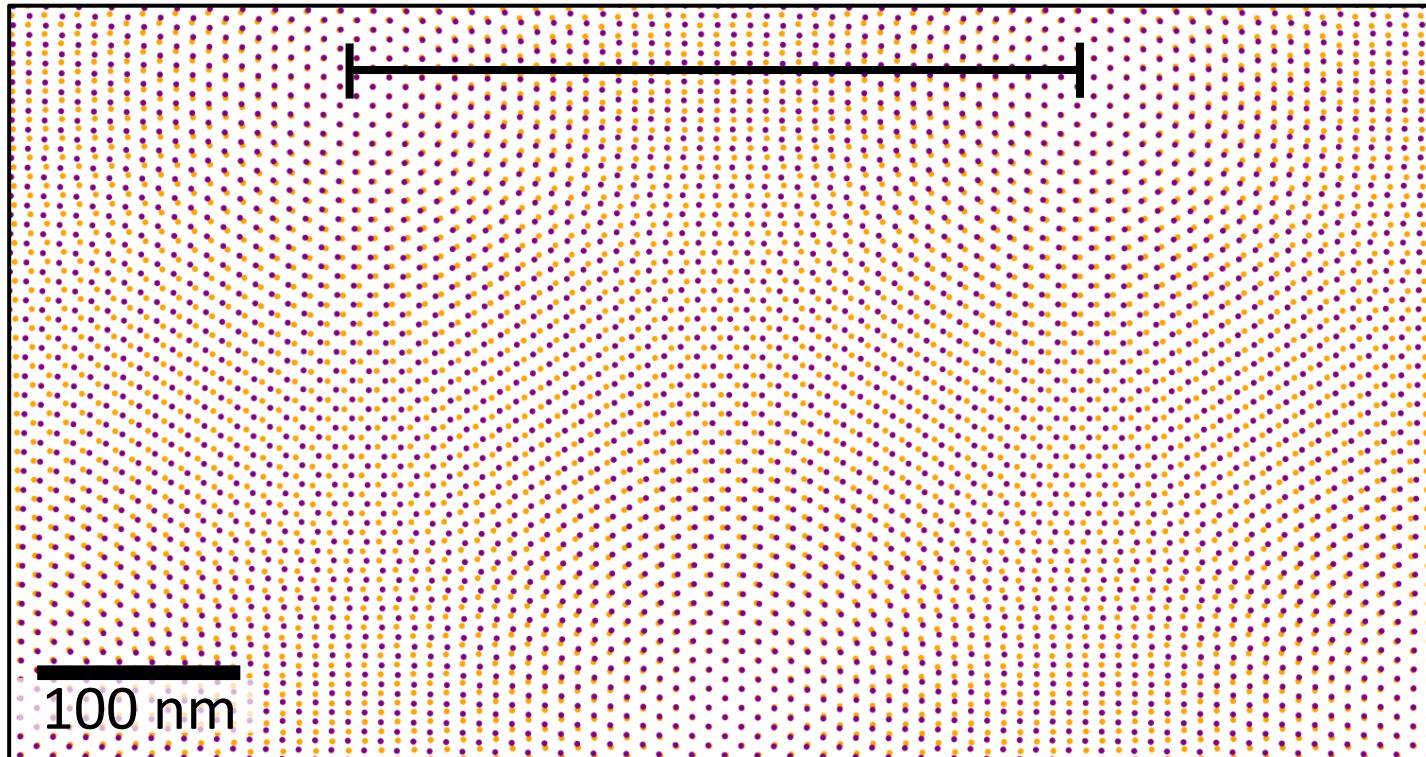
Yang, et al. arXiv:2310.12961
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Bistritzer and MacDonald, PNAS (2011)

● AA12 ● AA23

From moiré to multimoiré



$$\text{Supermoiré: } \lambda_{sm} = \frac{\lambda_m}{\theta} = \frac{a}{\theta^2}$$



Yang, et al. arXiv:2310.12961

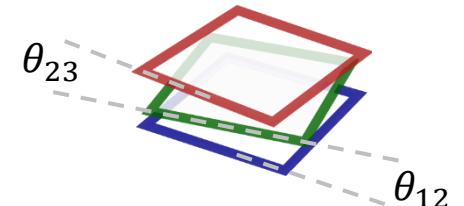
Nakatsuji, et al. PRX (2023)

Foo, et al. PRR (2024)

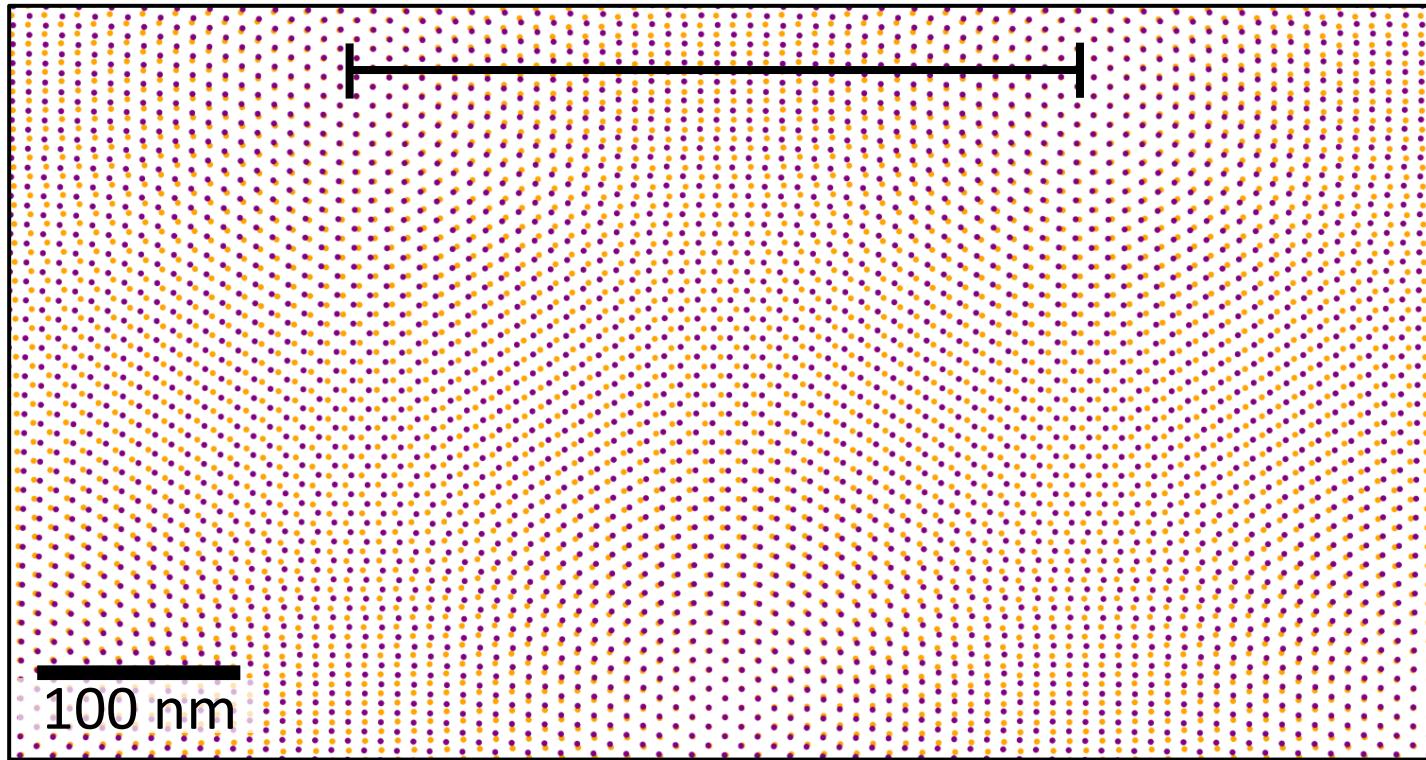
Bistritzer and MacDonald, PNAS (2011)

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From moiré to multimoiré



$$\text{Supermoiré: } \lambda_{sm} = \frac{\lambda_m}{\theta} = \frac{a}{\theta^2}$$



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Nakatsuji, et al. PRX (2023)

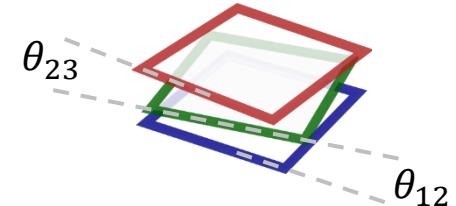
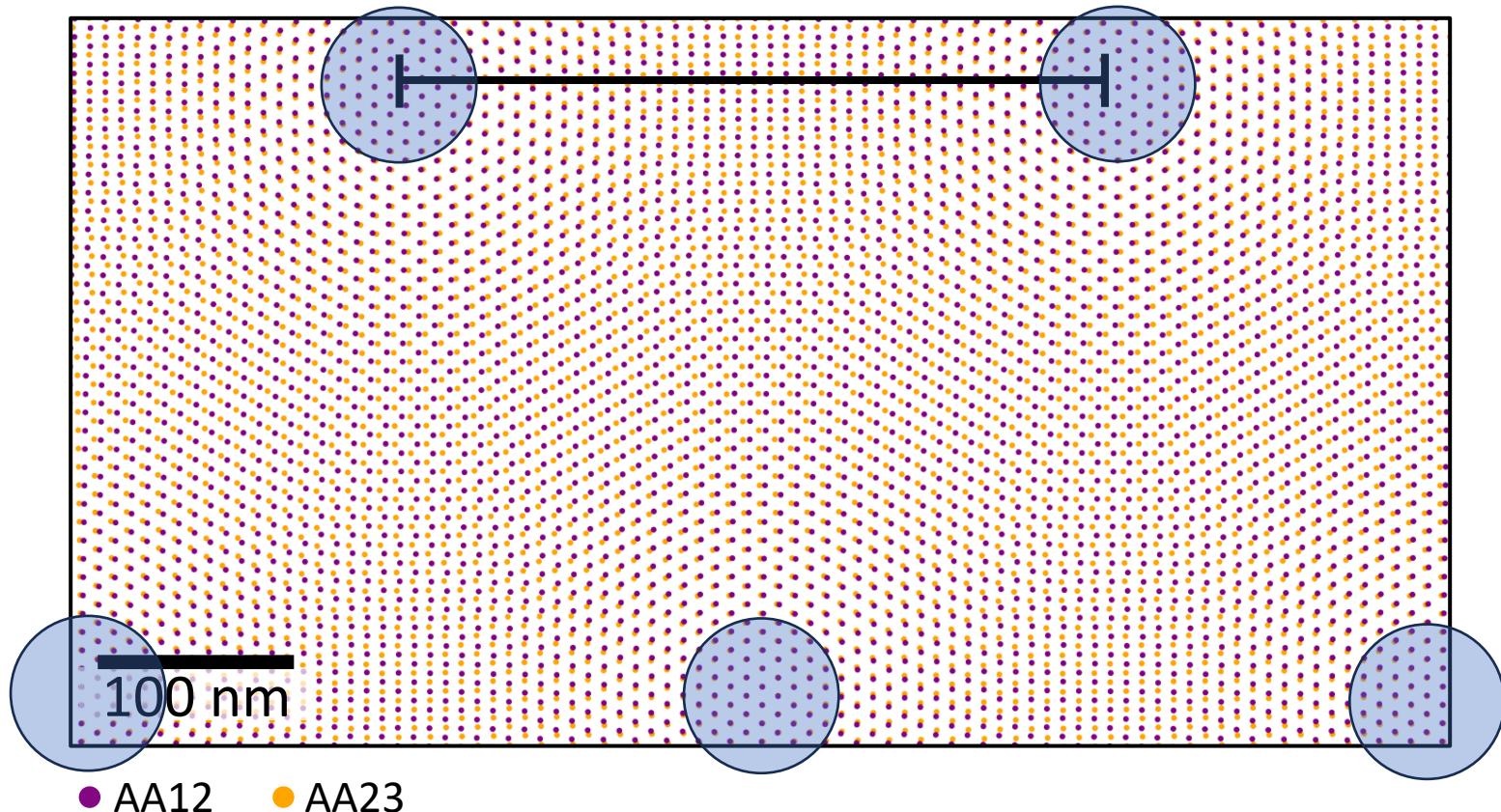
Foo, et al. PRR (2024)

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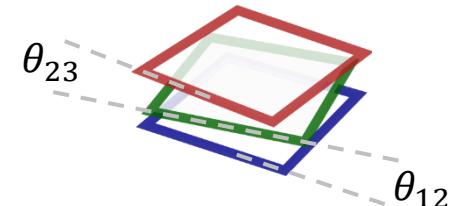
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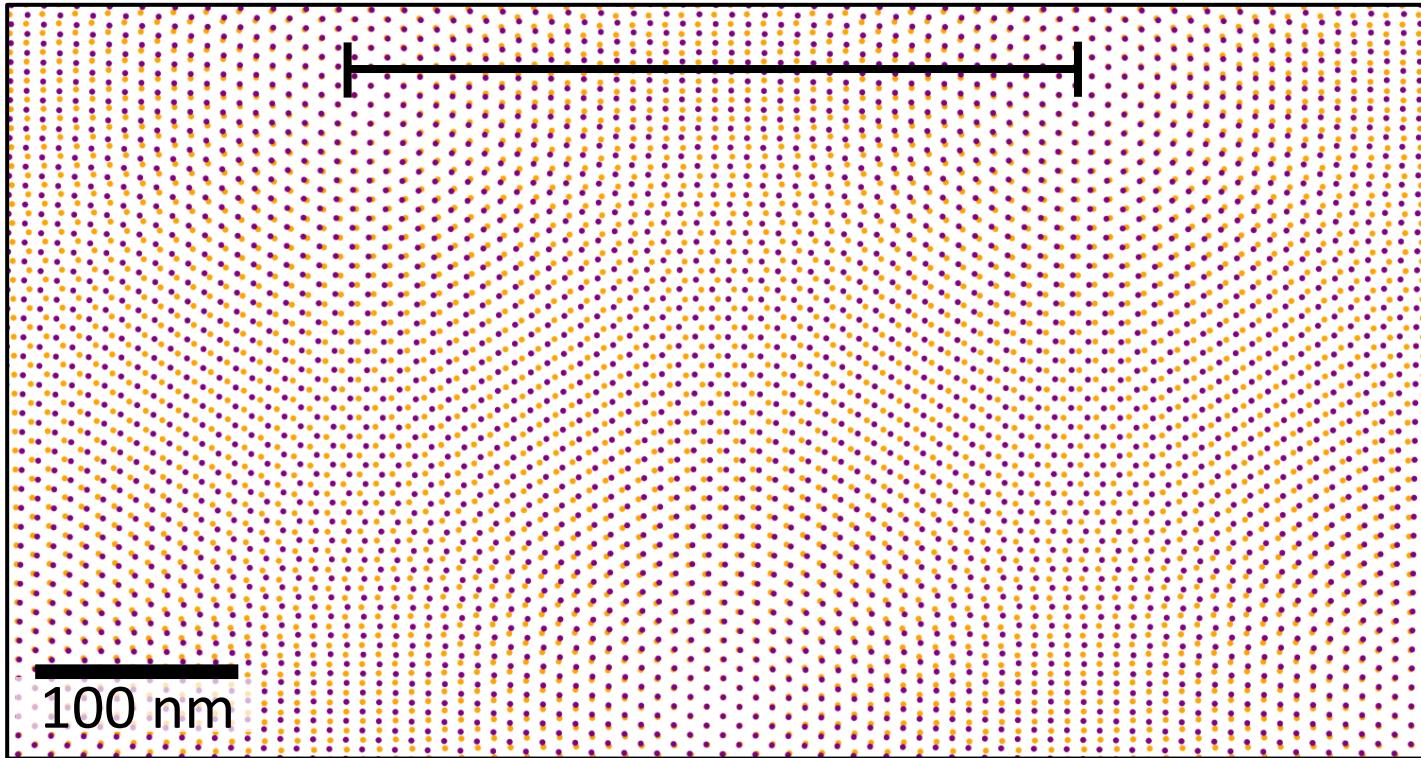
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Yang, et al. arXiv:2310.12961

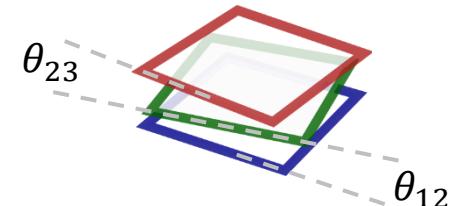
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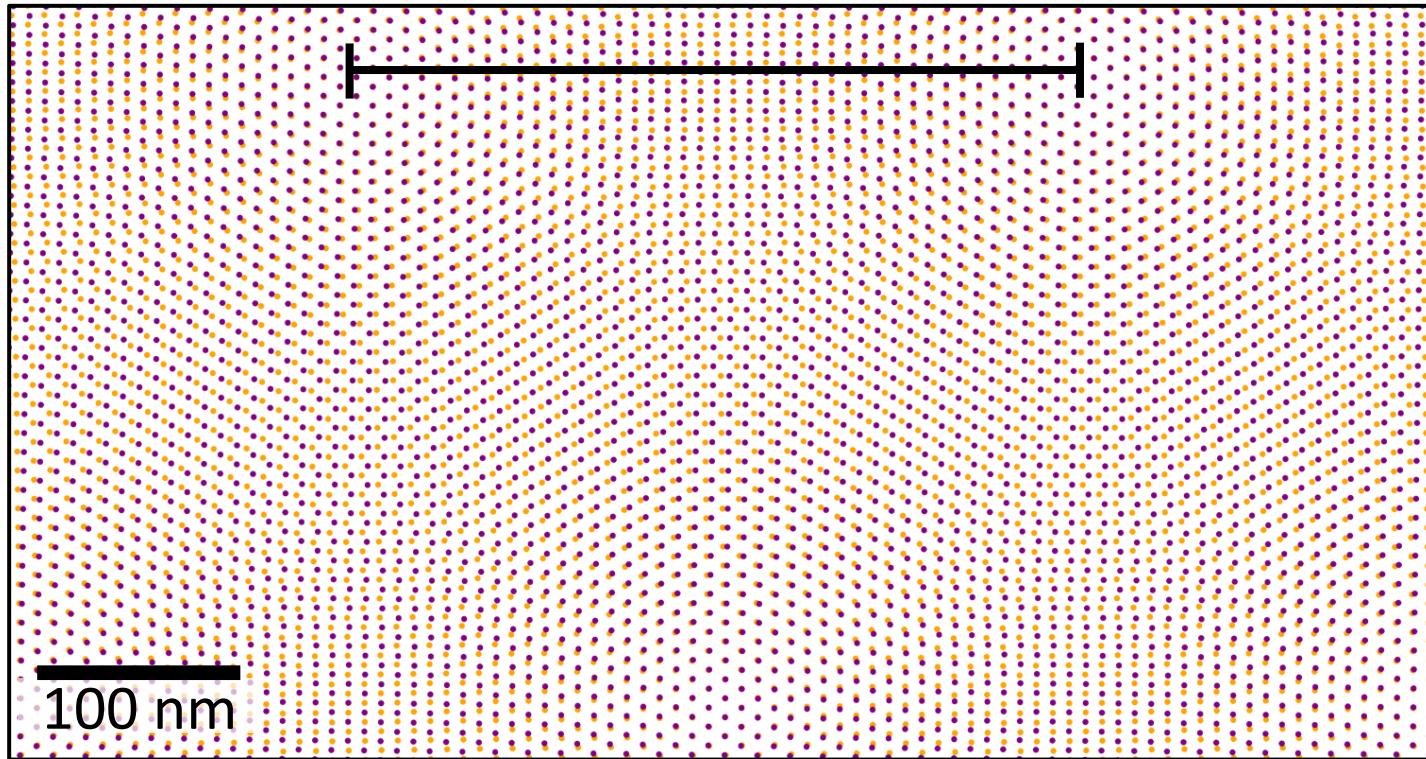
Bistritzer and MacDonald, PNAS (2011)

