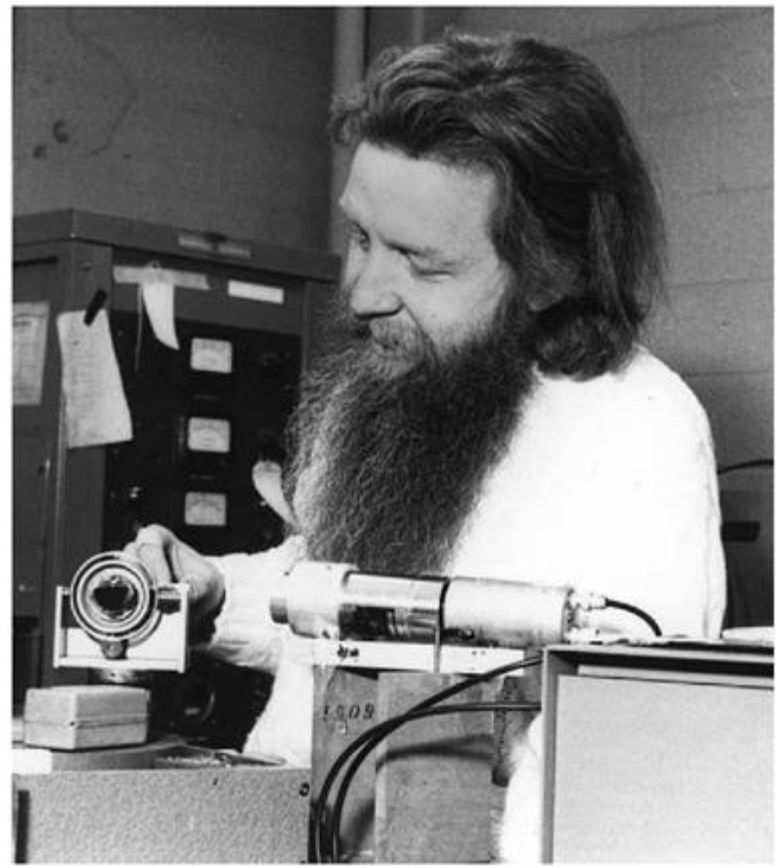
Making Carbon Magnetic

Aaron Sharpe

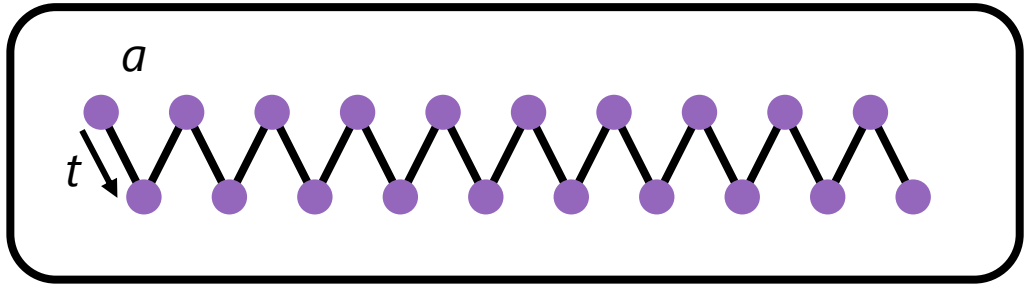
Bill McMillan Colloquium

October 2nd, 2024

Slides available @ aaronsharpe.science



Zig-Zag Chain



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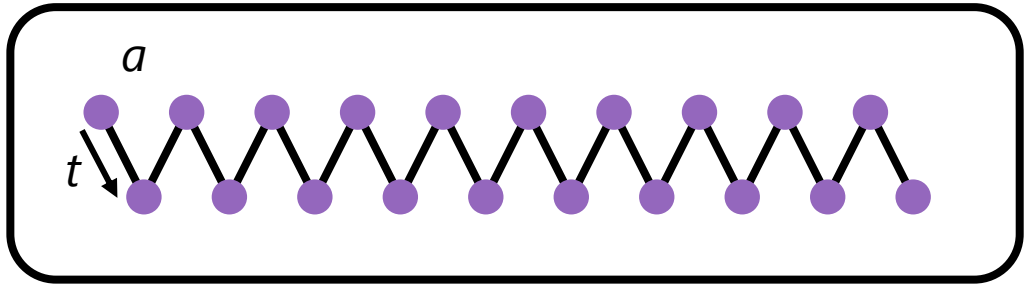

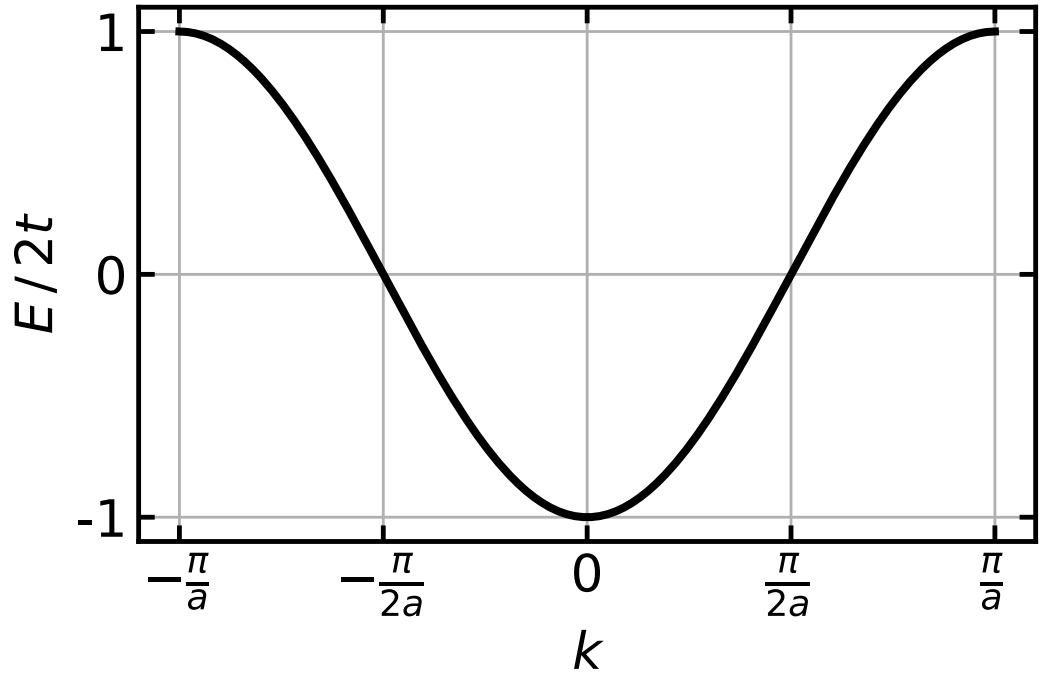

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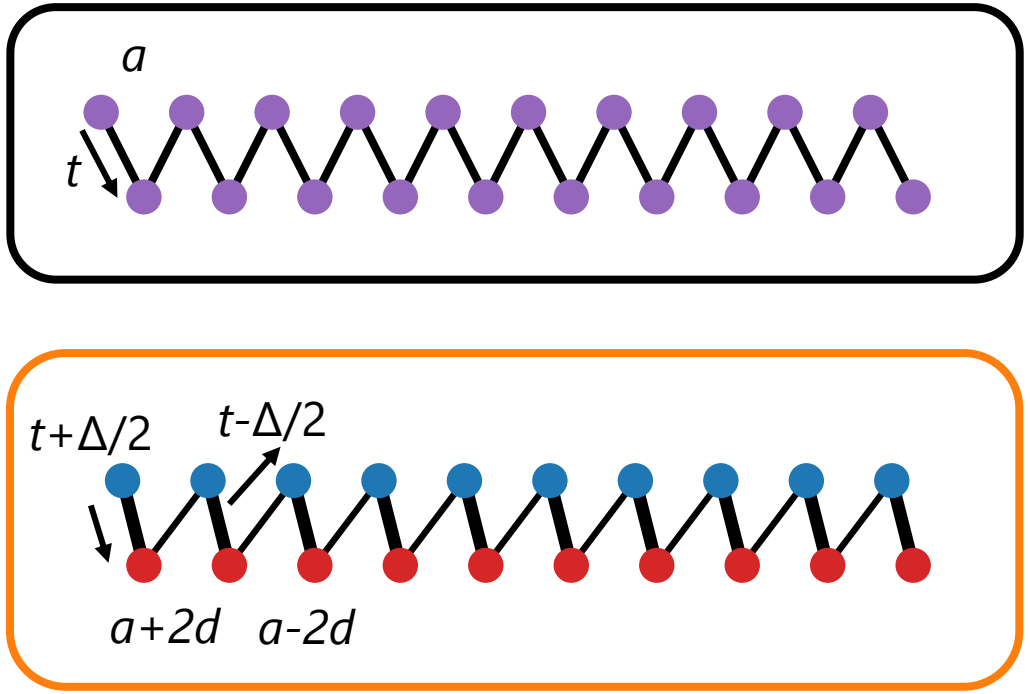

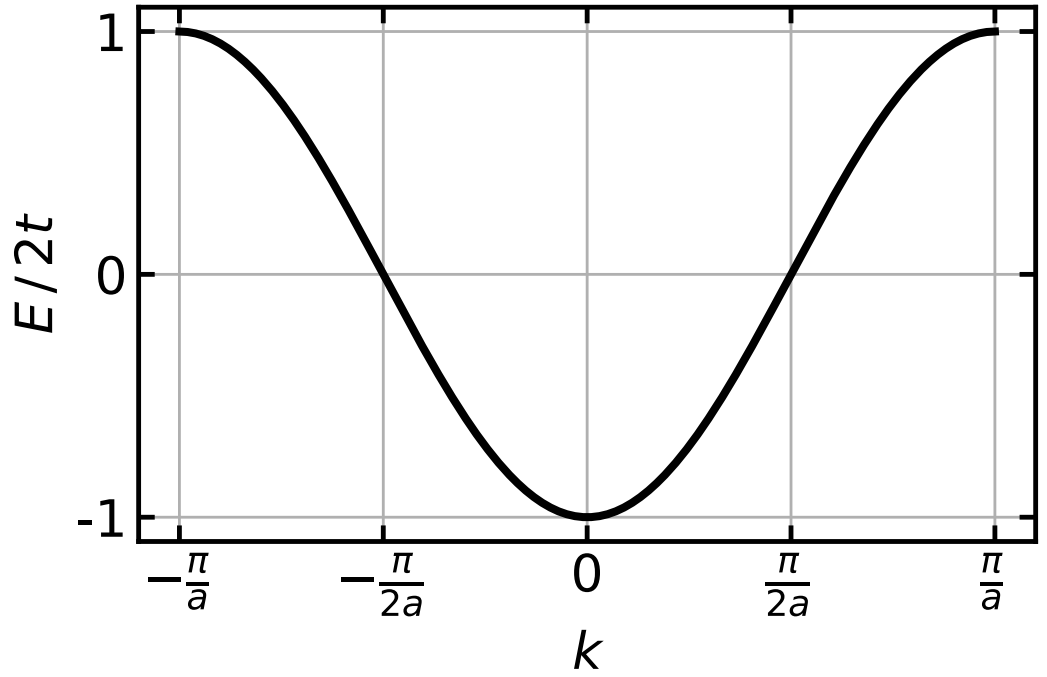

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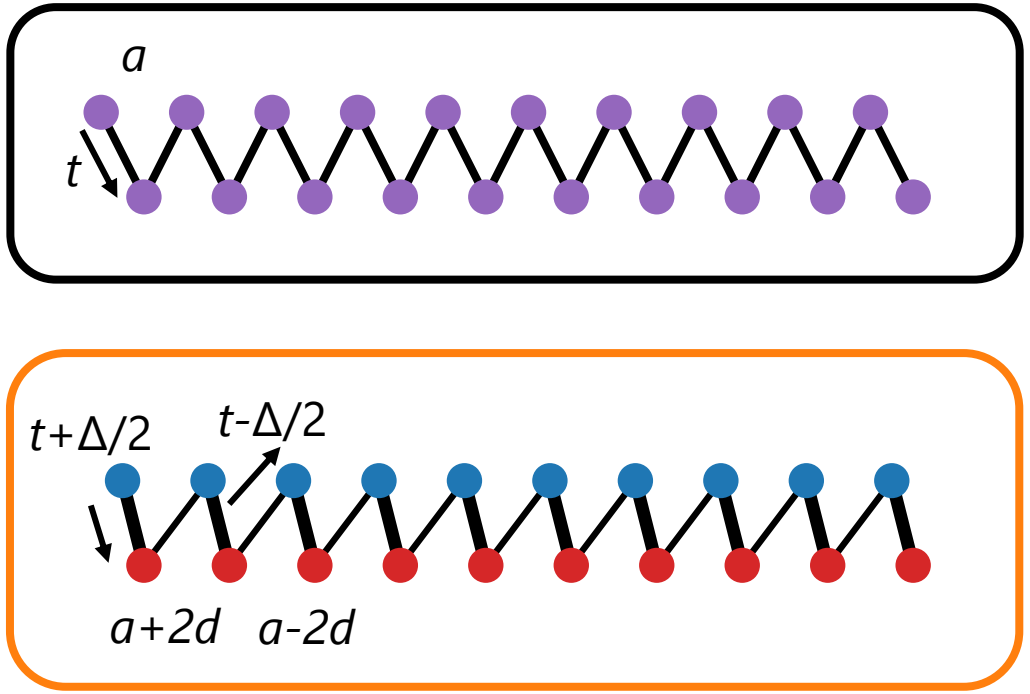

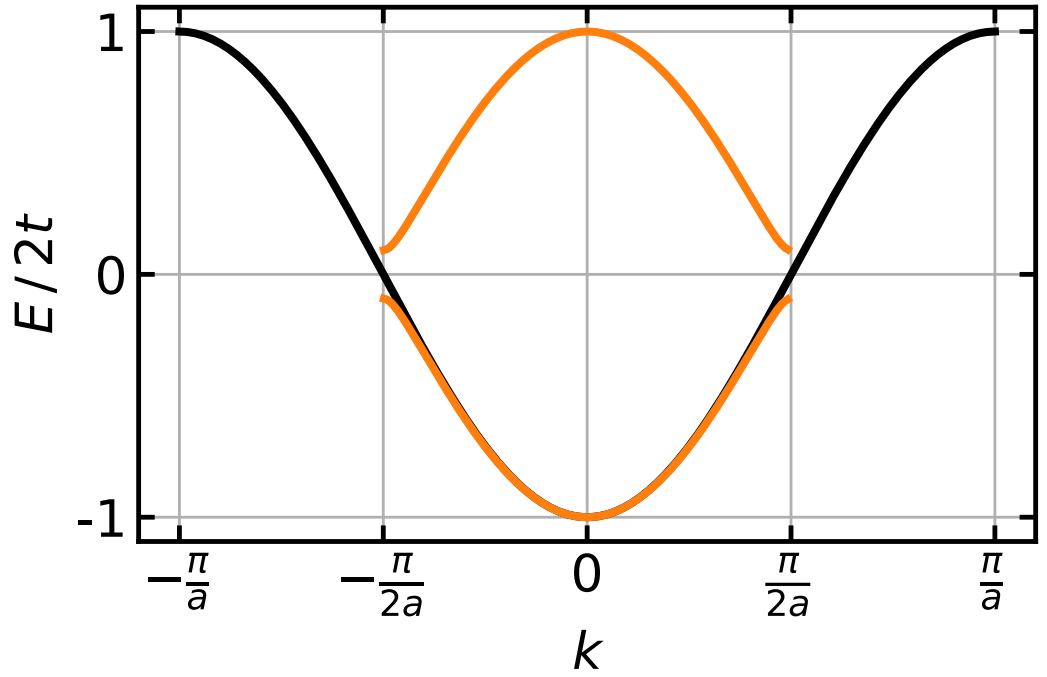

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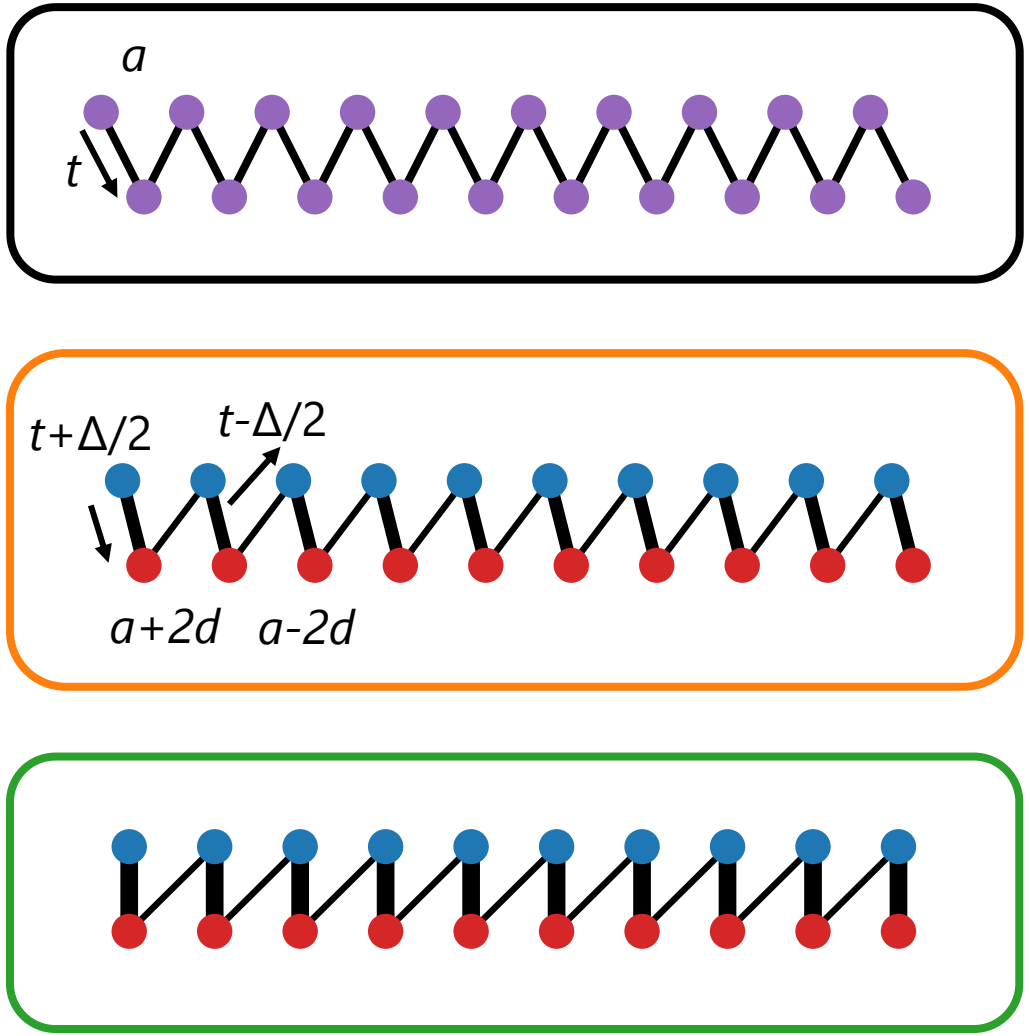

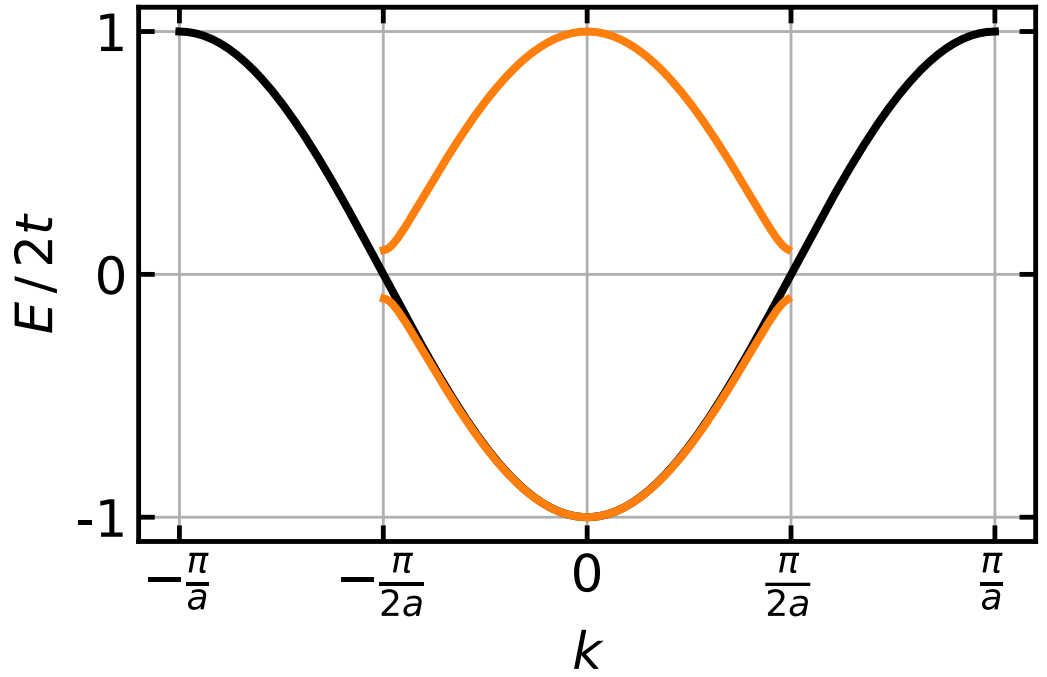

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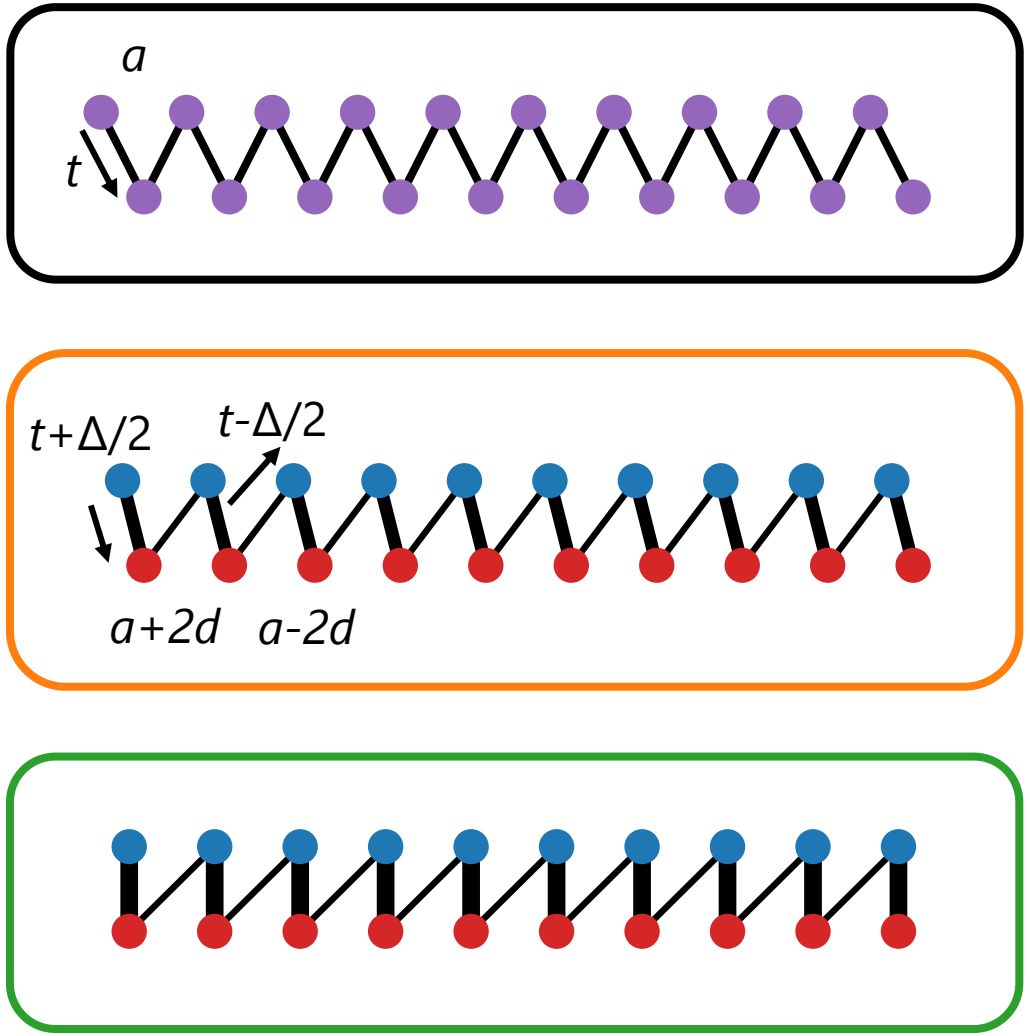
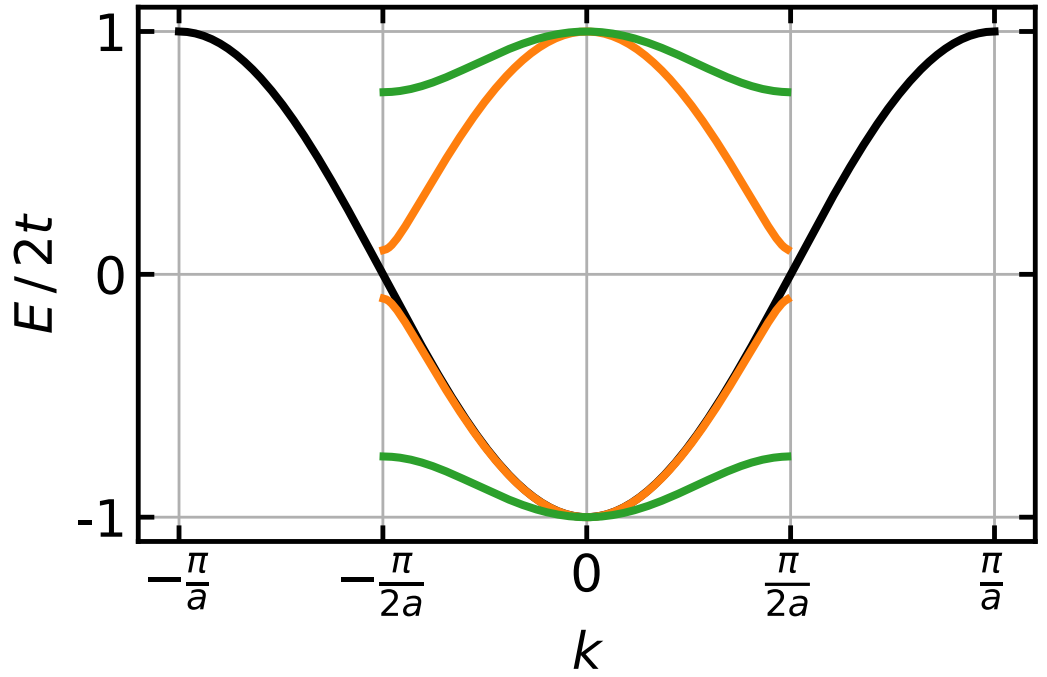
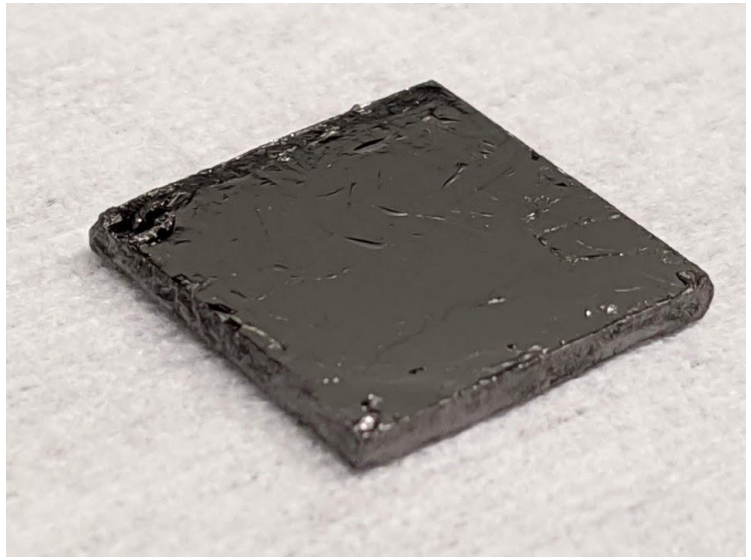
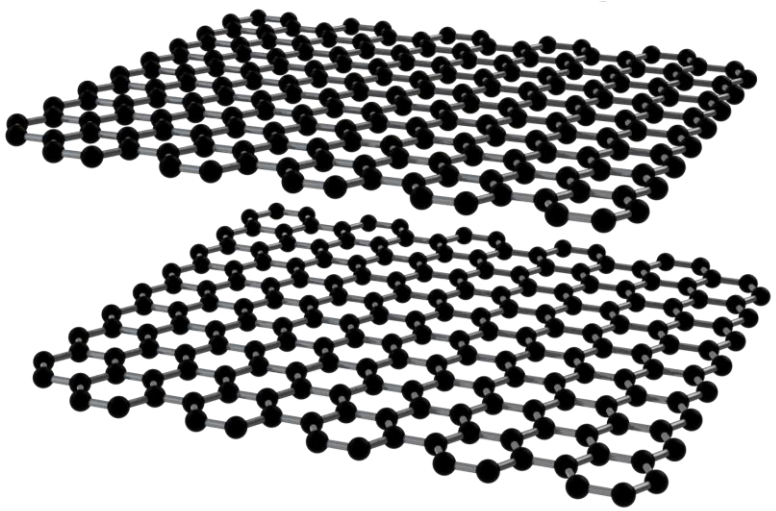


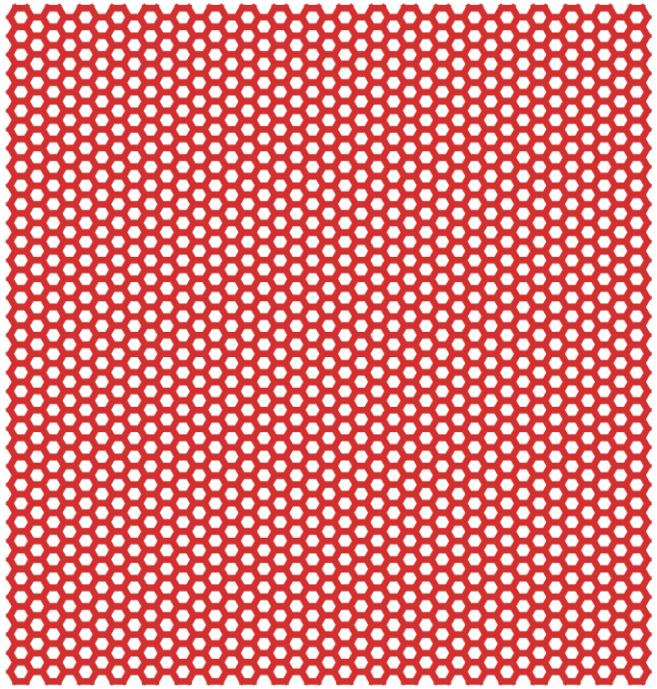


Graphene





Bandstructure engineering with moirés



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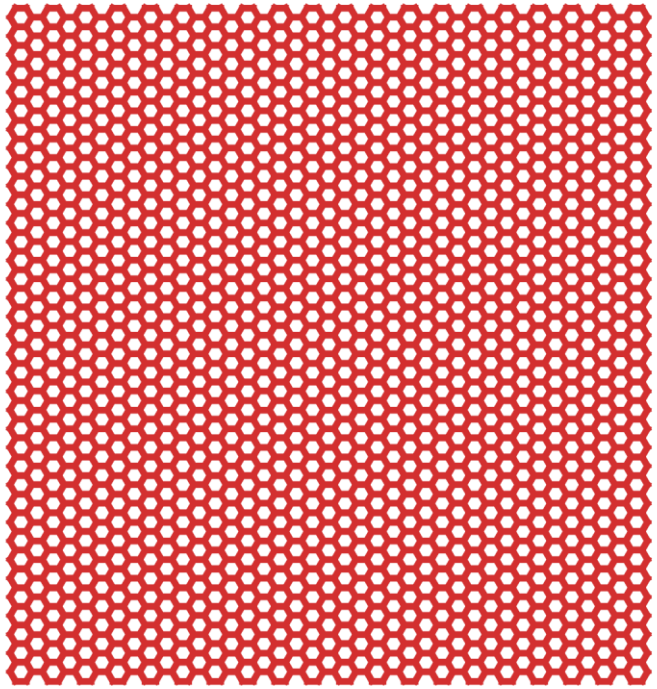

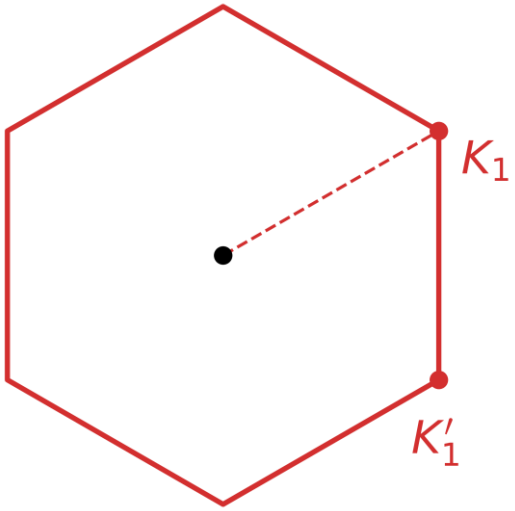

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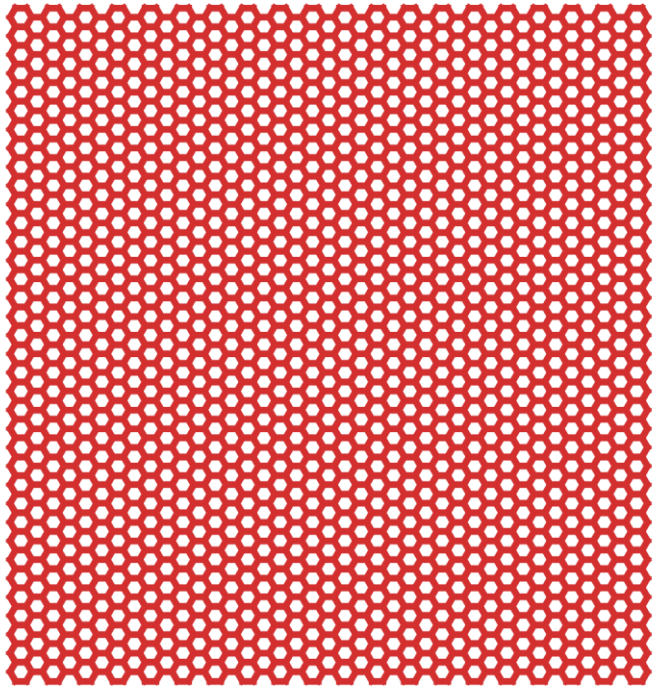
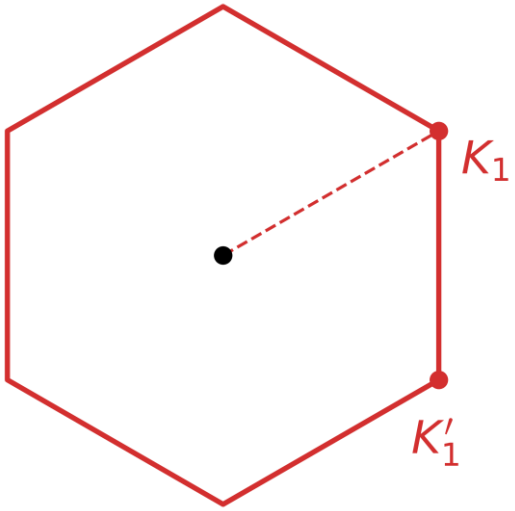
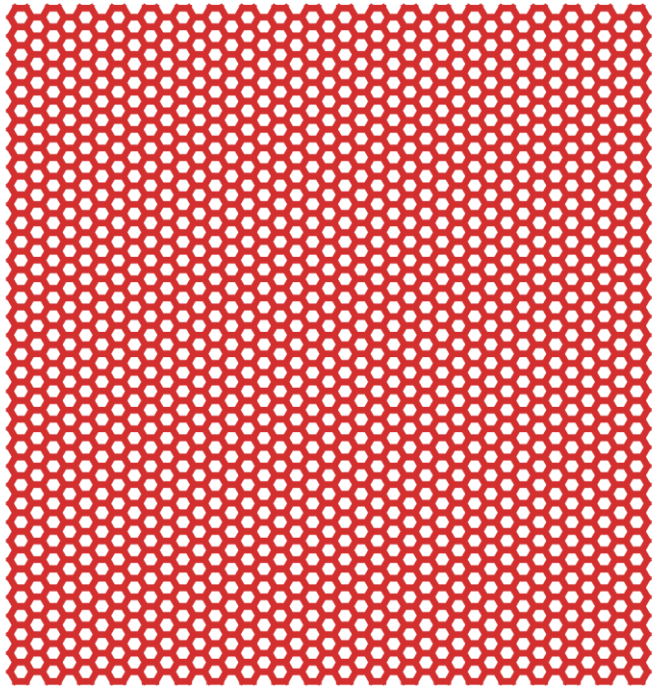
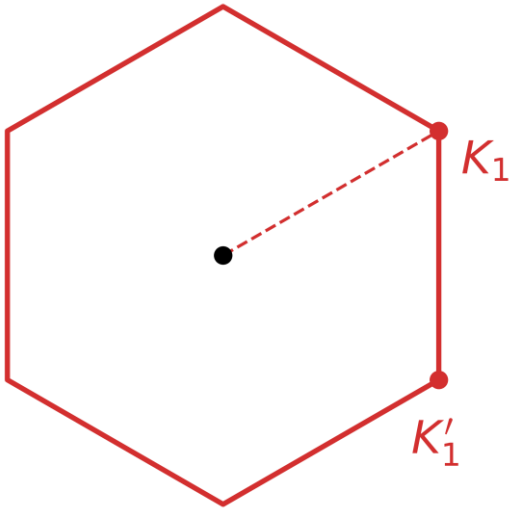