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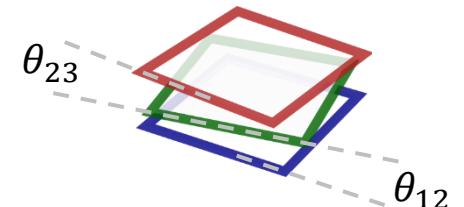
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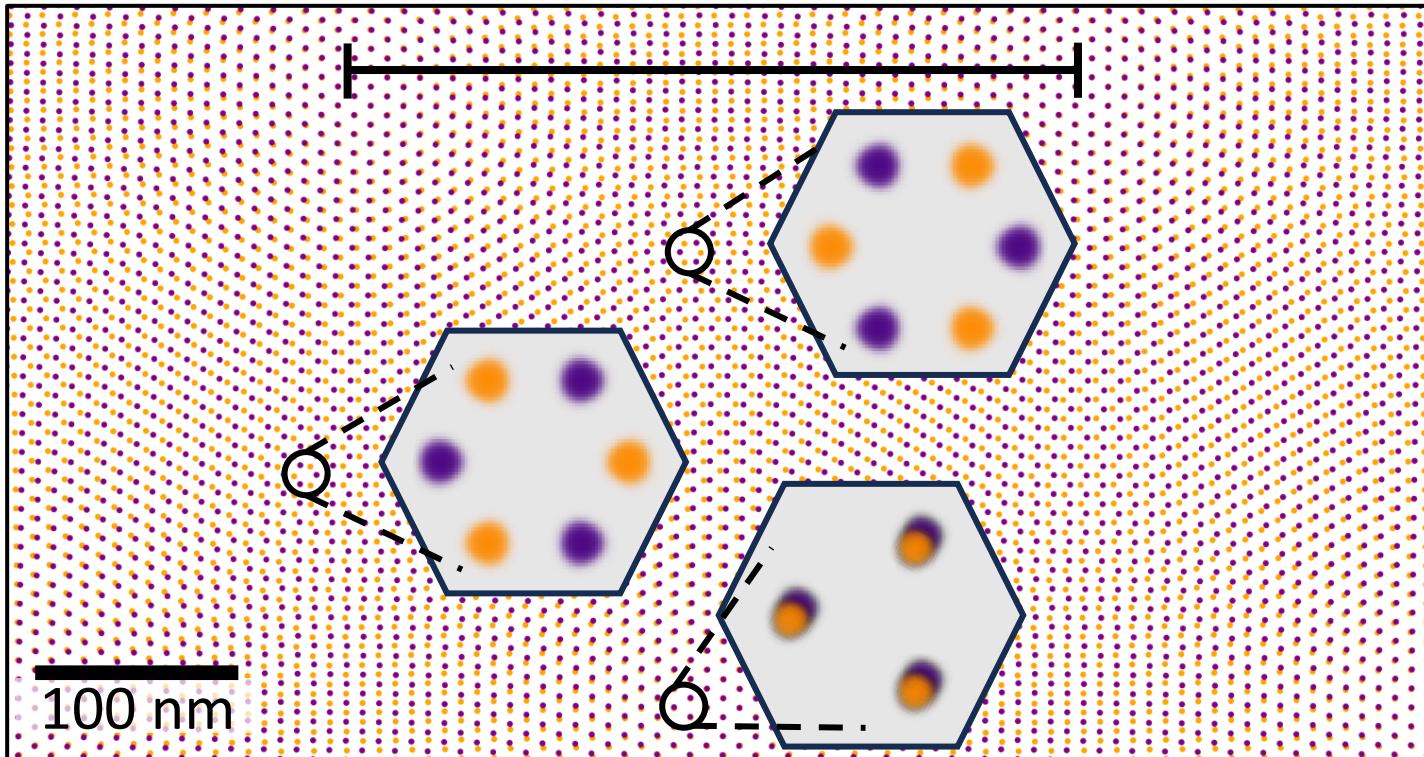
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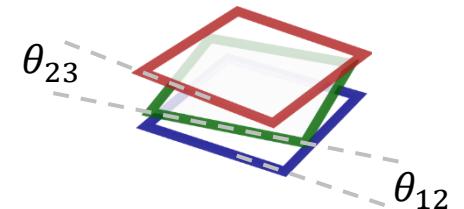
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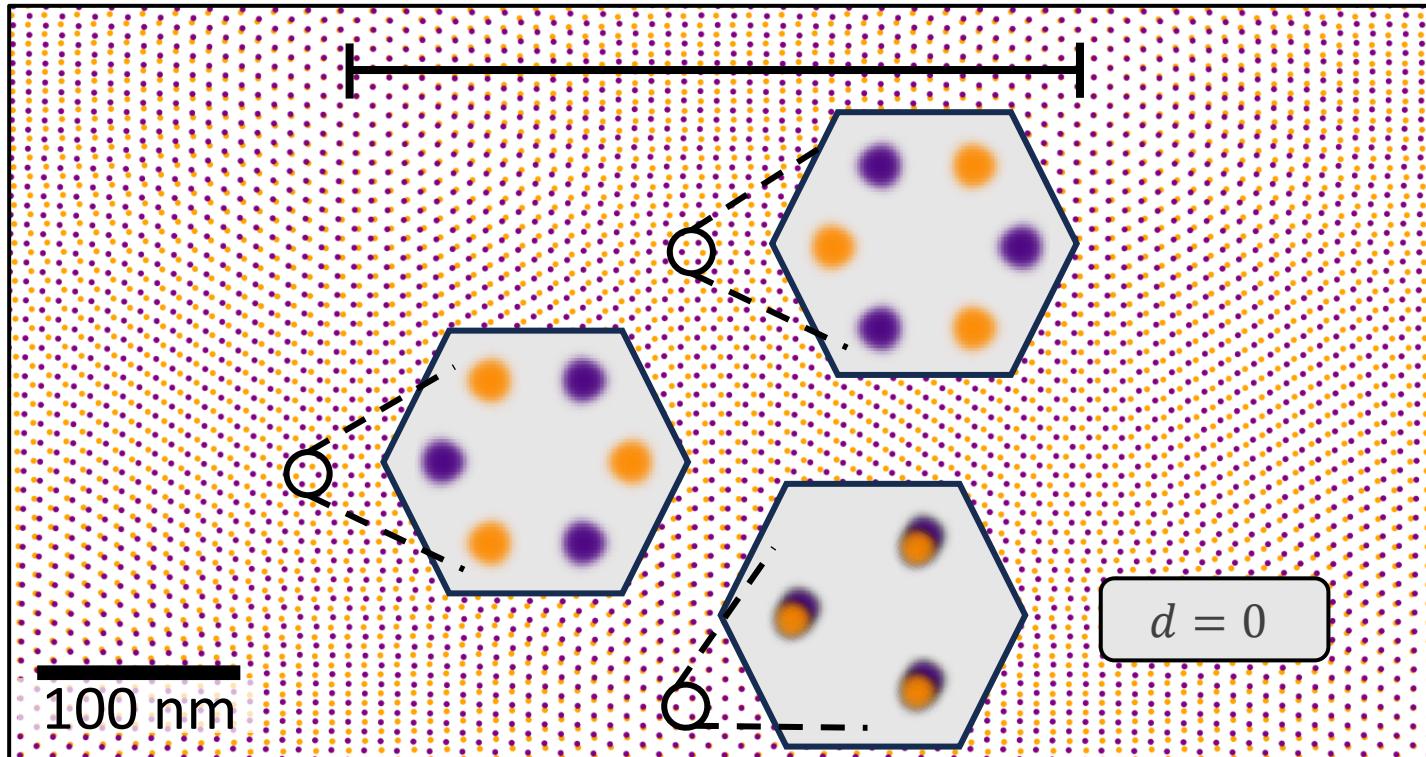
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From moiré to multimoiré



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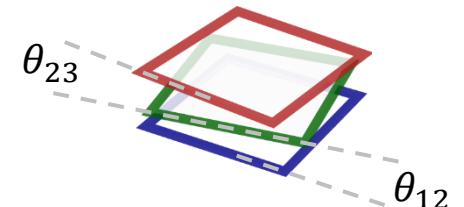
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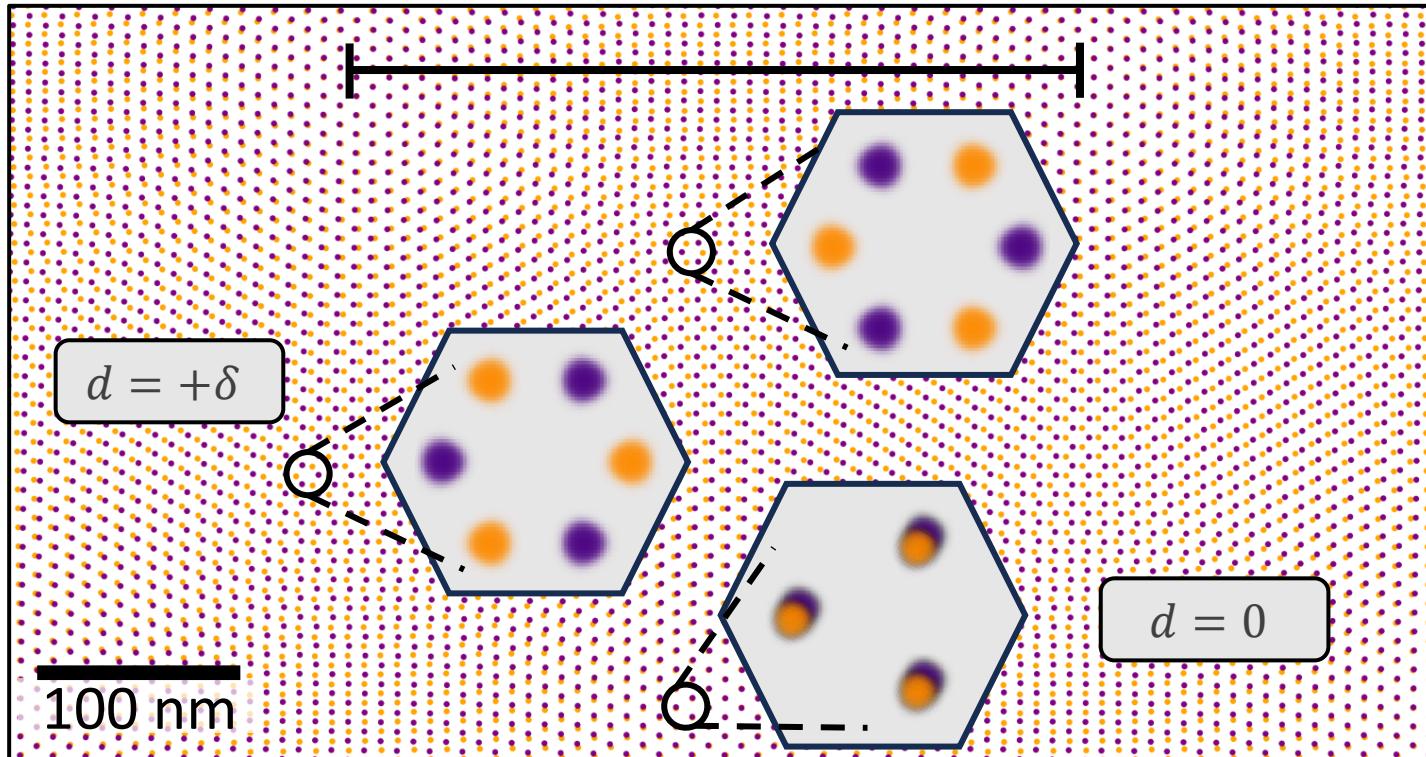
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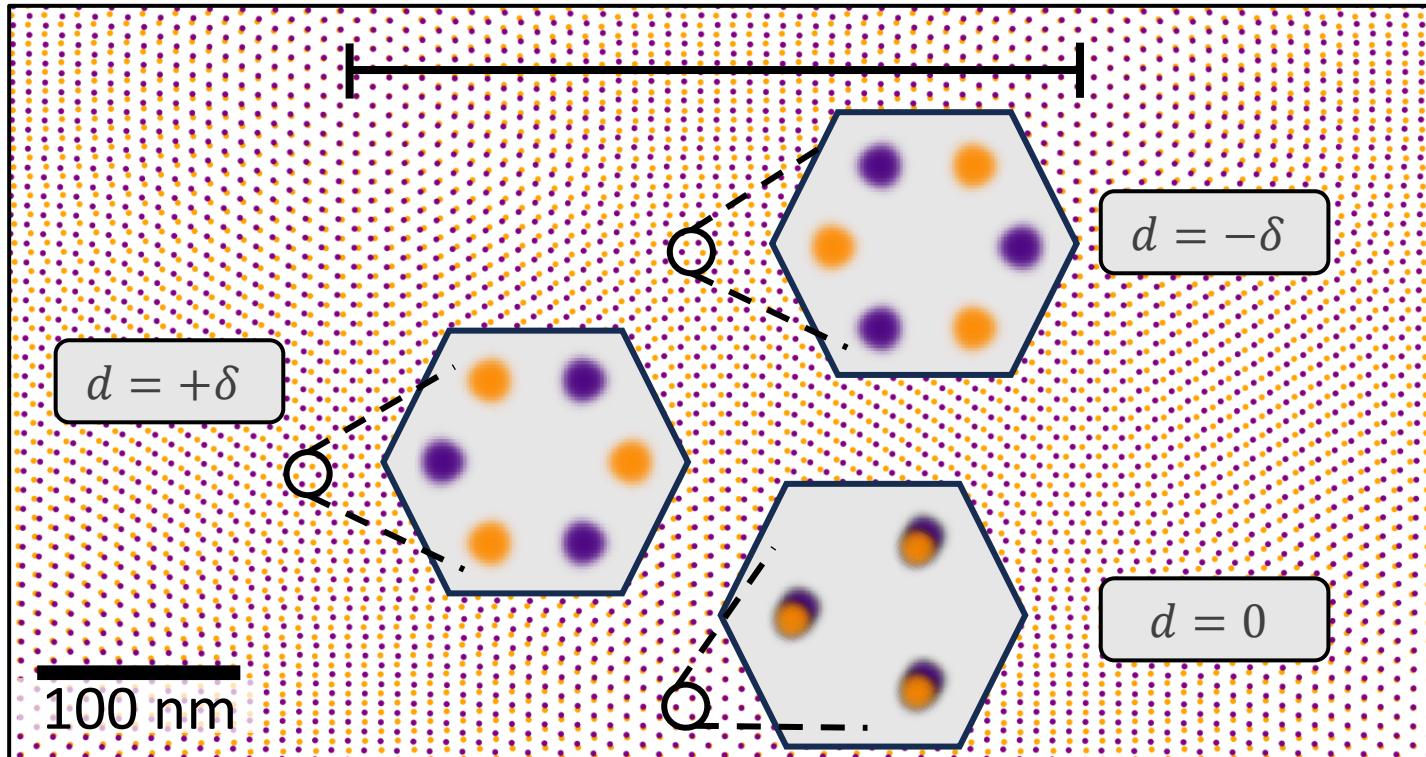
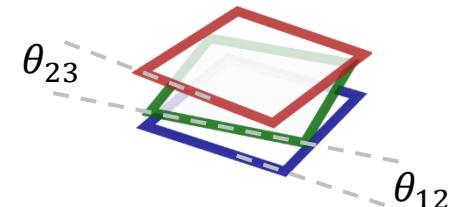
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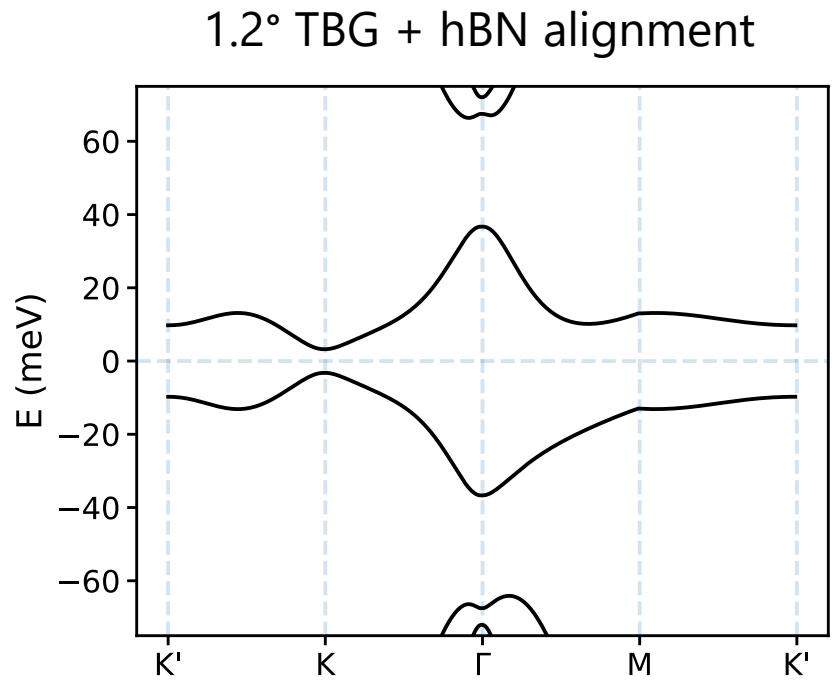
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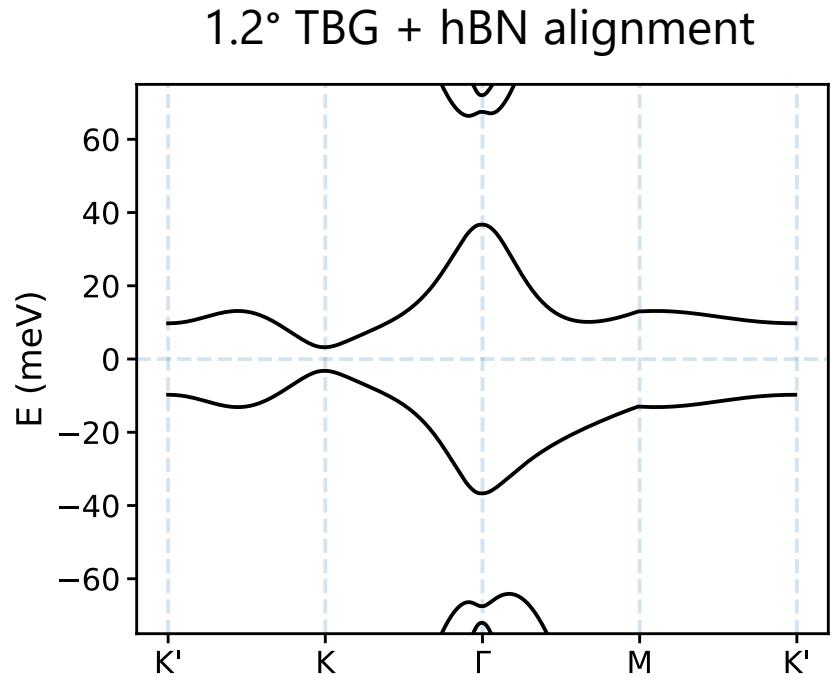
Bistritzer and MacDonald, PNAS (2011)

● AA12 ● AA23

TBG/hBN multimoiré



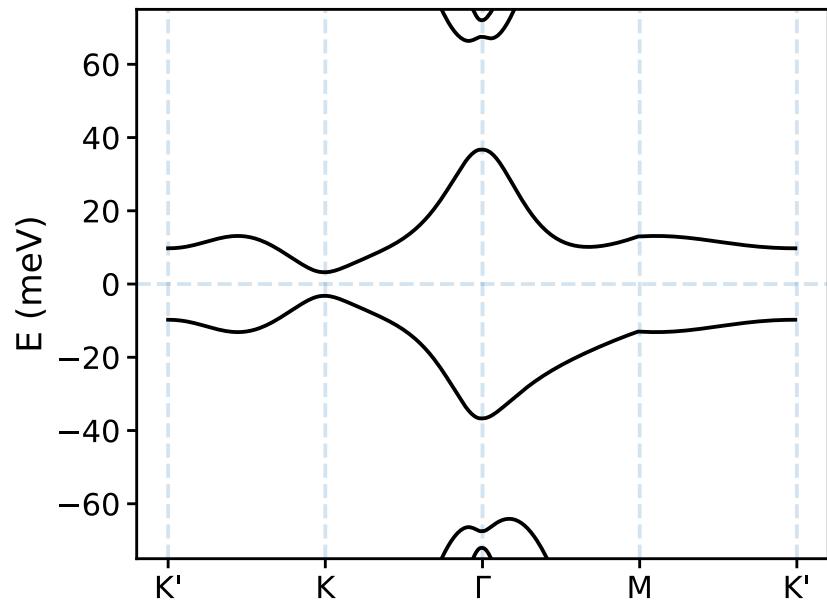
TBG/hBN multimoiré



$$V_{BN} = \sum_{\mathbf{k}} \psi_{1\mathbf{k}}^\dagger(m_0 \sigma^z) \psi_{1\mathbf{k}}$$

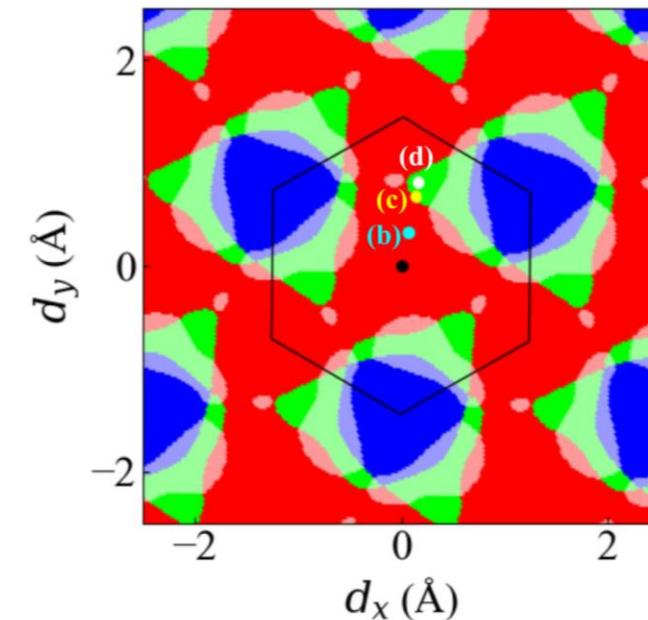
TBG/hBN multimoiré

1.2° TBG + hBN alignment



$$V_{BN} = \sum_{\mathbf{k}} \psi_{1\mathbf{k}}^\dagger (m_0 \sigma^z) \psi_{1\mathbf{k}}$$

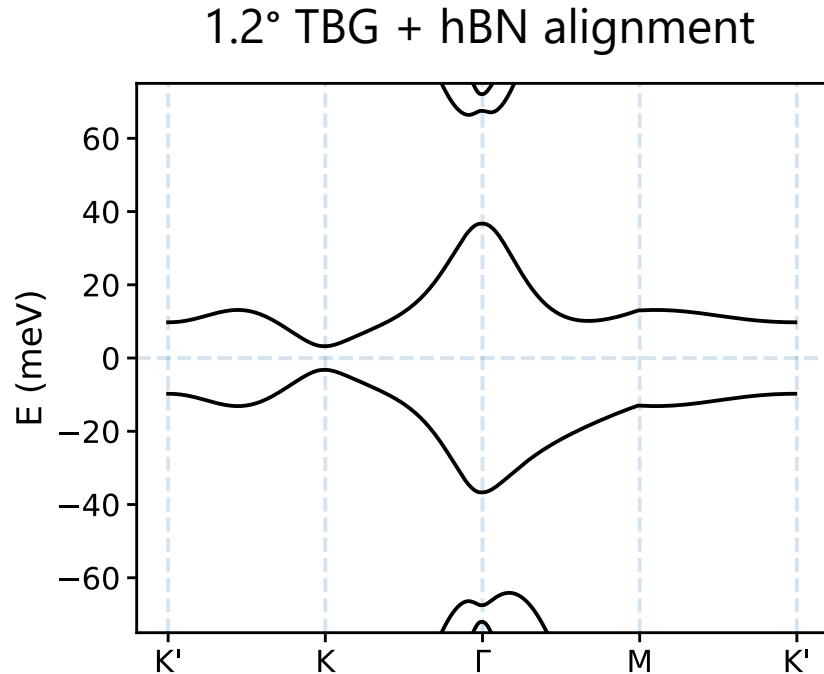
120° commensurate, $U = 0$



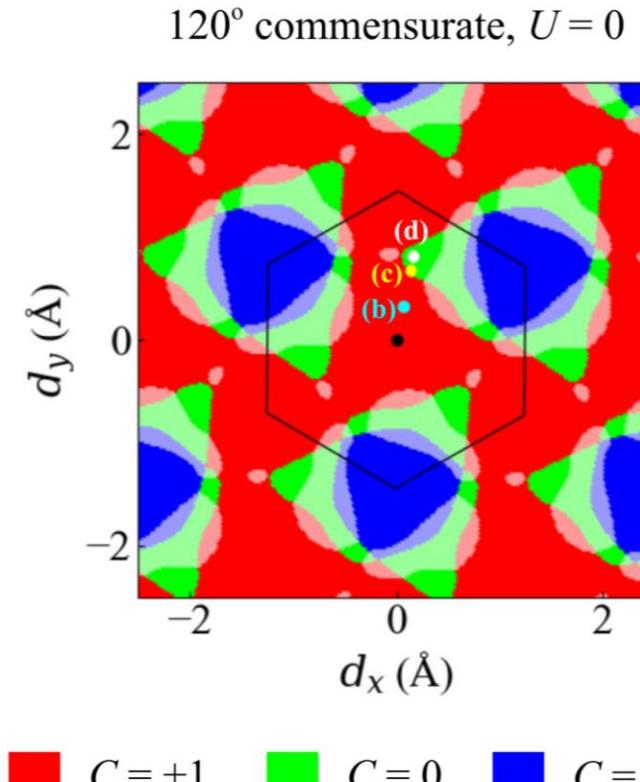
	$C = +1$		$C = 0$		$C = -1$
--	----------	--	---------	--	----------

$$V_{BN}(\mathbf{d}) = \sum_{\mathbf{k}} \left(\psi_{1\mathbf{k}}^\dagger (m_0 \sigma^z) \psi_{1\mathbf{k}} + \sum_{j=1} \psi_{1\mathbf{k}}^\dagger V_j(\mathbf{d}) \psi_{1(\mathbf{k}+\mathbf{g}_j)} \right)$$

TBG/hBN multimoiré



$$V_{BN} = \sum_{\mathbf{k}} \psi_{1\mathbf{k}}^\dagger (m_0 \sigma^z) \psi_{1\mathbf{k}}$$



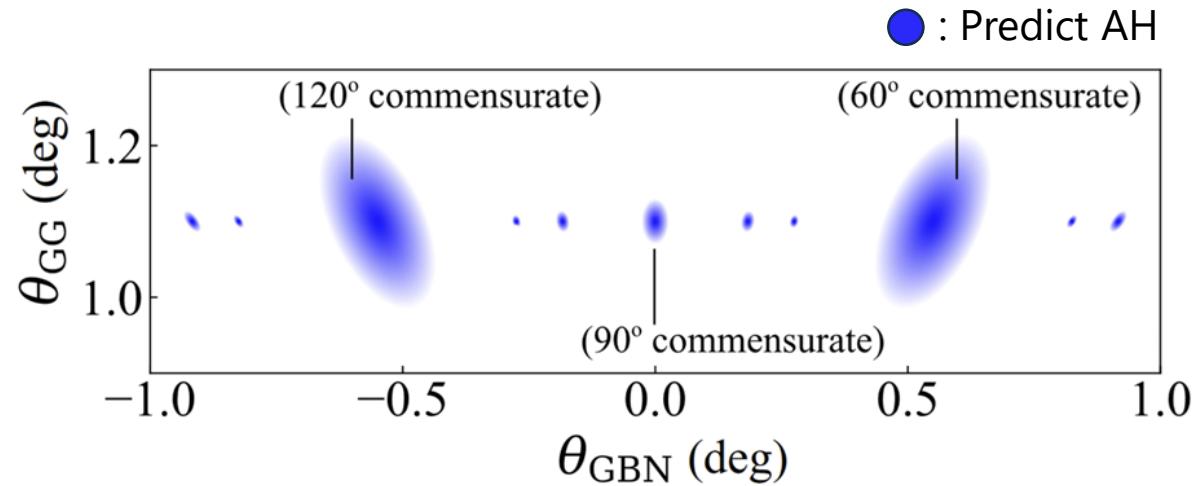
	$C = +1$		$C = 0$		$C = -1$
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$$V_{BN}(\mathbf{d}) = \sum_{\mathbf{k}} \left(\psi_{1\mathbf{k}}^\dagger (m_0 \sigma^z) \psi_{1\mathbf{k}} + \sum_{j=1} \psi_{1\mathbf{k}}^\dagger V_j(\mathbf{d}) \psi_{1(\mathbf{k}+\mathbf{g}_j)} \right)$$

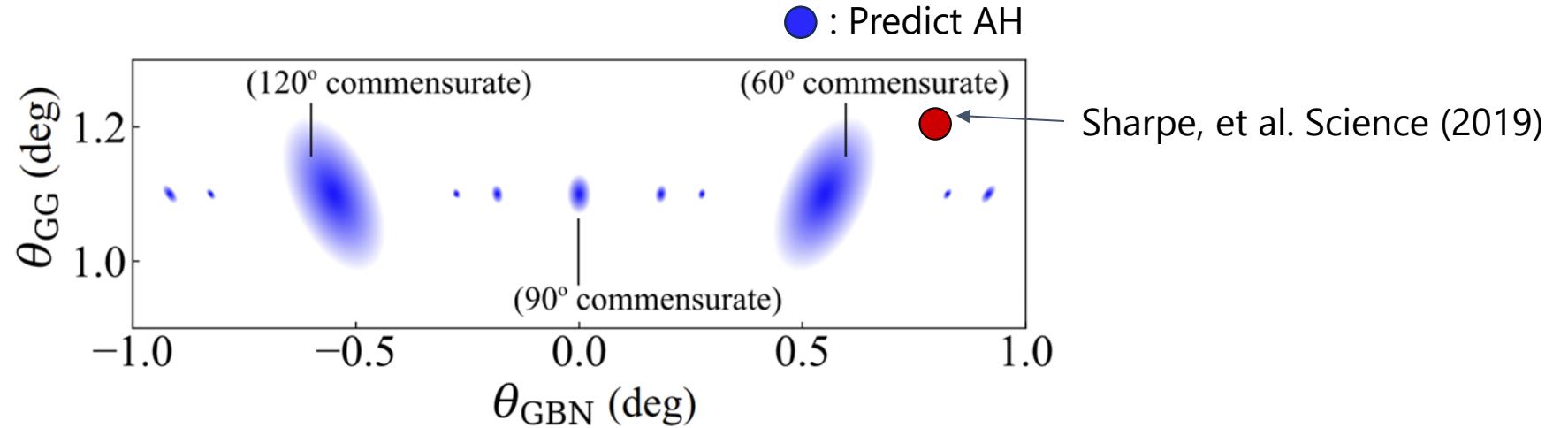
We must take the mesoscopics seriously!
Previous treatment too simplistic

TBG/hBN multimoiré

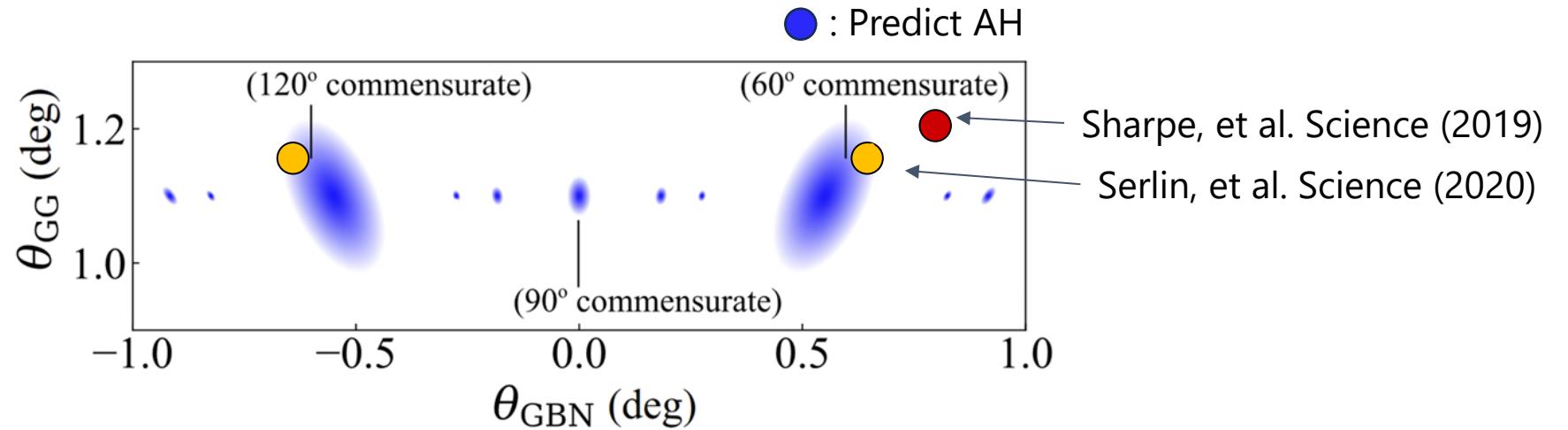
TBG/hBN multimoiré



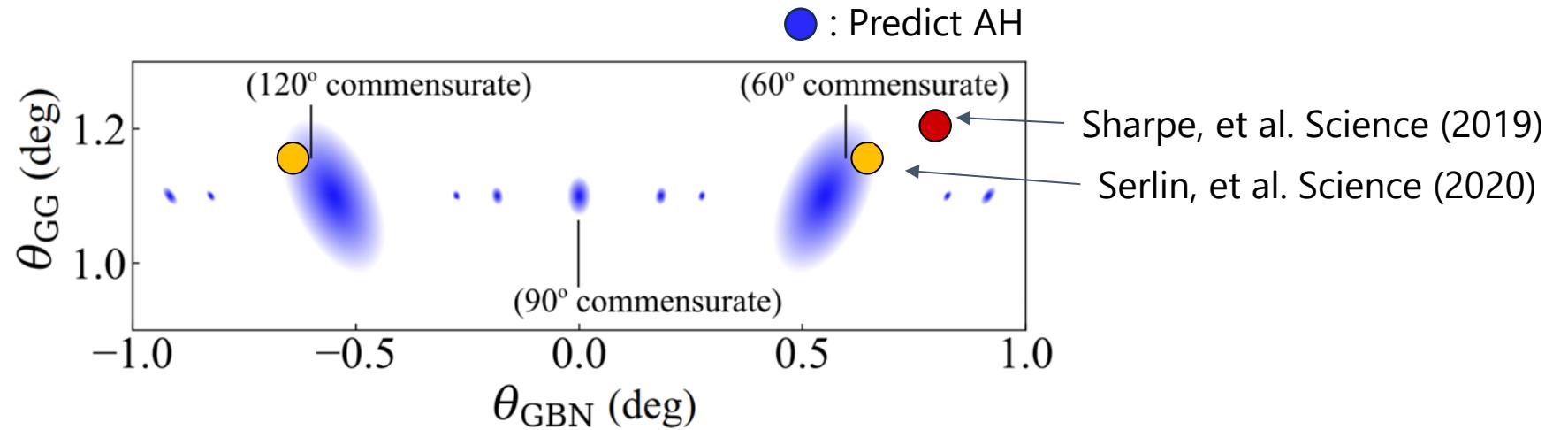
TBG/hBN multimoiré



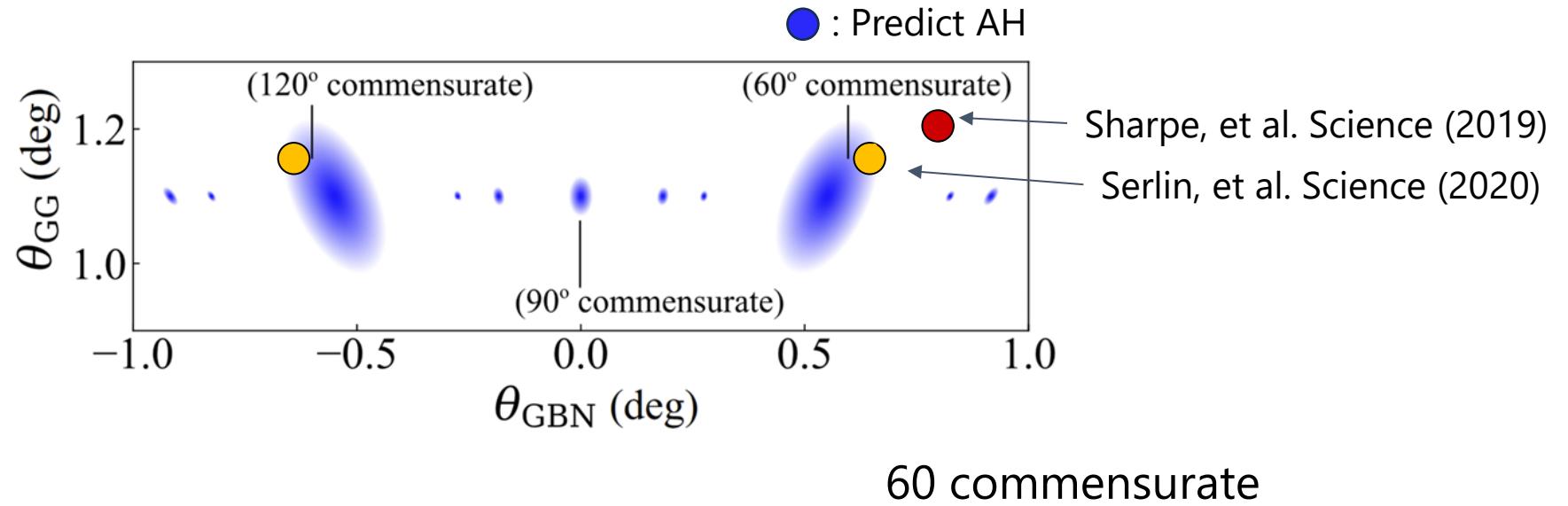
TBG/hBN multimoiré



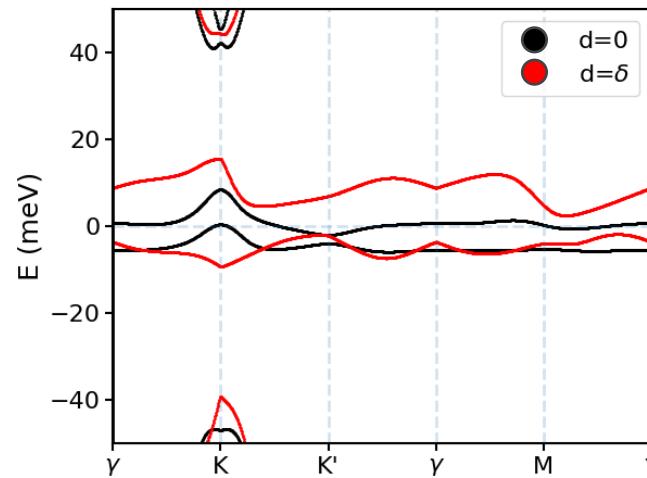
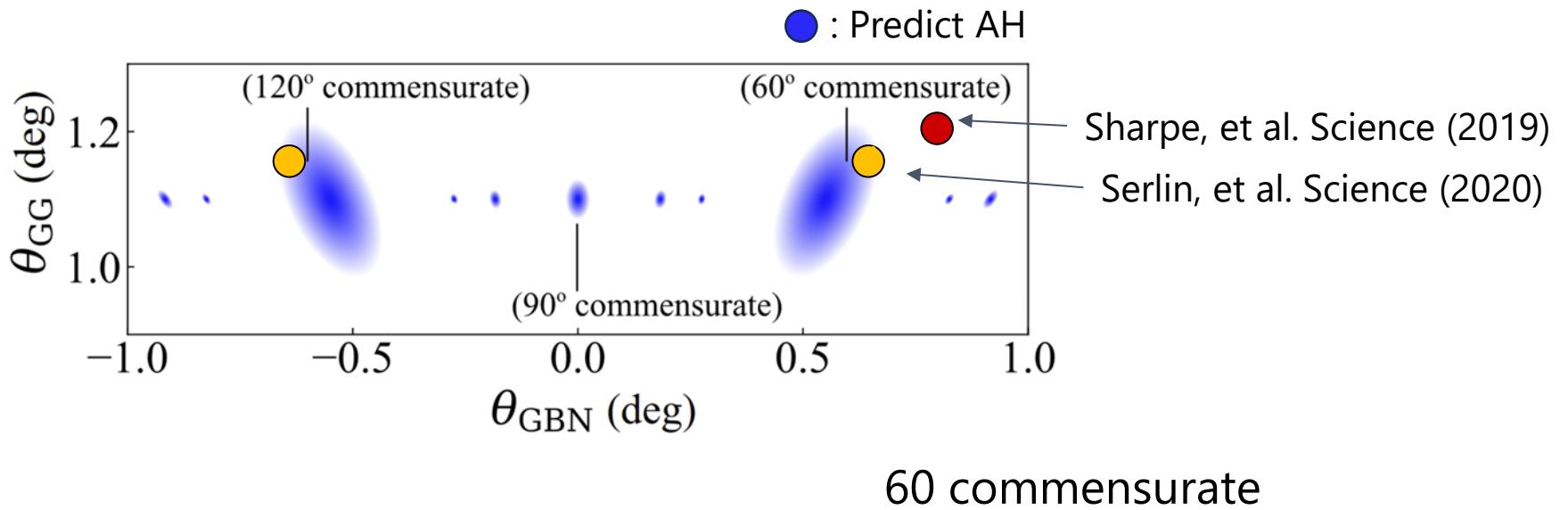
TBG/hBN multimoiré