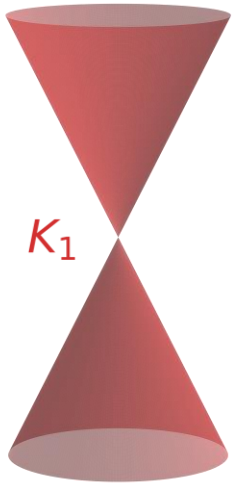


4-fold degenerate
spin x valley

Bandstructure engineering with moirés







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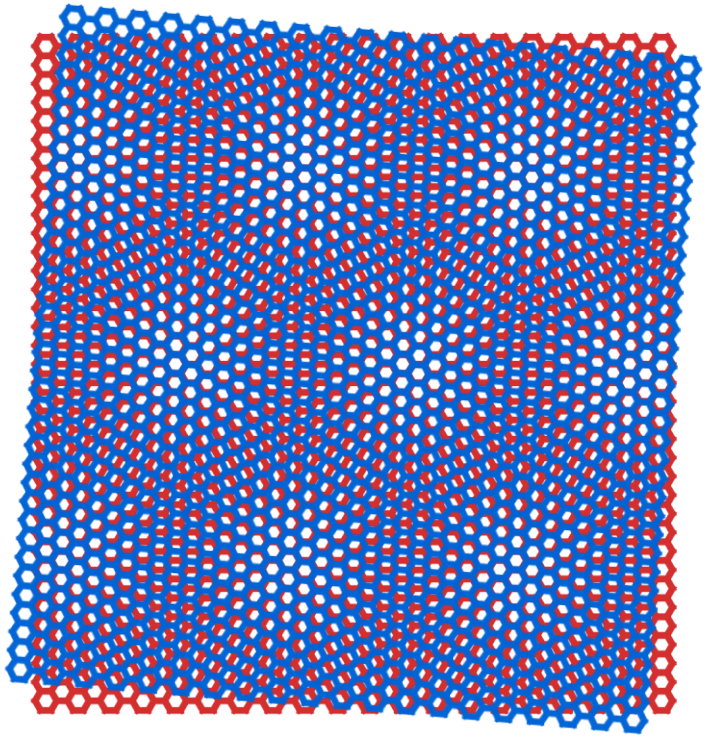

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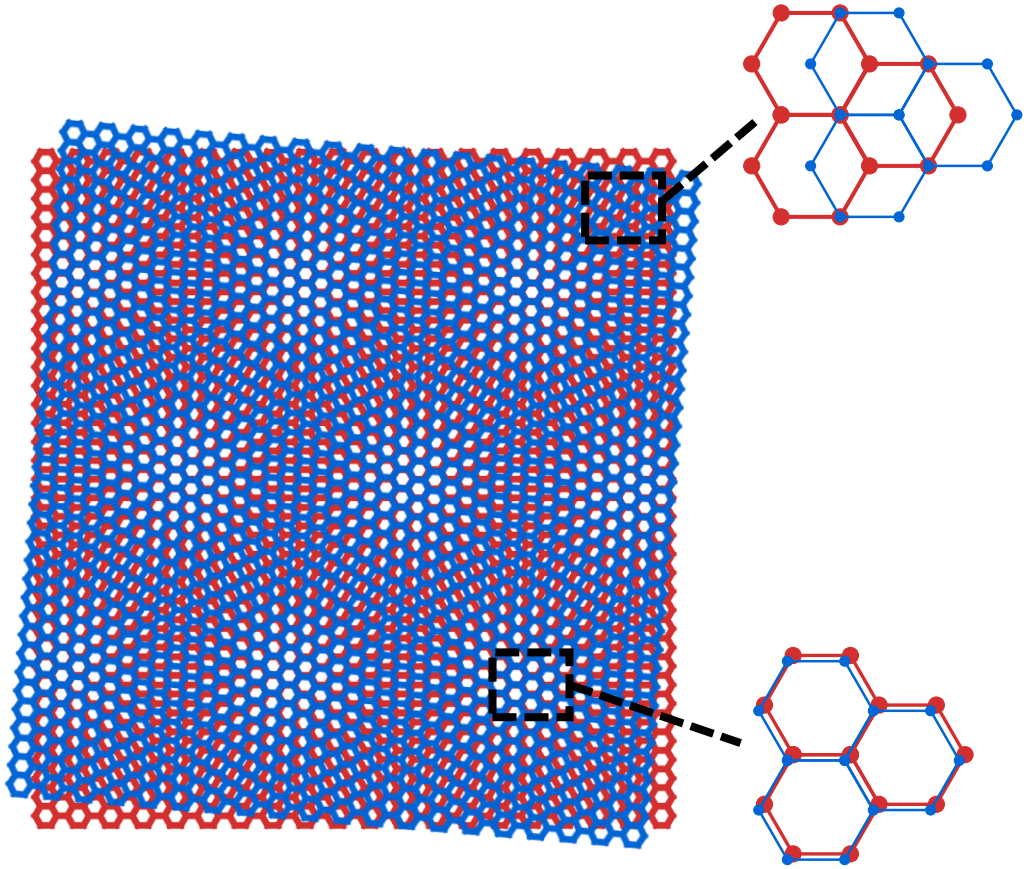
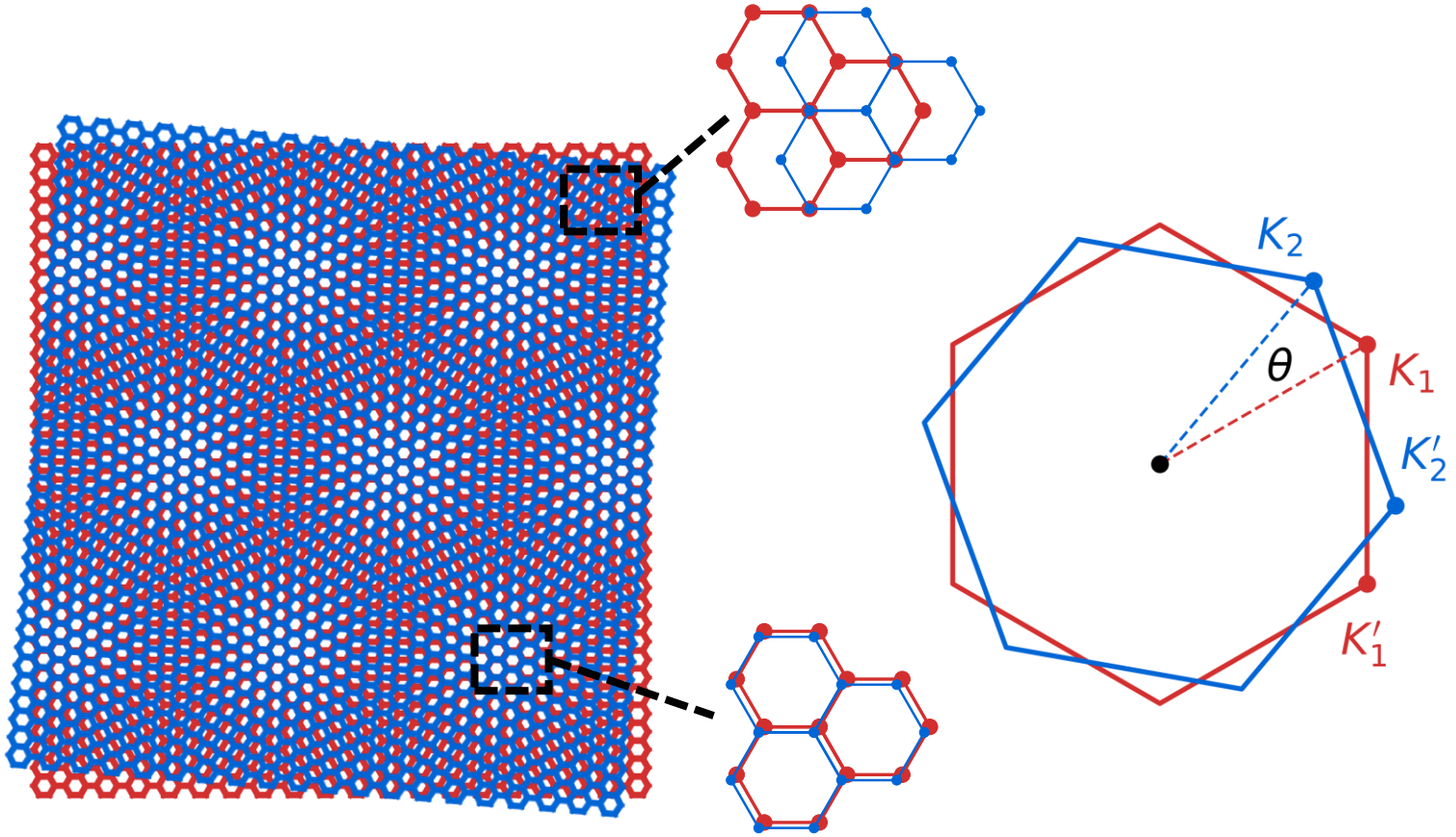


Bandstructure engineering with moirés



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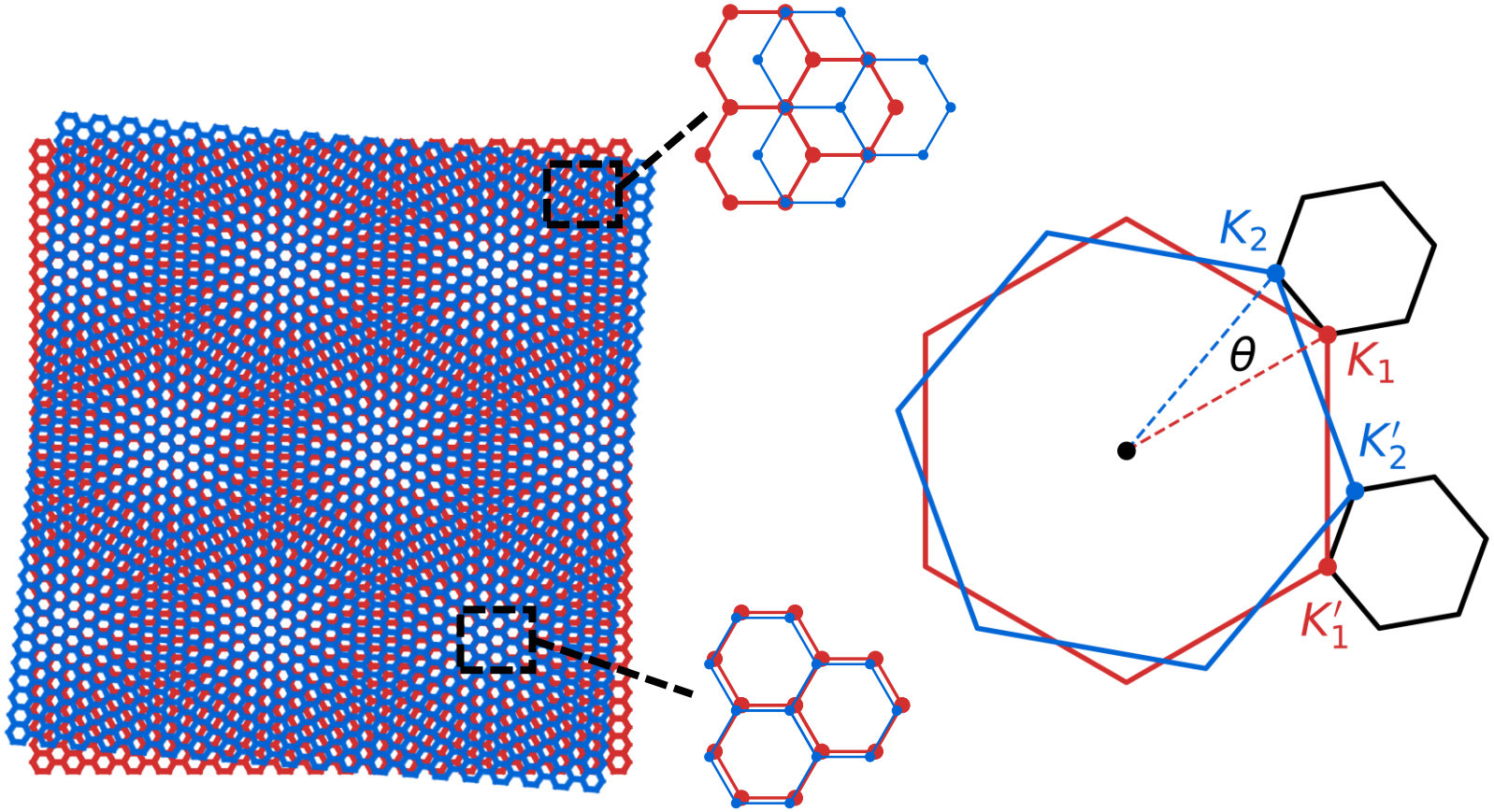

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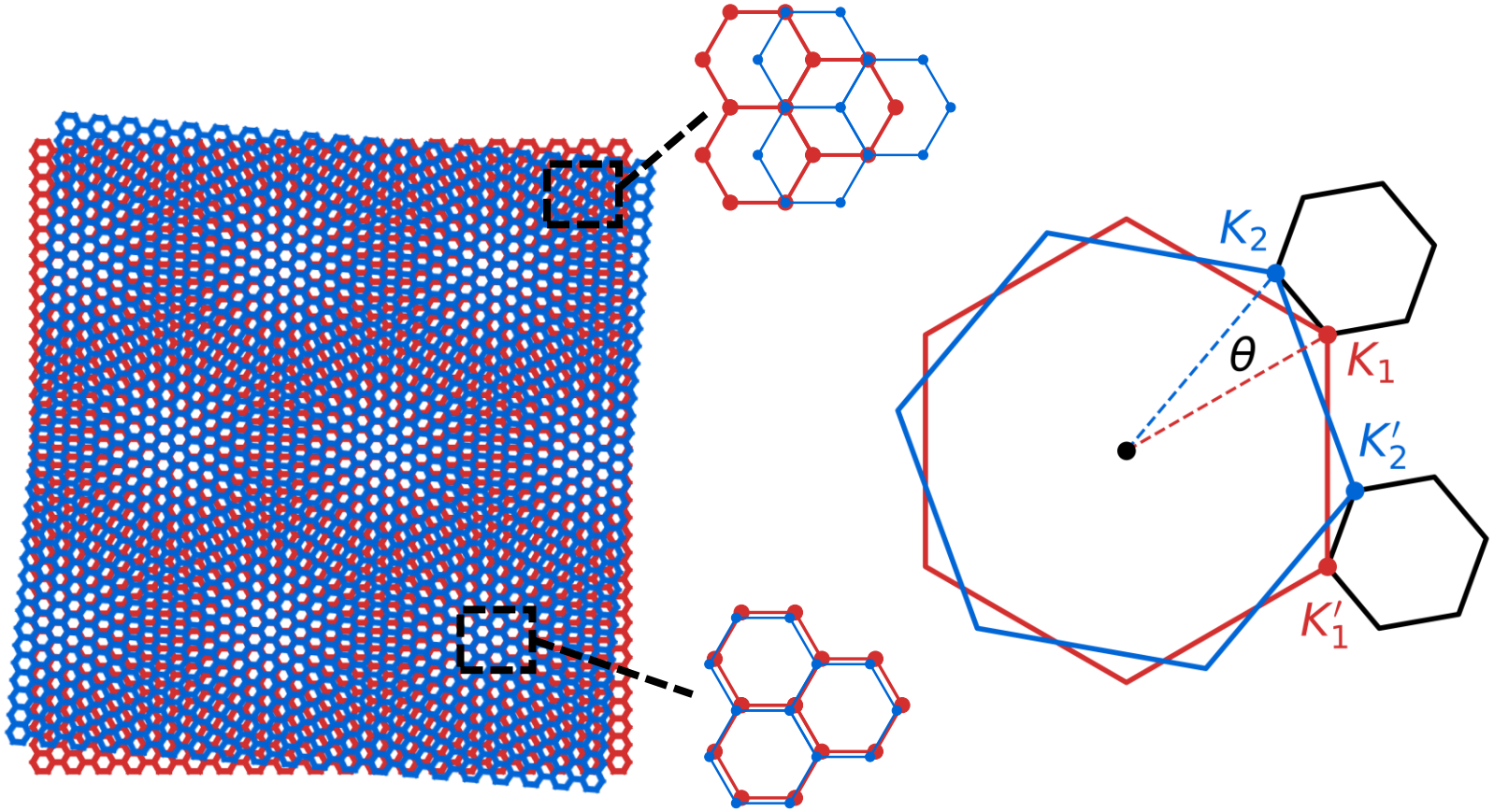

4-fold degenerate
spin x valley

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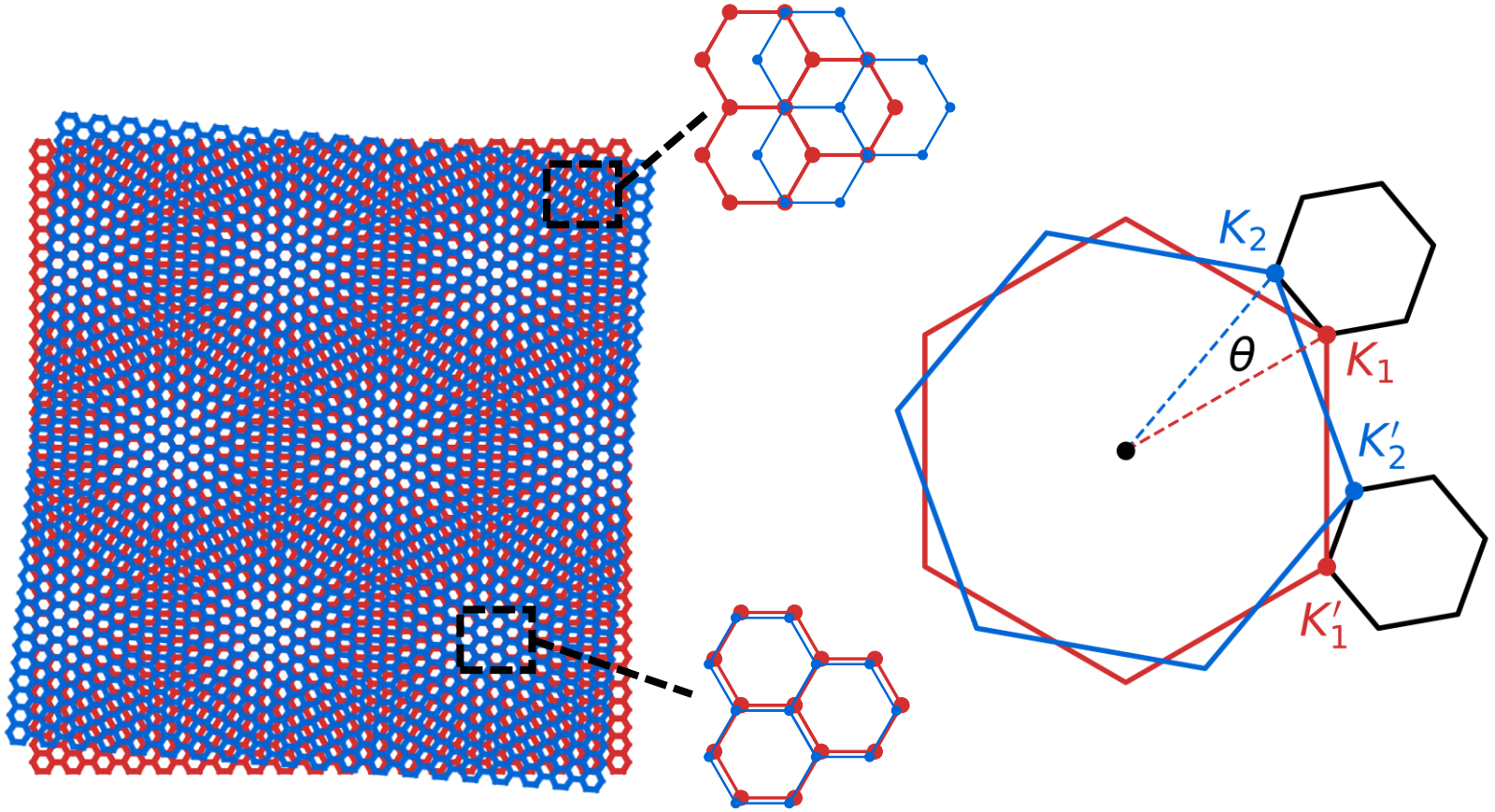
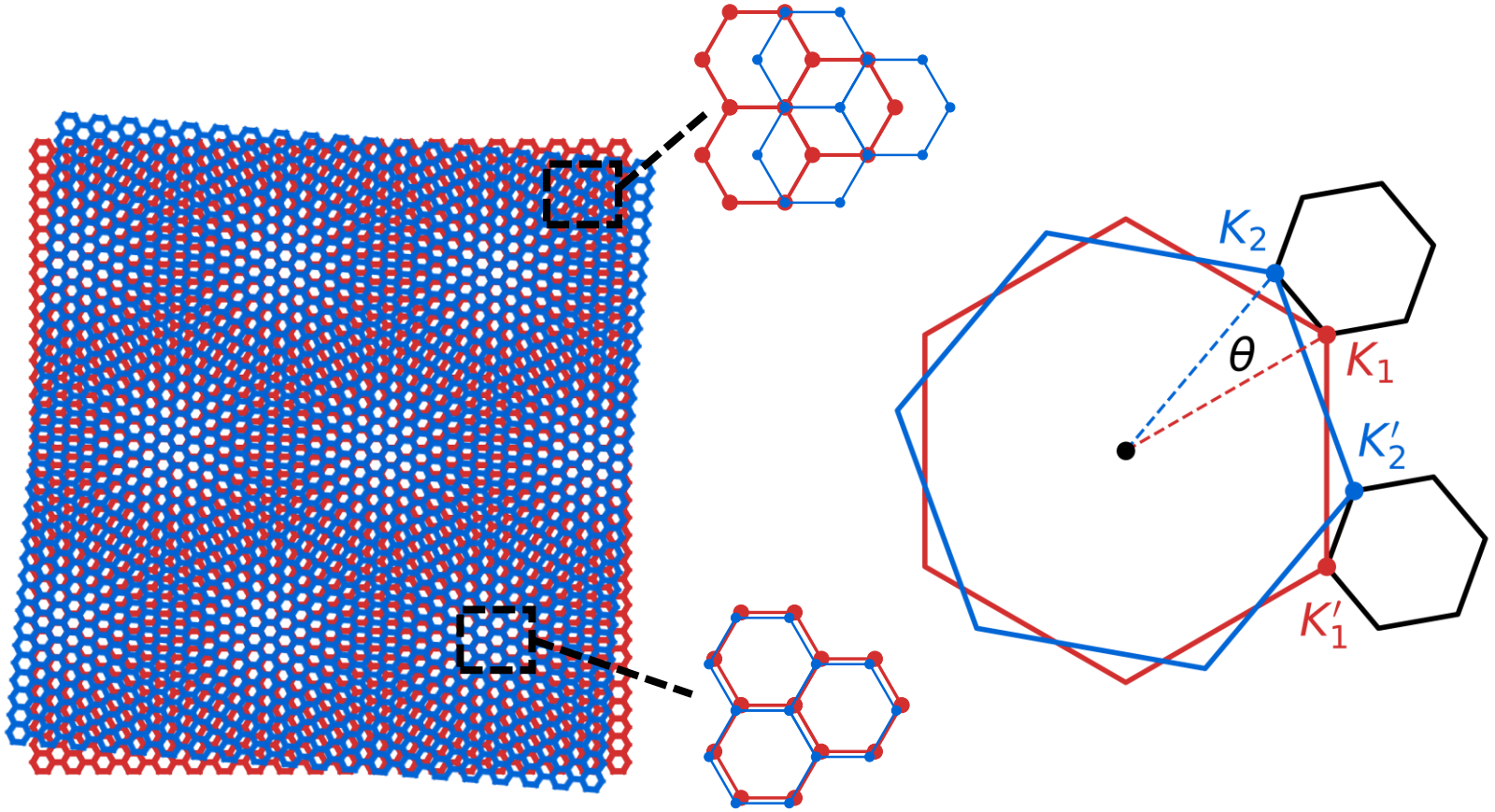
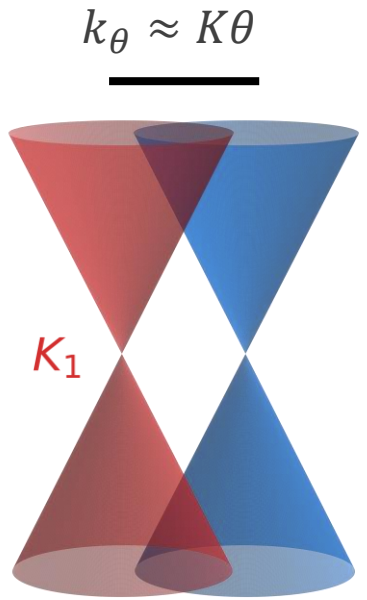


$k_\theta \approx K\theta$

4-fold degenerate
spin x valley

Bandstructure engineering with moirés





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spin x valley

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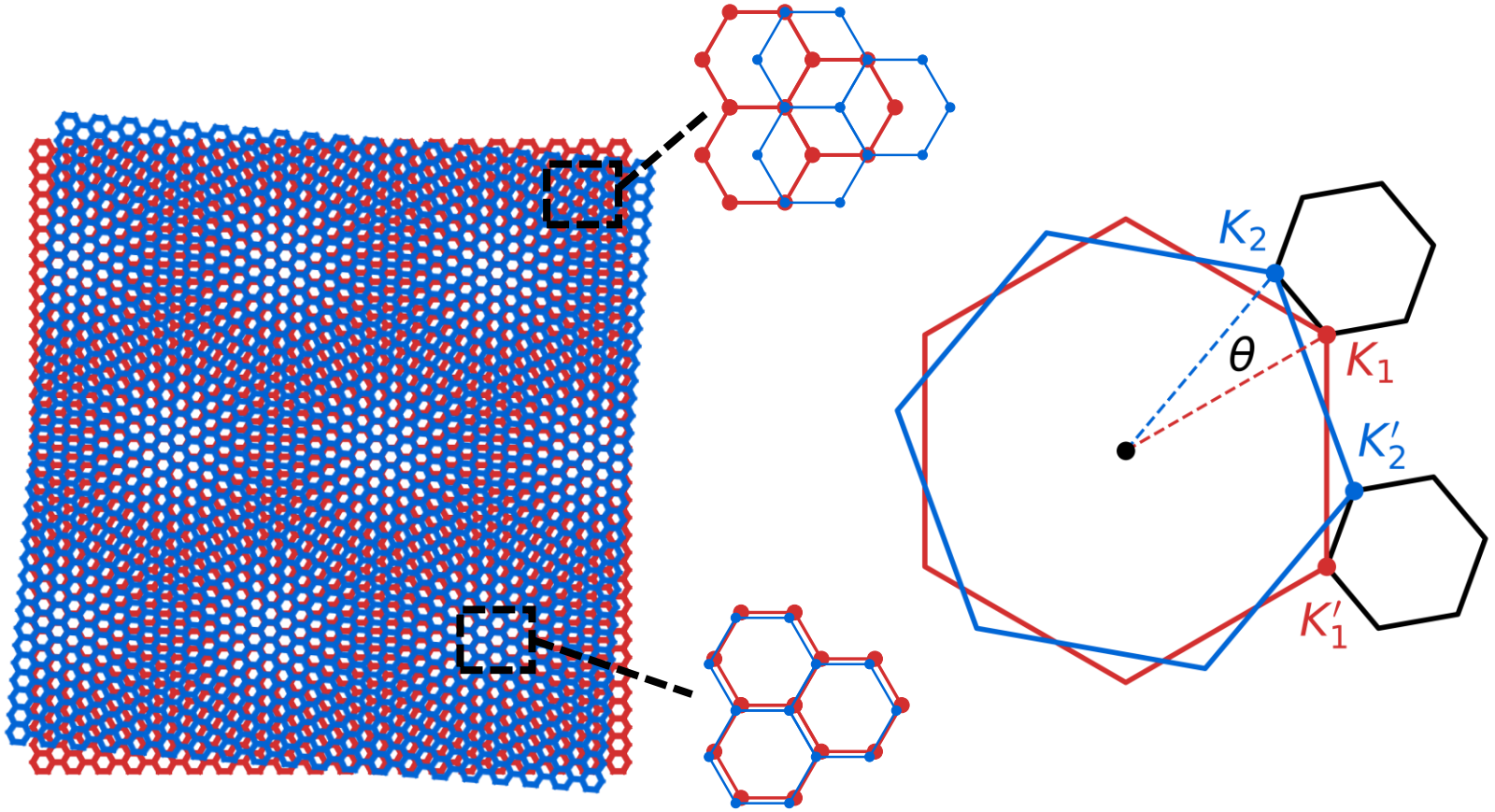

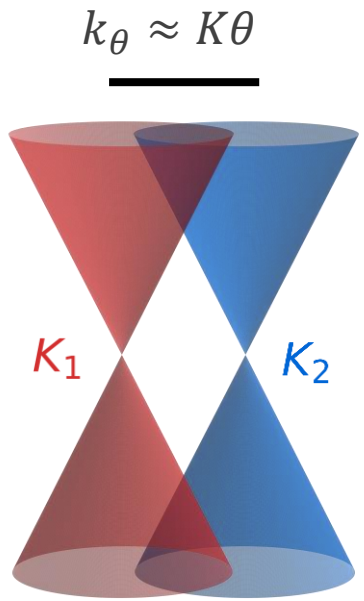

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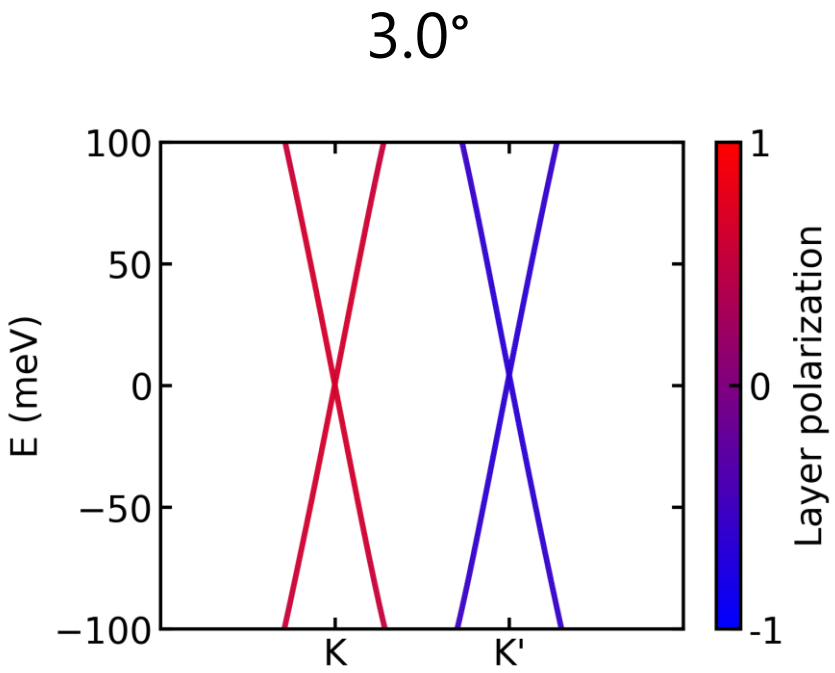
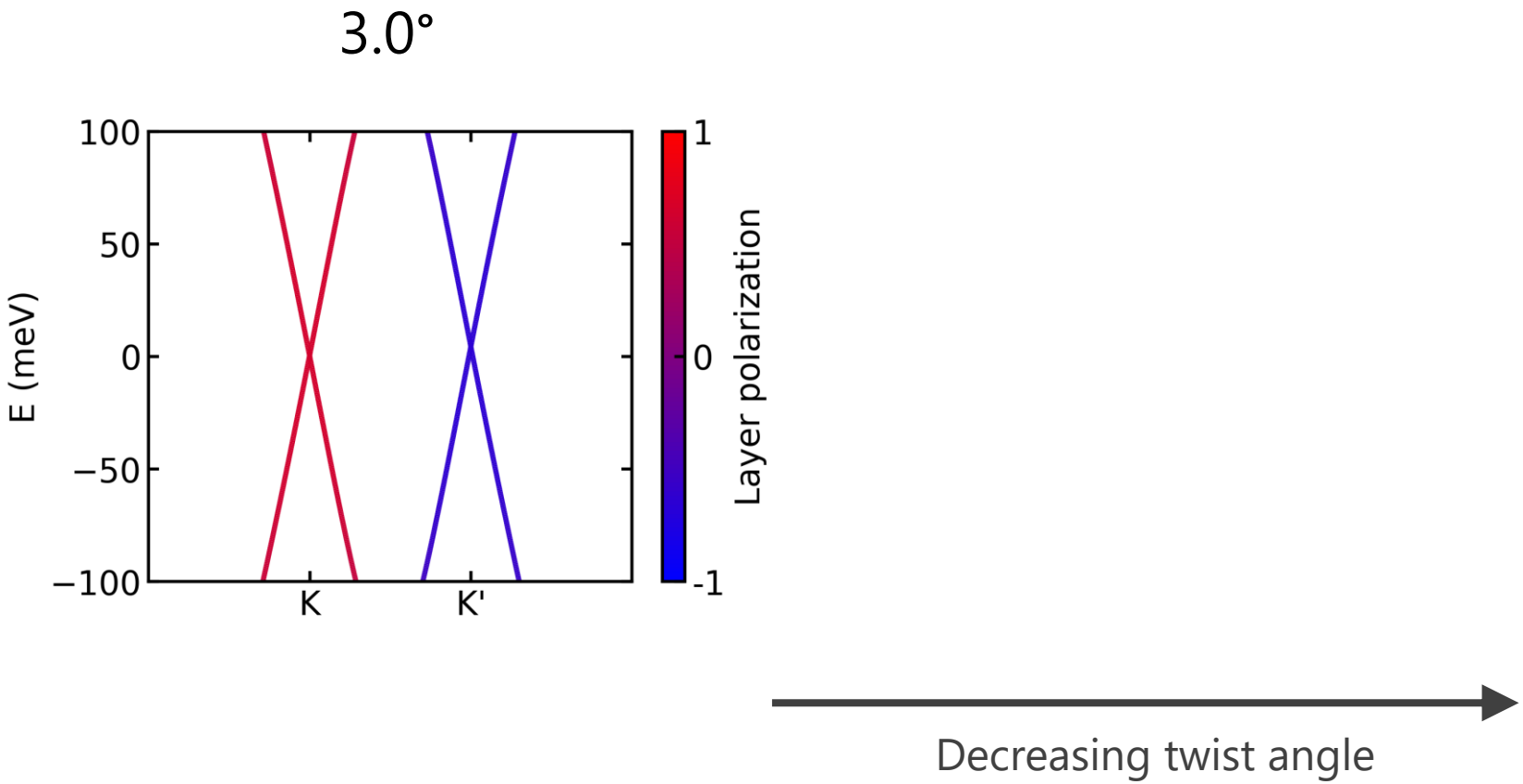
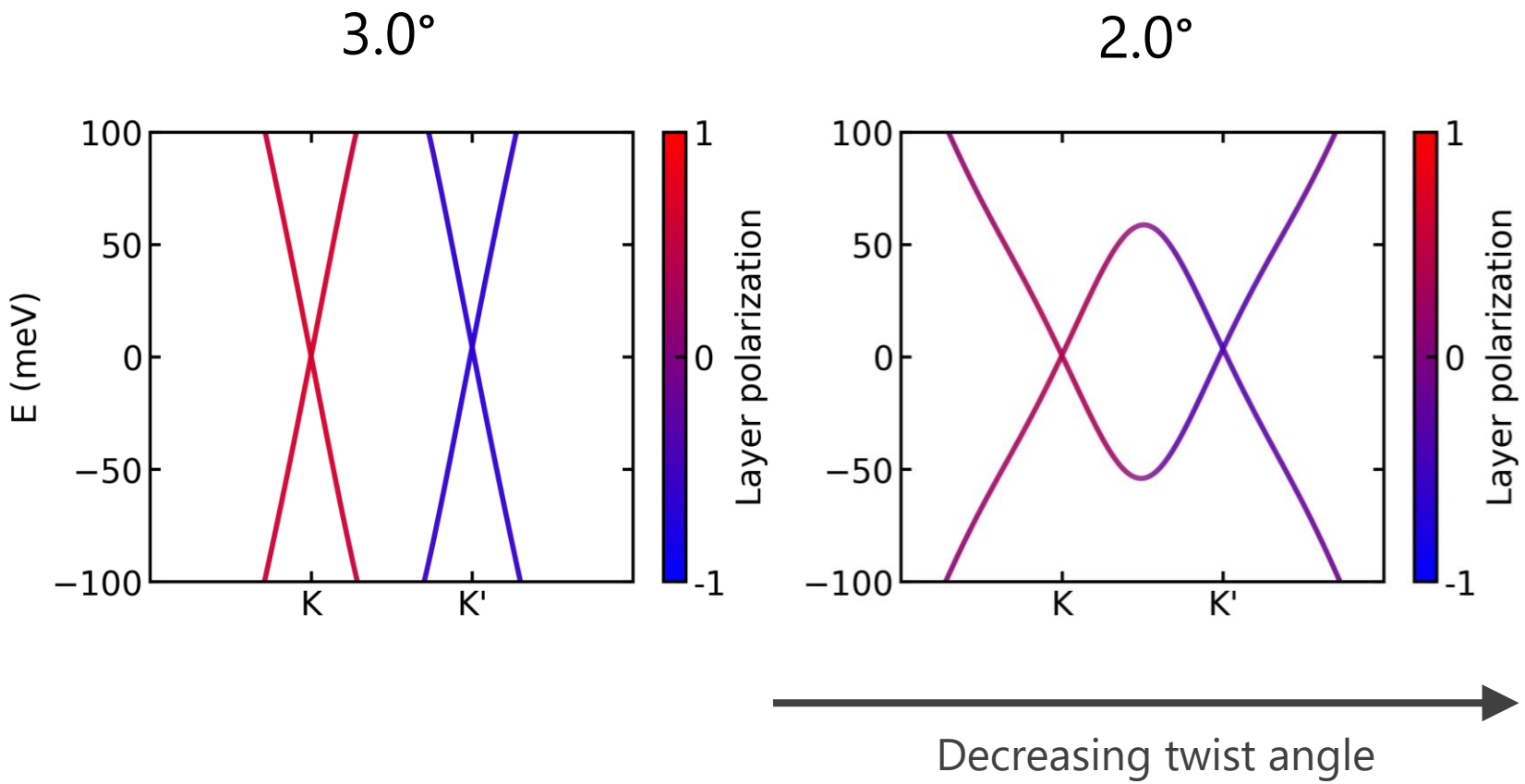