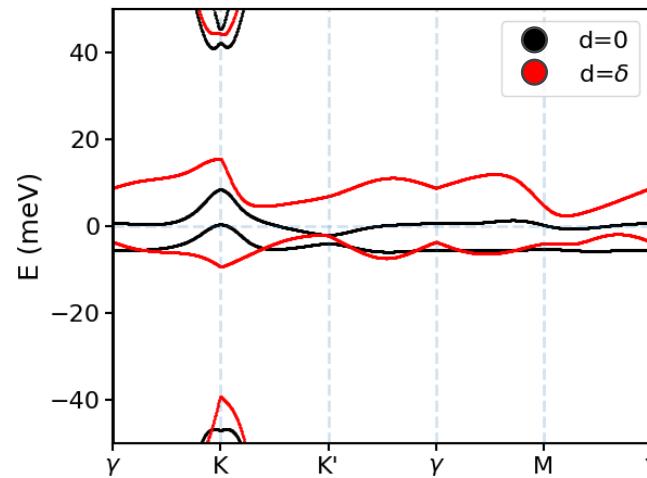
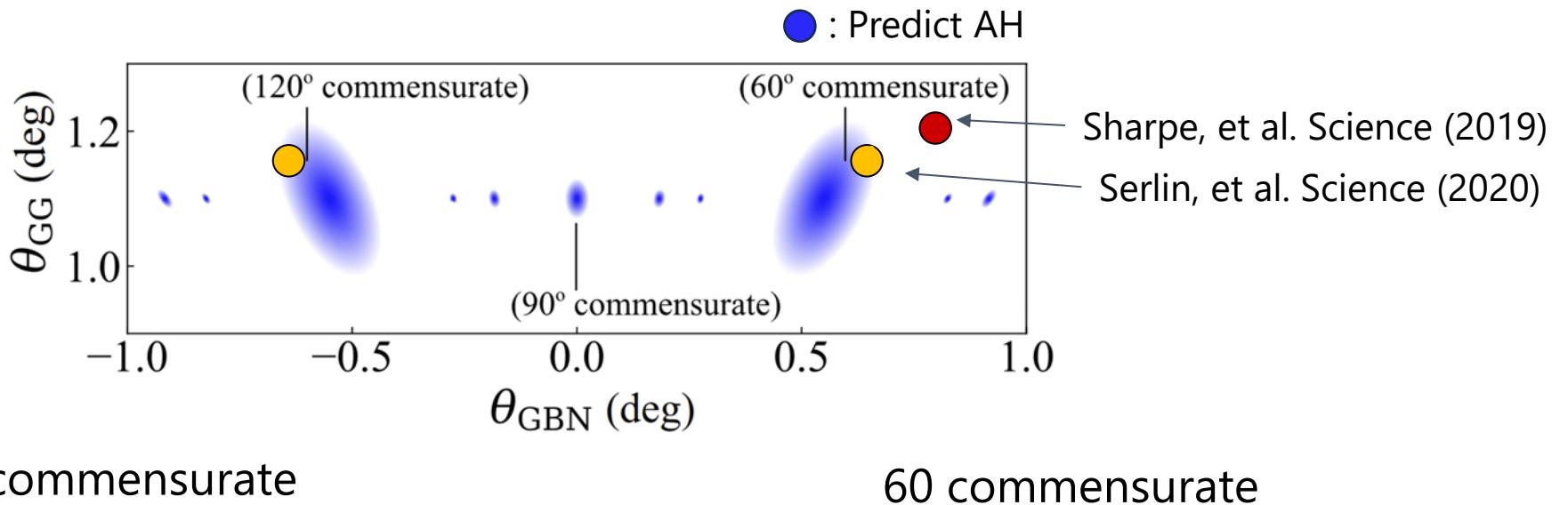
TBG/hBN multimoiré



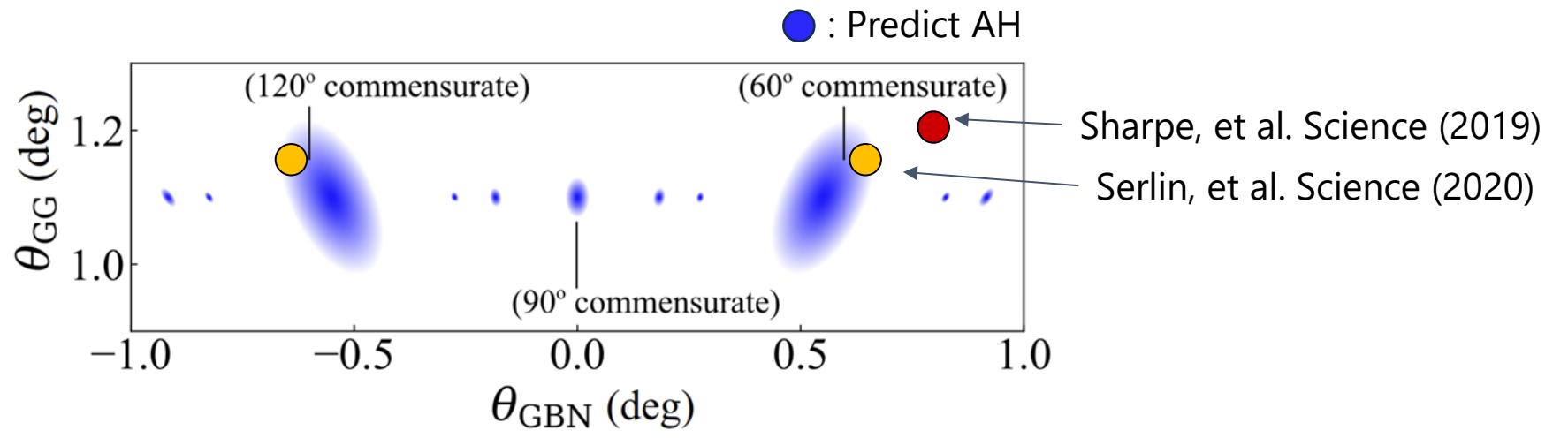
TBG/hBN multimoiré



TBG/hBN multimoiré

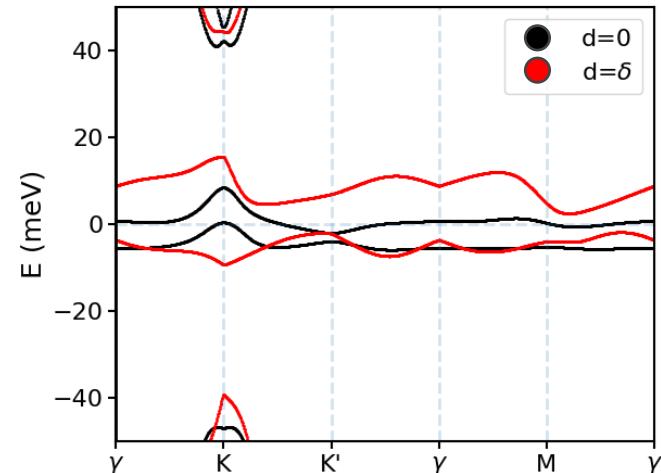
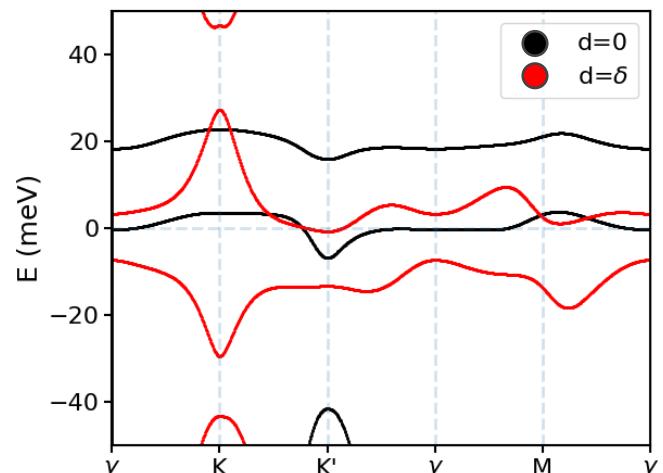


TBG/hBN multimoiré

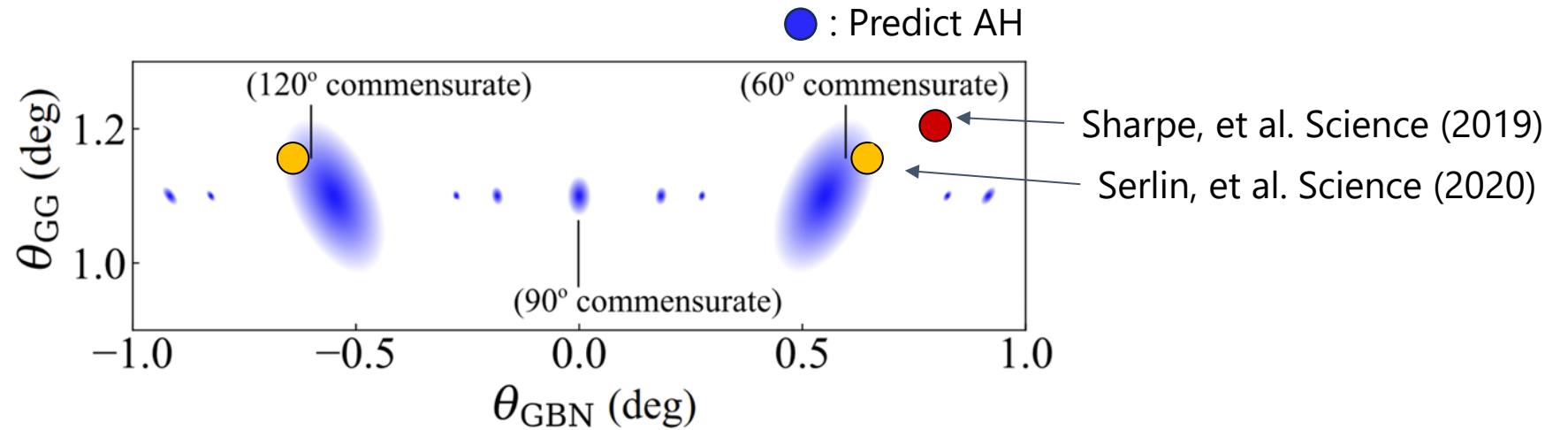


120 commensurate

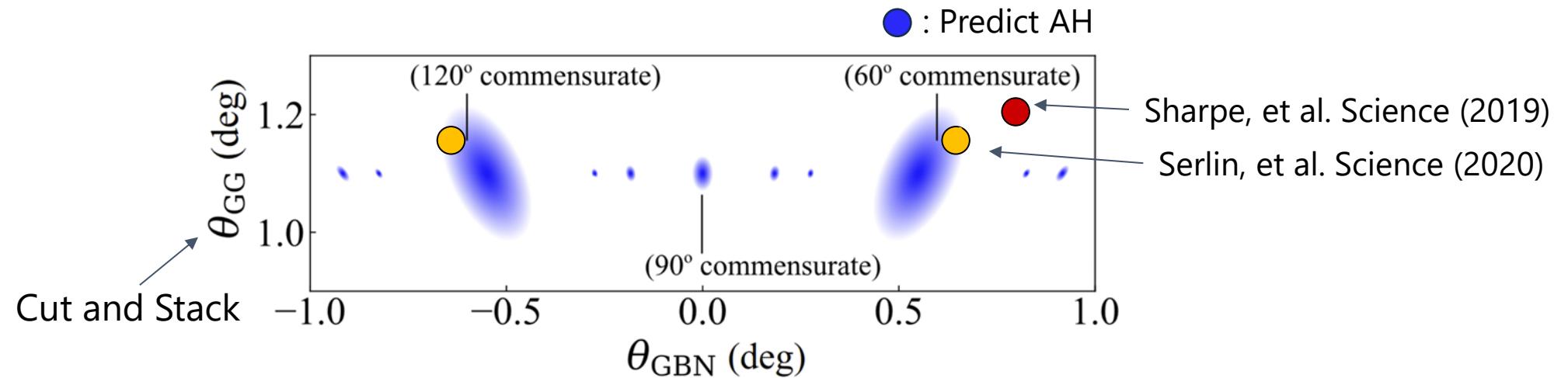
60 commensurate



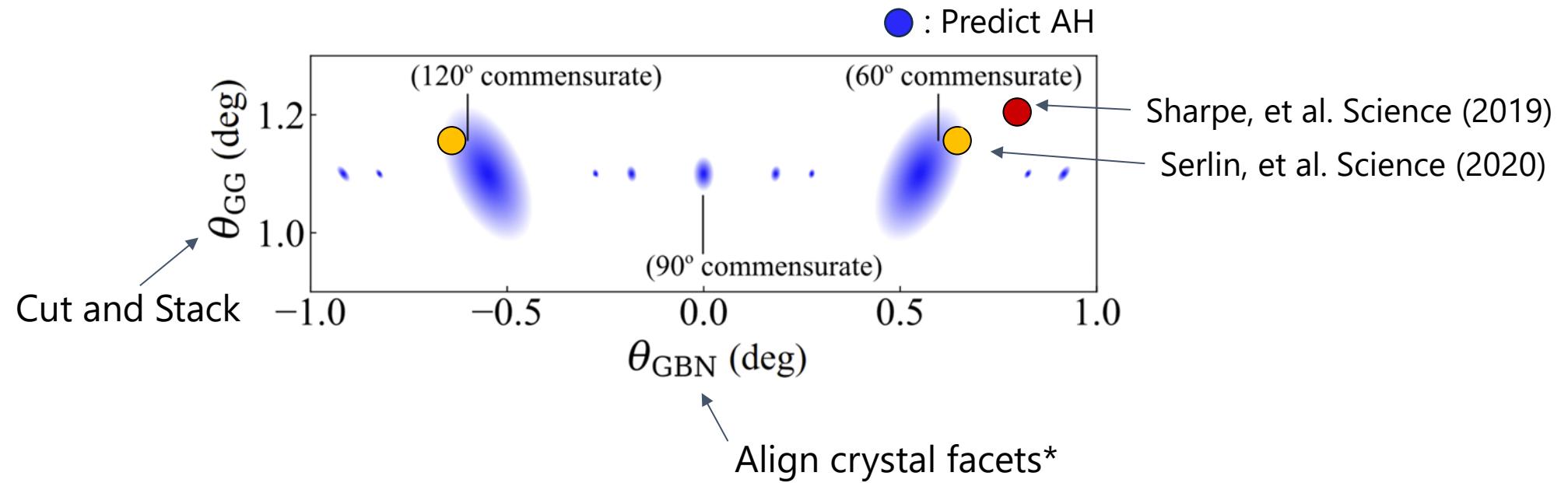
TBG/hBN multimoiré



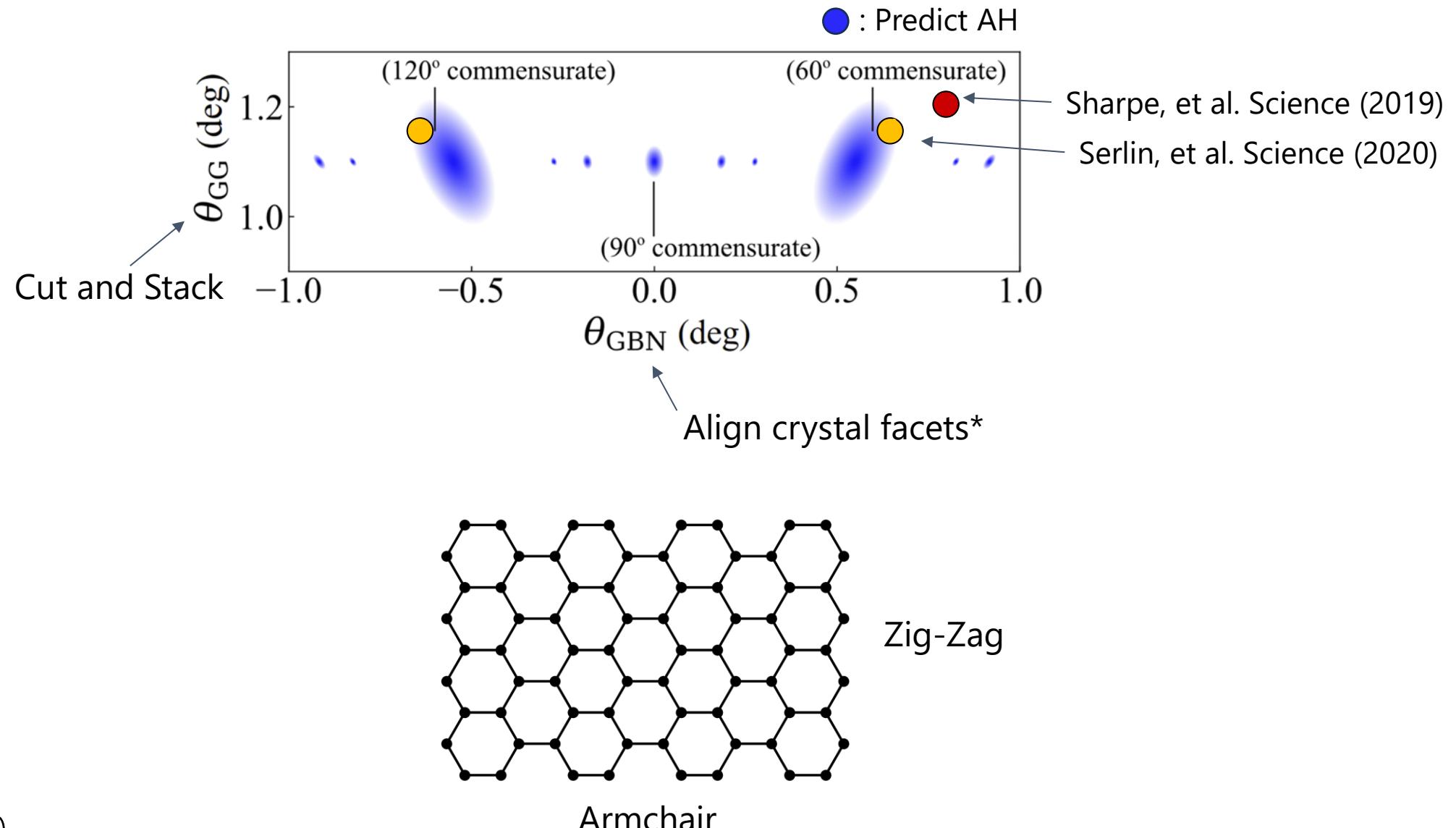
TBG/hBN multimoiré



TBG/hBN multimoiré

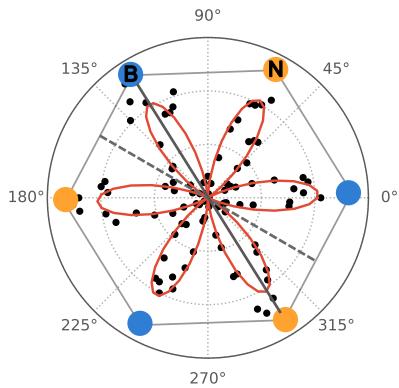
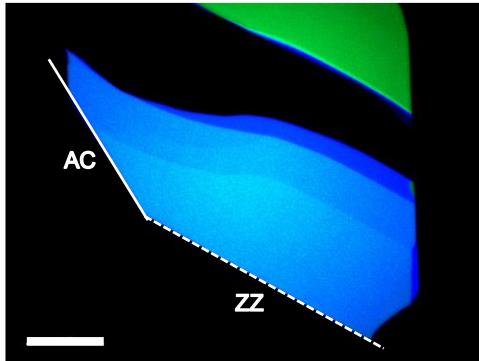