git.sr.ht/~spxtr/bm_model

Bandstructure engineering with moirés



Bandstructure engineering with moirés



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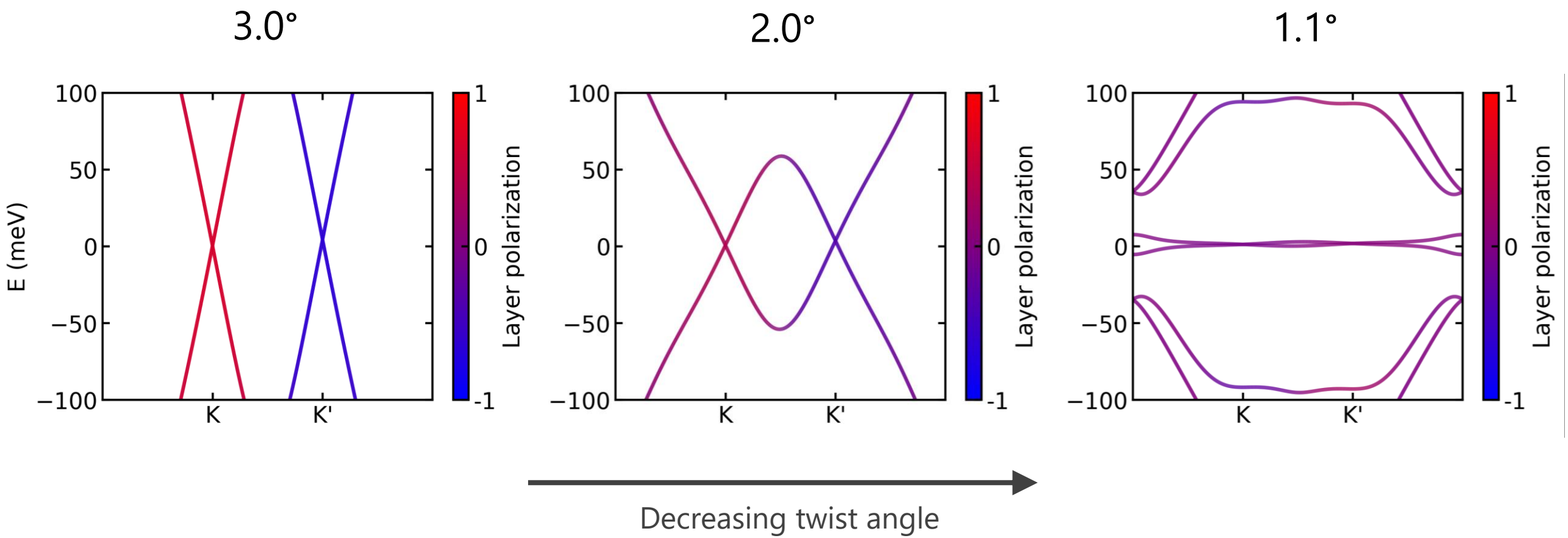
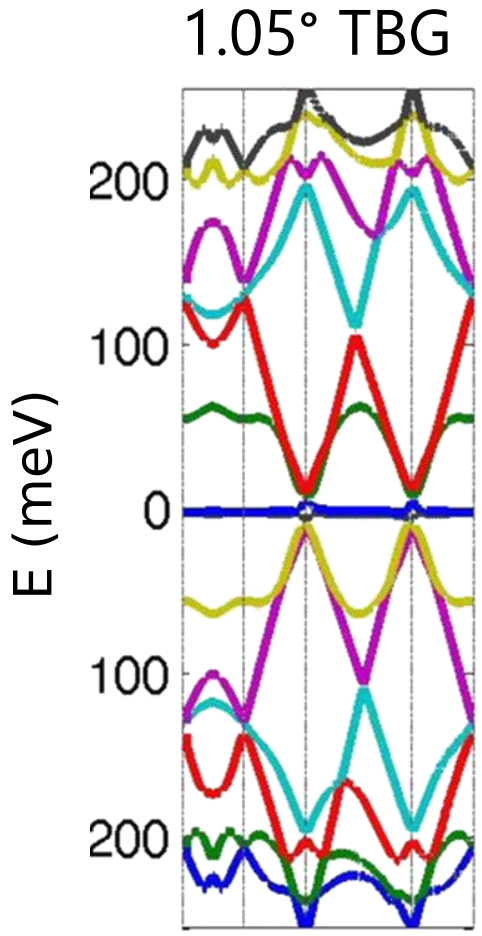


First experimental realization of TBG



Bistritzer and MacDonald, PNAS (2011)
Cao, et al. Nature (2018)

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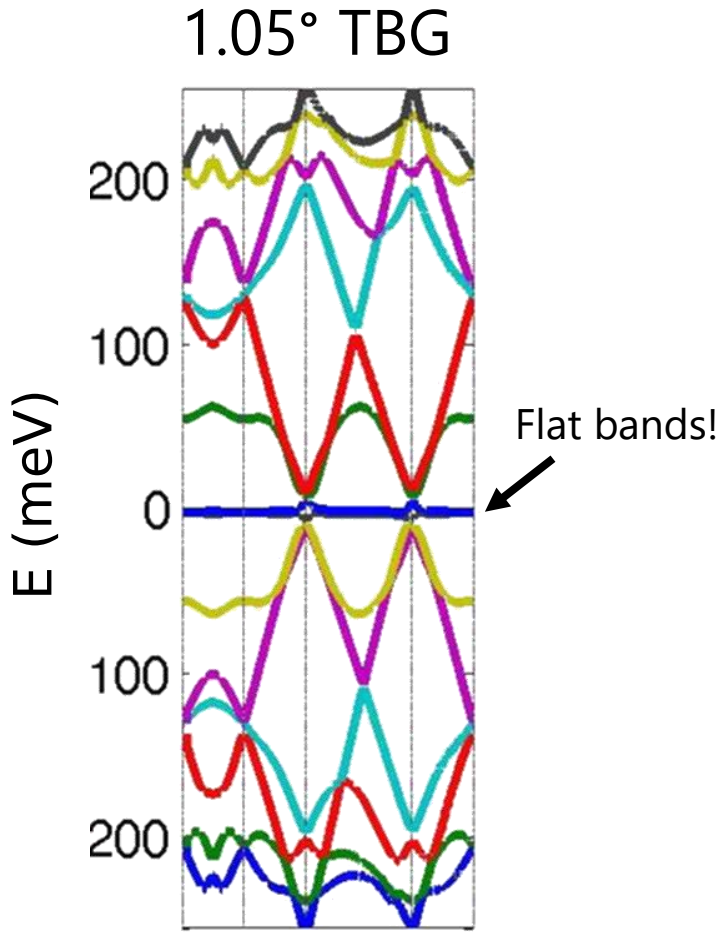

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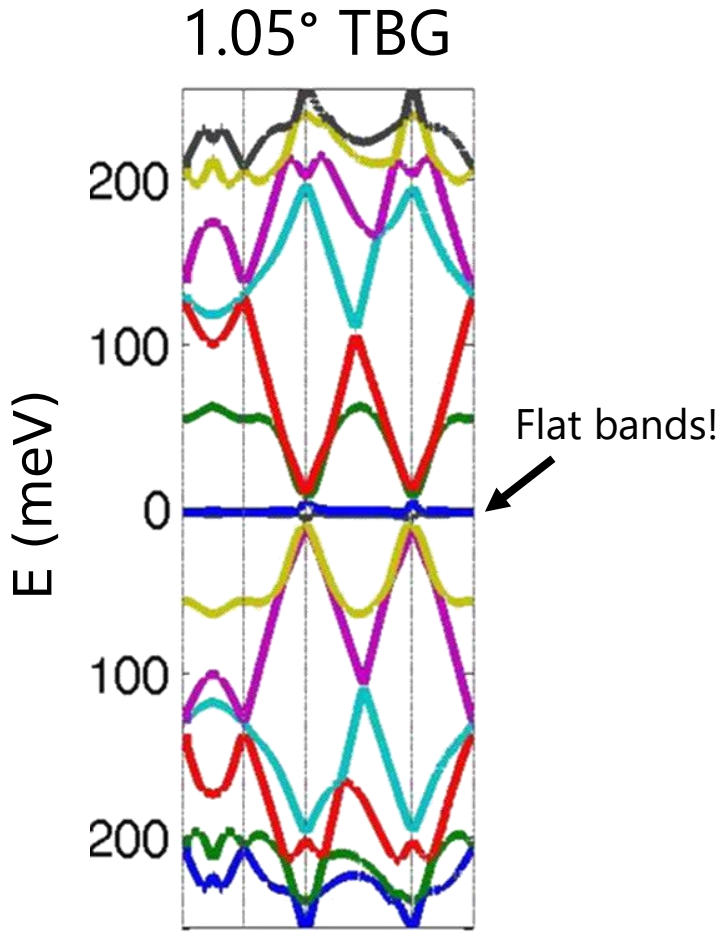

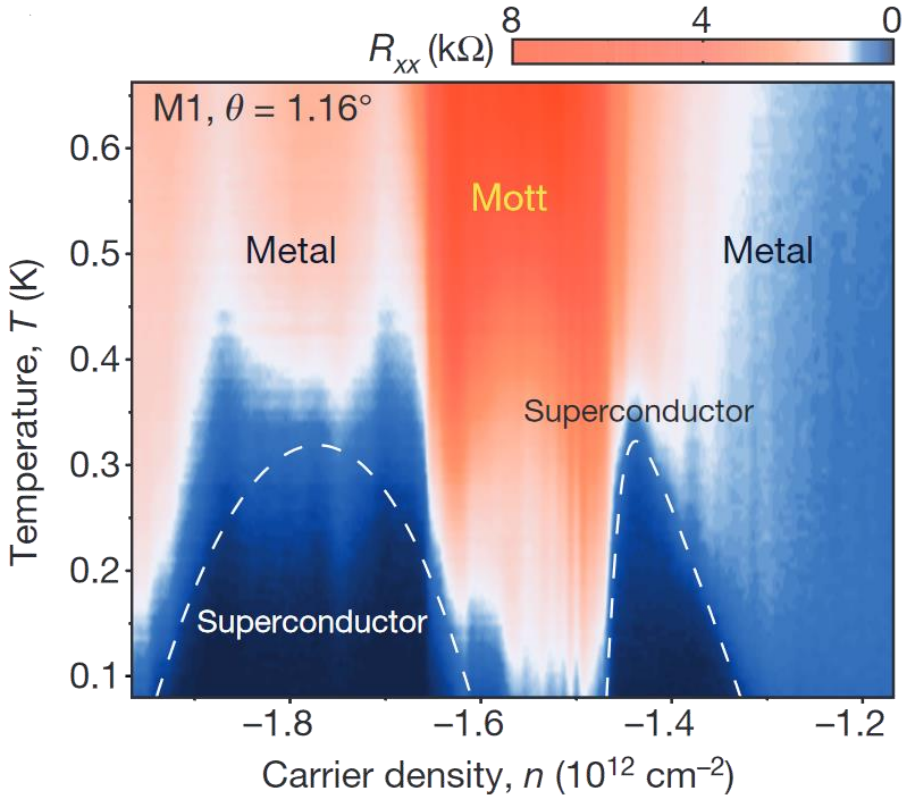

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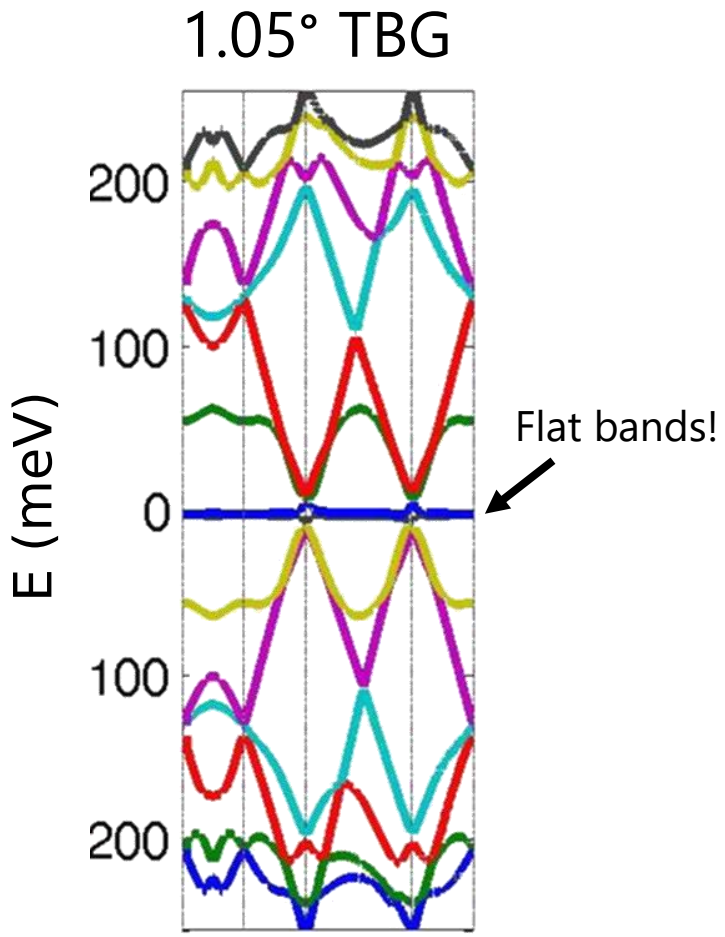
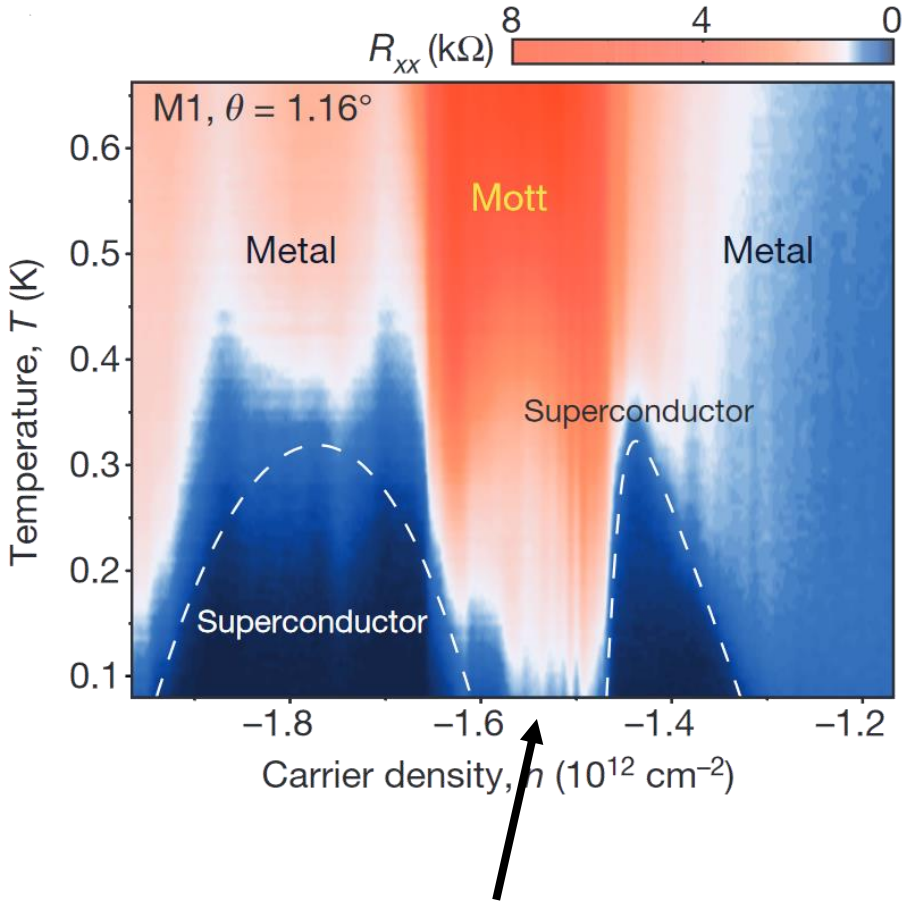




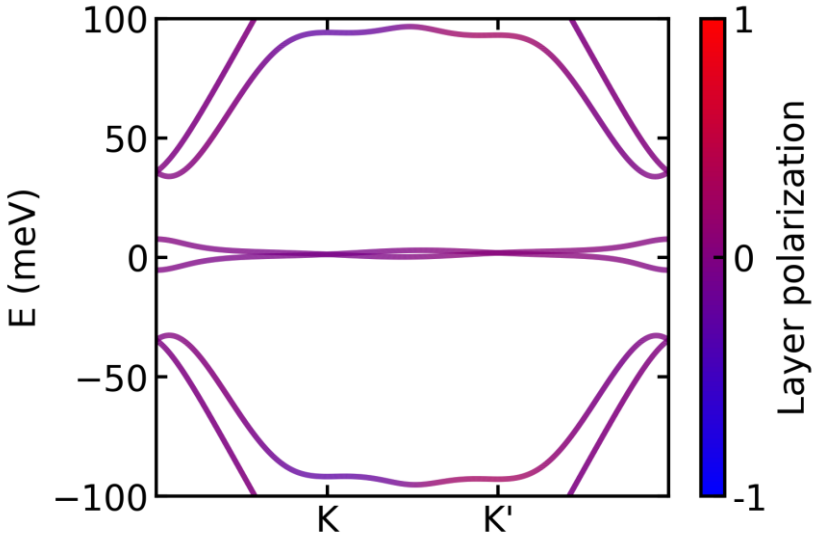
Half filled band – symmetry broken phase
Think Hund's rules

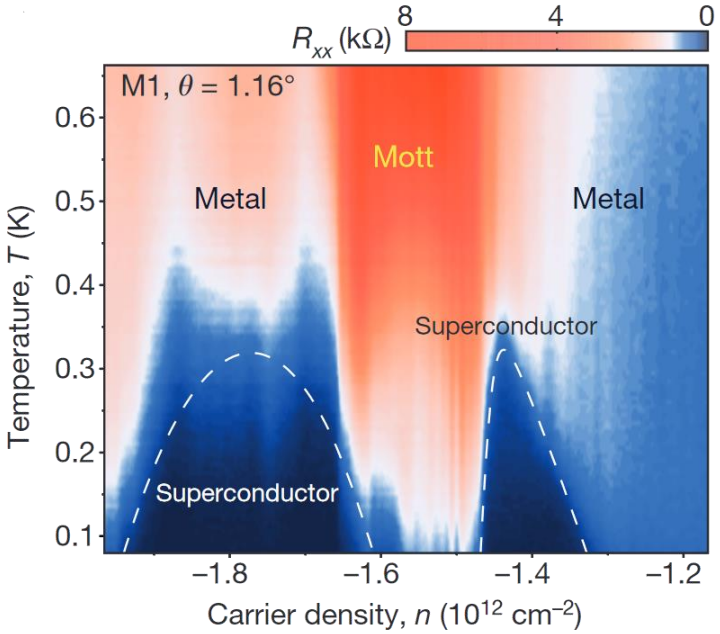
Bistritzer and MacDonald, PNAS (2011)
Cao, et al. Nature (2018)

Summary

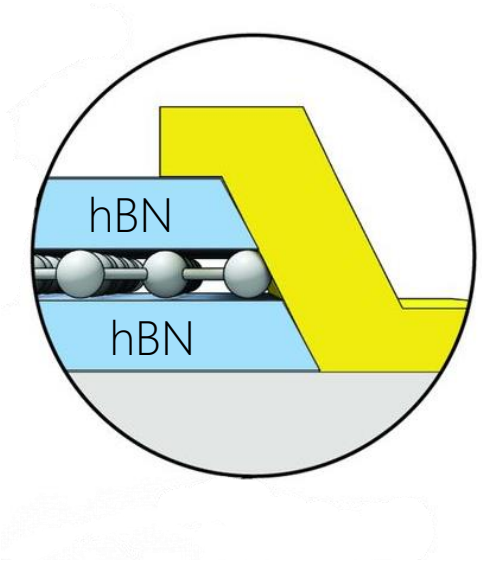
By controlling twist angle, graphene multilayers can become strongly correlated!

Let's make some samples!





Typical twisted bilayer graphene (TBG)



Wang, et al. Science (2013)
Sharpe, et al. Science (2019)

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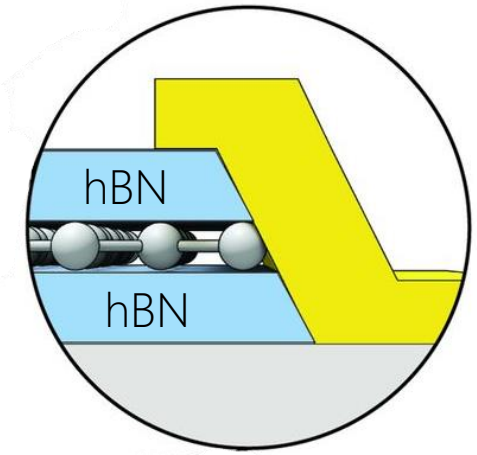

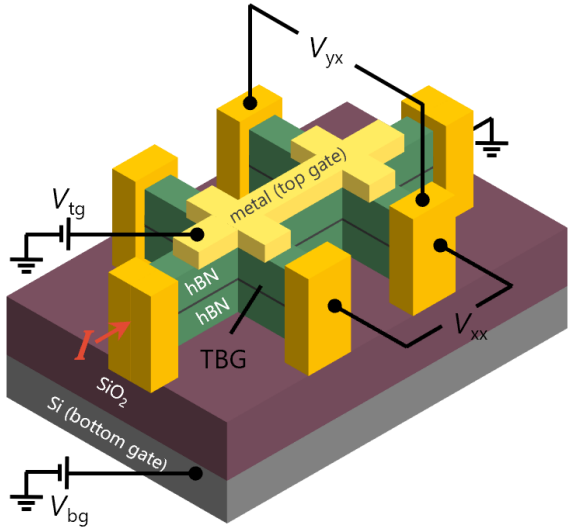

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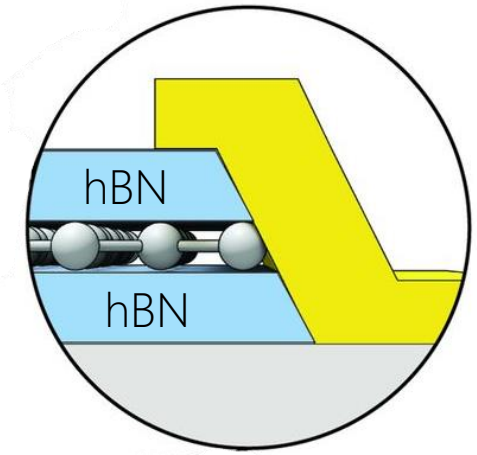

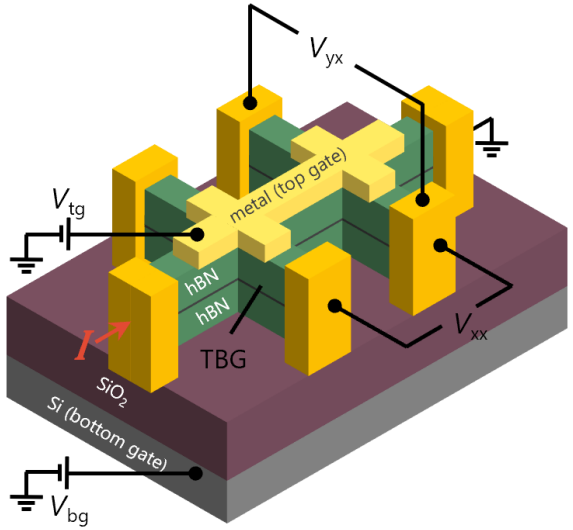

$$n \propto V_{tg}/d_{tg} + V_{bg}/d_{bg}$$
$$D \propto V_{tg}/d_{tg} - V_{bg}/d_{bg}$$

Wang, et al. Science (2013)
Sharpe, et al. Science (2019)

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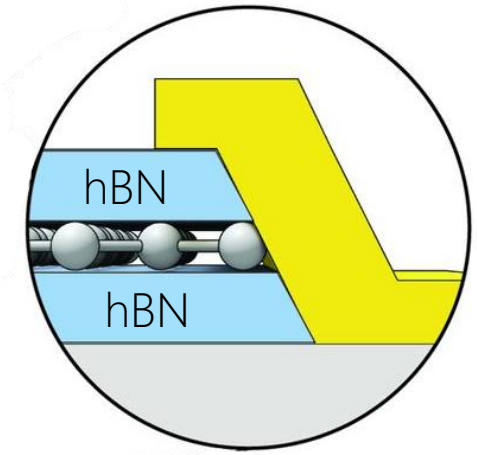

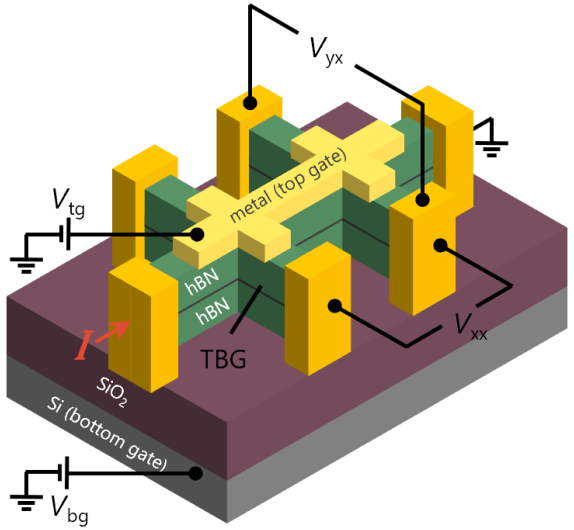

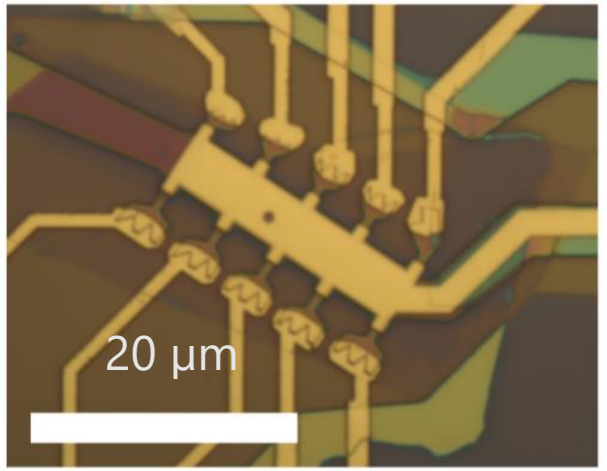

$$n \propto V_{tg}/d_{tg} + V_{bg}/d_{bg}$$
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Wang, et al. Science (2013)
Sharpe, et al. Science (2019)

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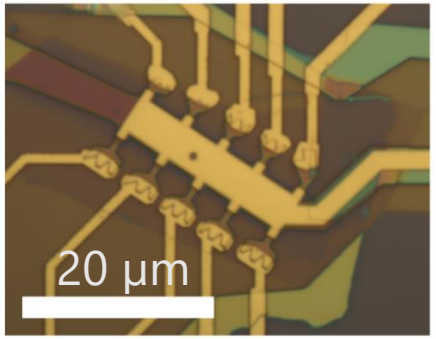

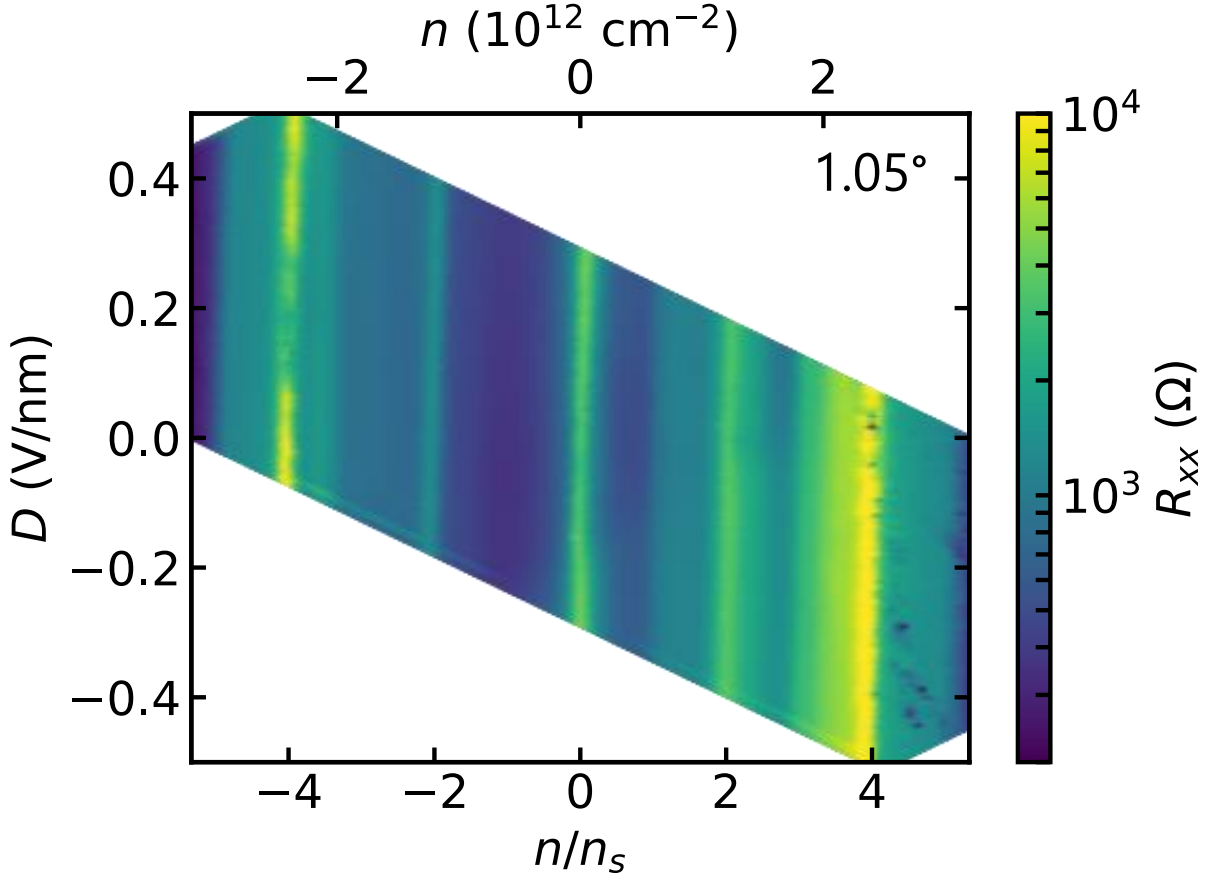

Sharpe, et al. Science (2019)

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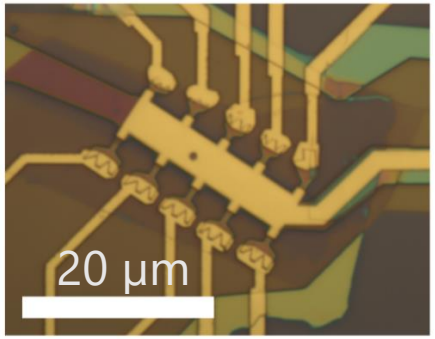

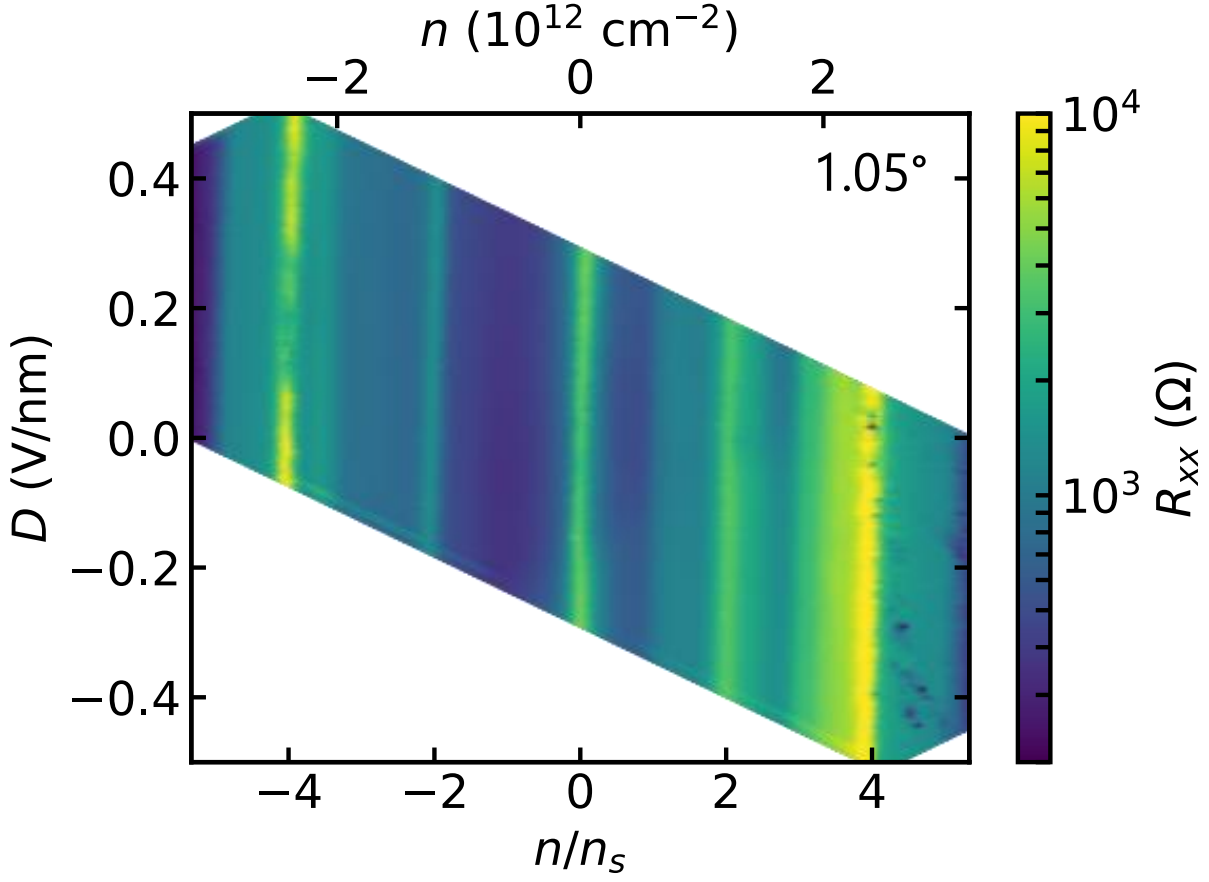

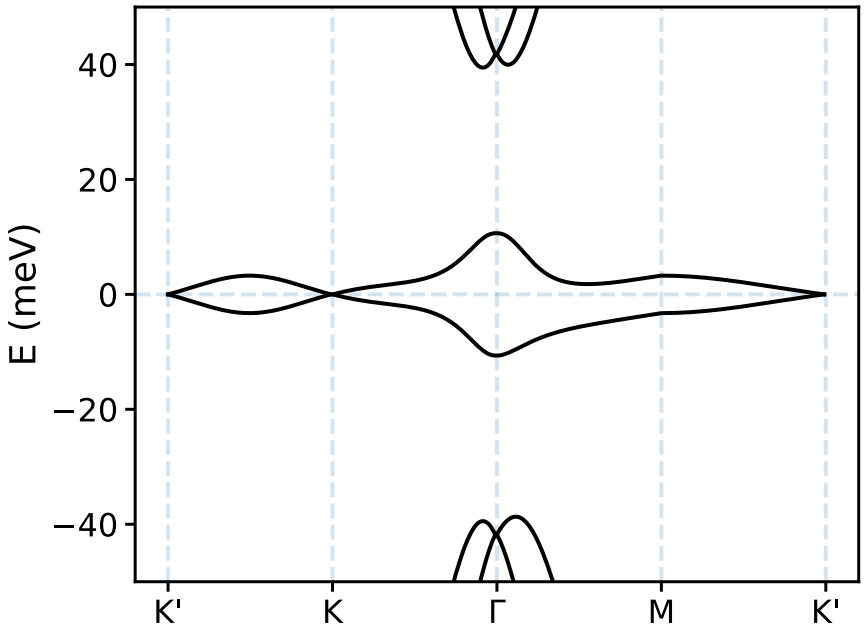

Sharpe, et al. Science (2019)

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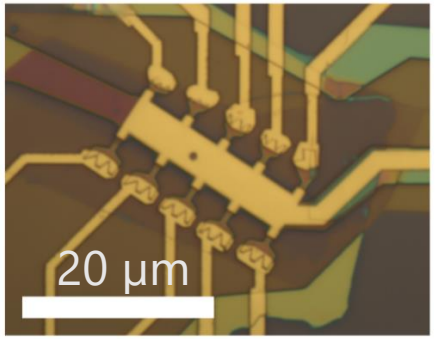

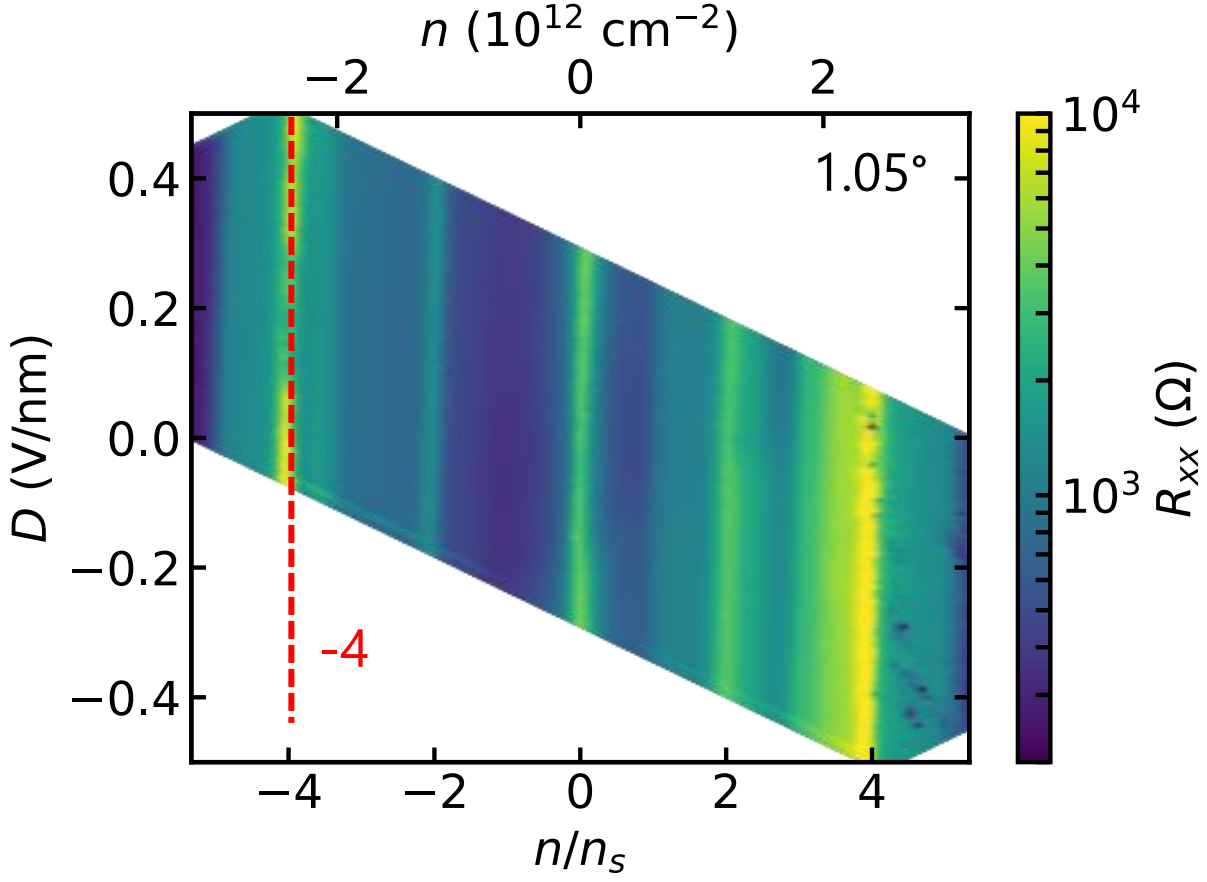

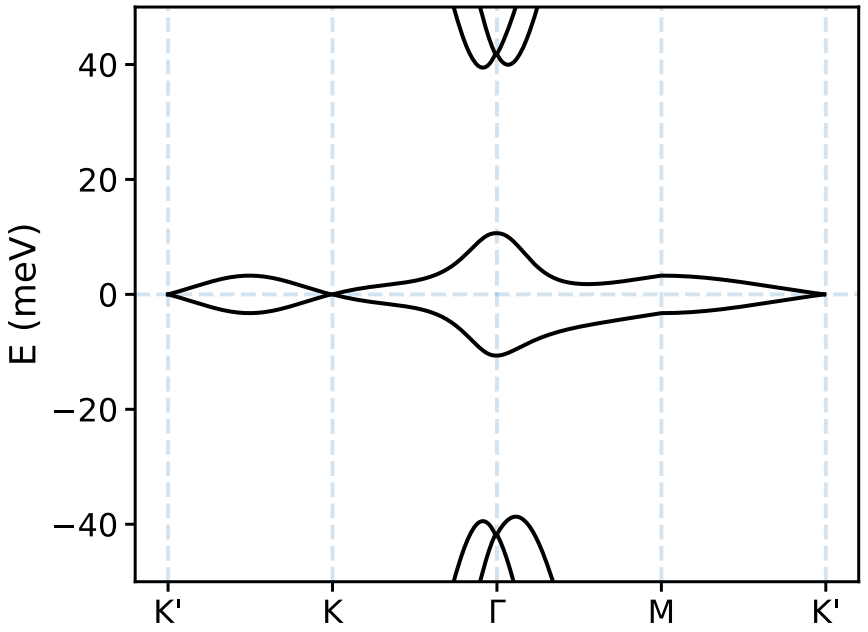

Sharpe, et al. Science (2019)

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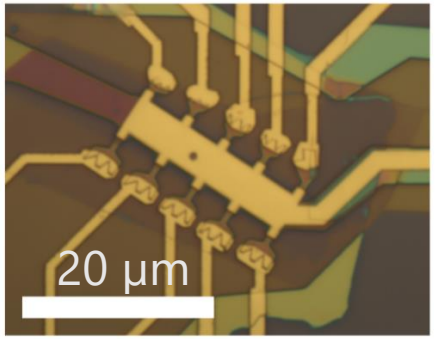

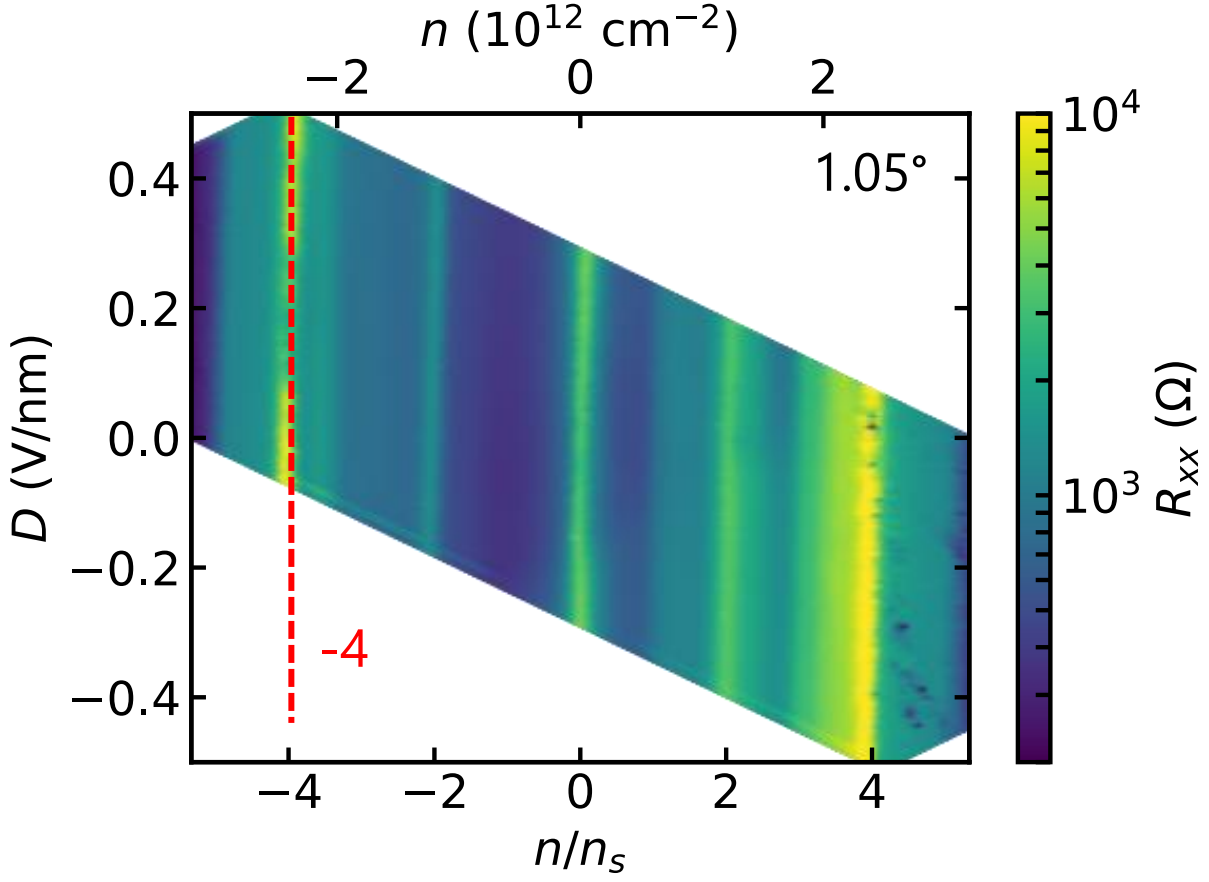

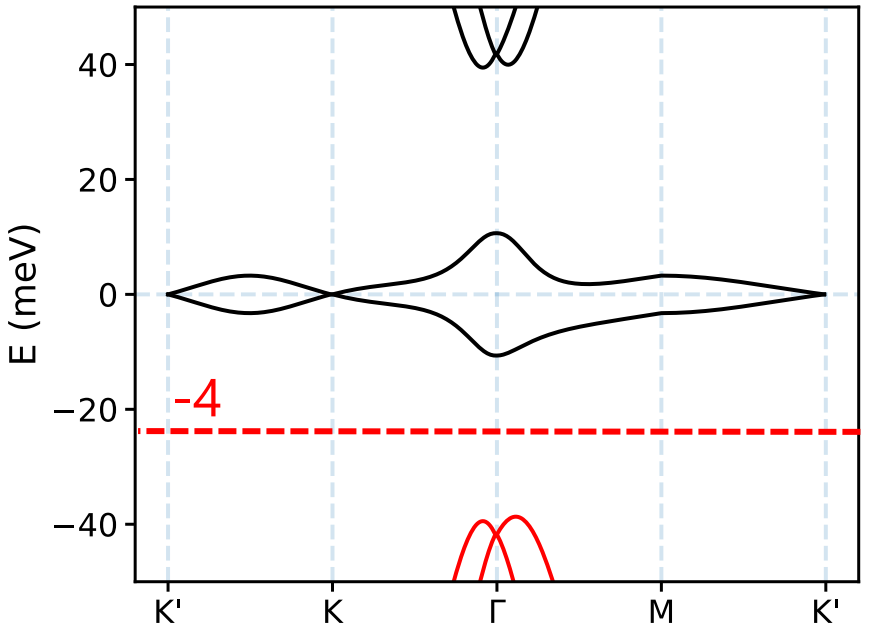

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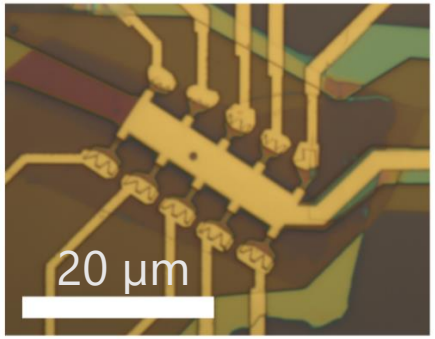

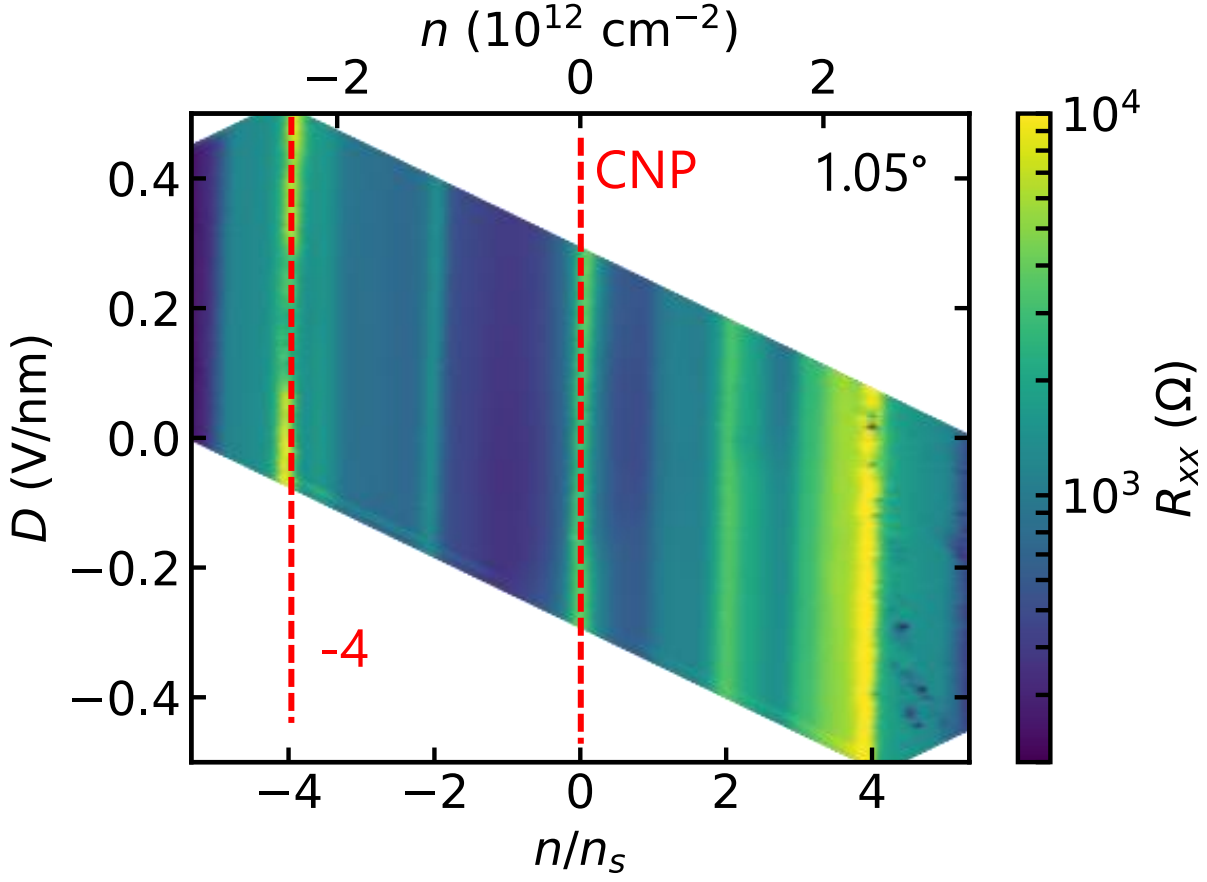

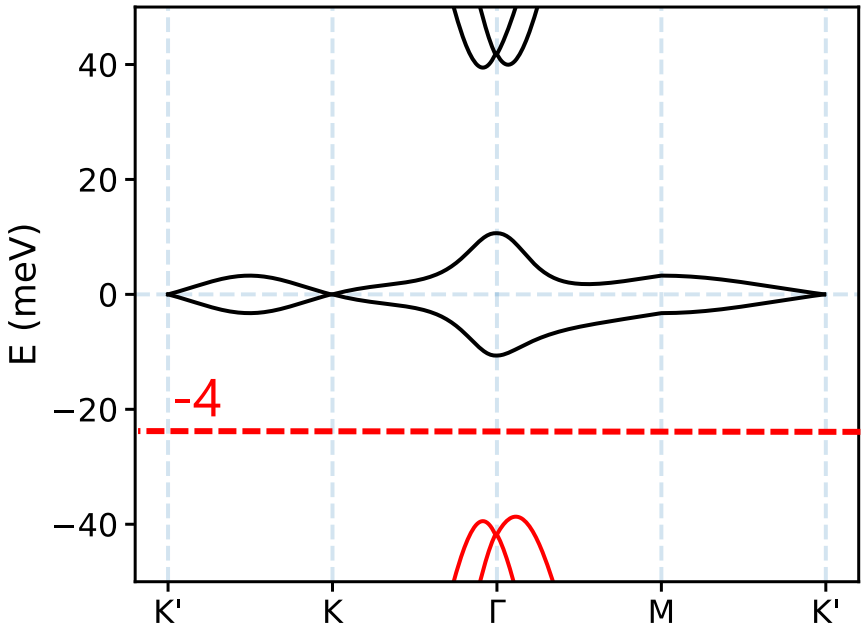

Sharpe, et al. Science (2019)

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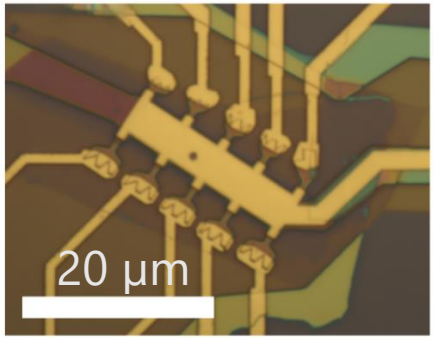

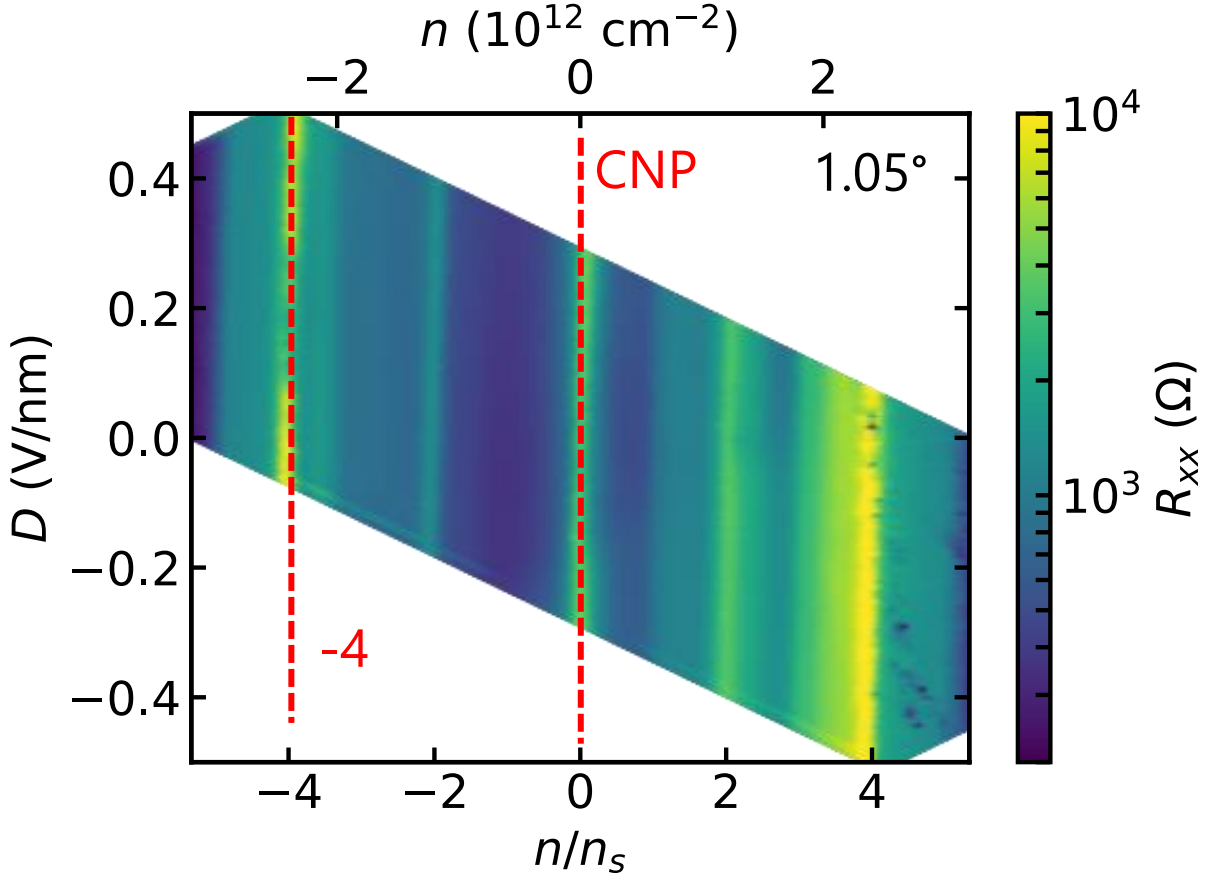

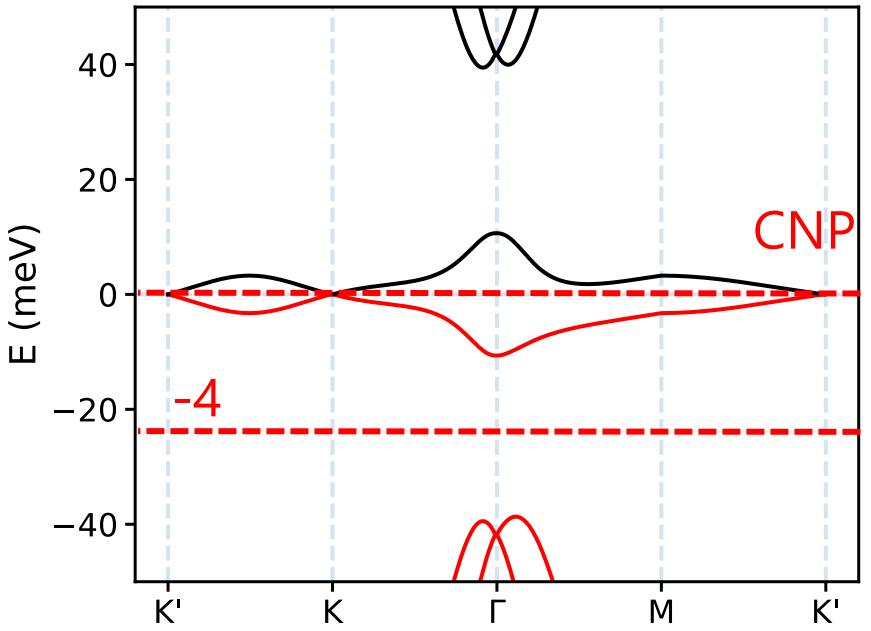

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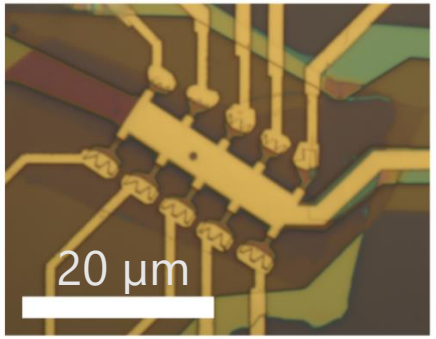

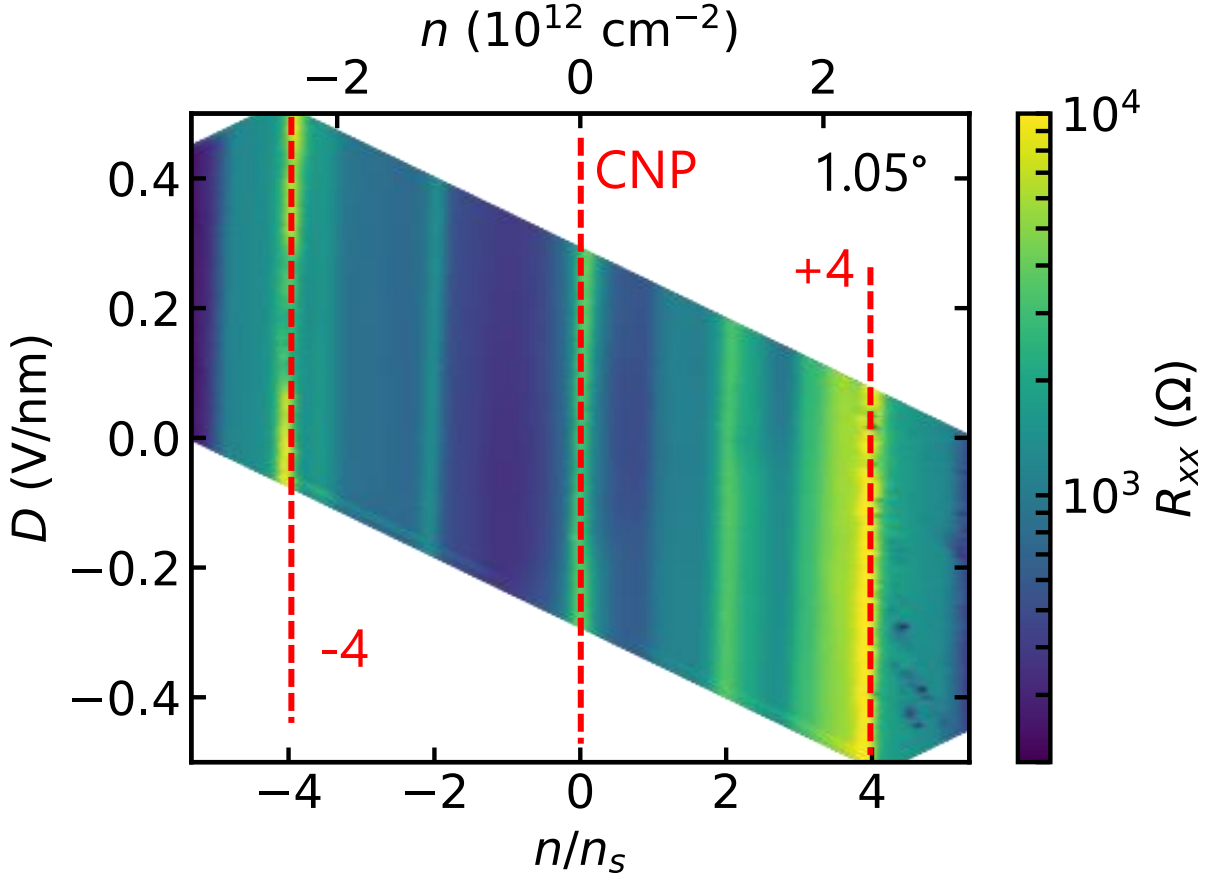

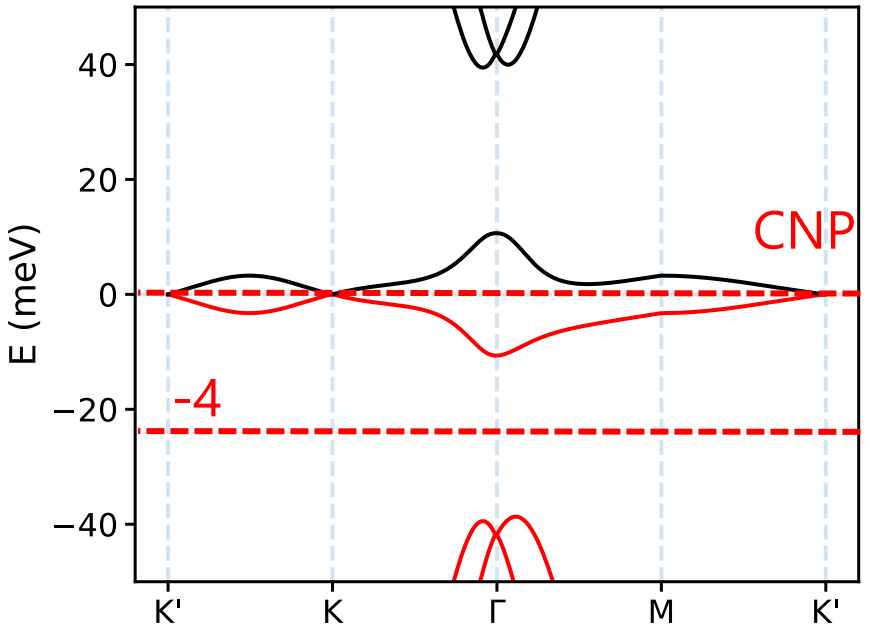

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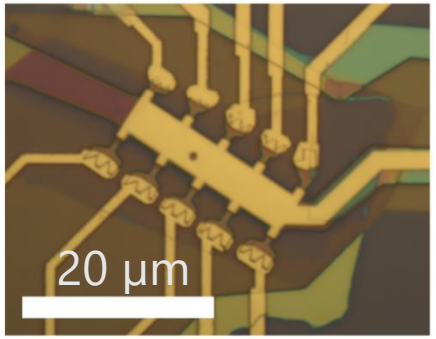

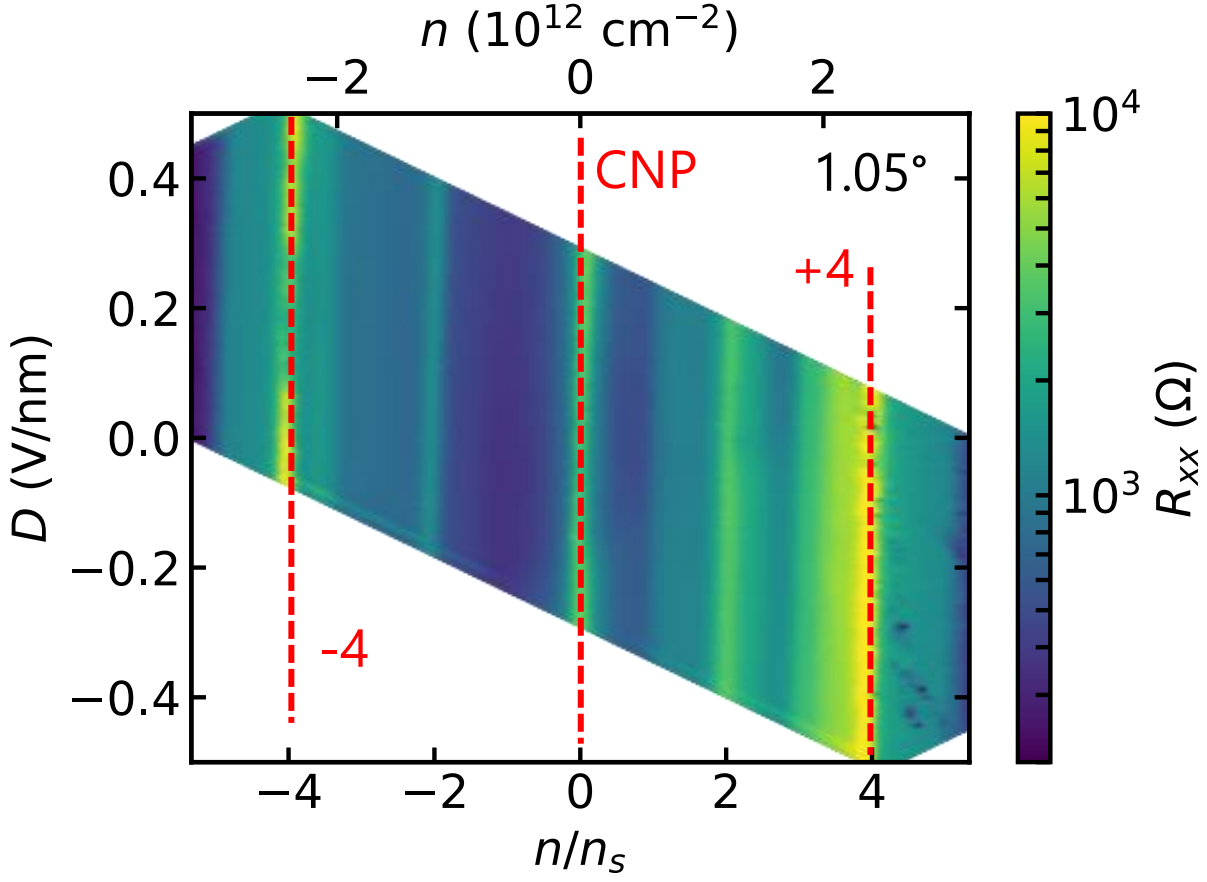

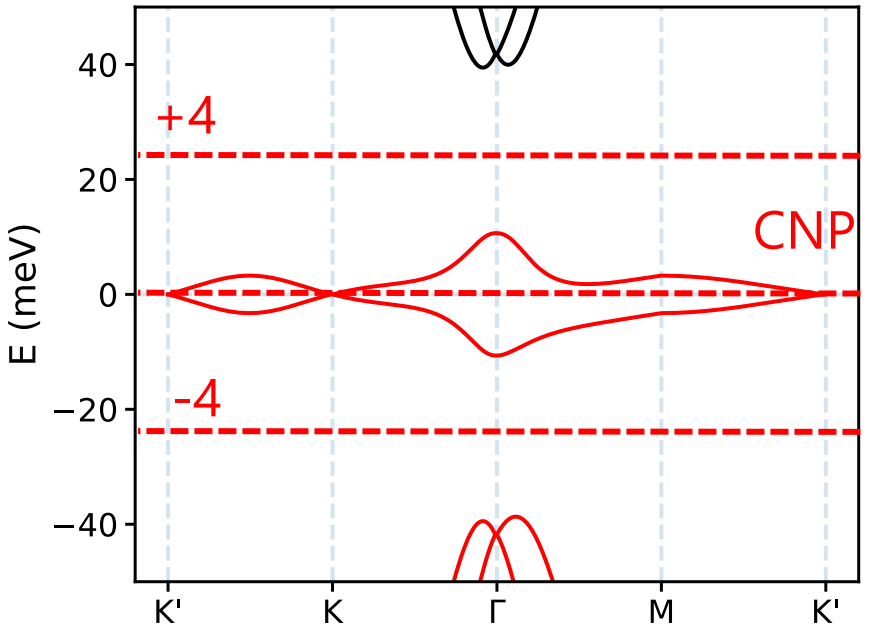

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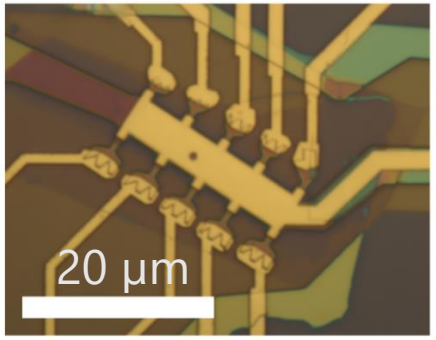