TBG/hBN multimoiré



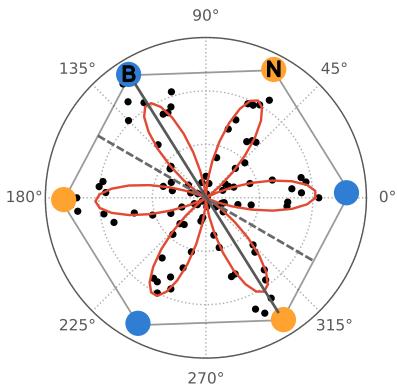
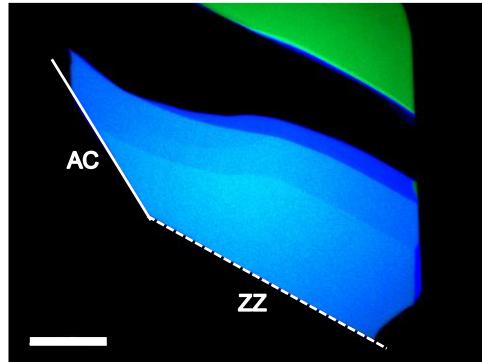
Deterministic fab with rapid feedback

Second harmonic generation for hBN

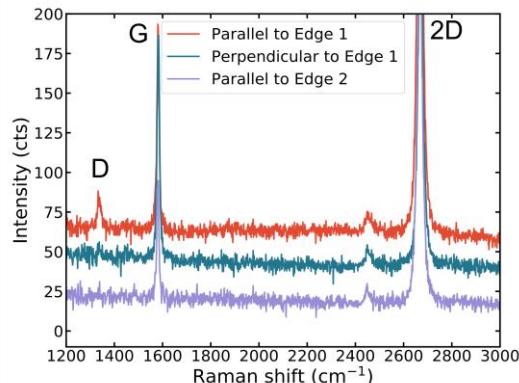
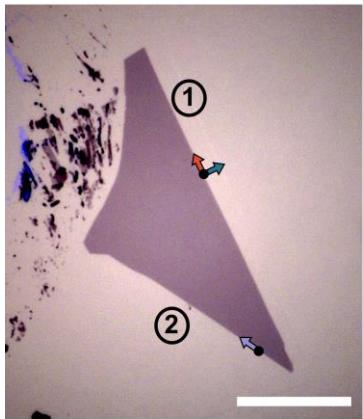


Deterministic fab with rapid feedback

Second harmonic generation for hBN

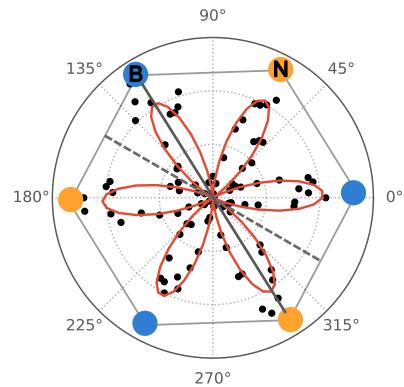
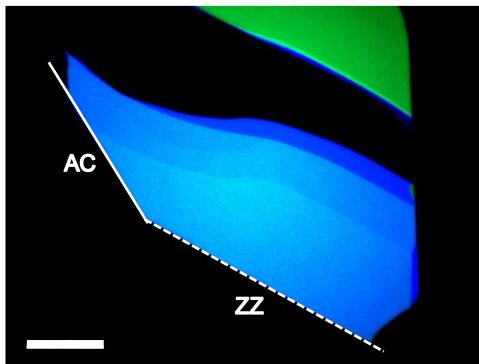


Polarized Raman for graphene

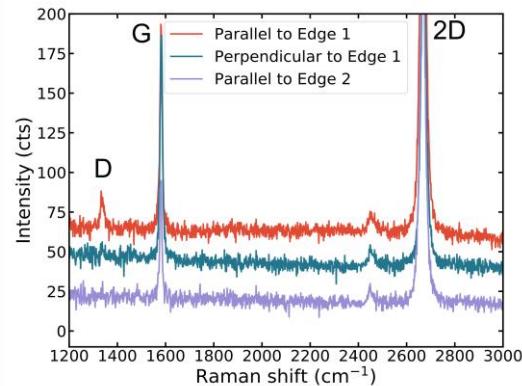
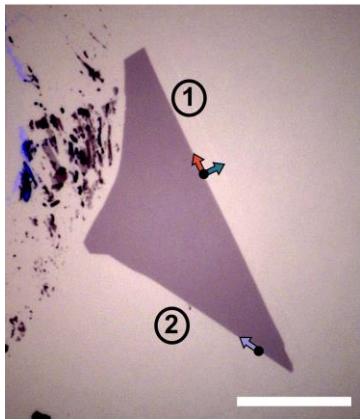


Deterministic fab with rapid feedback

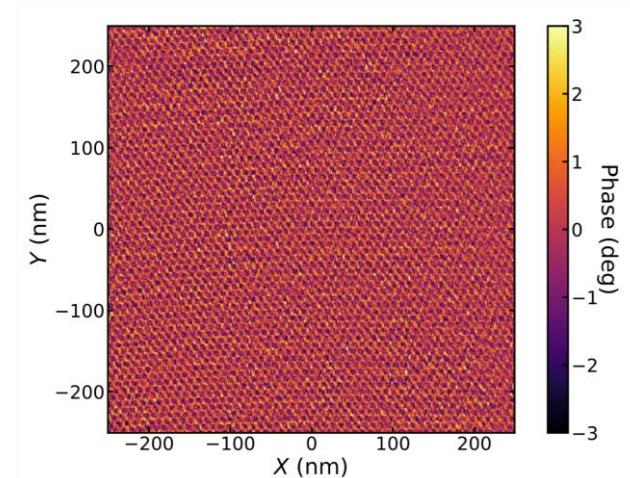
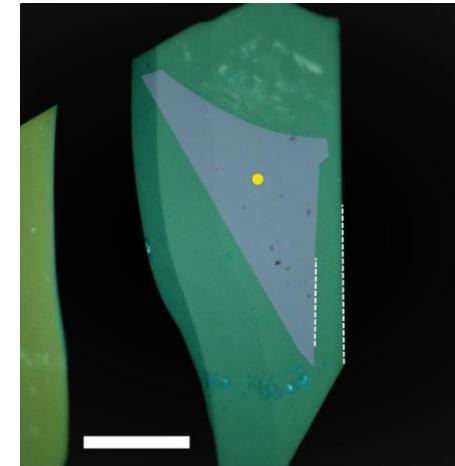
Second harmonic generation for hBN



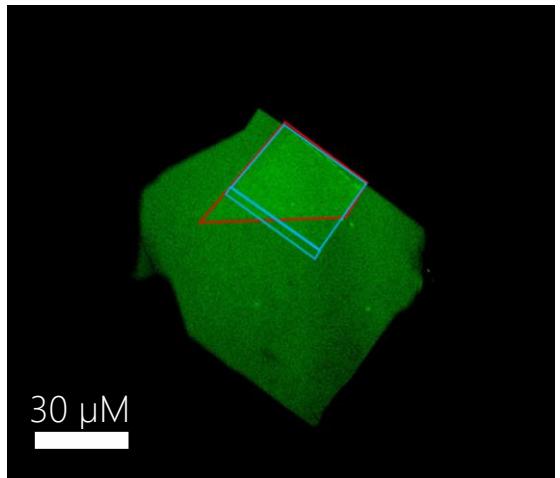
Polarized Raman for graphene



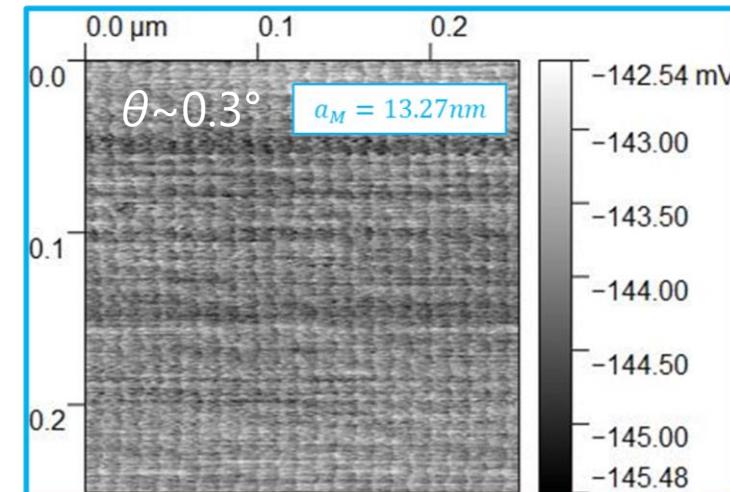
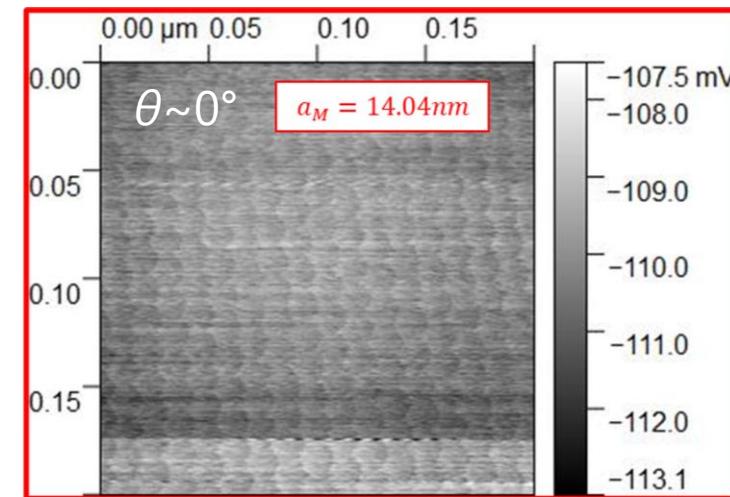
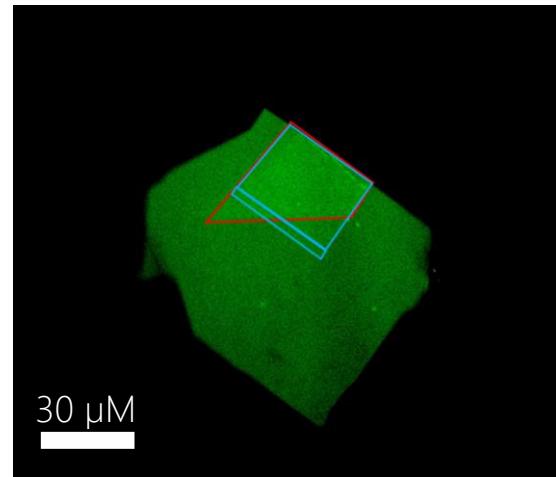
Torsional force microscopy for moirés



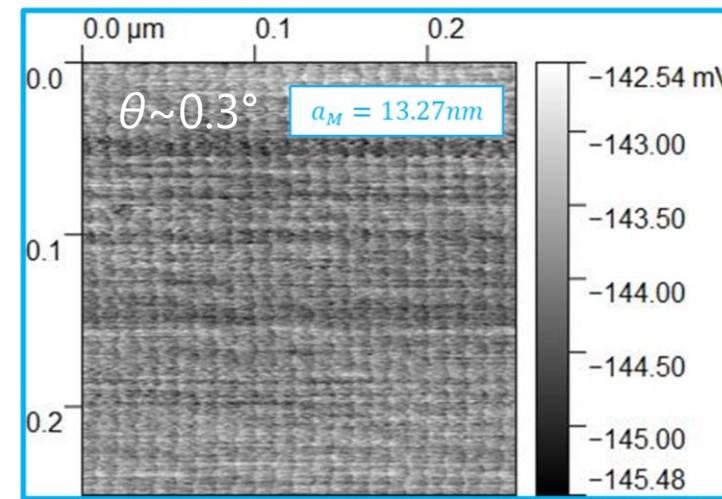
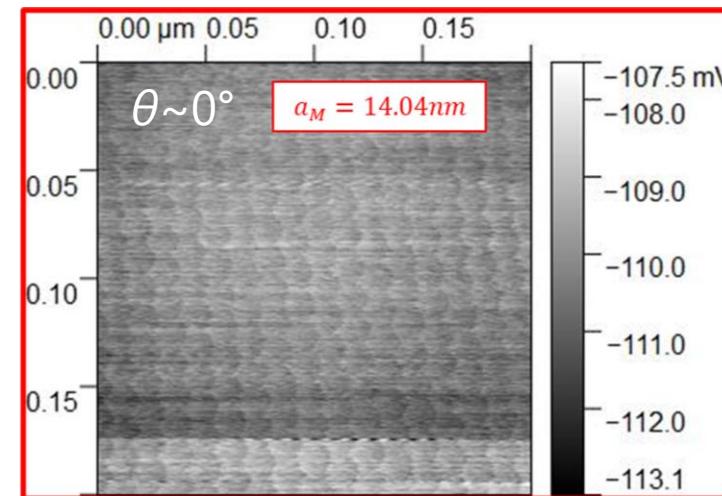
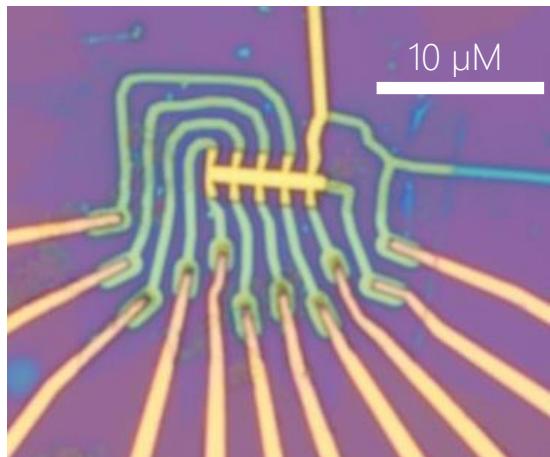
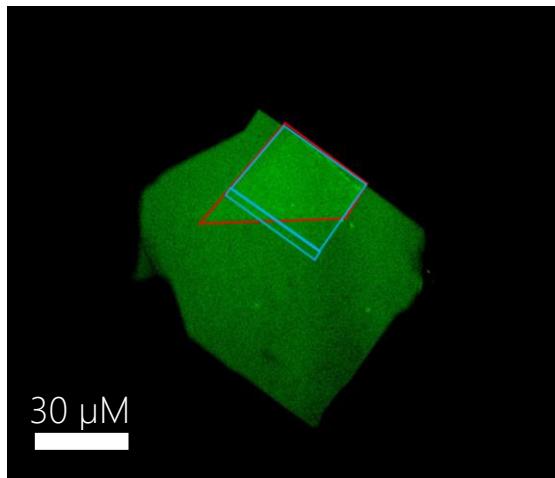
Deterministic fab with rapid feedback