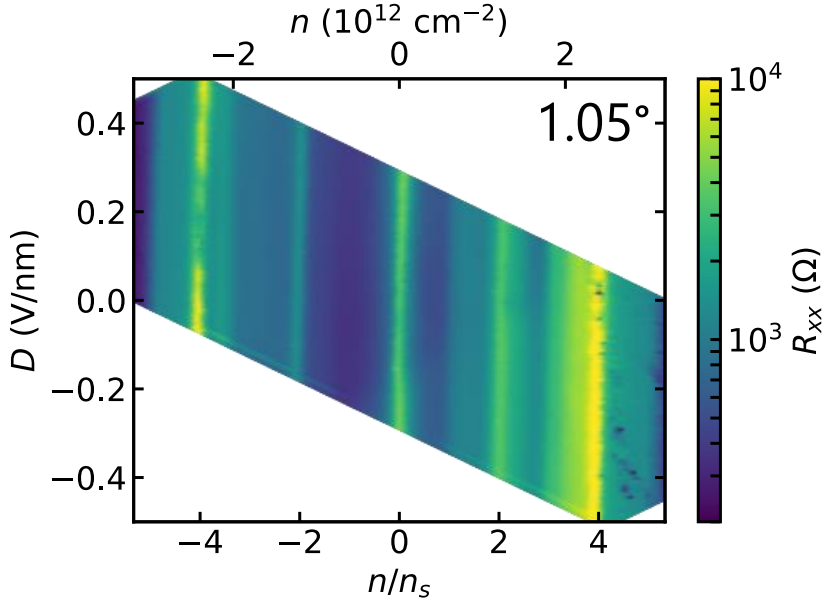

Atypical TBG

Sharpe, et al. Science (2019)

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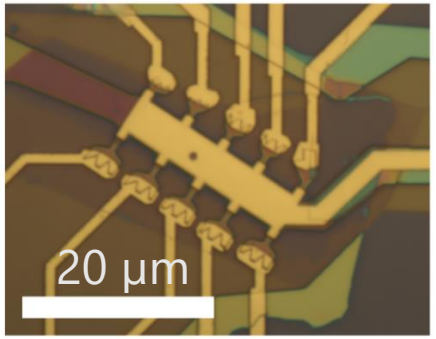

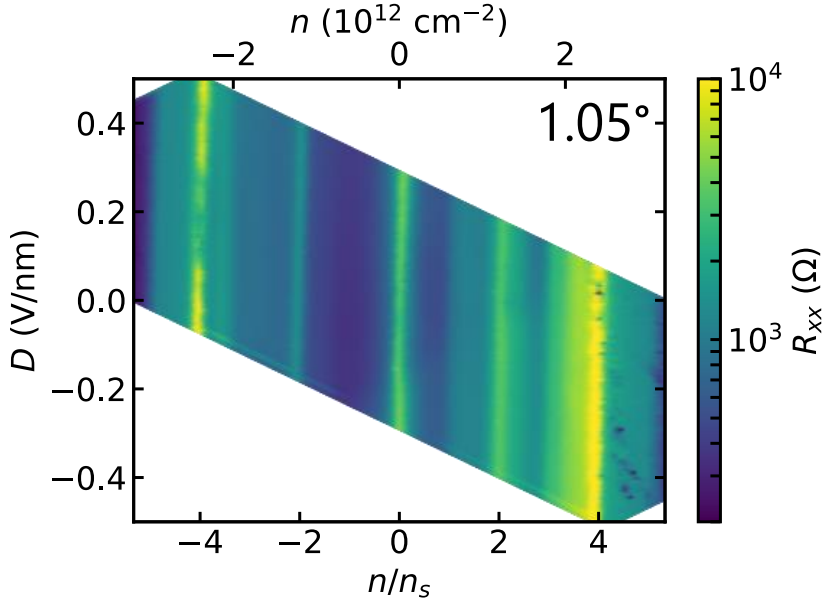

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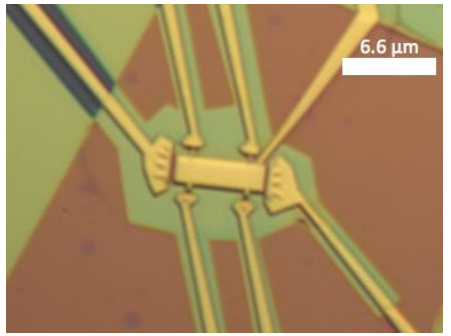

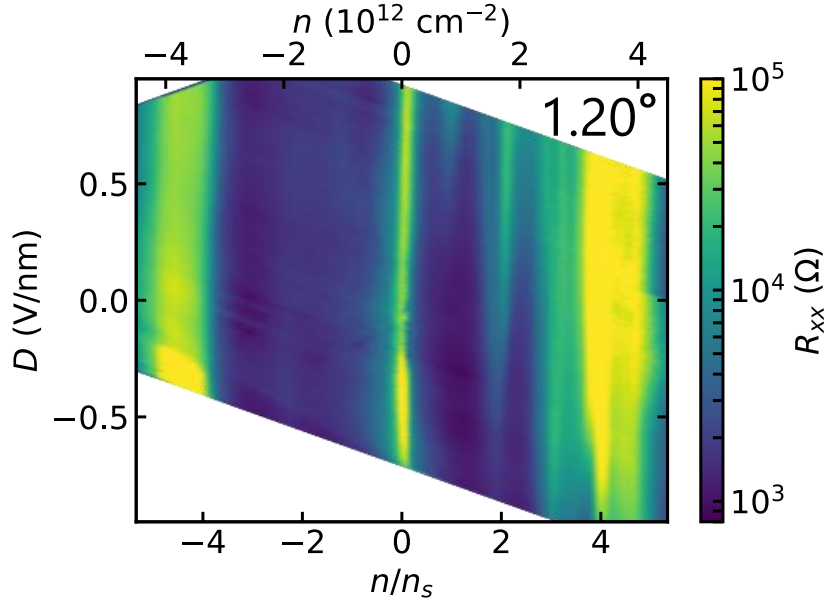

Sharpe, et al. Science (2019)

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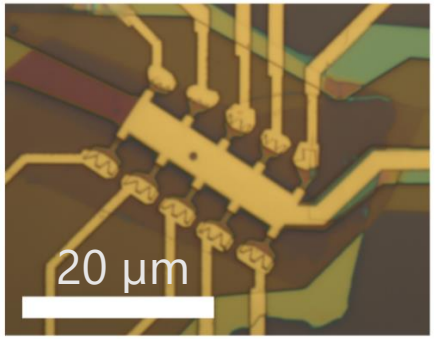

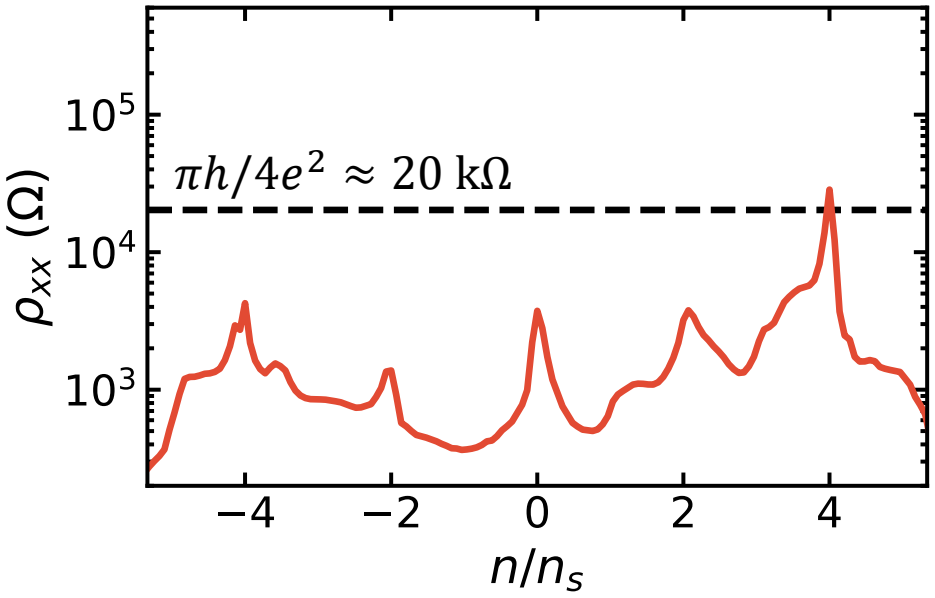

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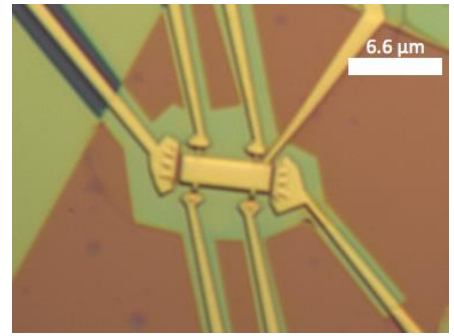
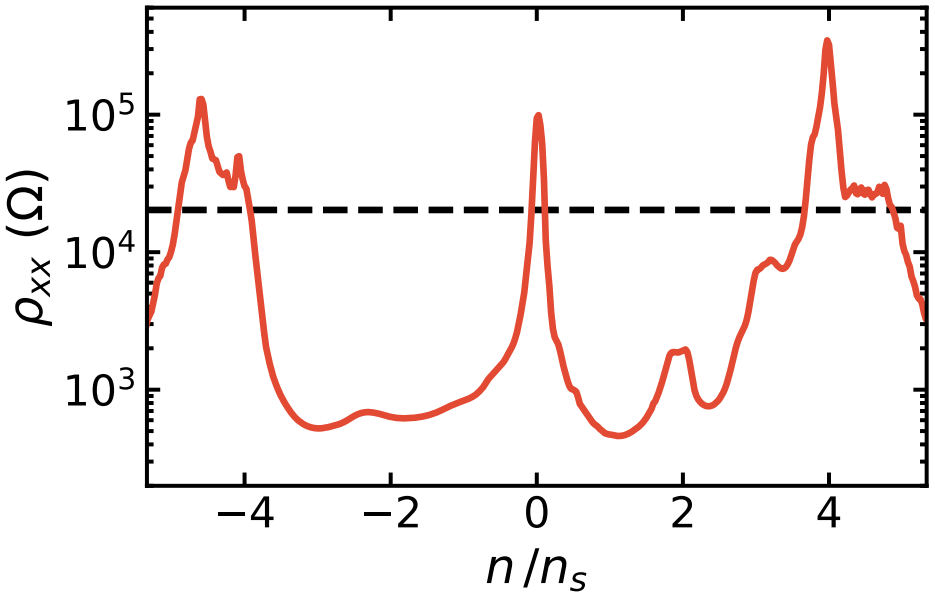
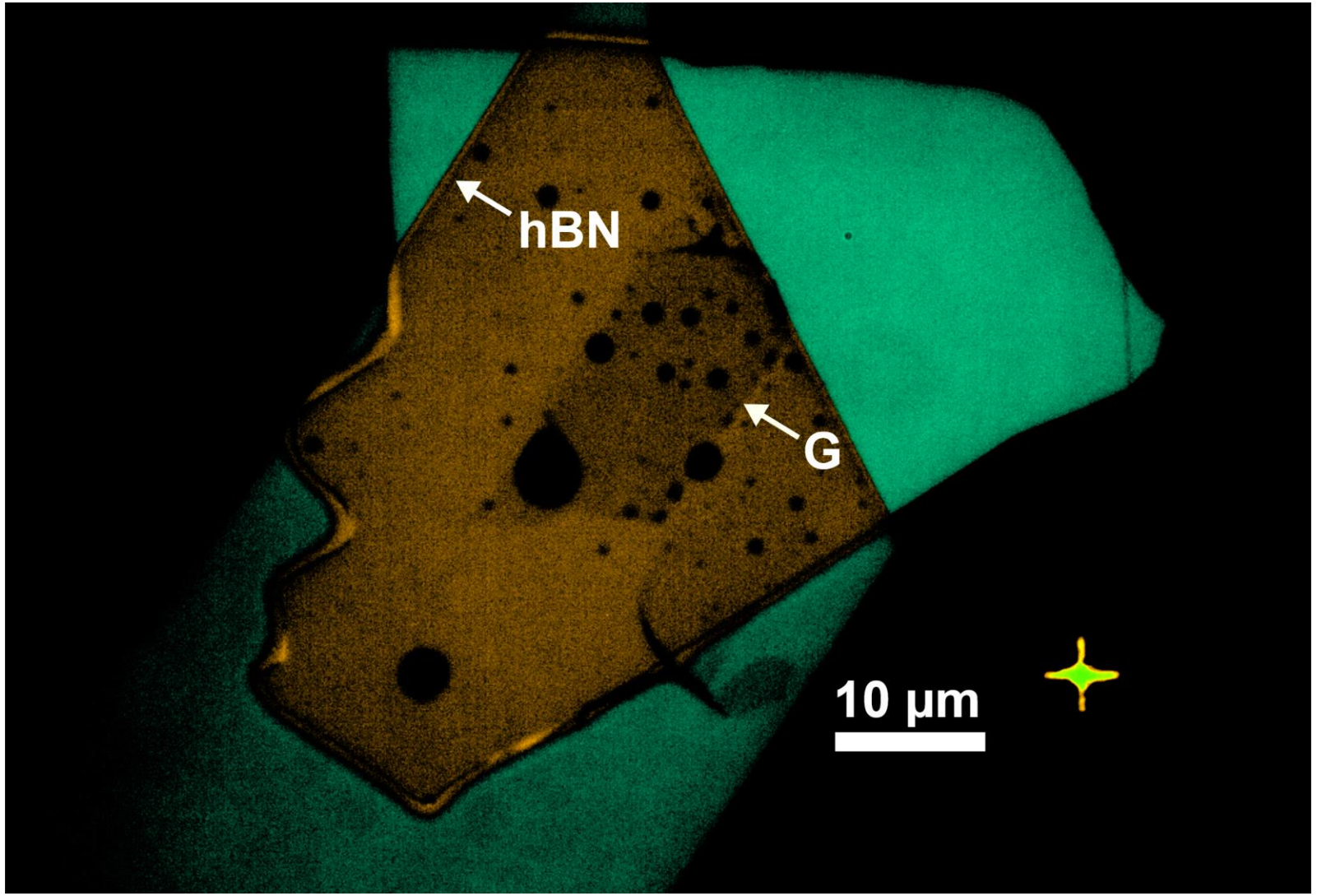




Sharpe, et al. Science (2019)

Visual hBN alignment



Amet et al., PRL (2013)

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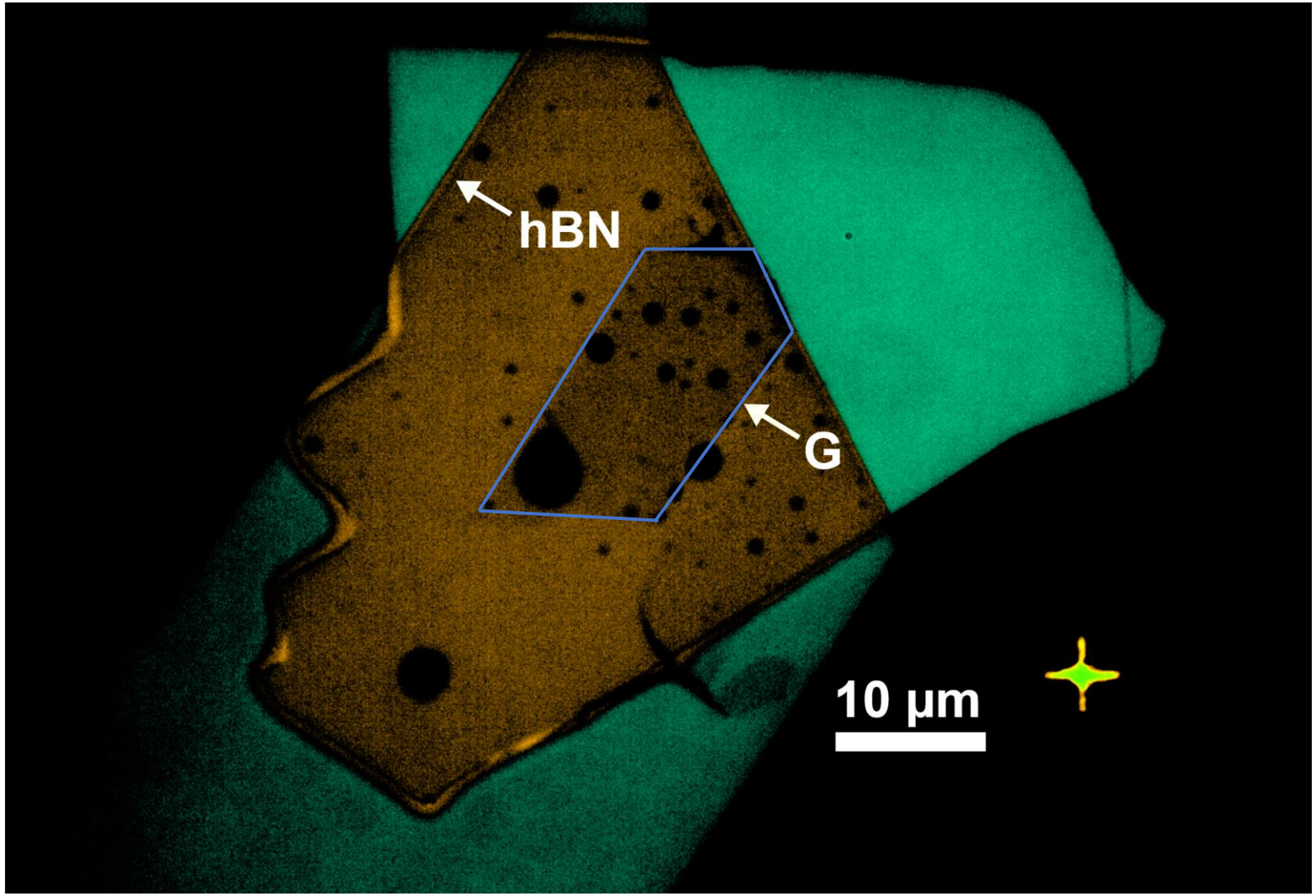

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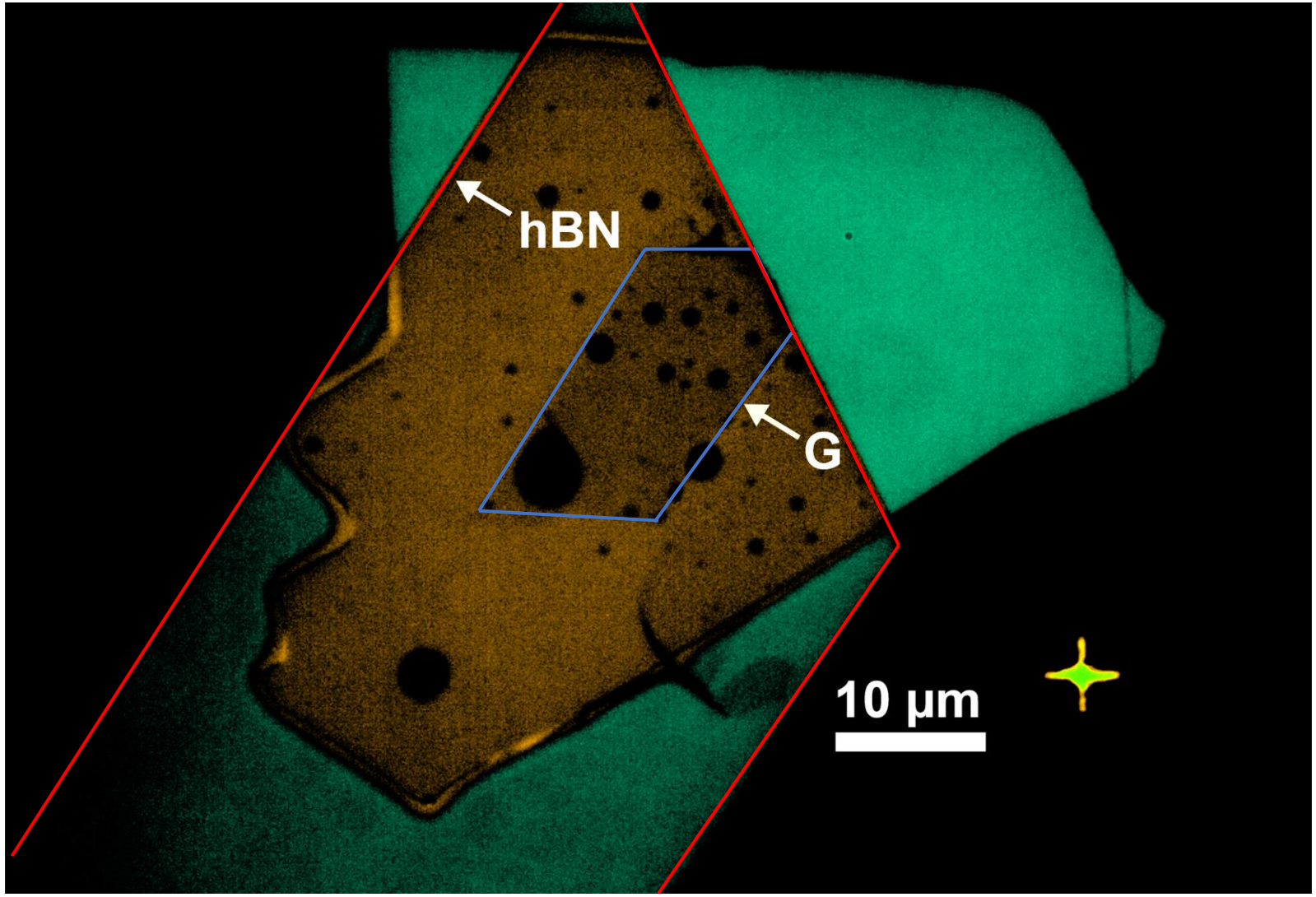

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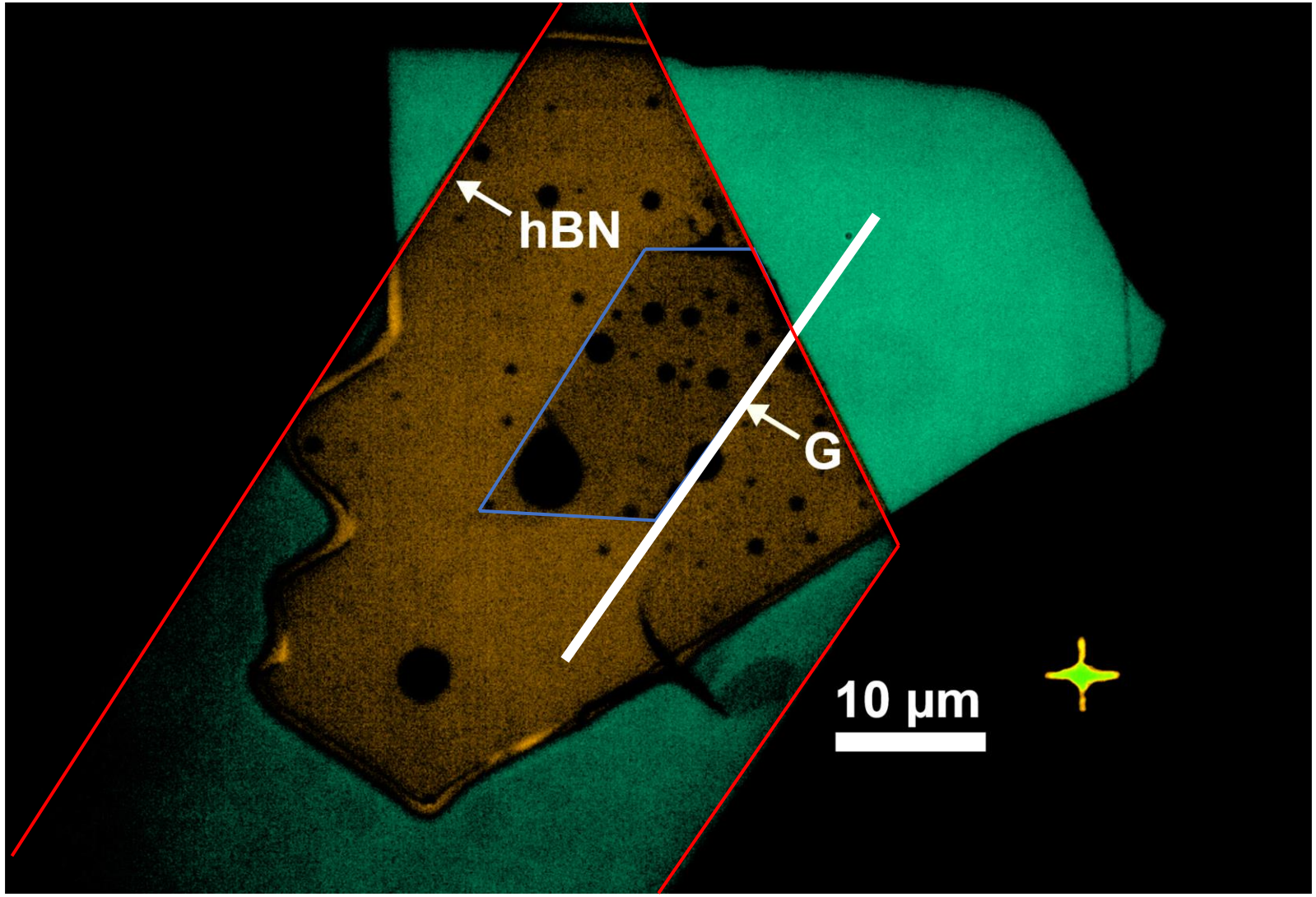

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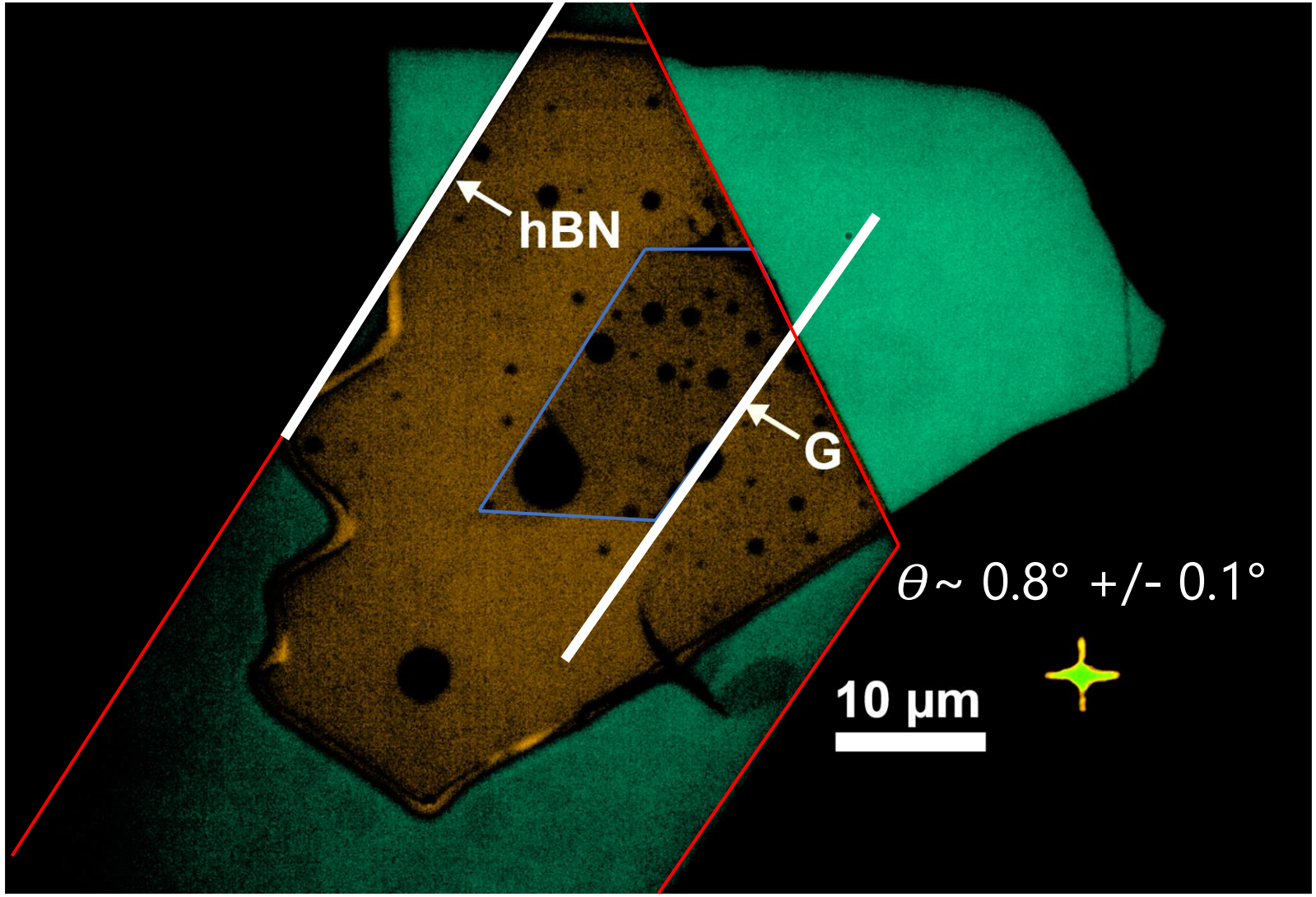

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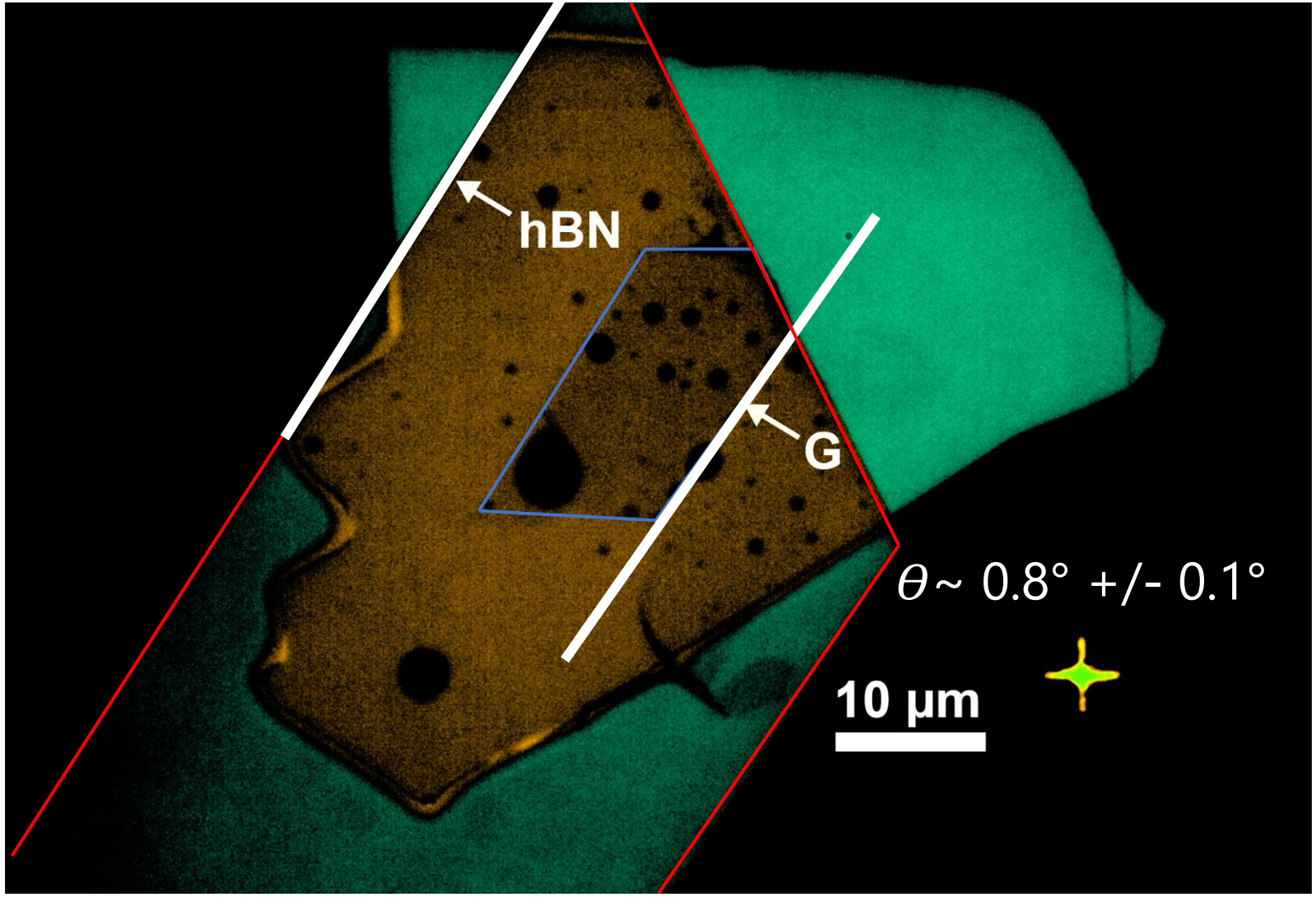