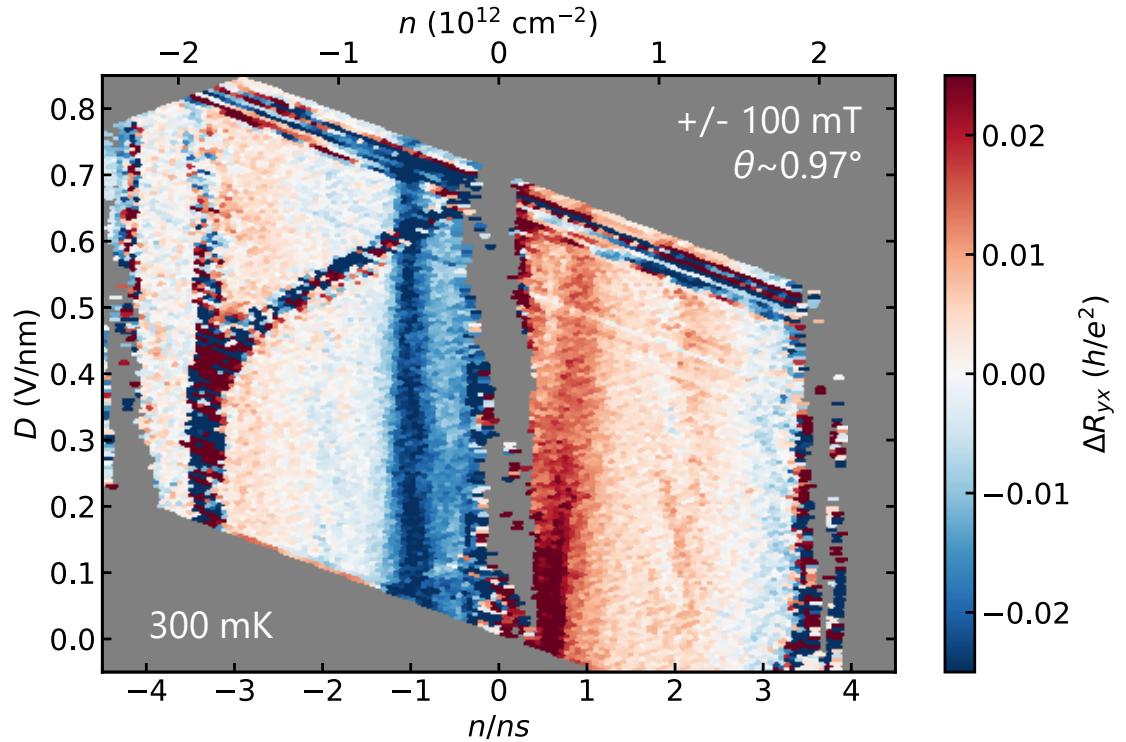
Deterministic fab with rapid feedback



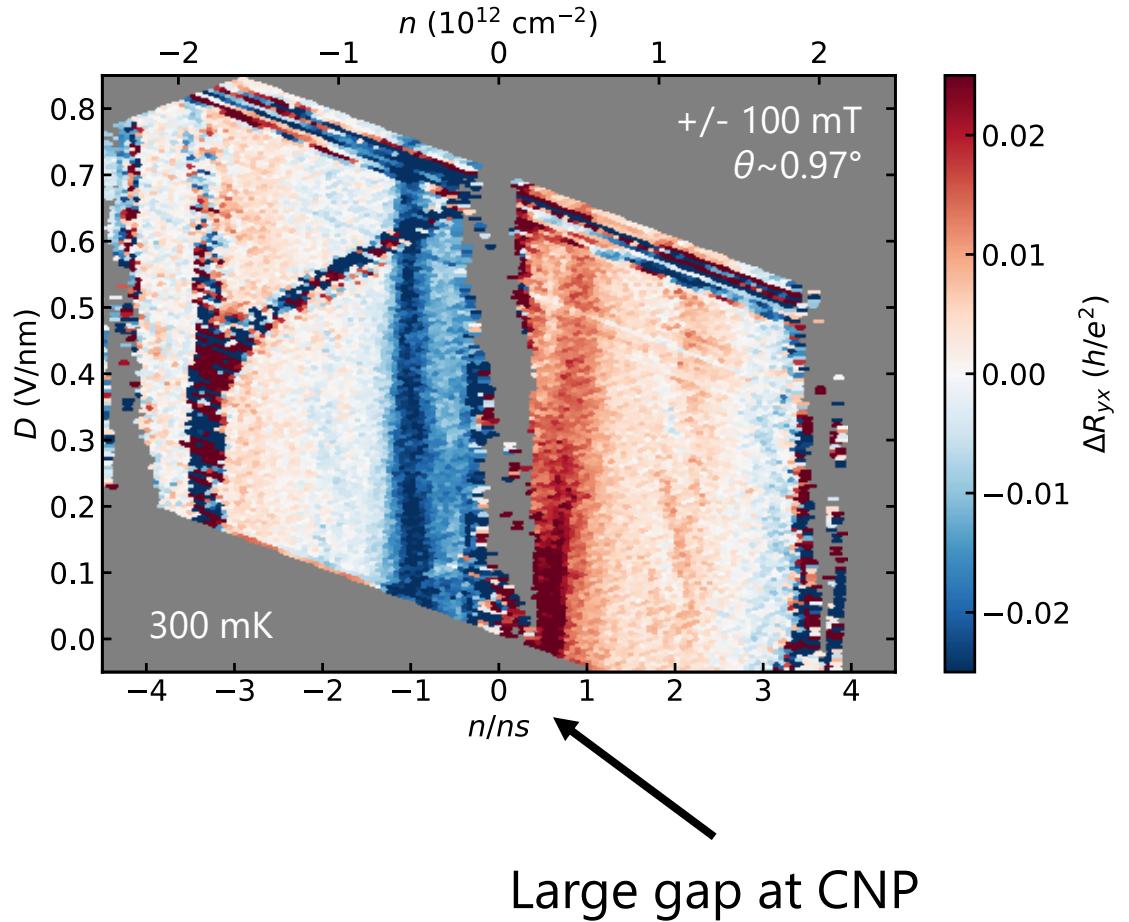
Deterministic fab with rapid feedback



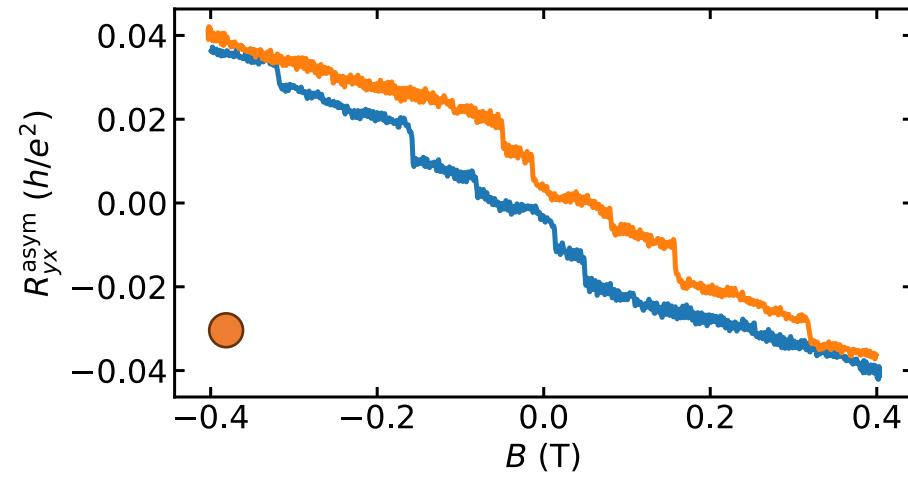
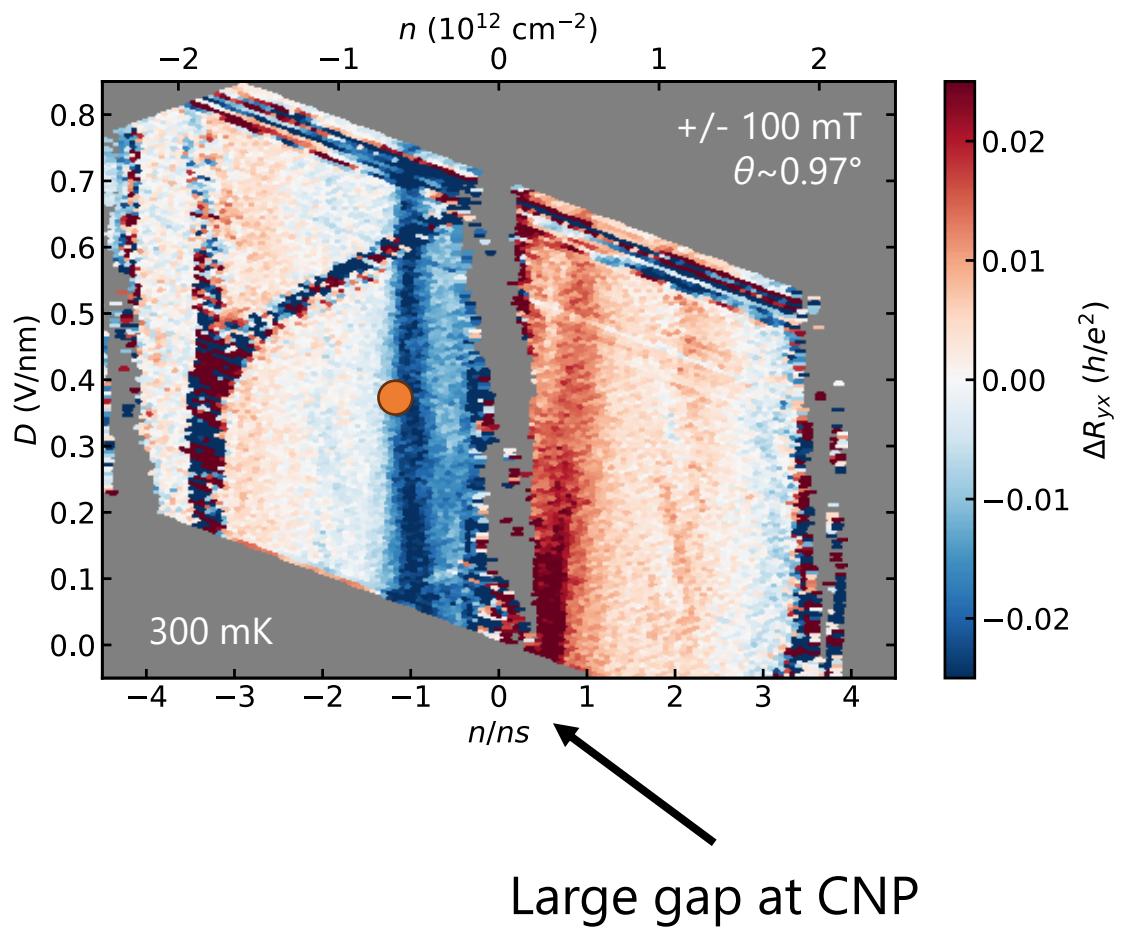
Anomalous Hall!



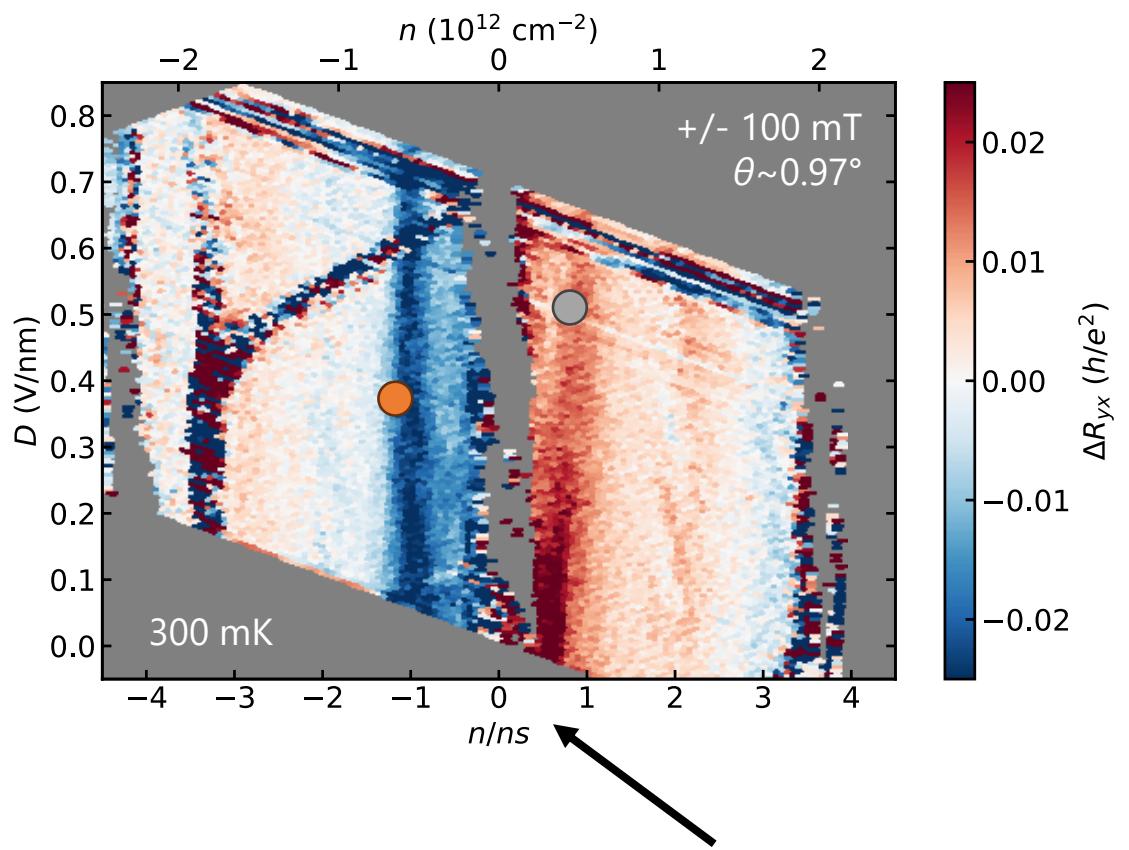
Anomalous Hall!



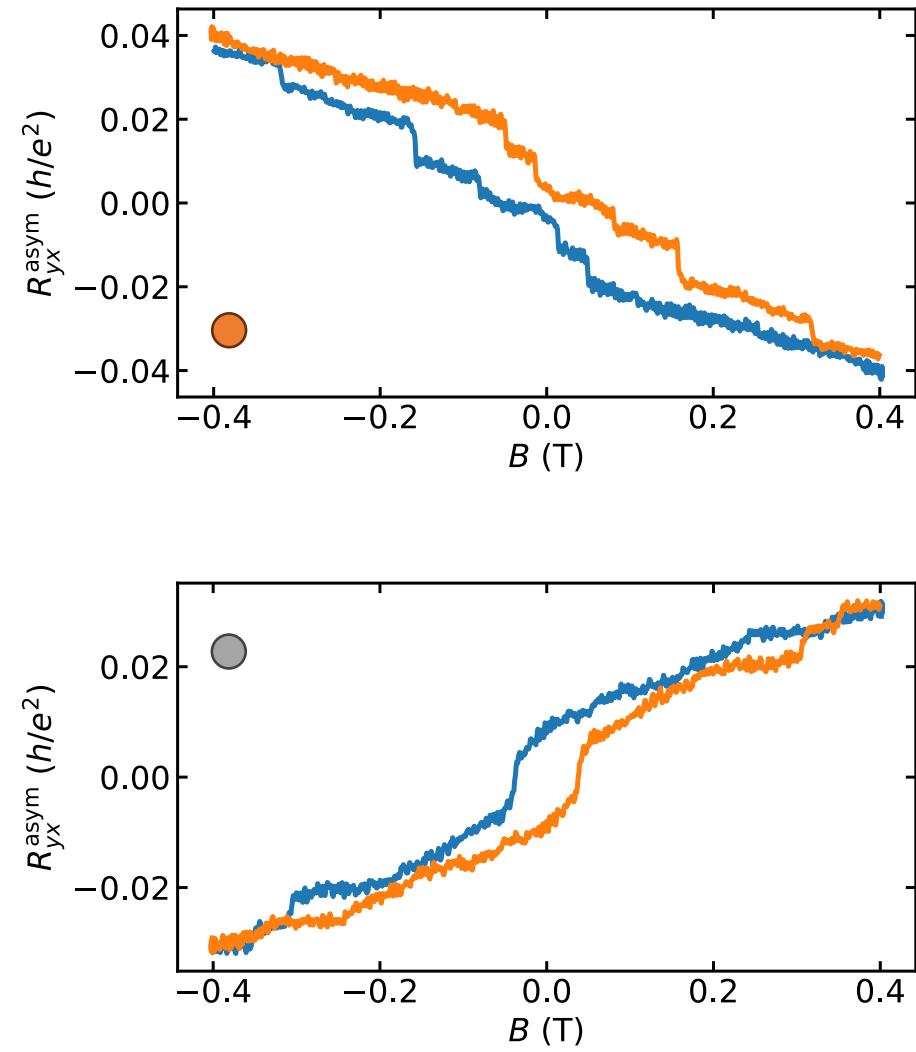
Anomalous Hall!



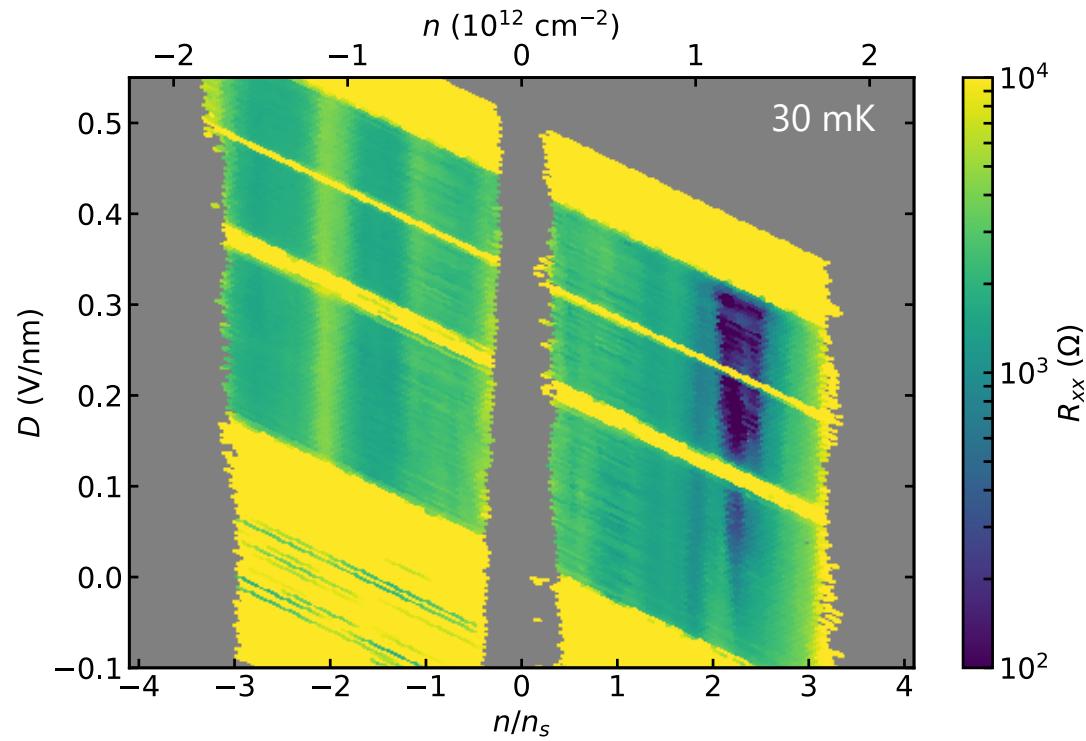
Anomalous Hall!



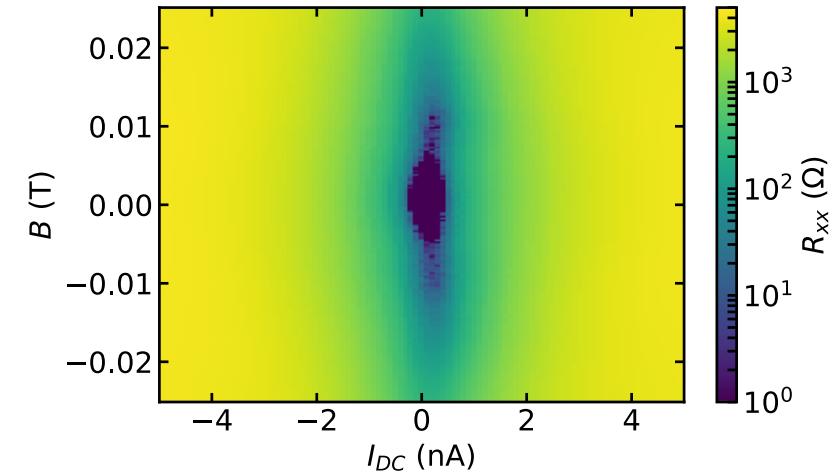
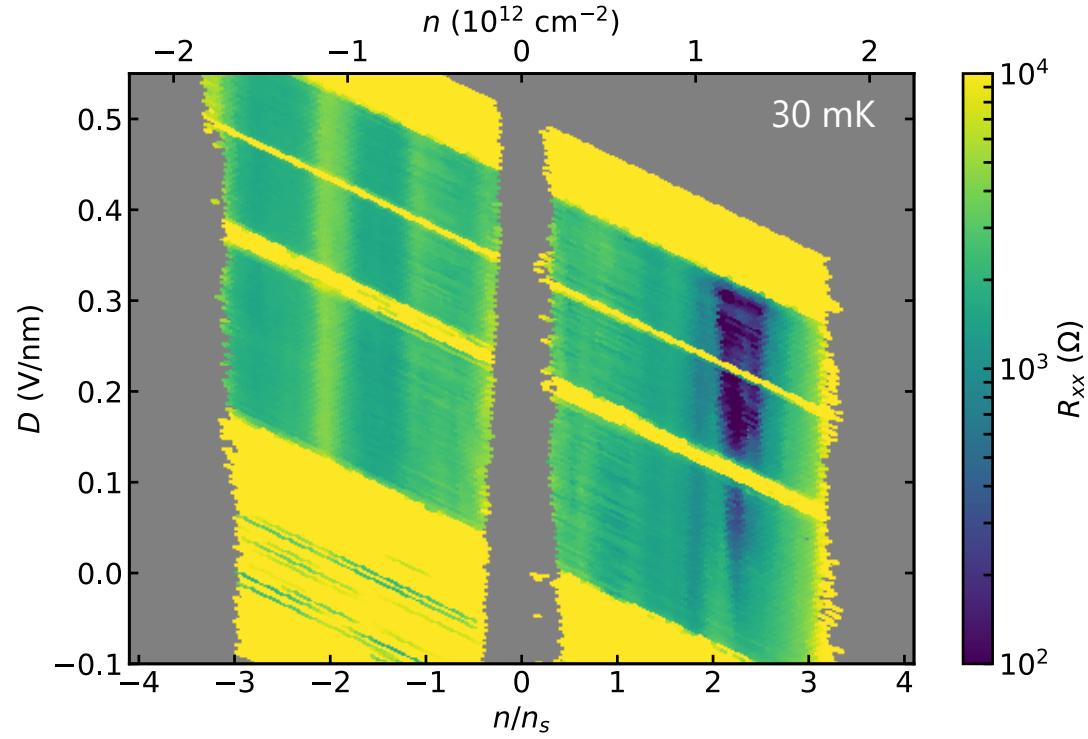
Large gap at CNP