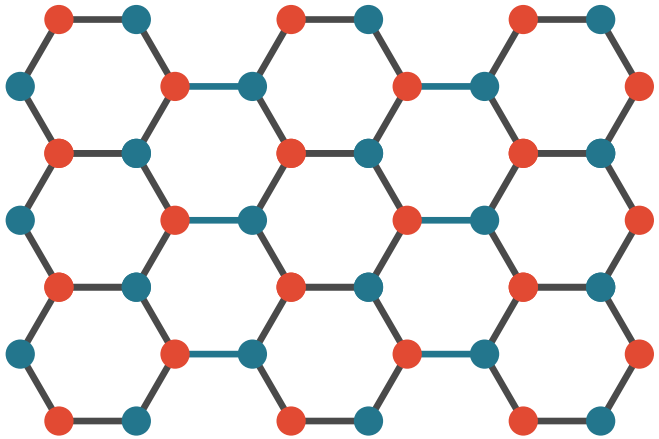

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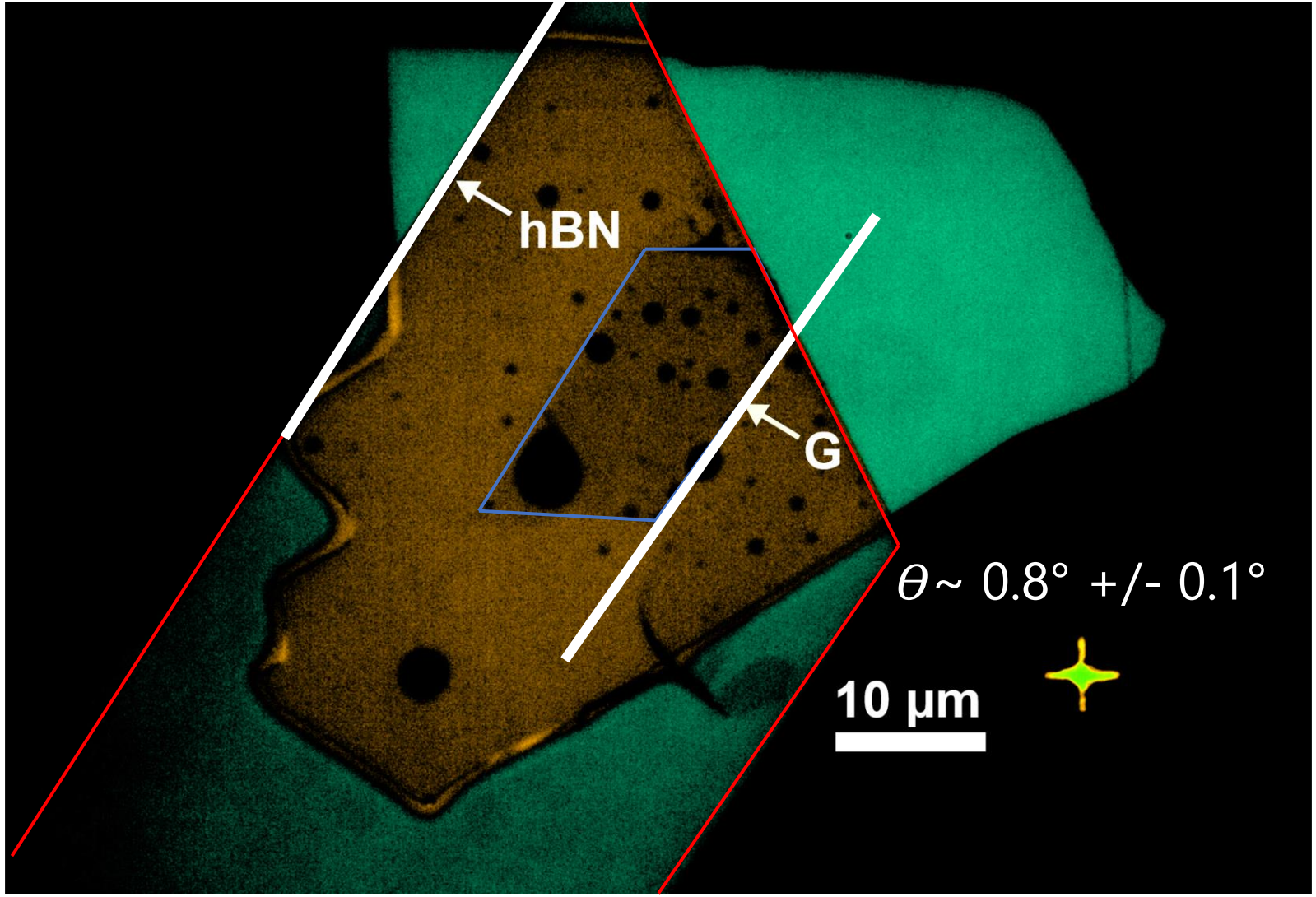
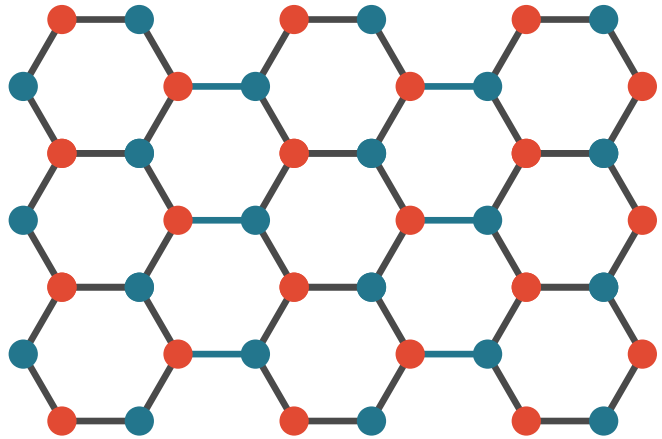
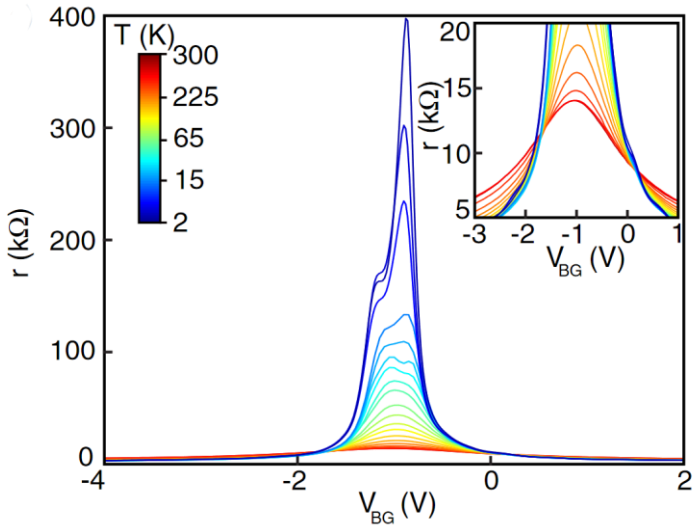
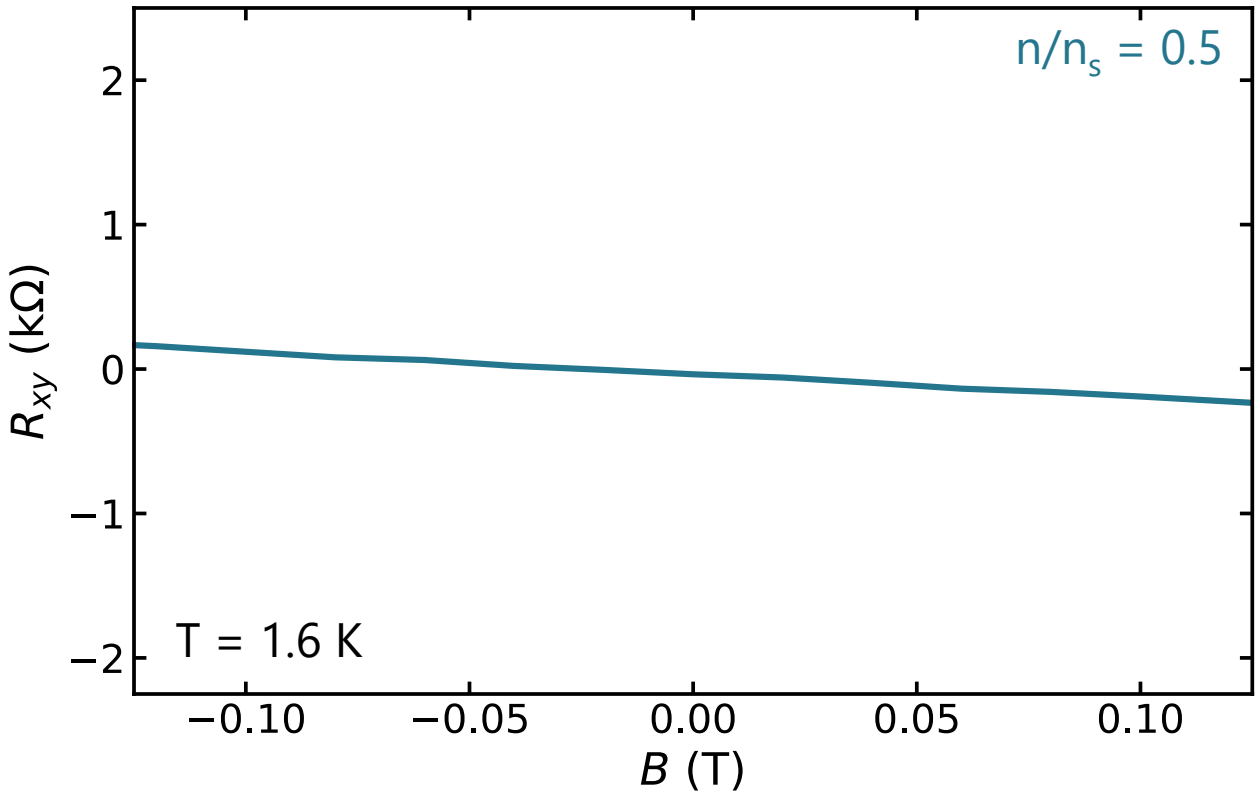






Amet et al., PRL (2013)

Hall slope density dependence



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

Sharpe, et al. Science (2019)

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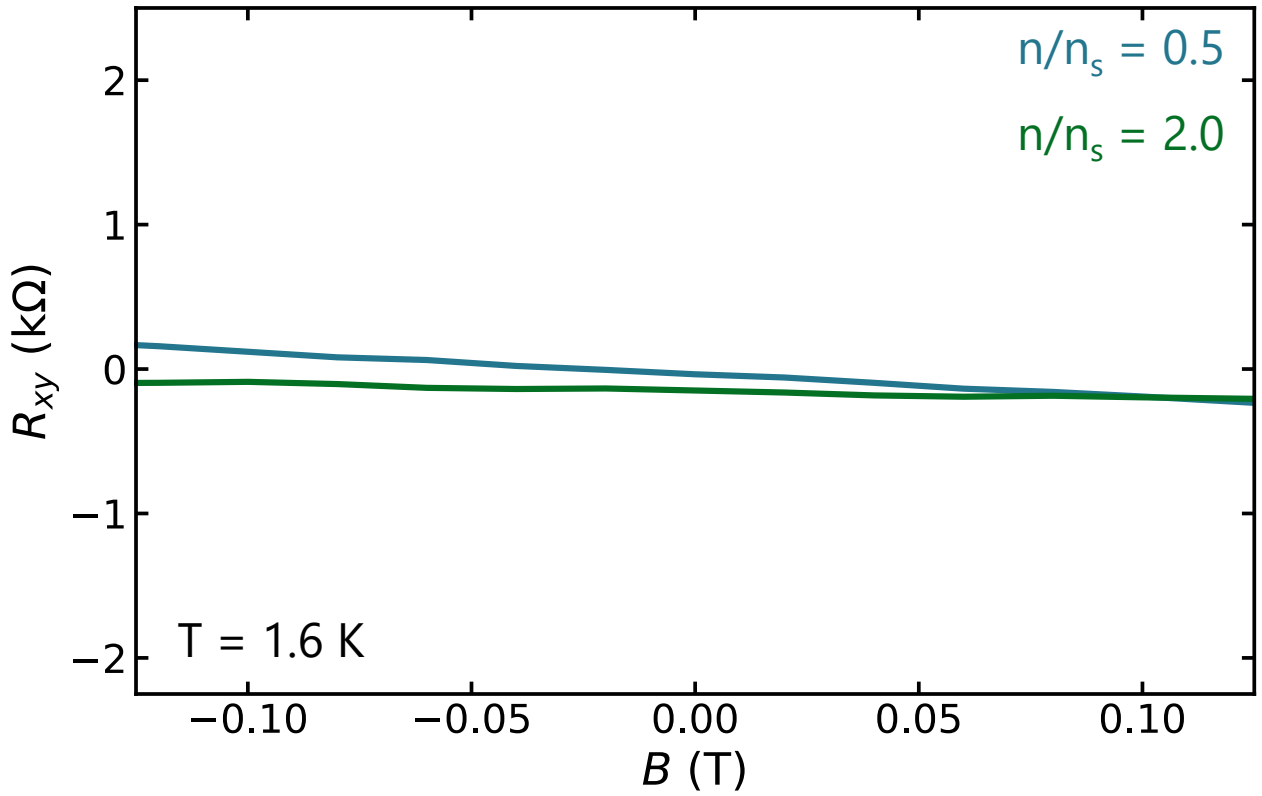

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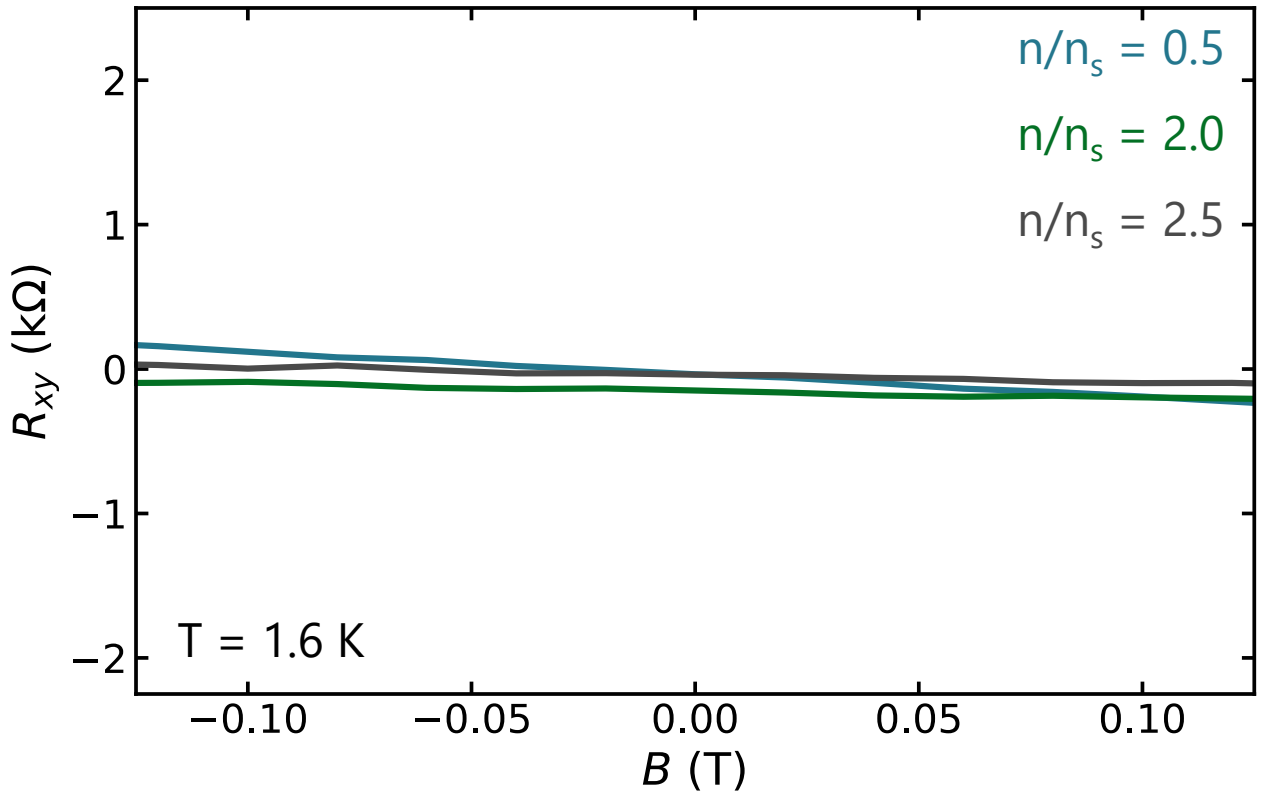

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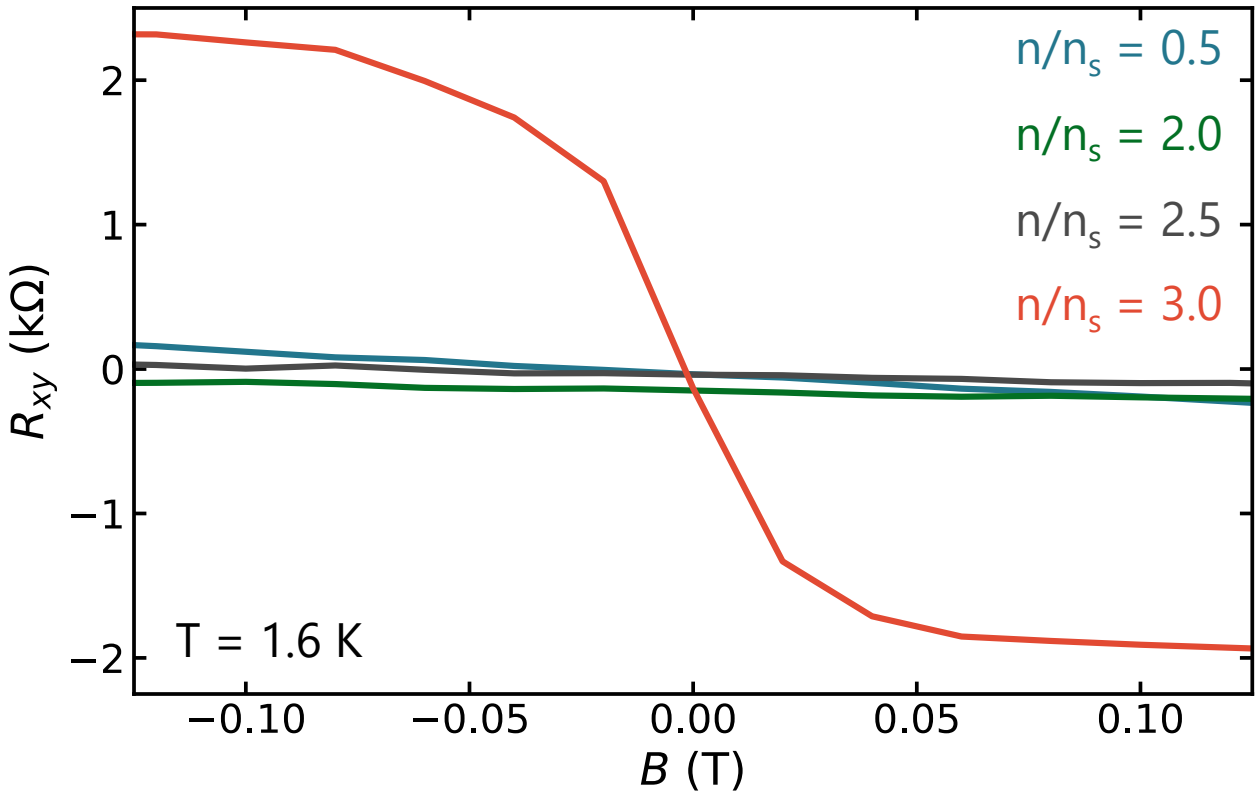

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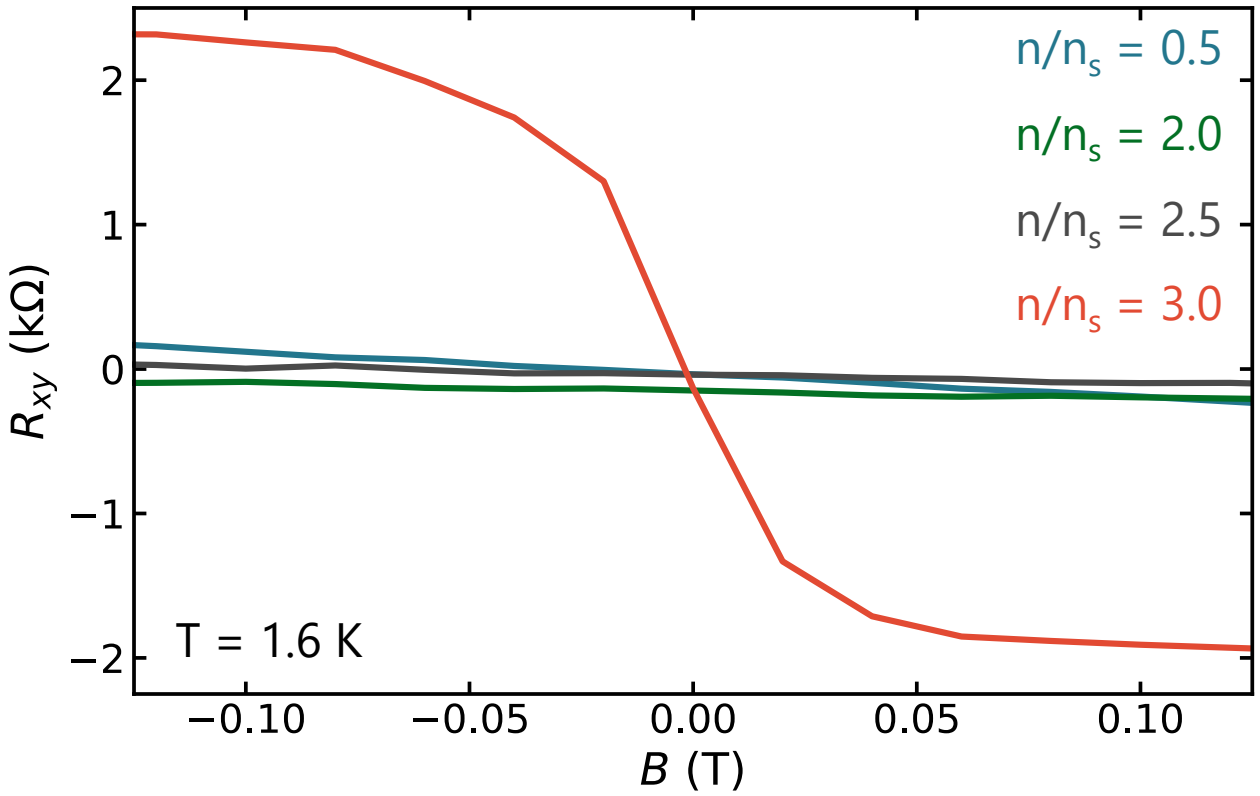

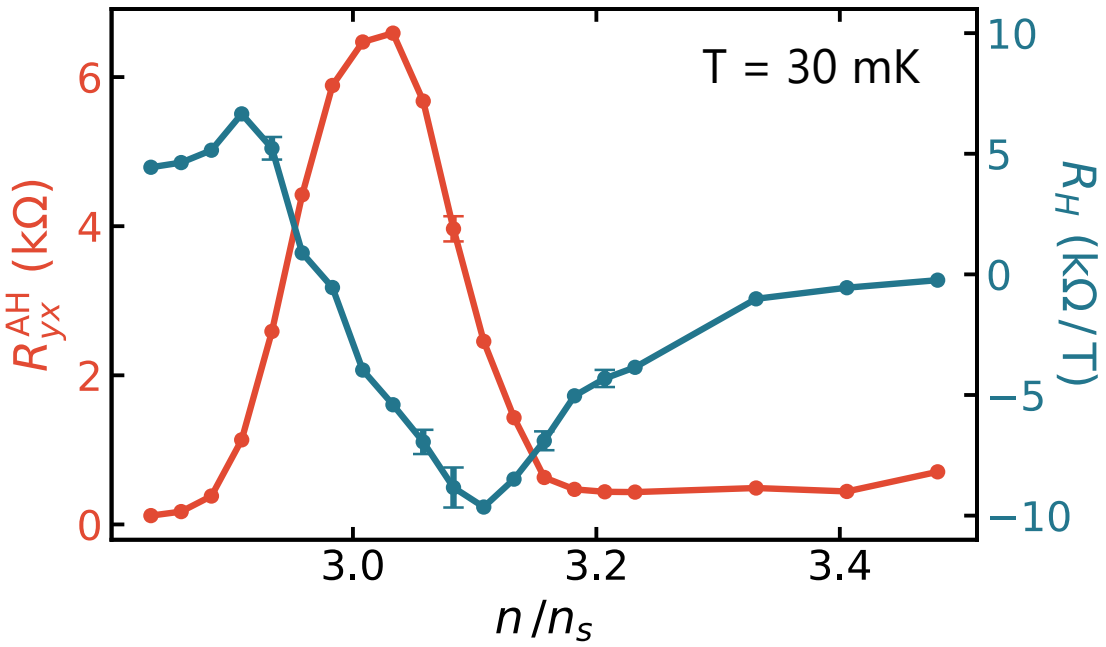

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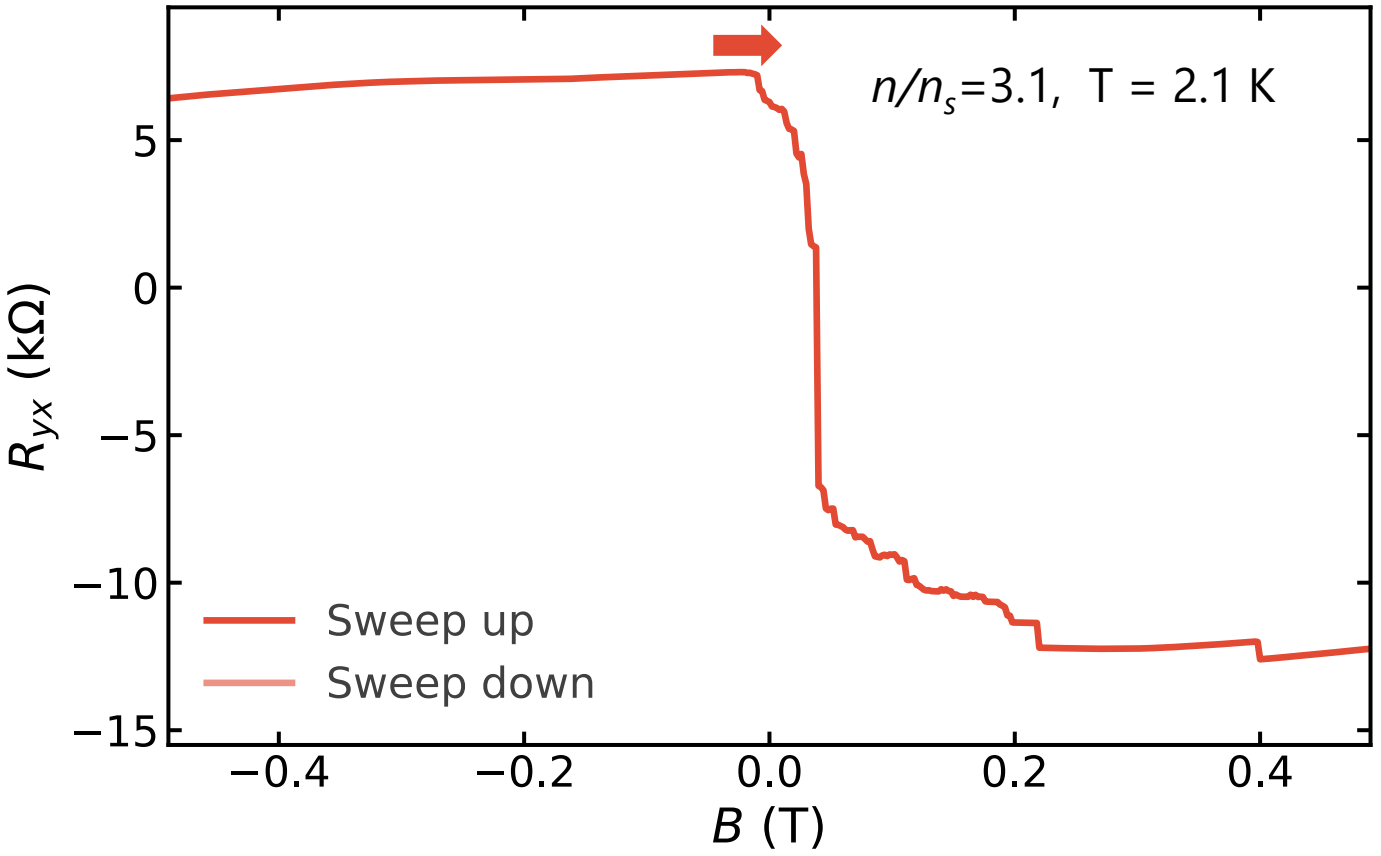
Sharpe, et al. Science (2019)

Anomalous Hall signal can be really large!



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

Sharpe, et al. Science (2019)

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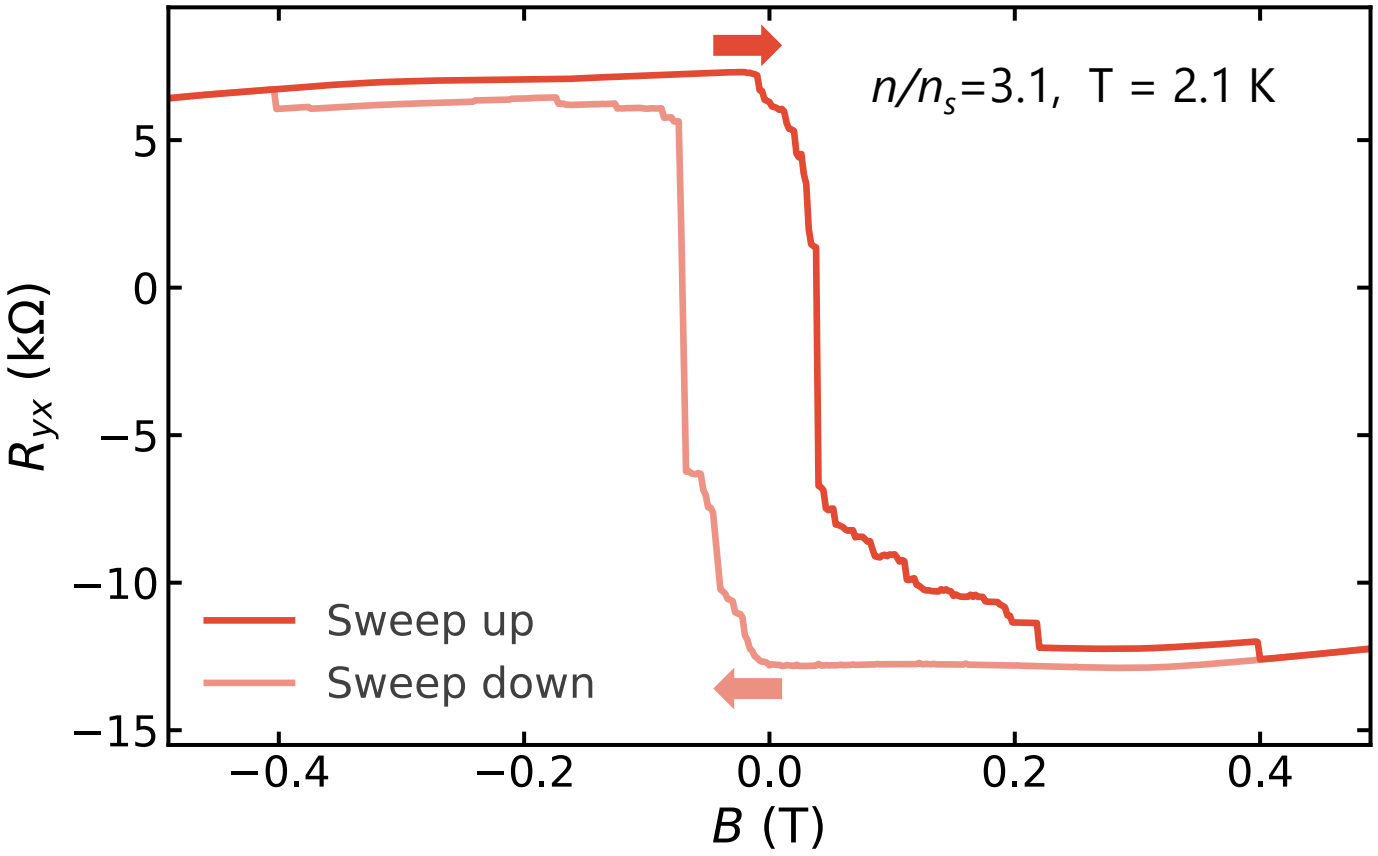

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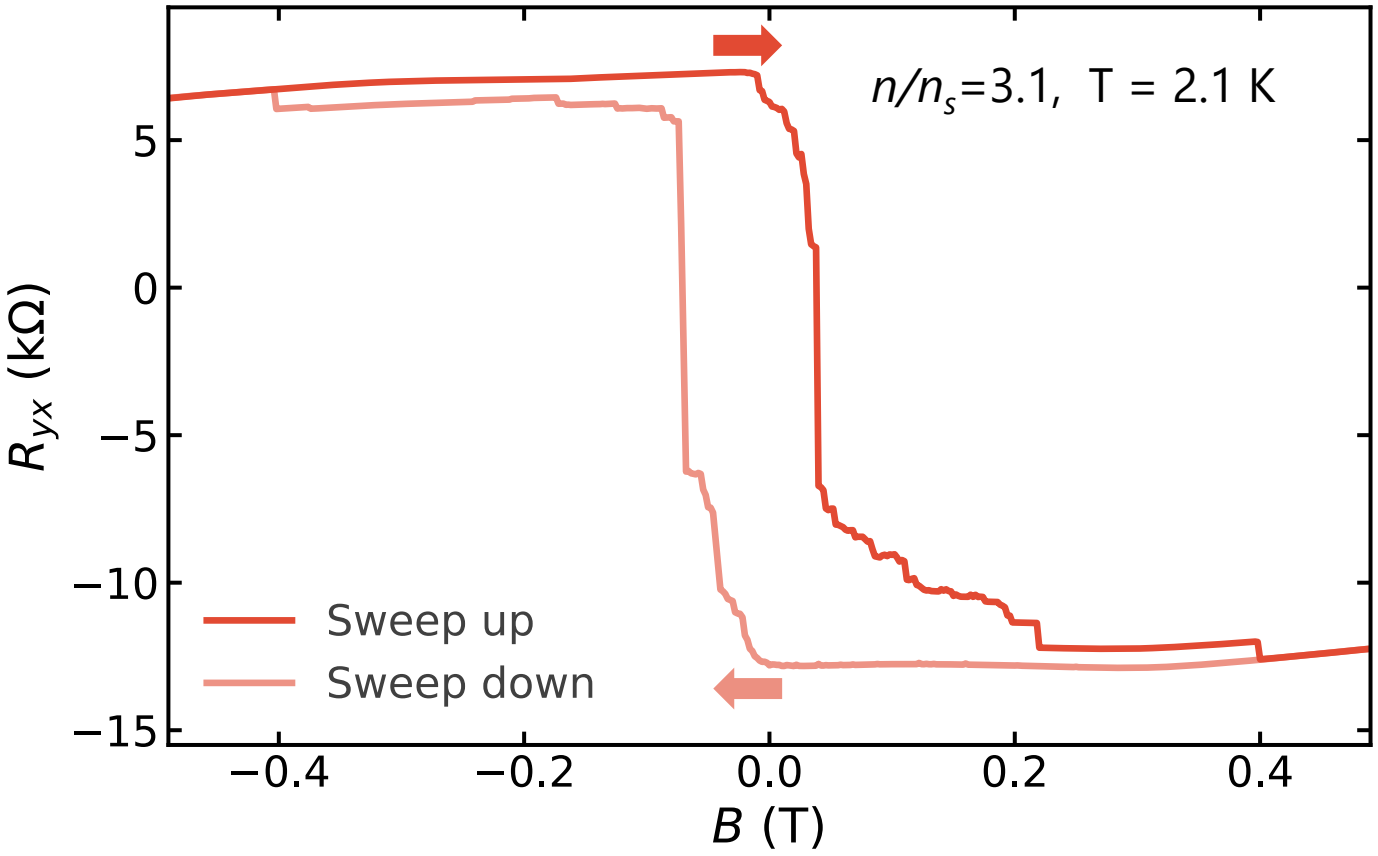

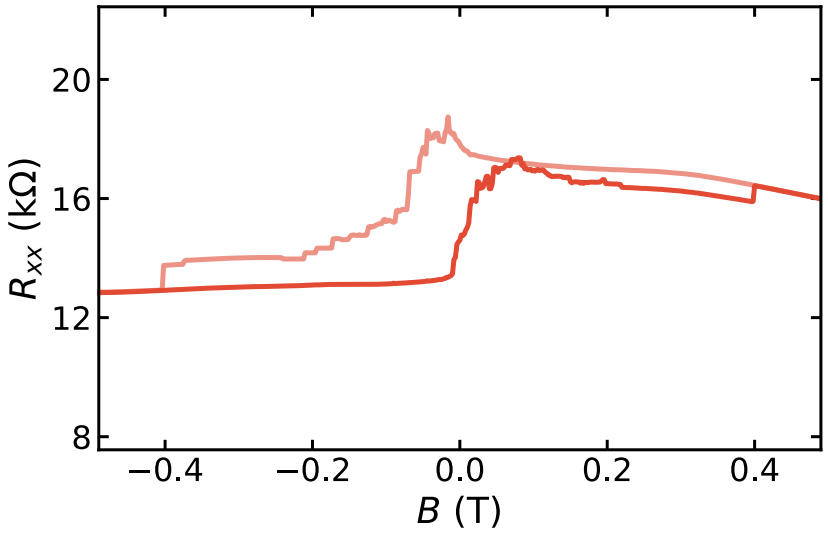

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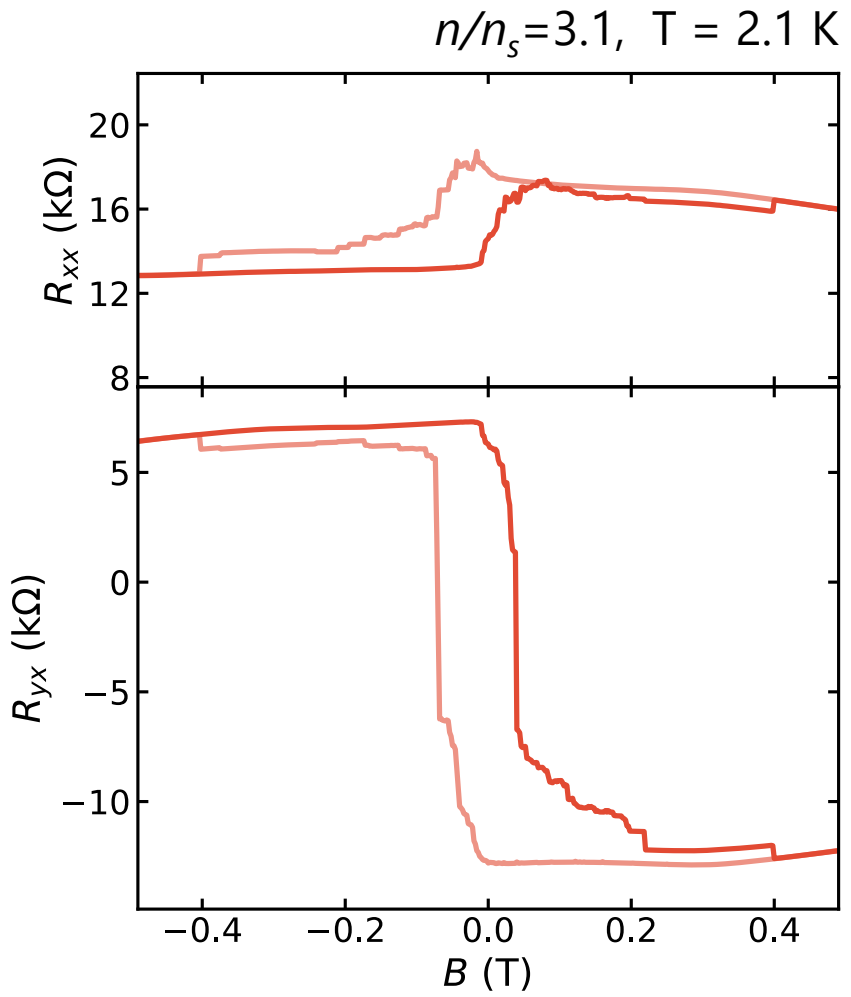
Sharpe, et al. Science (2019)

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 ~5K
- Evidence of domains

Reminiscent of early Magnetic Tis →Chern insulator?



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Ideally: $\rho_{xx} = 0$
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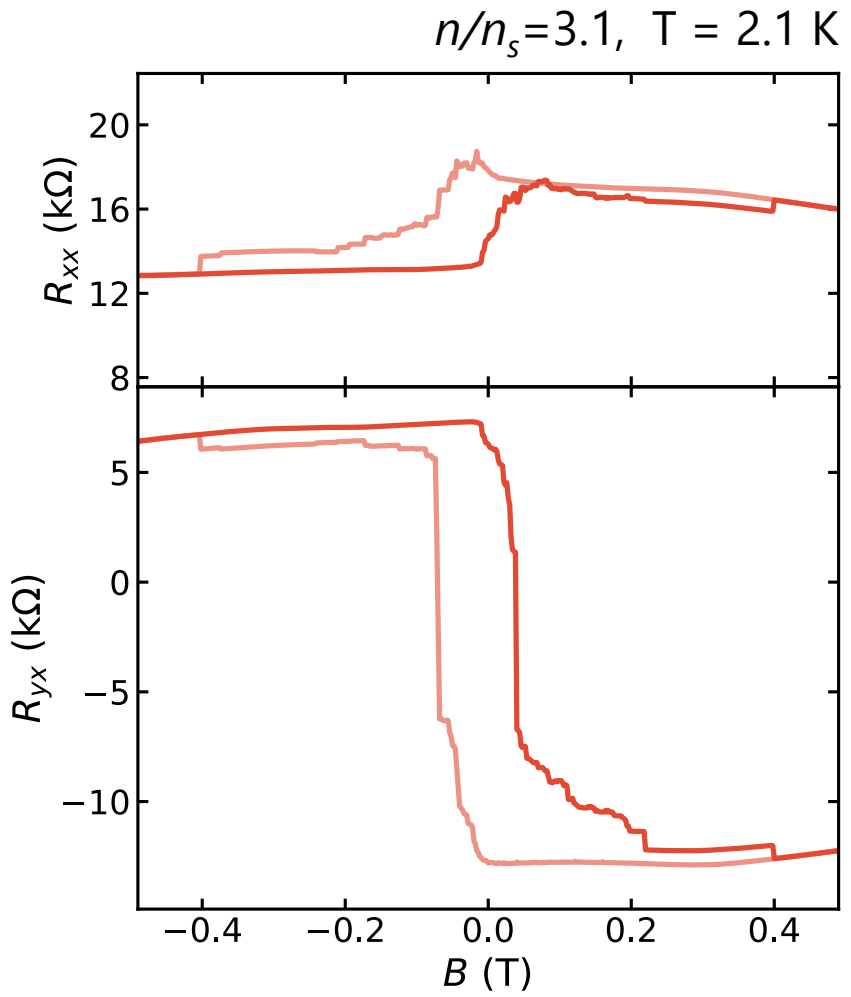

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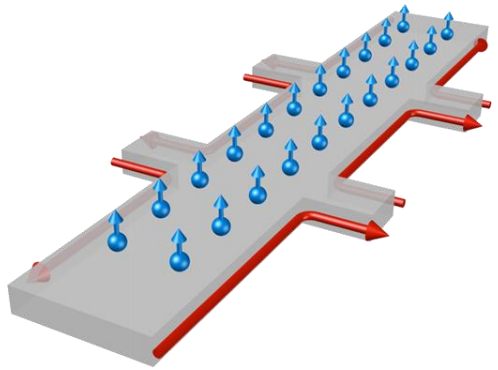
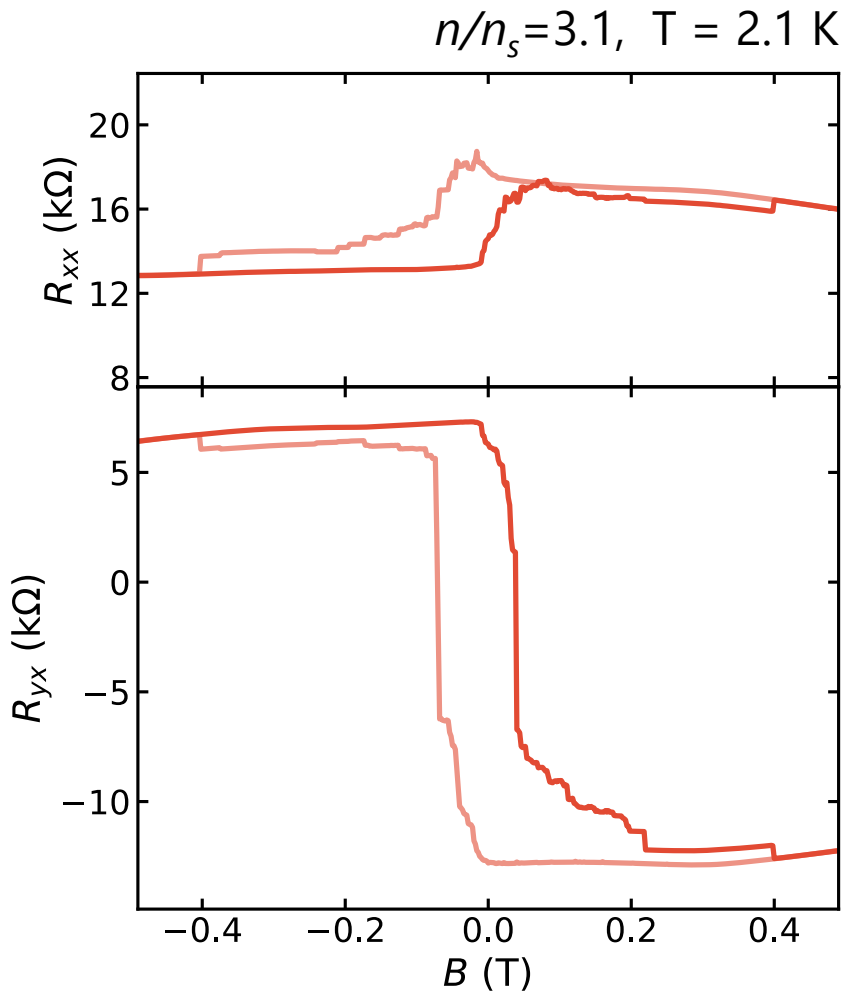
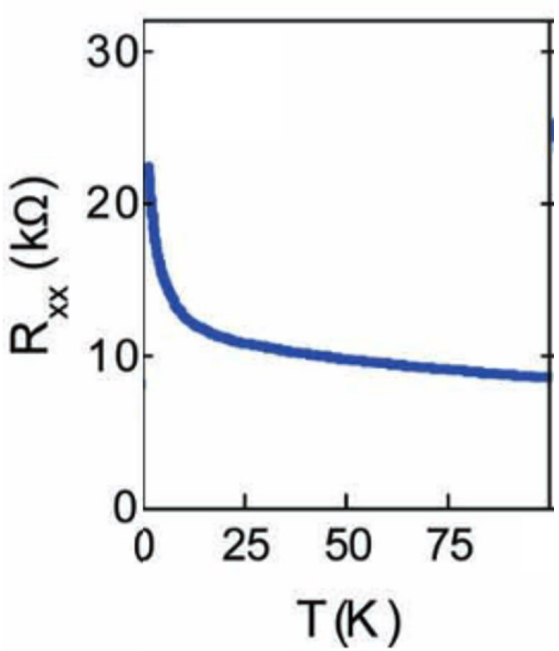
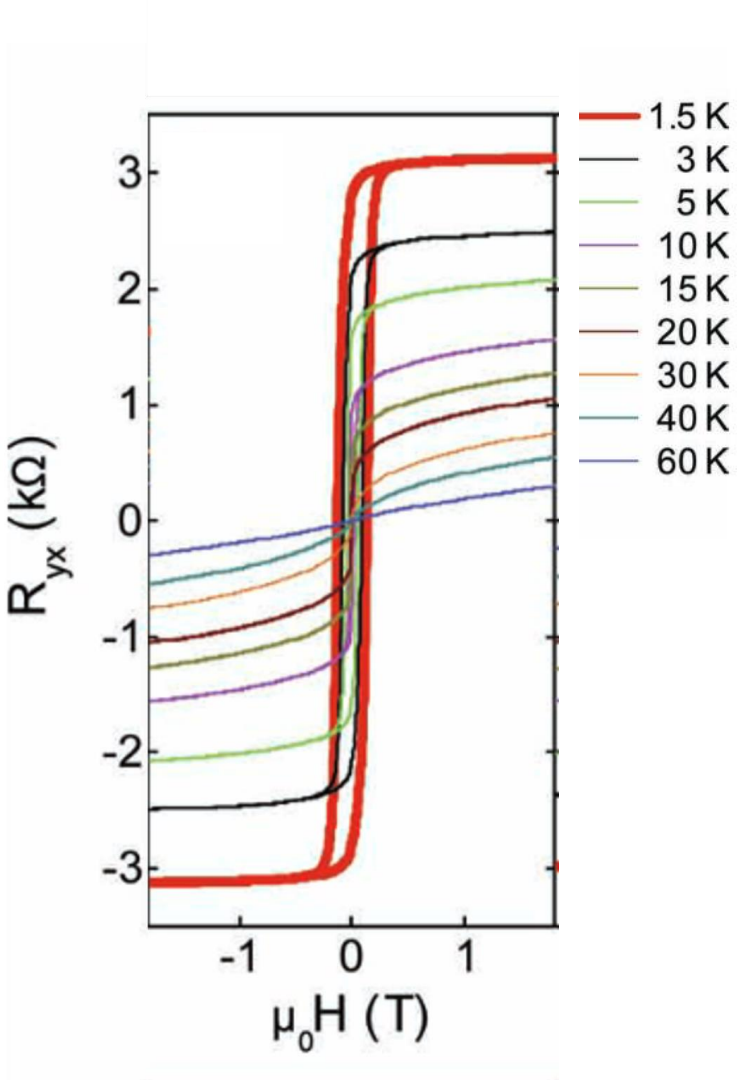




Sharpe, et al. Science (2019)

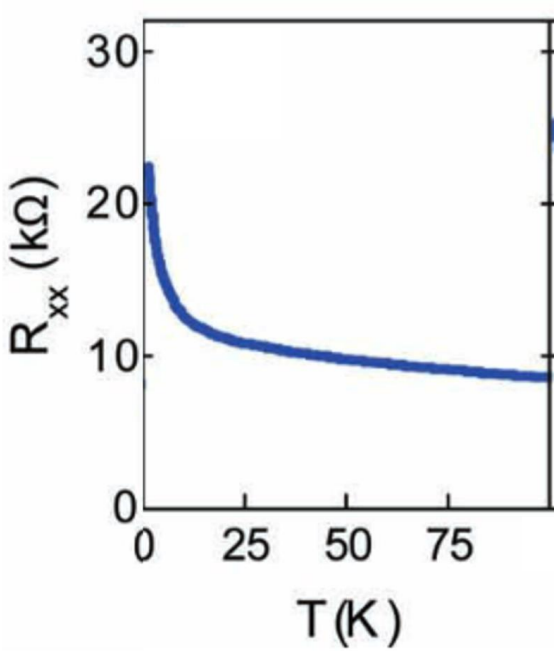
Early magnetically doped topological insulators

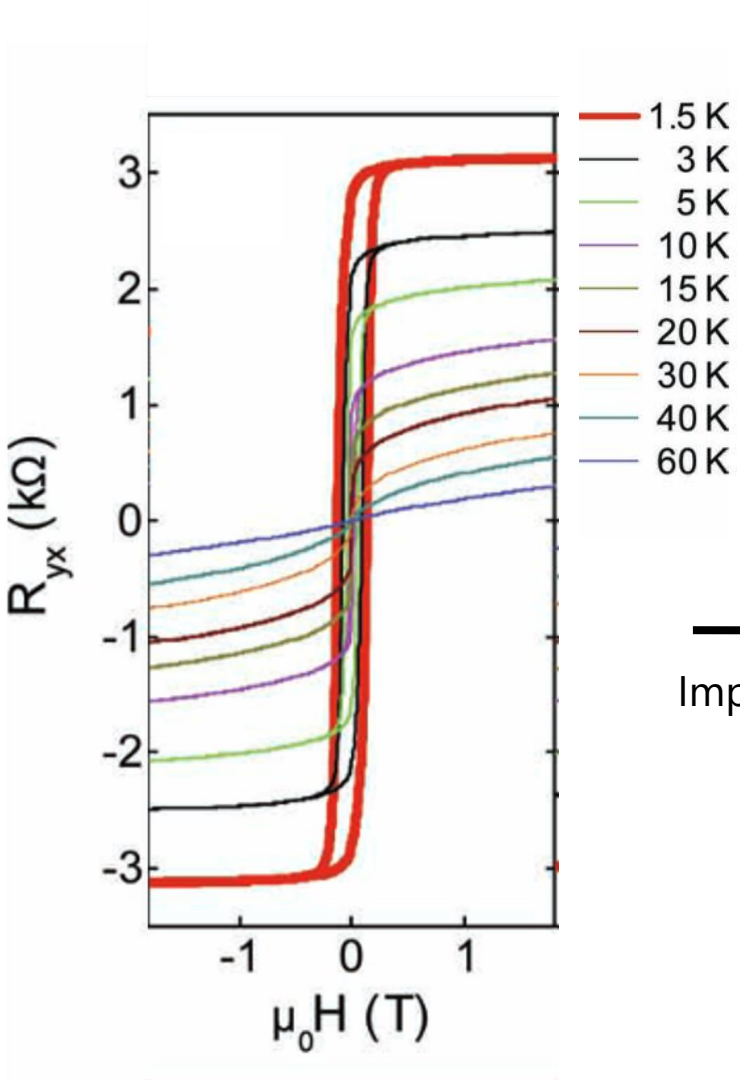




Chang, et al. Adv. Mater (2013)
Fox, et al., PRB (2018)

Early magnetically doped topological insulators

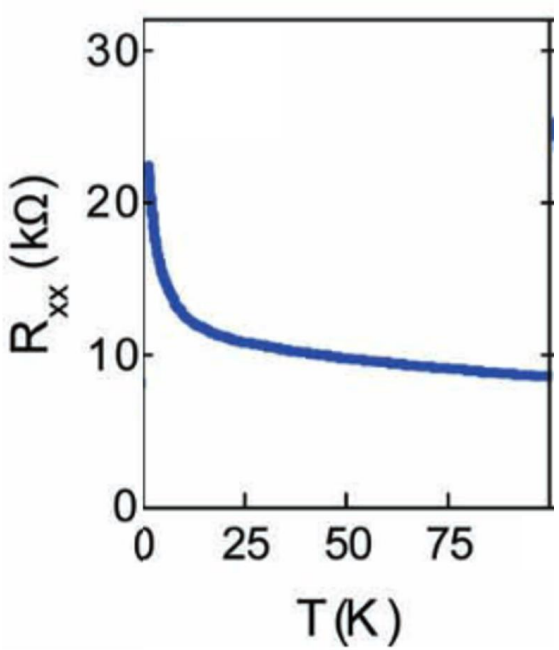


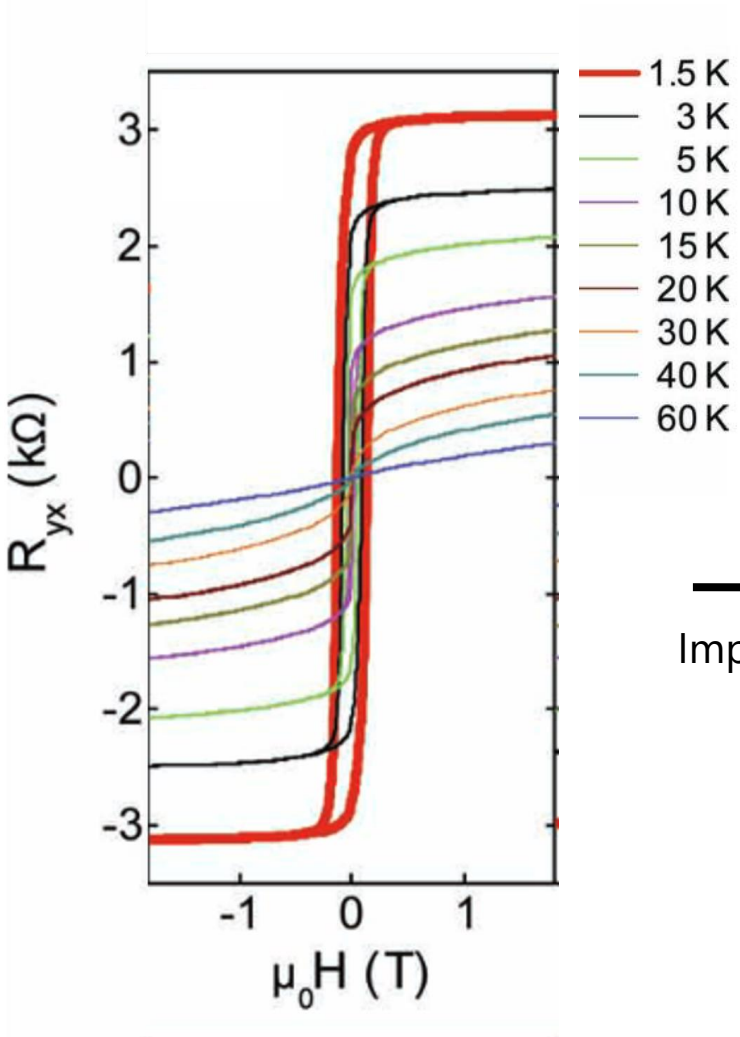


Improve films

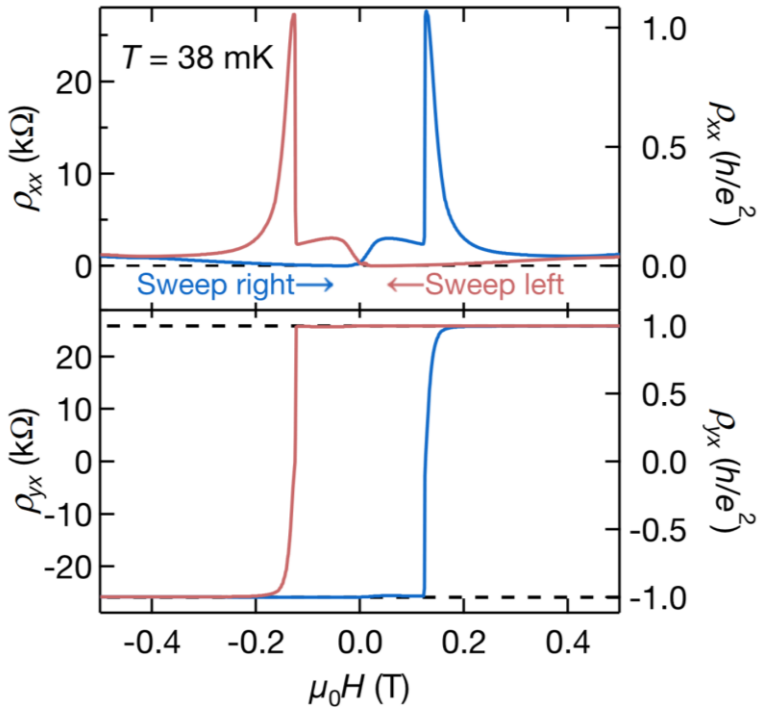
Chang, et al. Adv. Mater (2013)
Fox, et al., PRB (2018)

Early magnetically doped topological insulators





Improve films



Chang, et al. Adv. Mater (2013)
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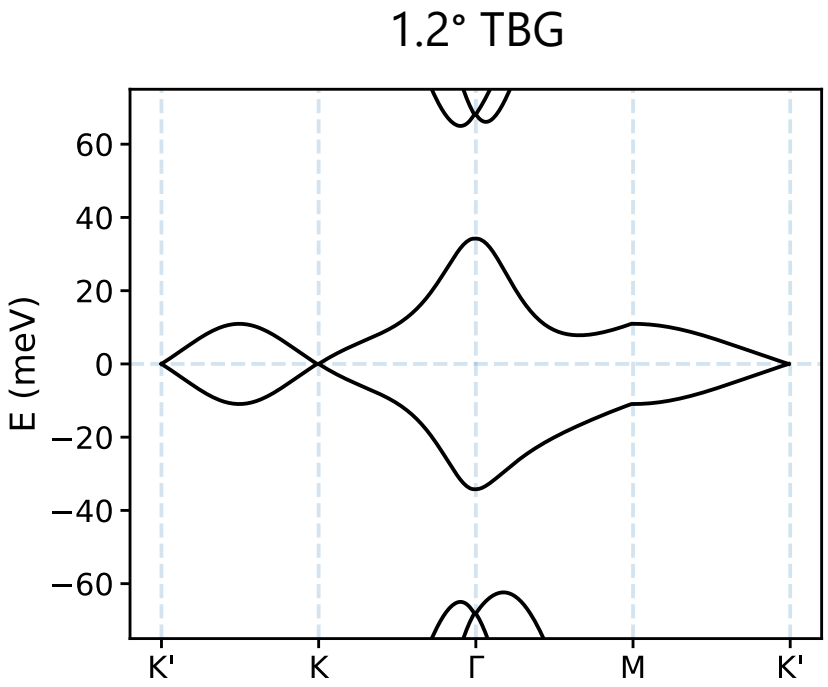
hBN alignment is the key for AH?



https://git.sr.ht/~spxtr/bm_model
Sharpe, et al. Science (2019)

Theory:
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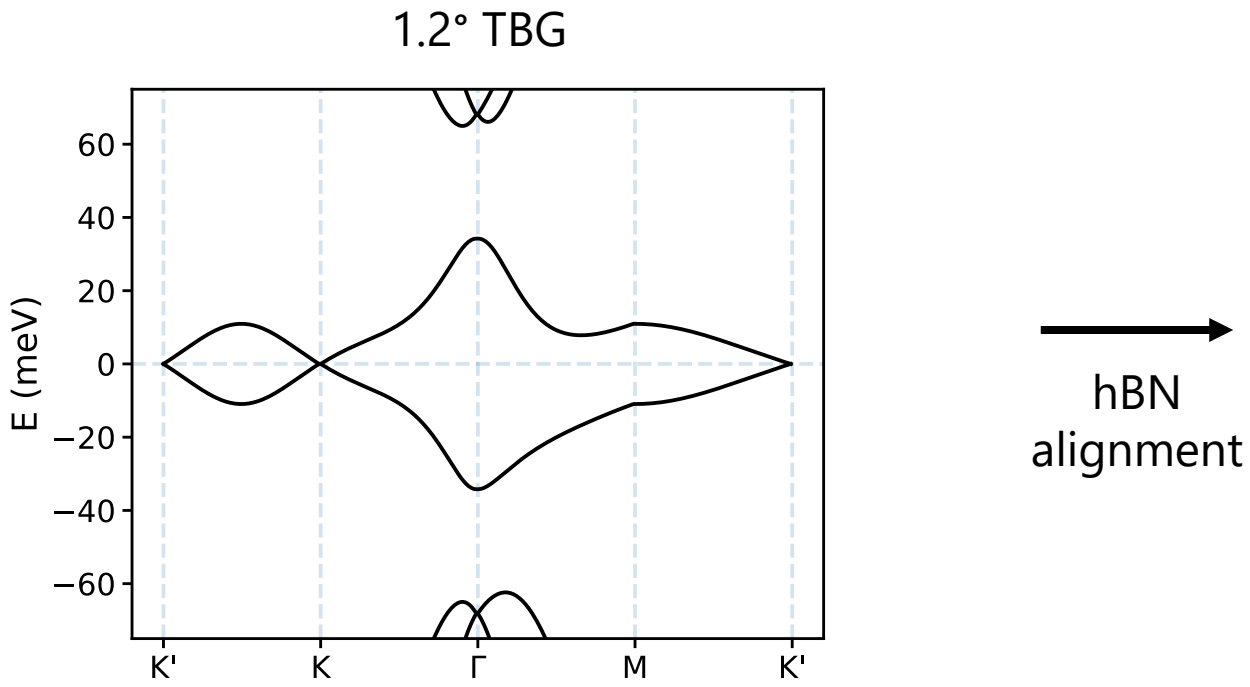

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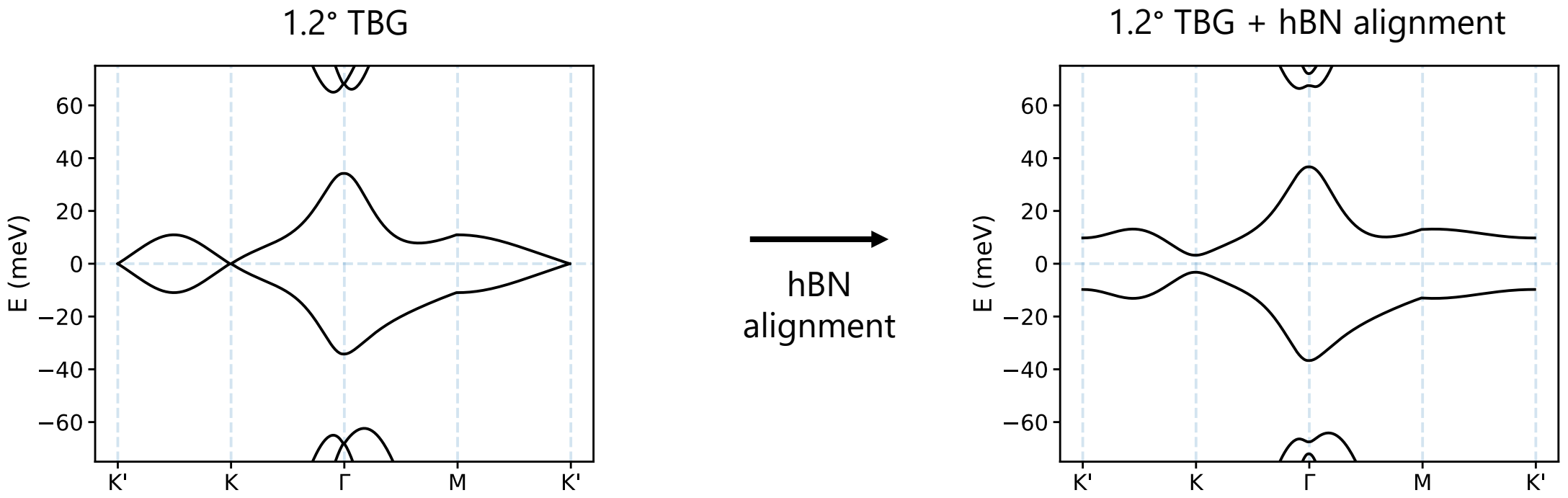

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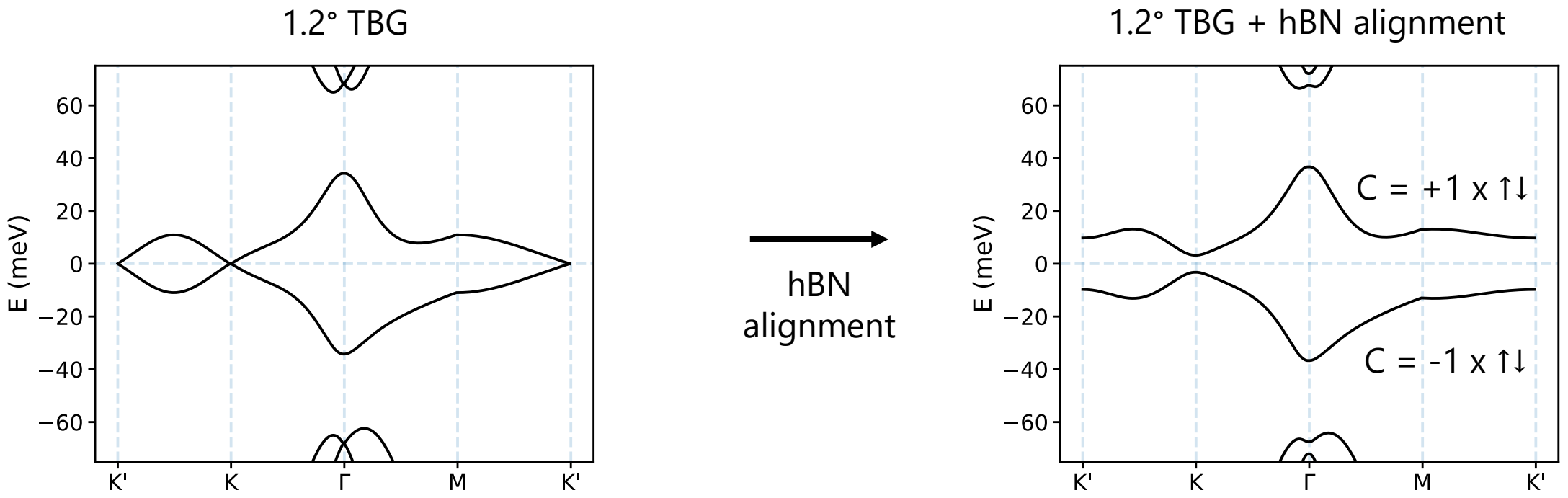

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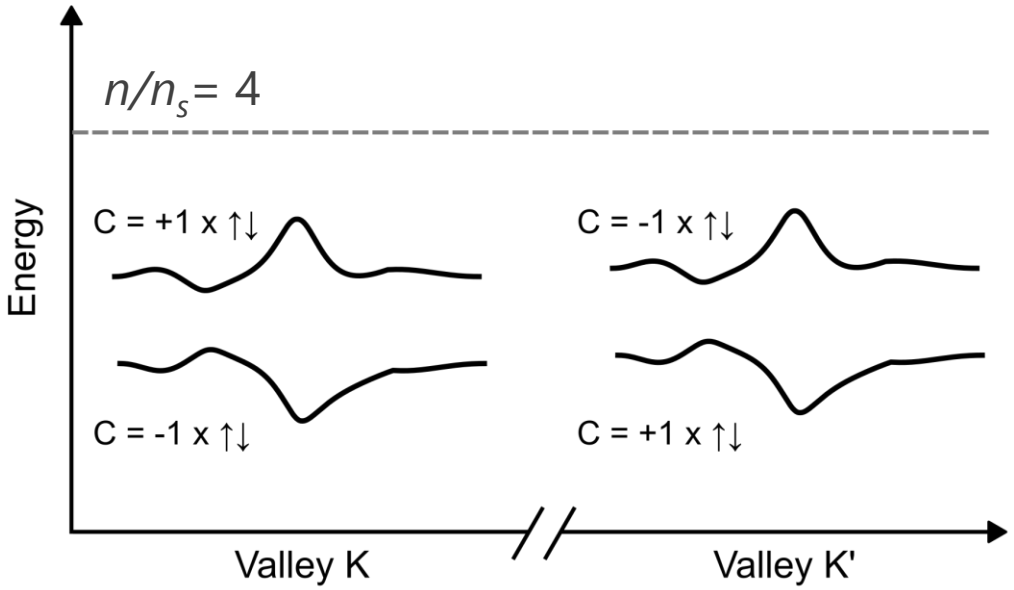

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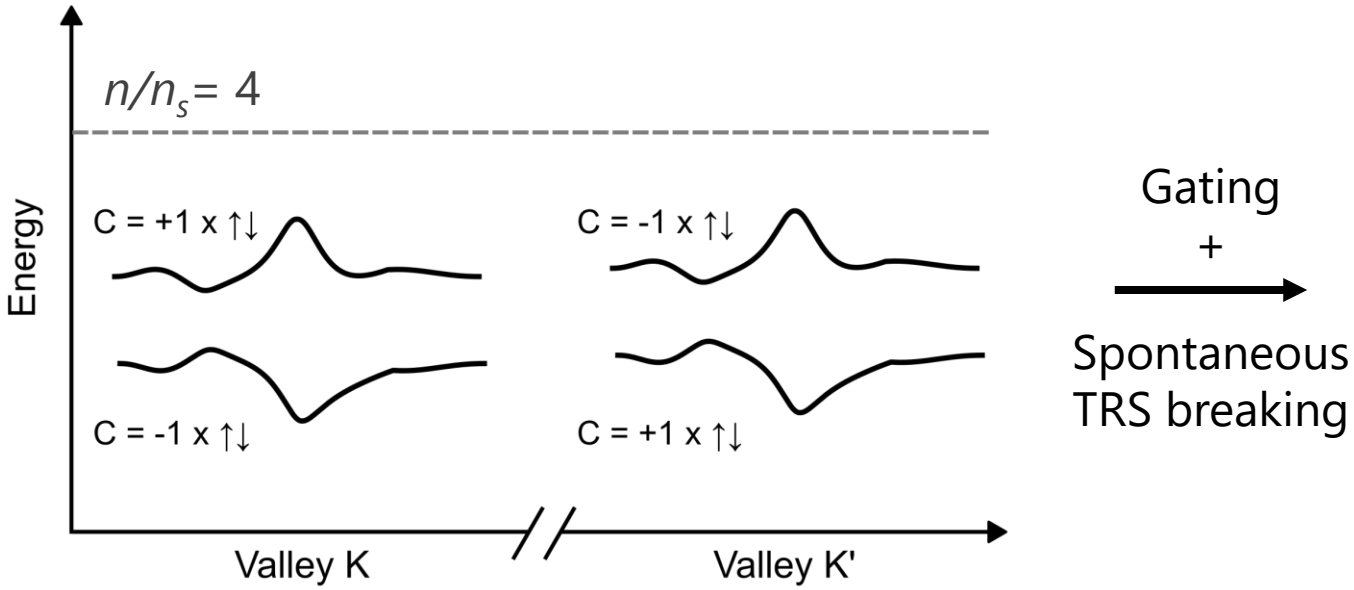