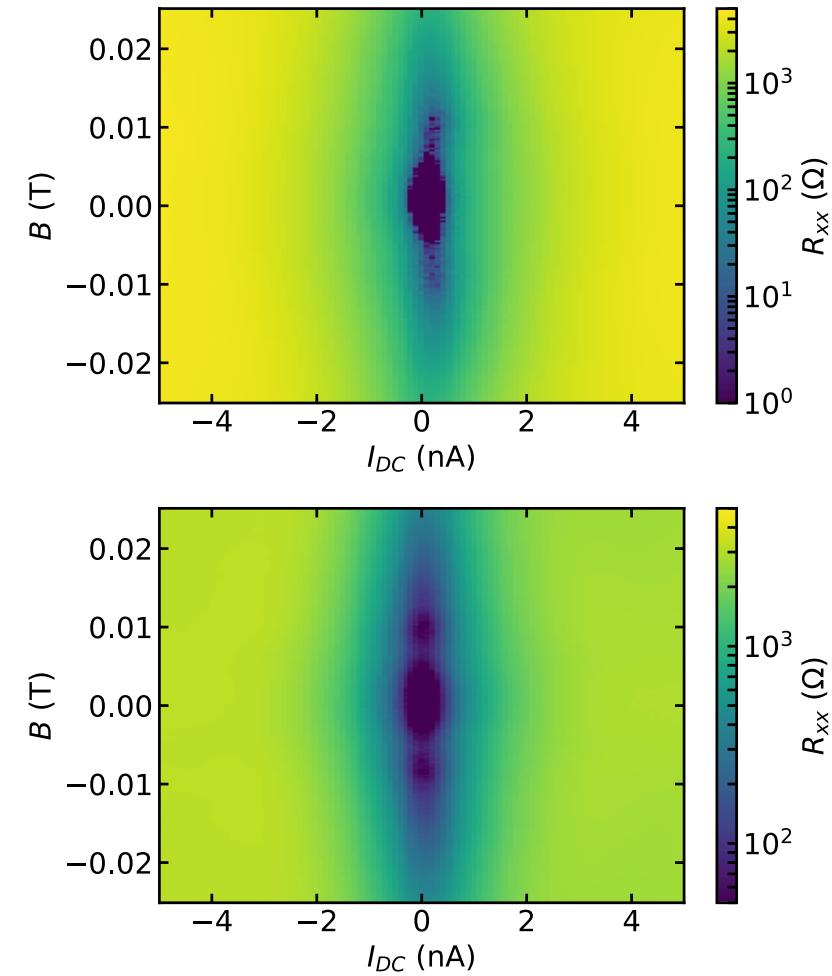
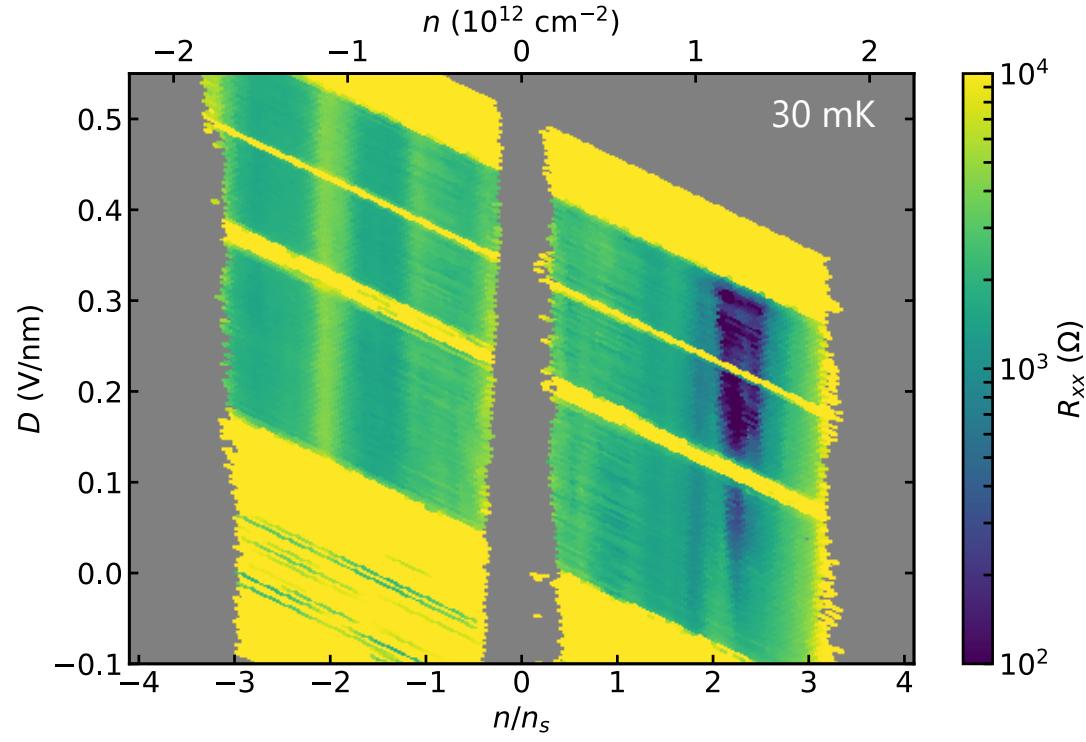
Cooling to 30 mK, superconductivity!?



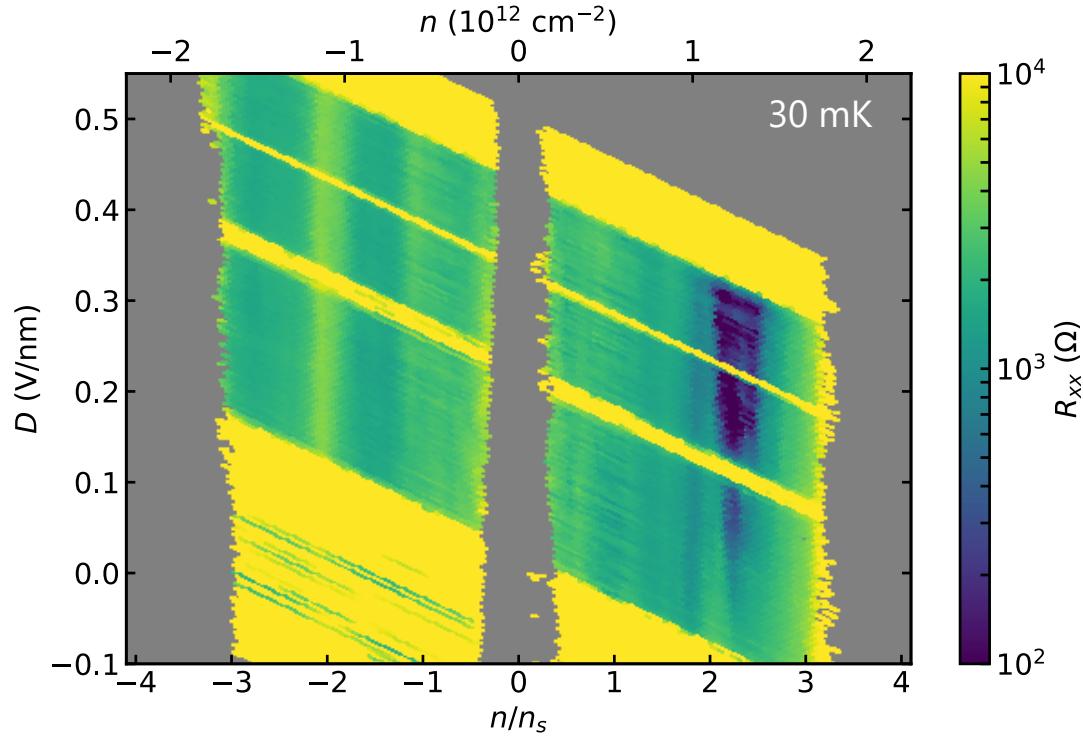
Cooling to 30 mK, superconductivity!?



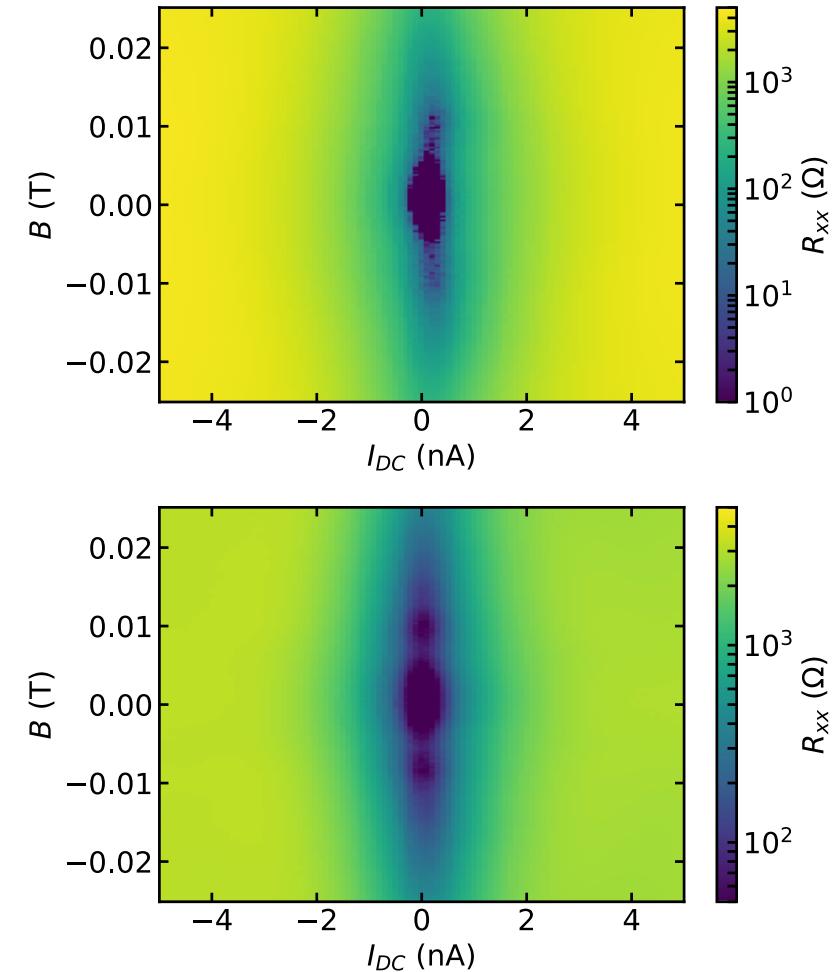
Cooling to 30 mK, superconductivity!?



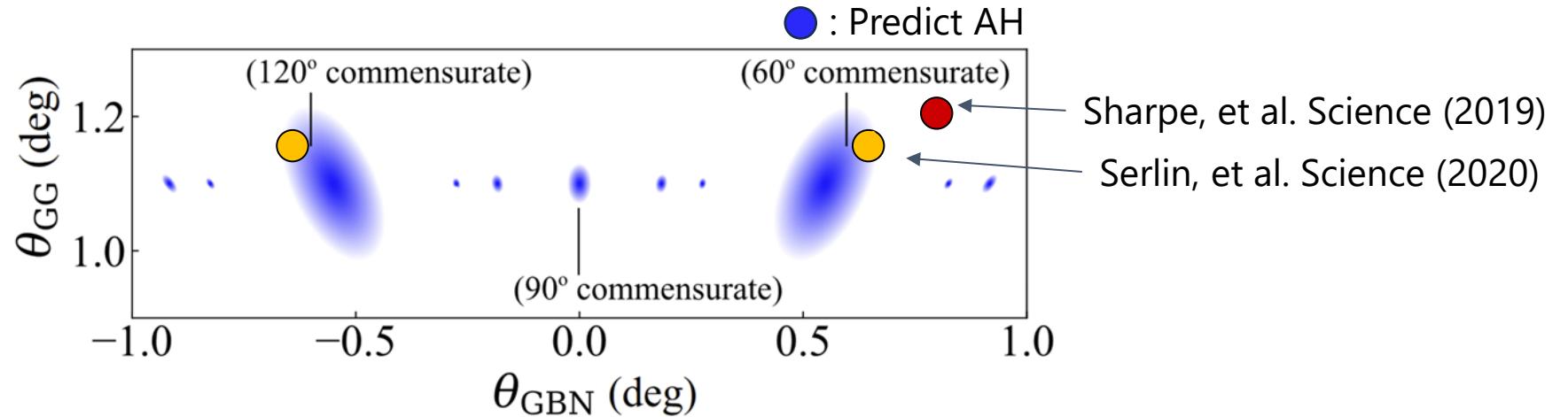
Cooling to 30 mK, superconductivity!?



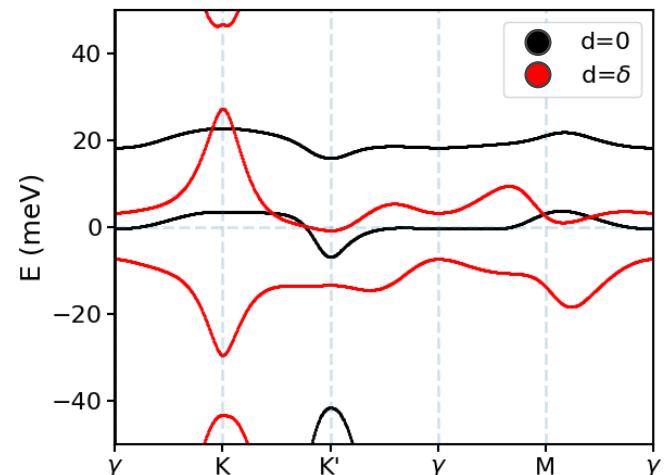
This device exhibits both magnetism and superconductivity!



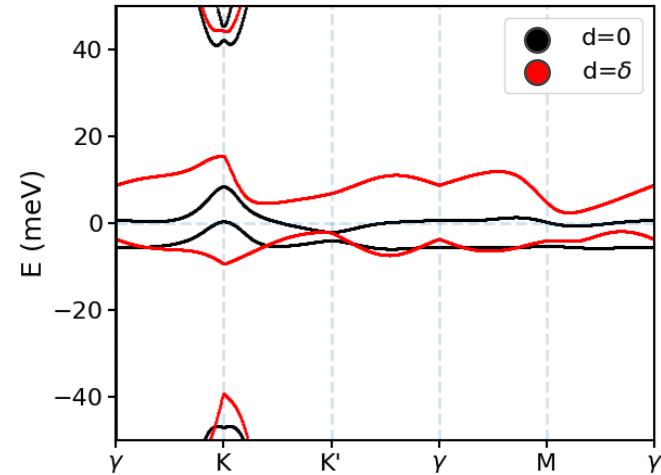
What is θ_{GBN} ?



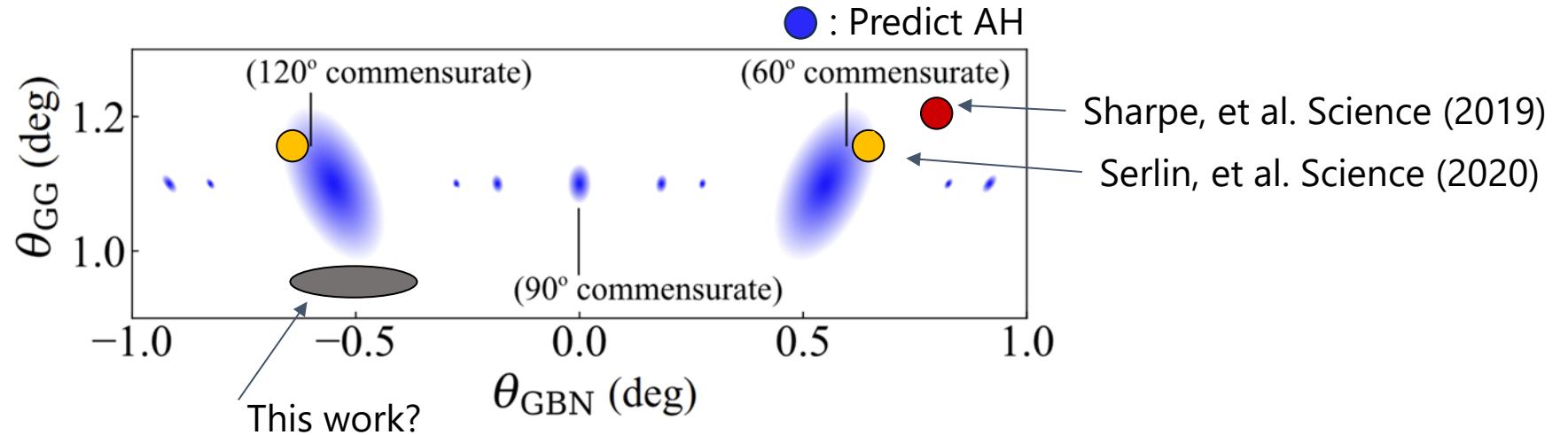
120 commensurate



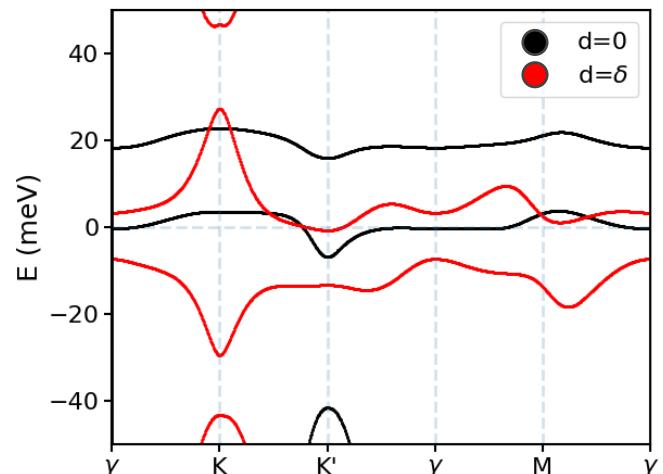
60 commensurate



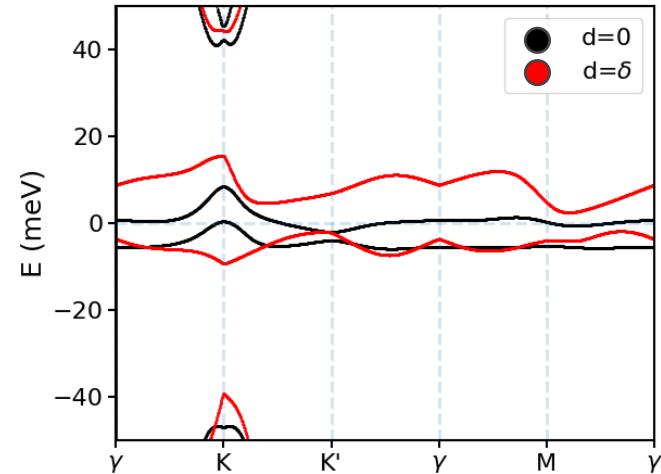
What is θ_{GBN} ?



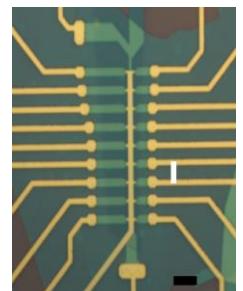
120 commensurate



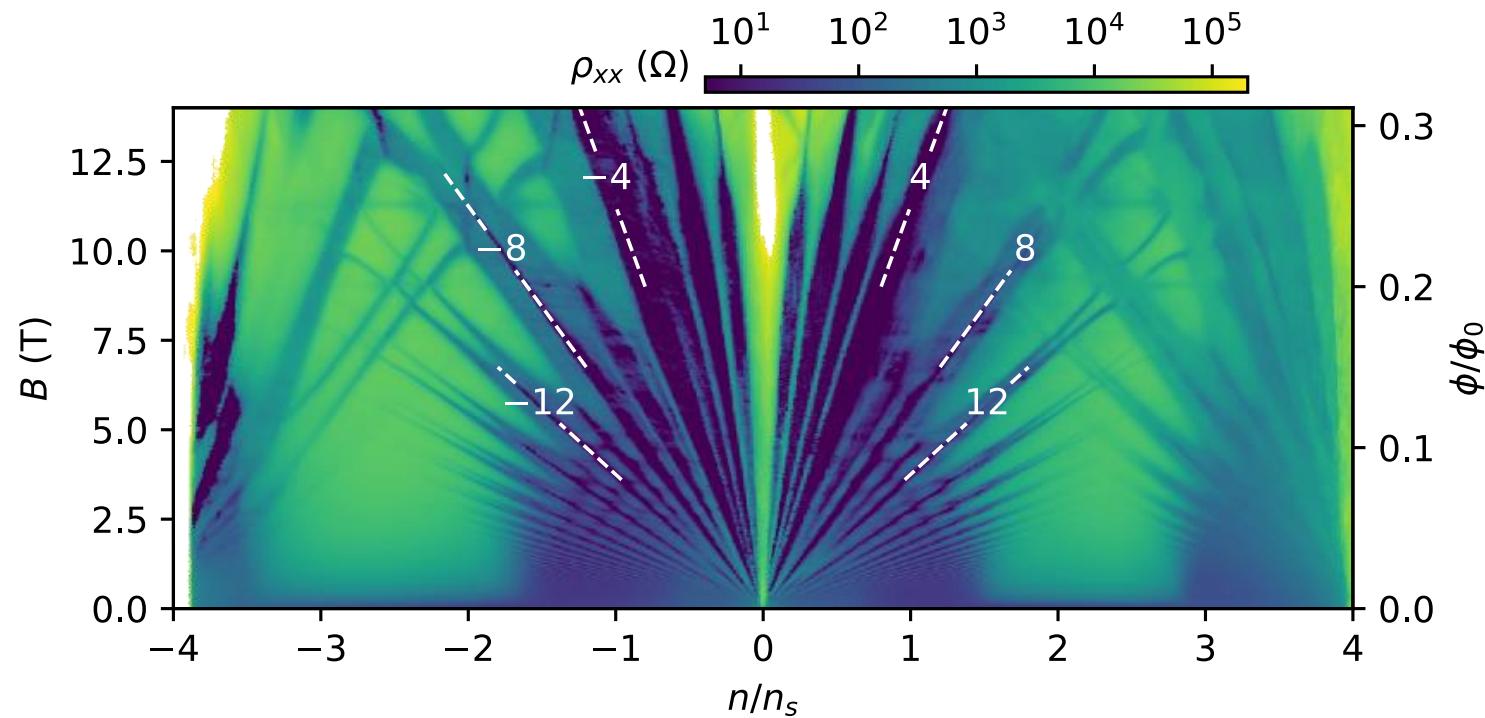
60 commensurate



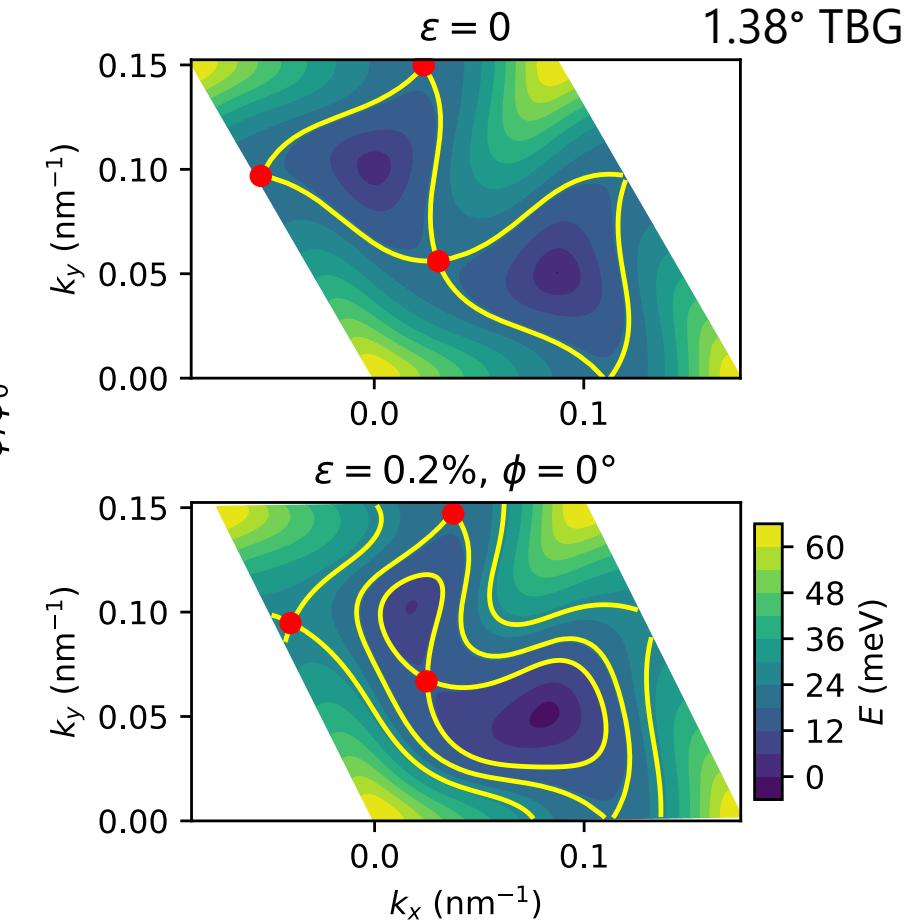
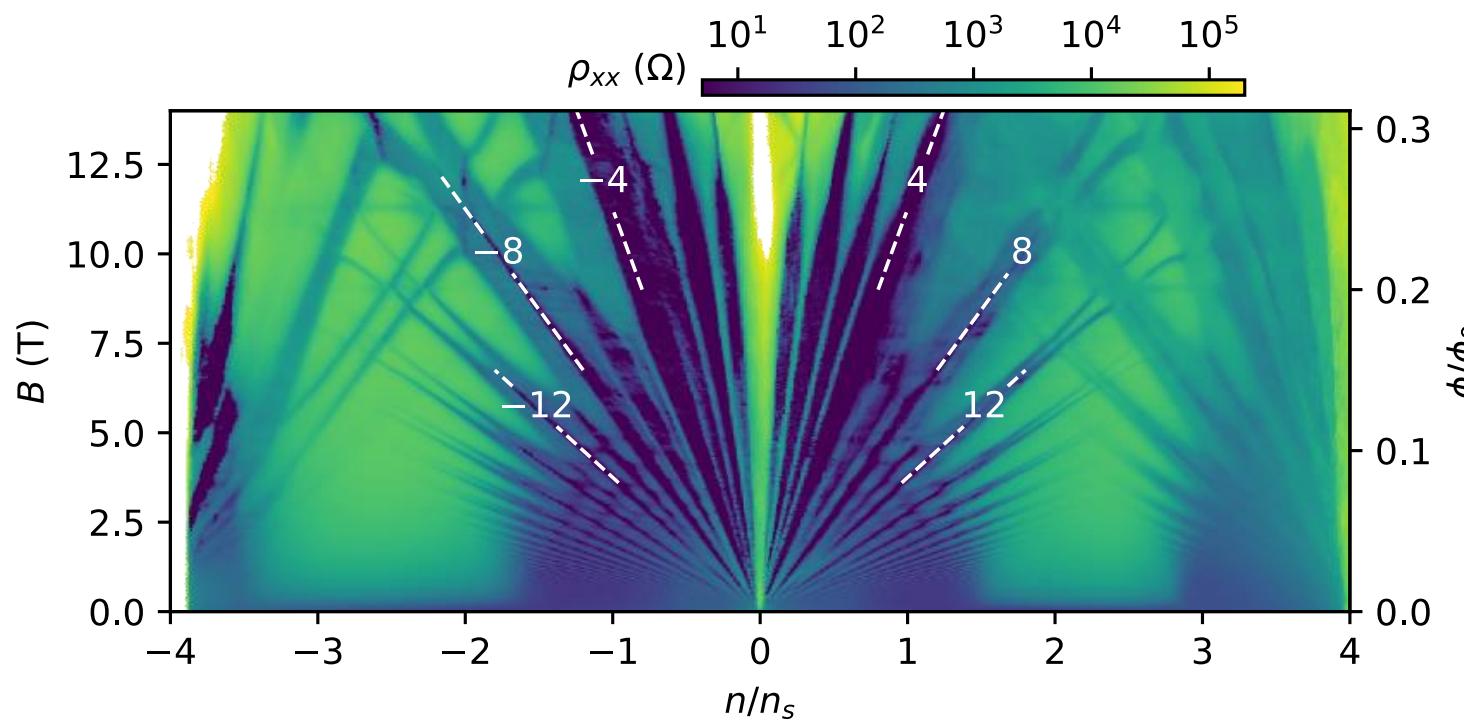
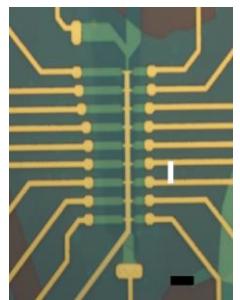
TBG is still hiding secrets



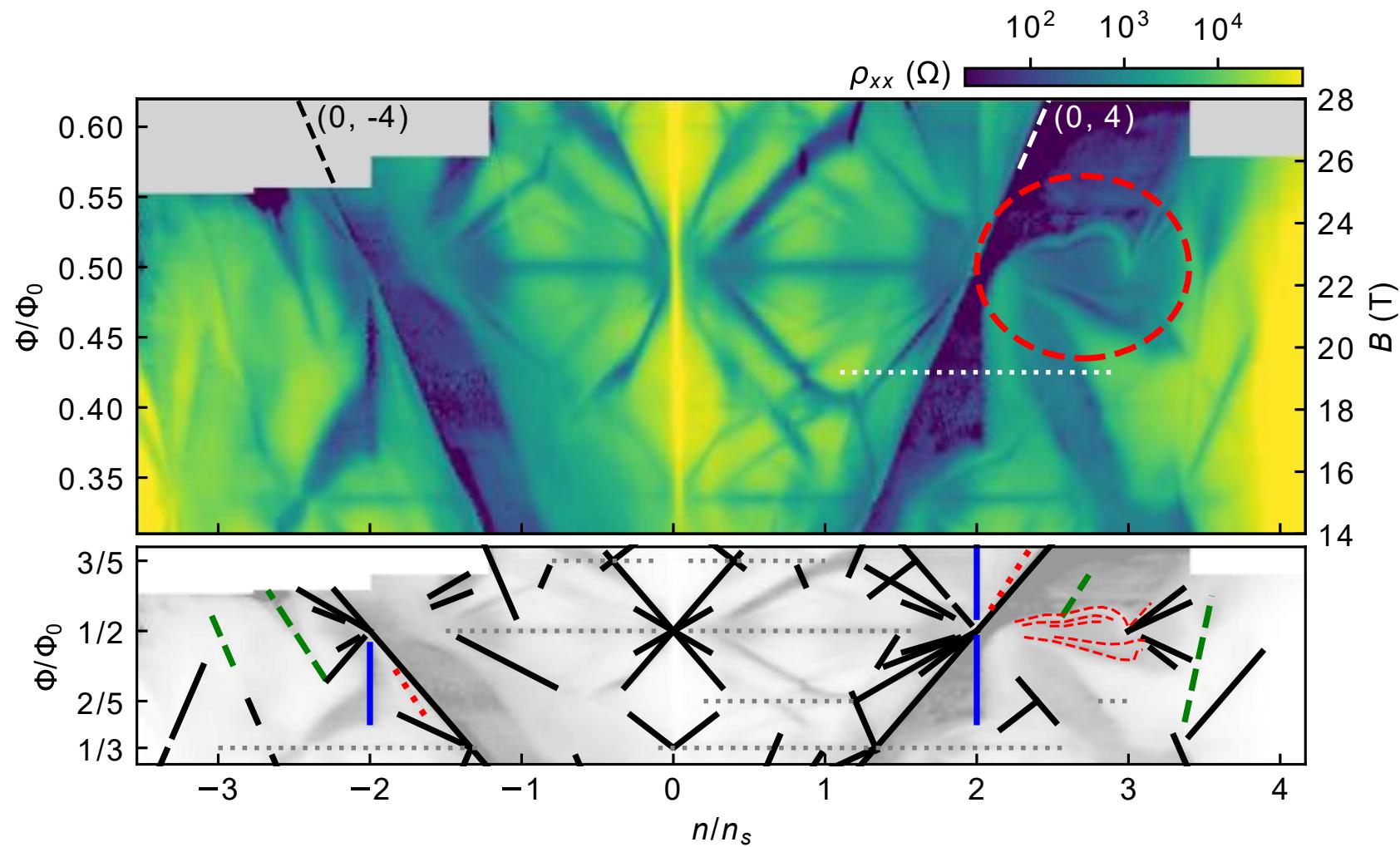
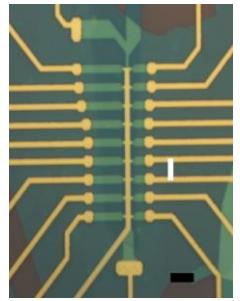
1.38° TBG



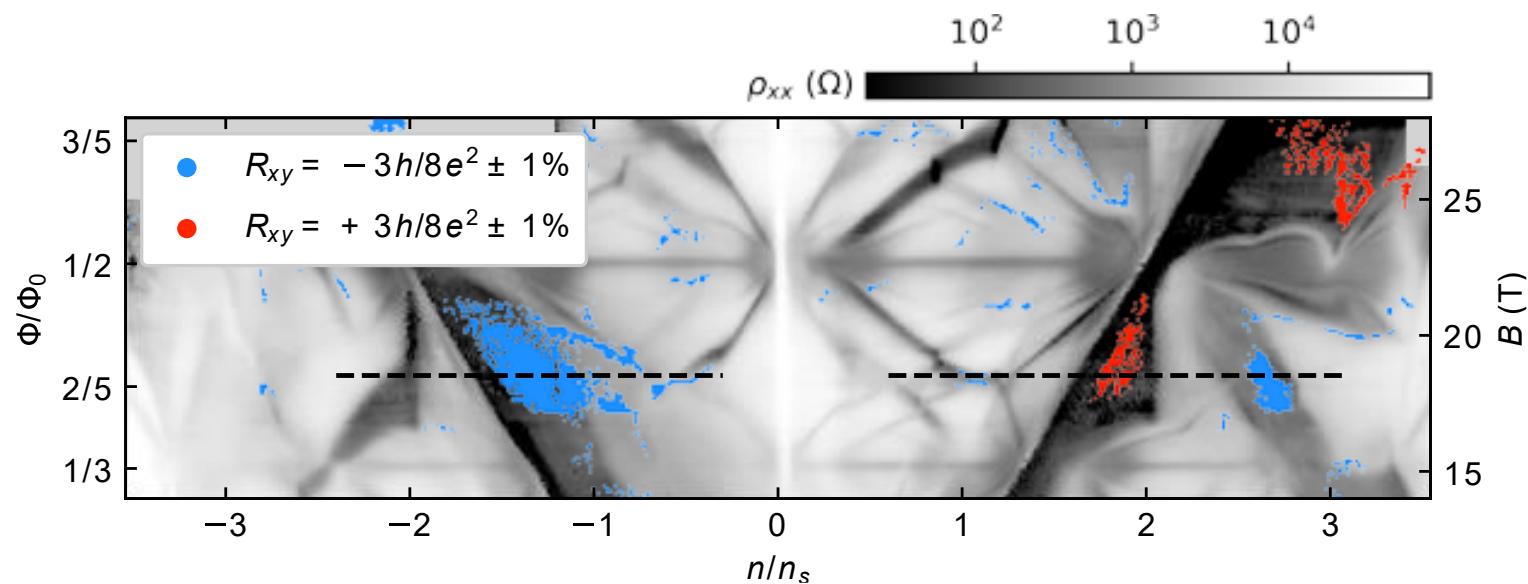
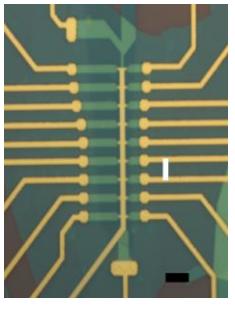
TBG is still hiding secrets



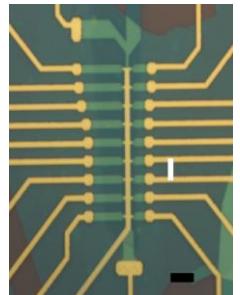
TBG is still hiding secrets



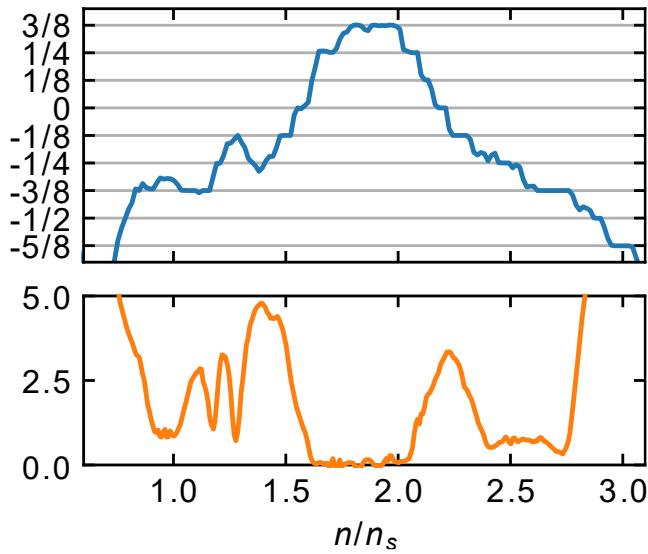
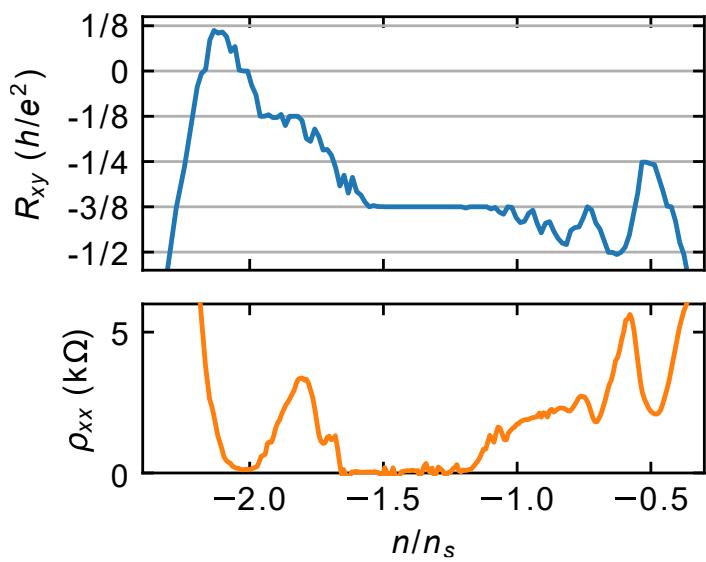
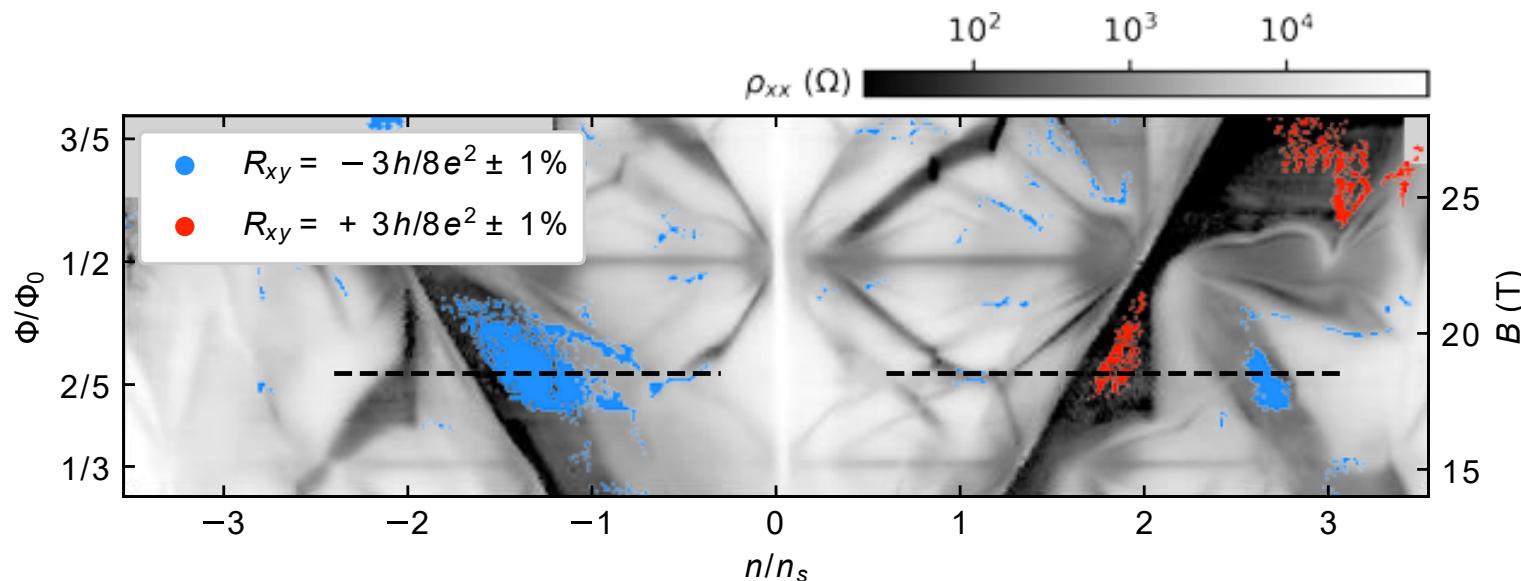
TBG is still hiding secrets



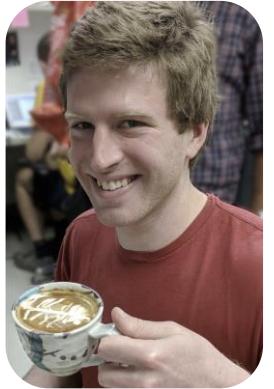
TBG is still hiding secrets



1.38° TBG



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Summary

By controlling twist angle, graphene multilayers can become strongly correlated!

TBG is a Chern insulator at filling factor 3!

Magnetism is orbital in character
hBN alignment may be crucial

Why is this orbital magnet so hard to reproduce in TBG?

(Q)AH appears to be quite ubiquitous in moirés
hBN's effect is shift dependent!

Expect AH near commensurate structures?

Using intermediate characterization during fab we:

Verified hBN alignment, AH, and superconductivity in a single sample

We still have much to learn about TBG. Explore:

G-G and G-hBN twist phase space
FCI states at high field