Gating
+

Spontaneous
TRS breaking

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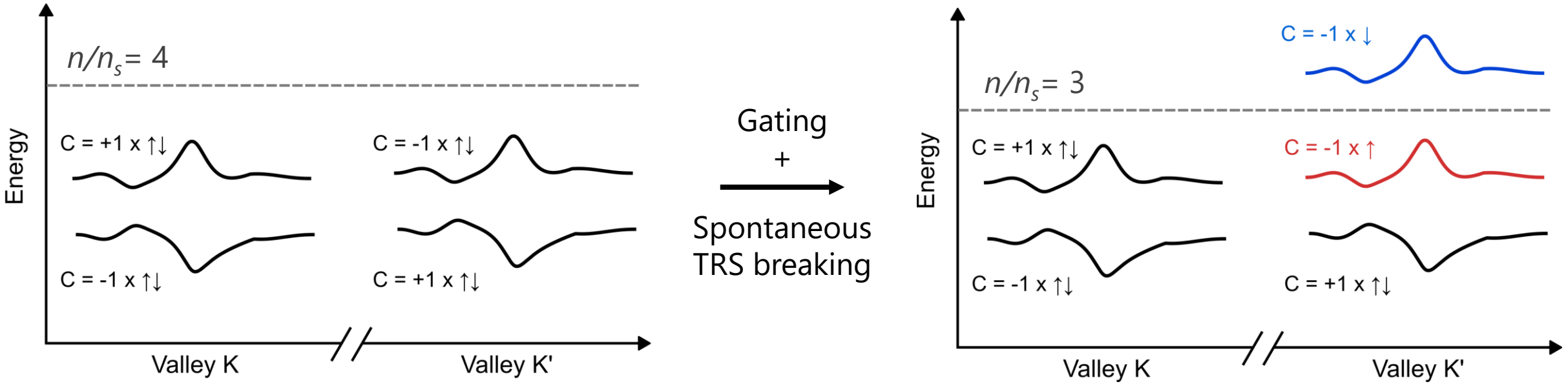

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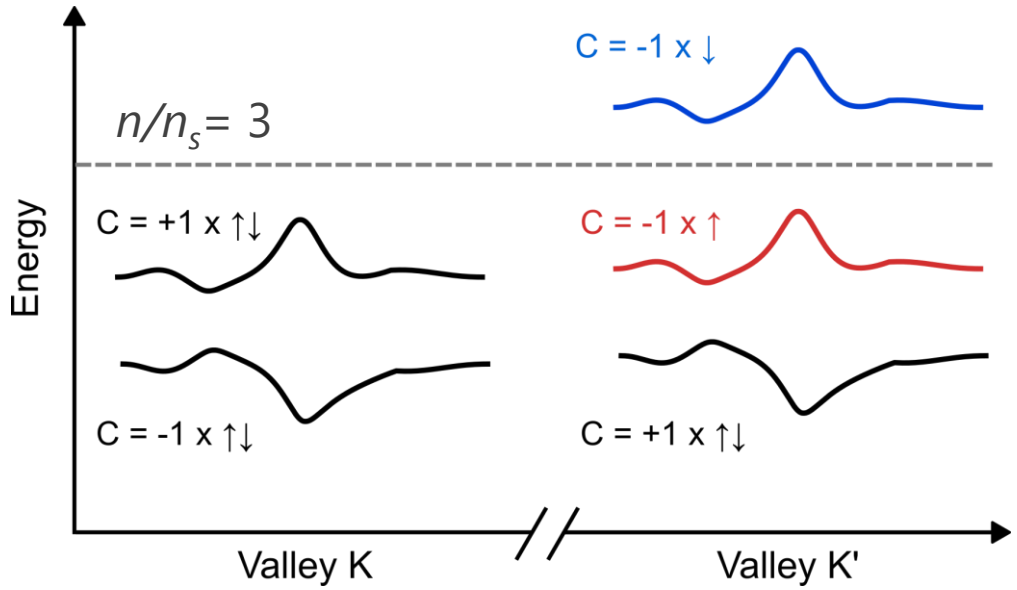
Possible Scenarios and in-plane response

Interaction driven spin/valley polarization



Xie et al. PRL (2020)
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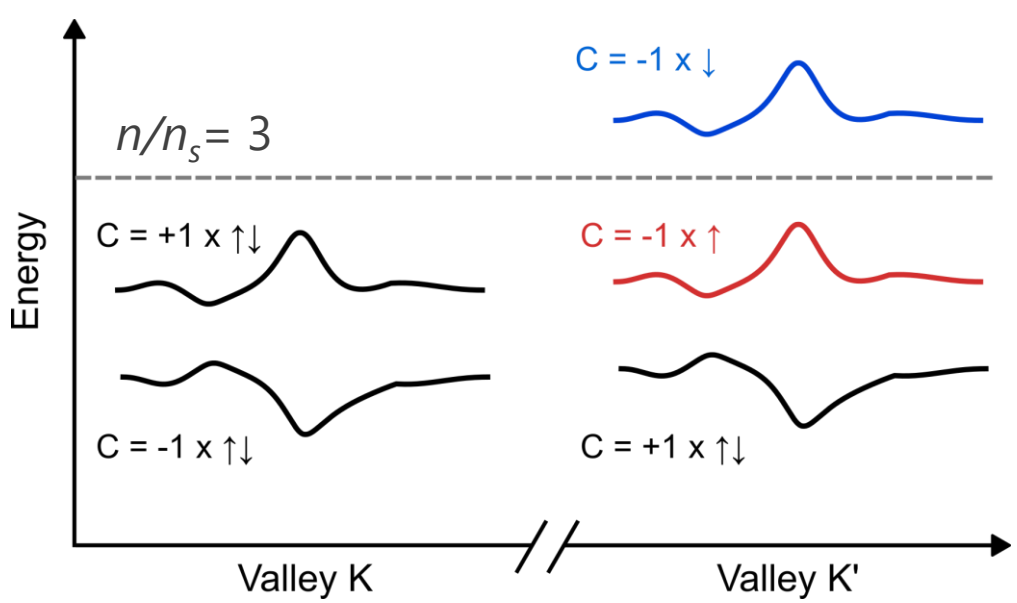

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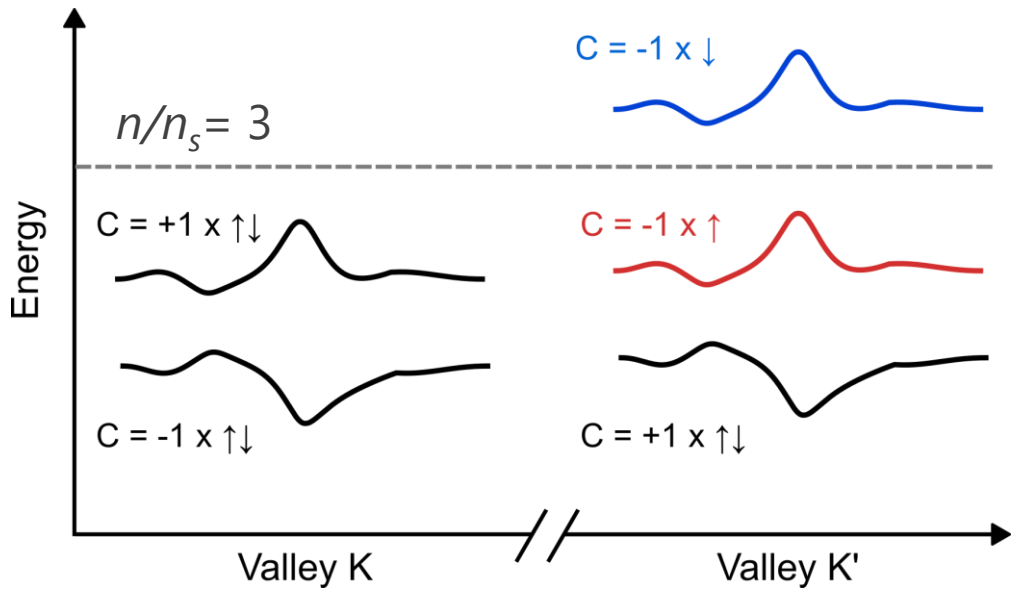

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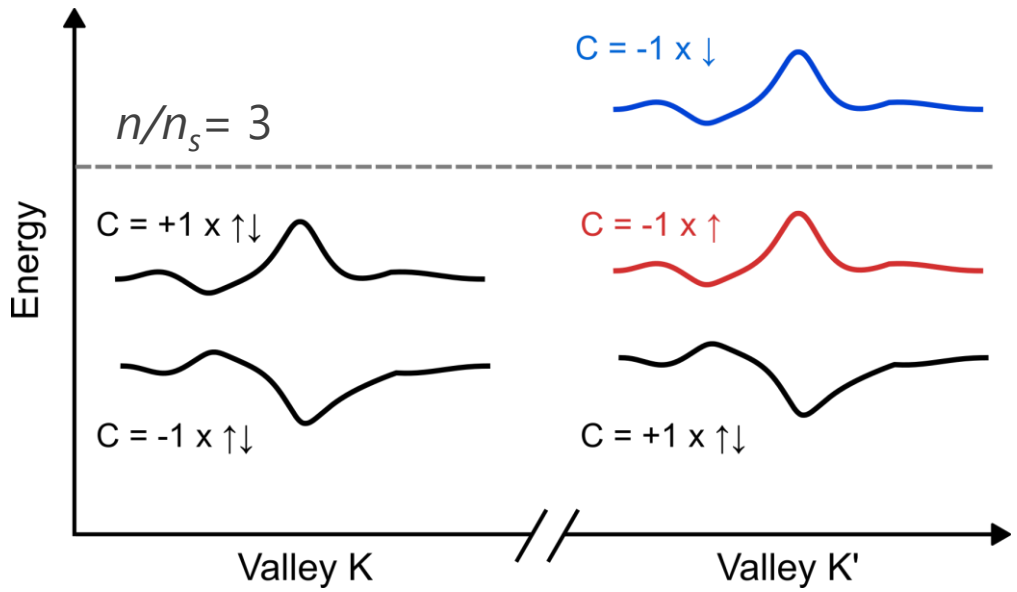

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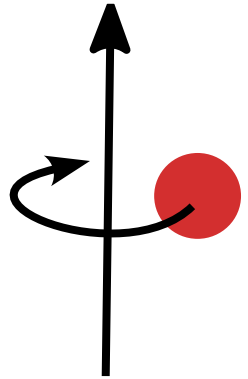

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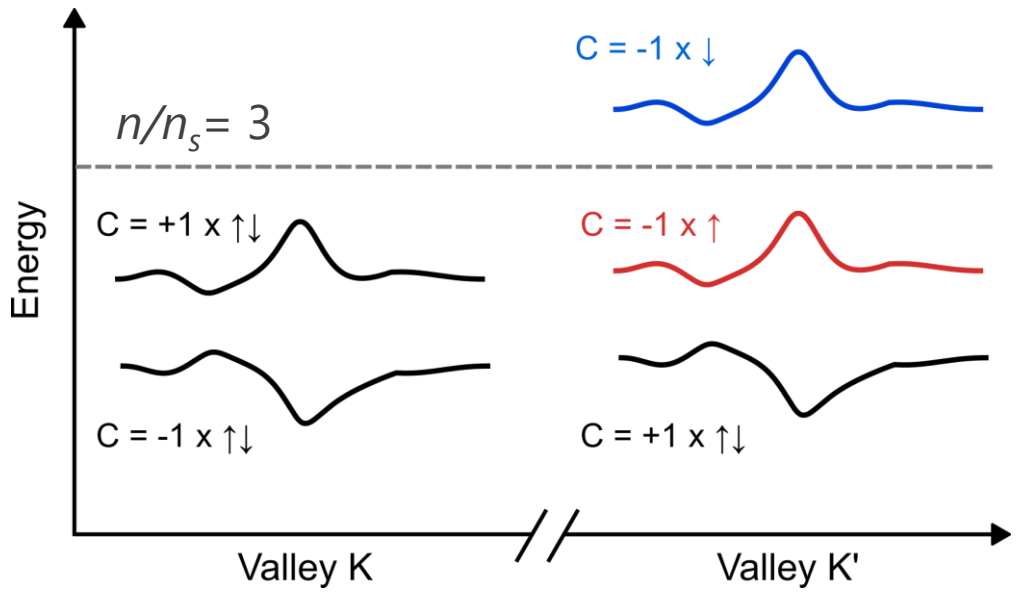

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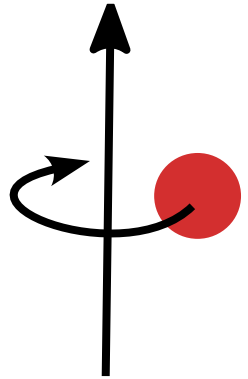

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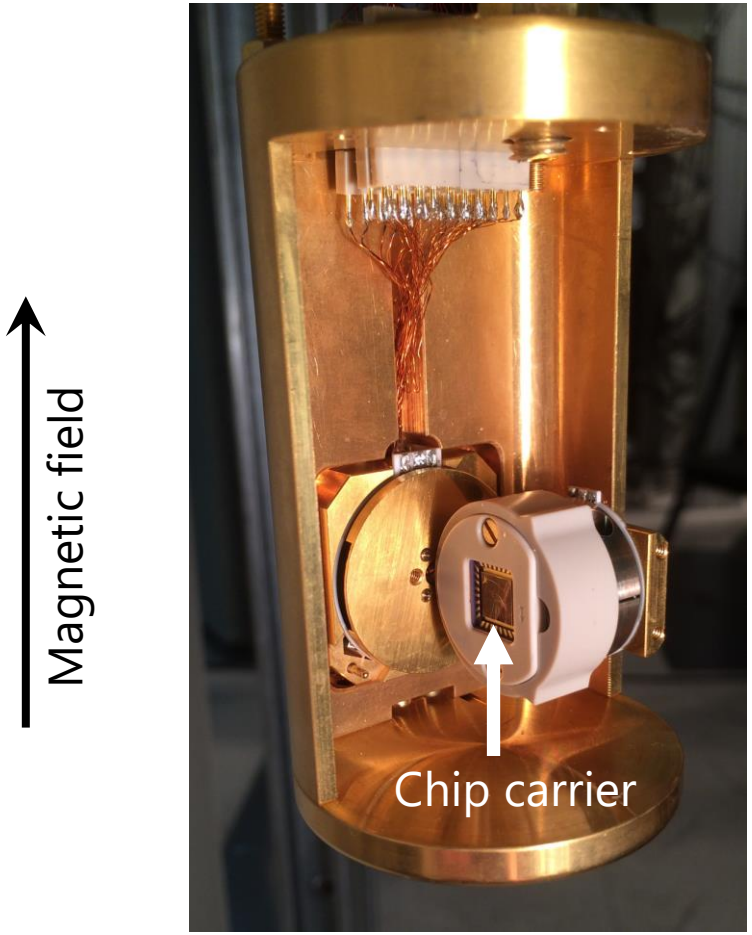
In-plane field can couple to valley!

Probing Nature of Magnetism



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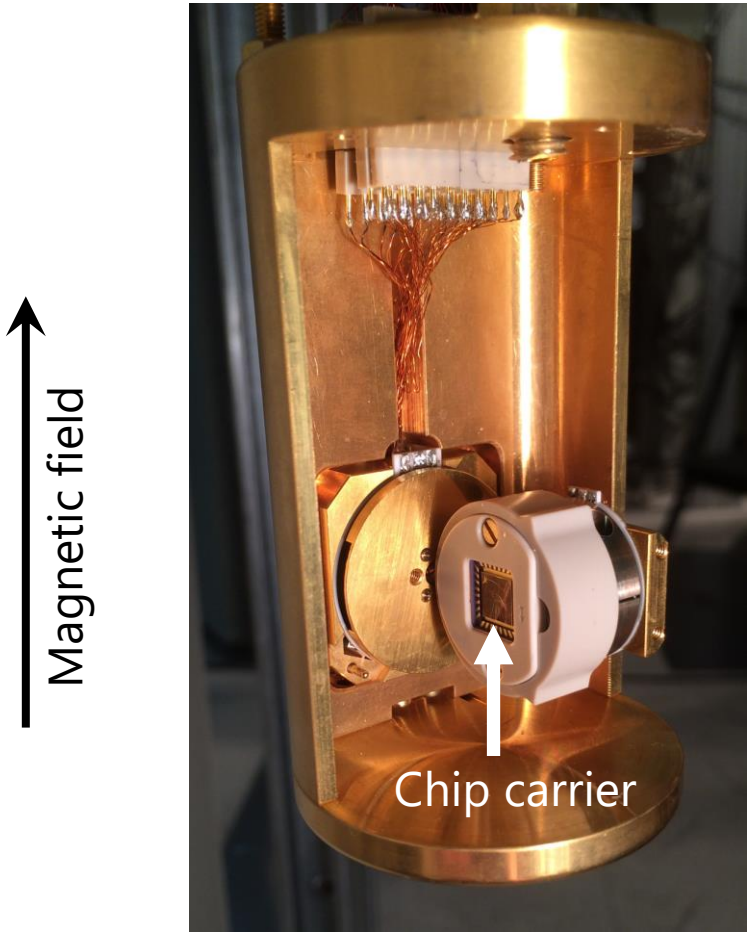

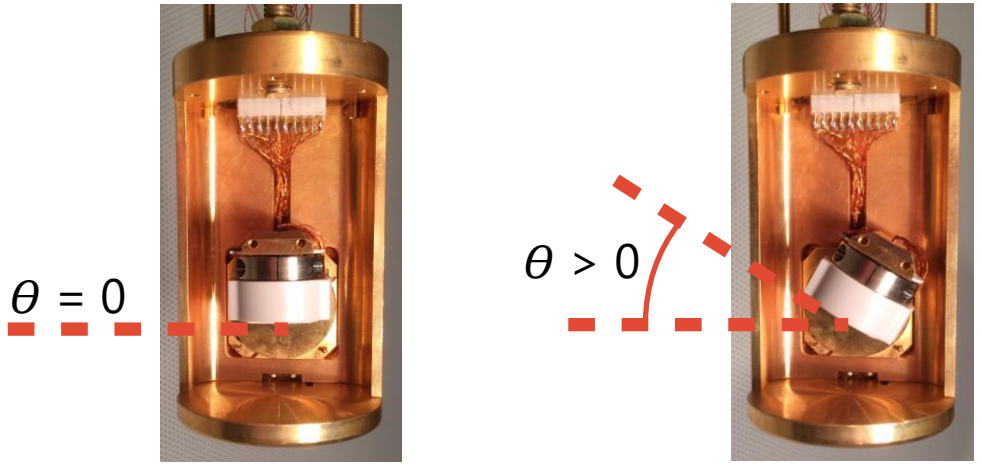

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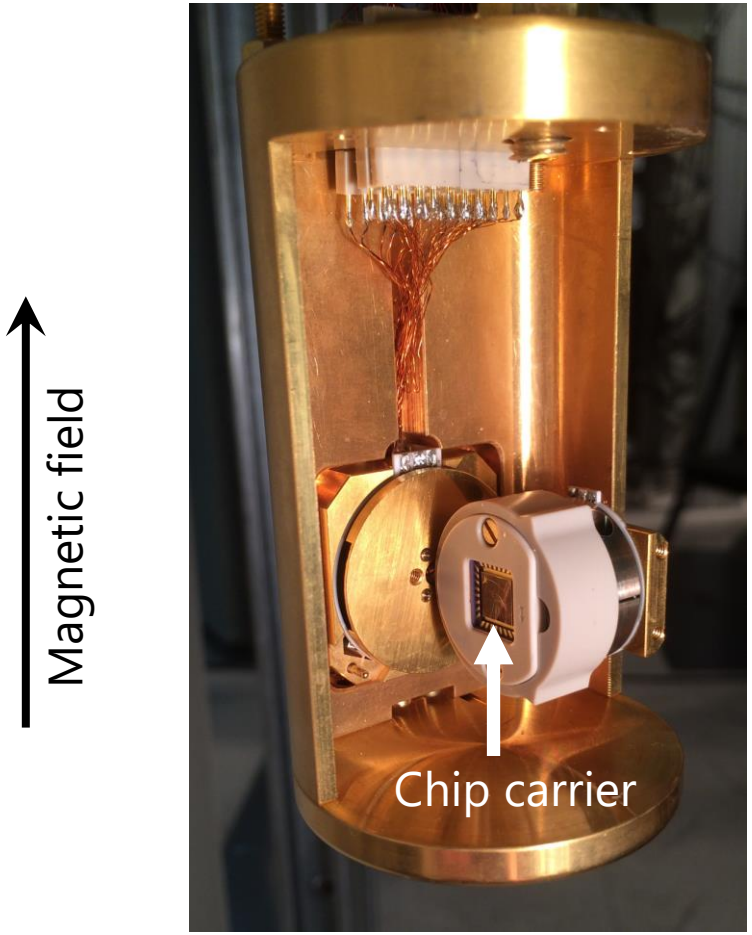
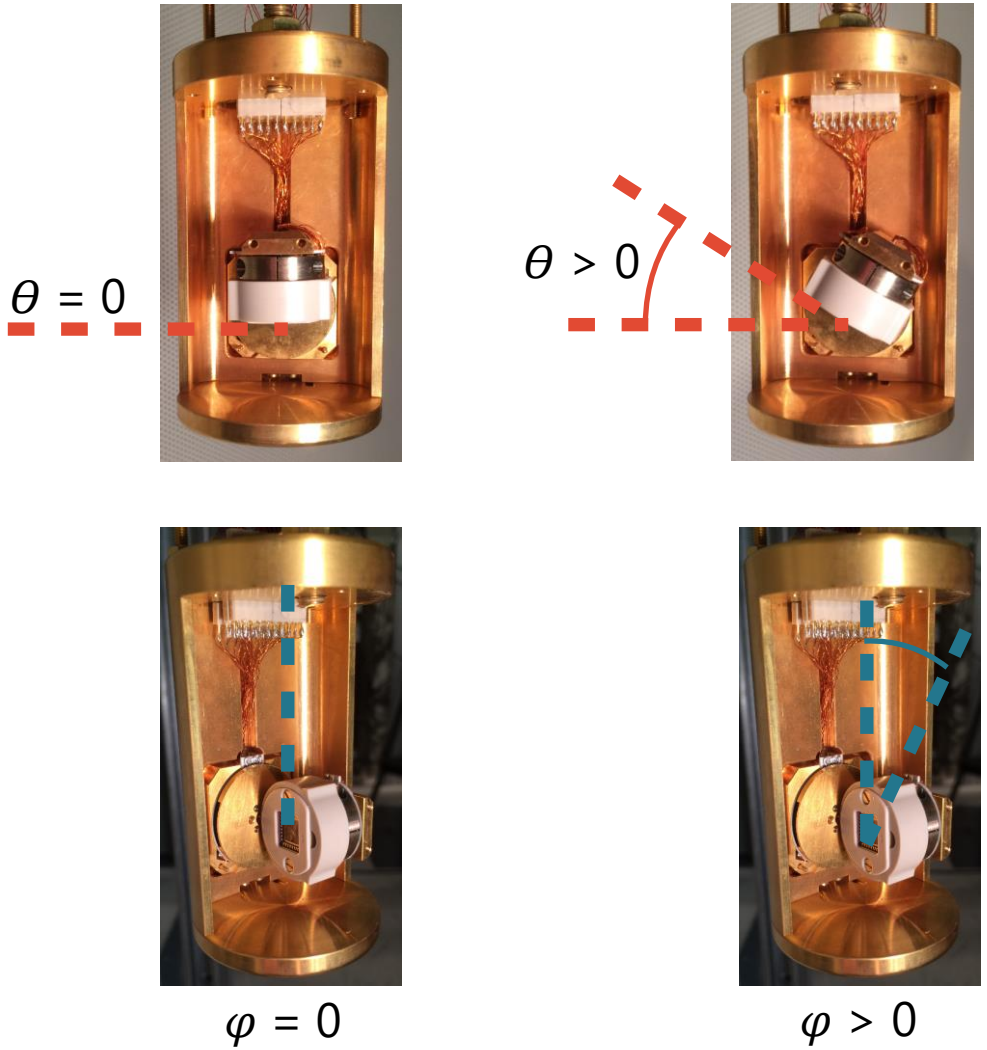
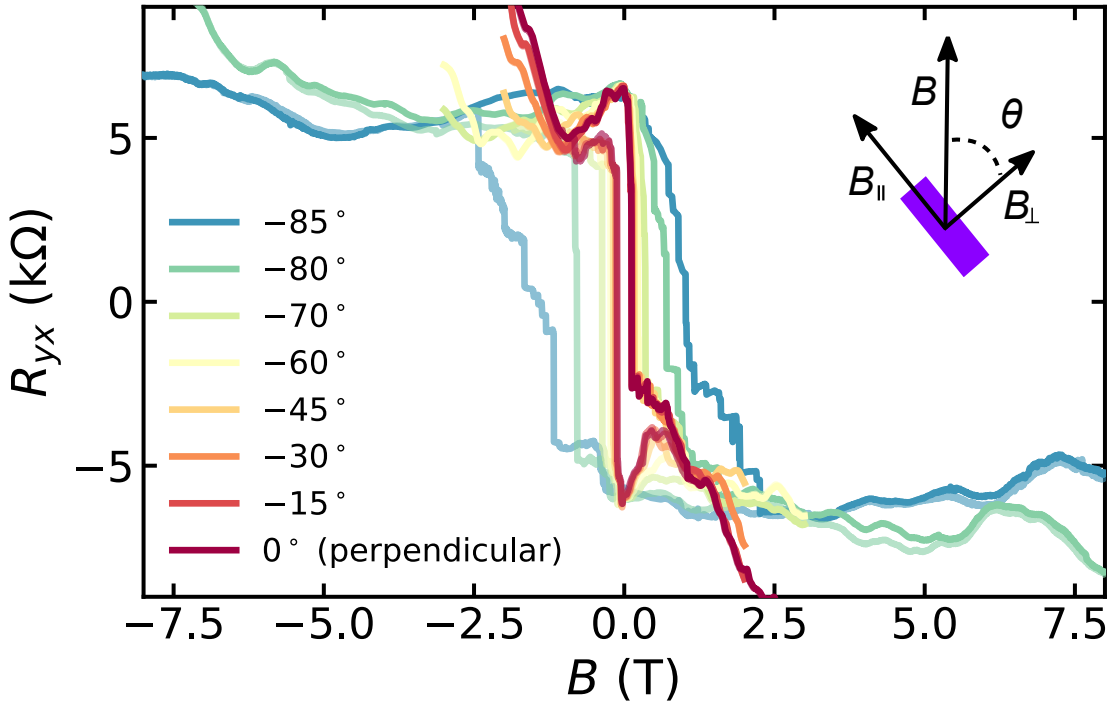




Hysteresis loops in tilted field



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

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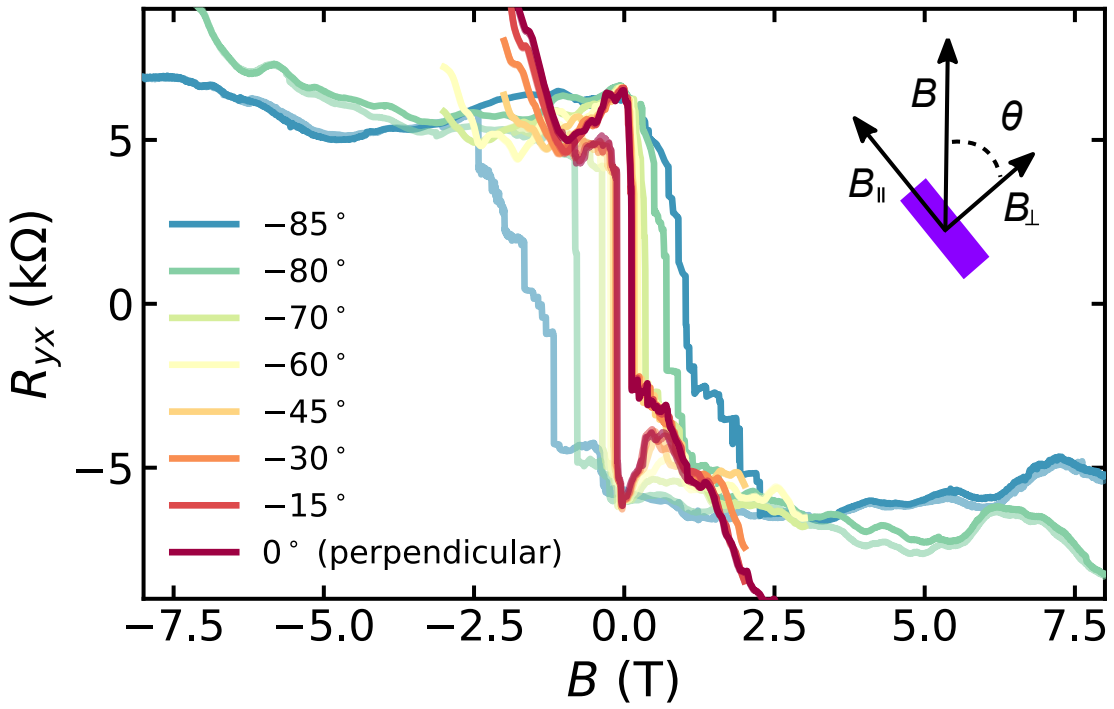

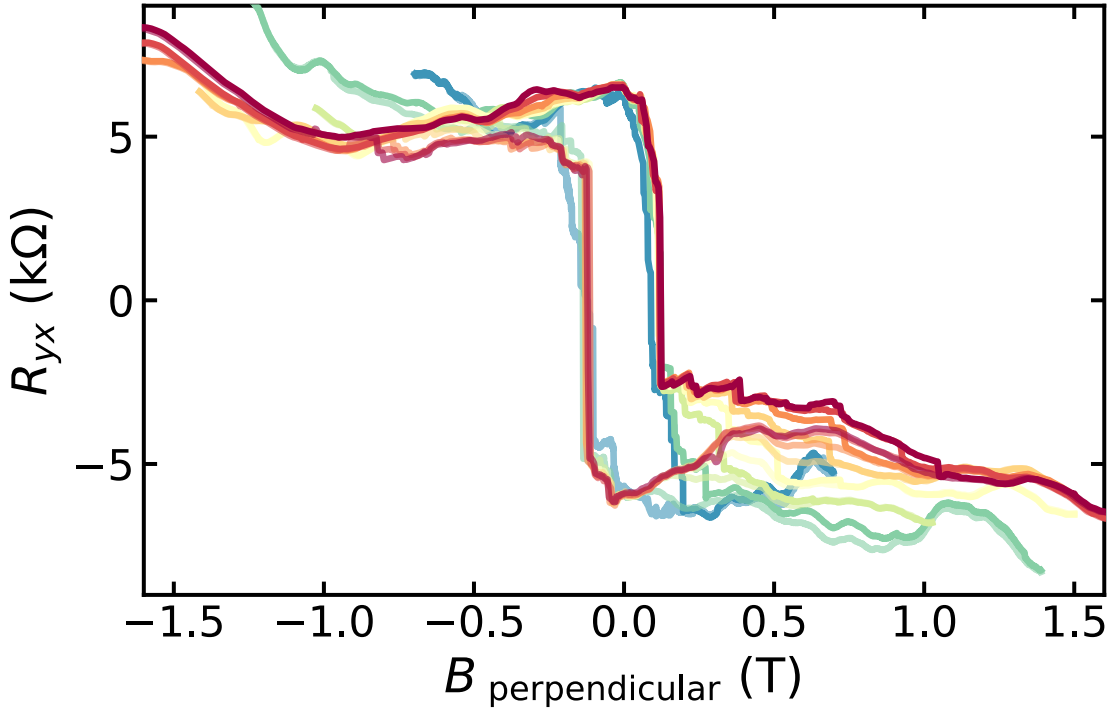

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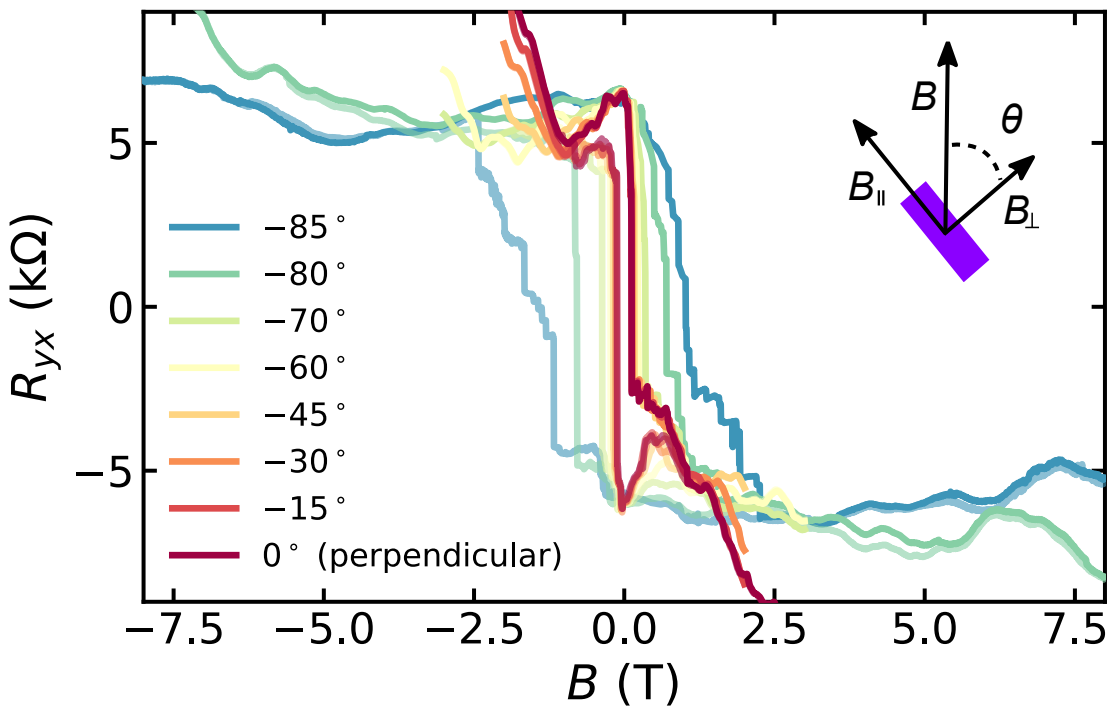

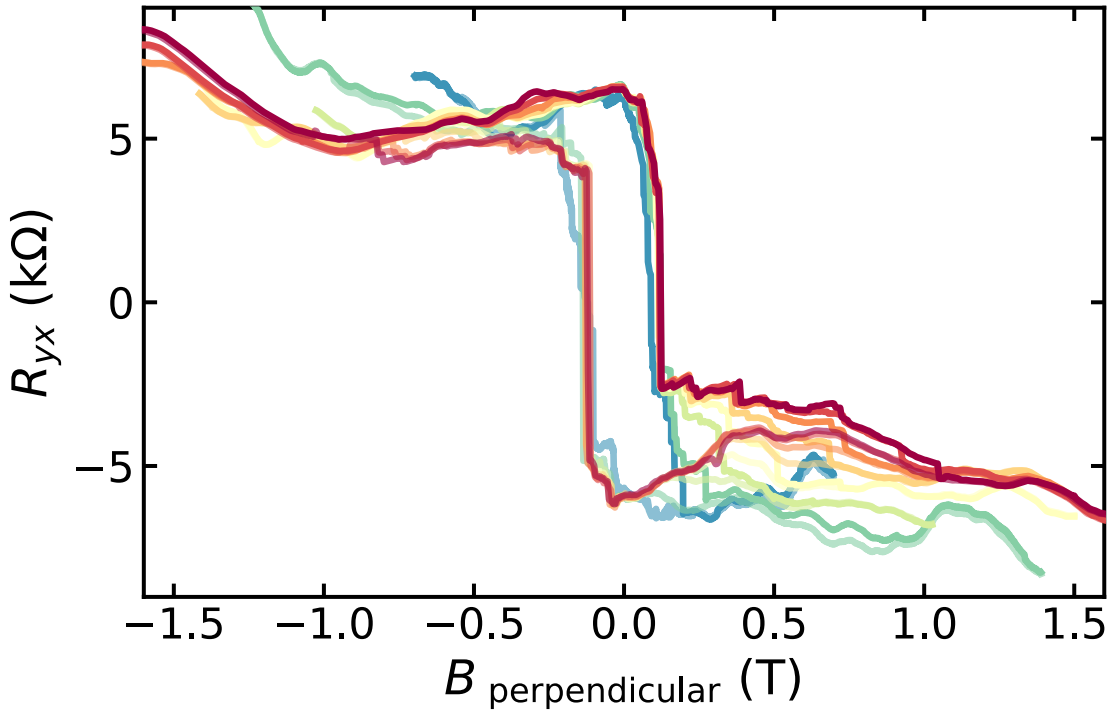

Data collapses when plotted vs B perpendicular!
Orbital Ferromagnet!

Similar conclusion: Tschirhart, et al. Science (2021)
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Quantum anomalous Hall in TBG

Serlin, et al. Science (2020)
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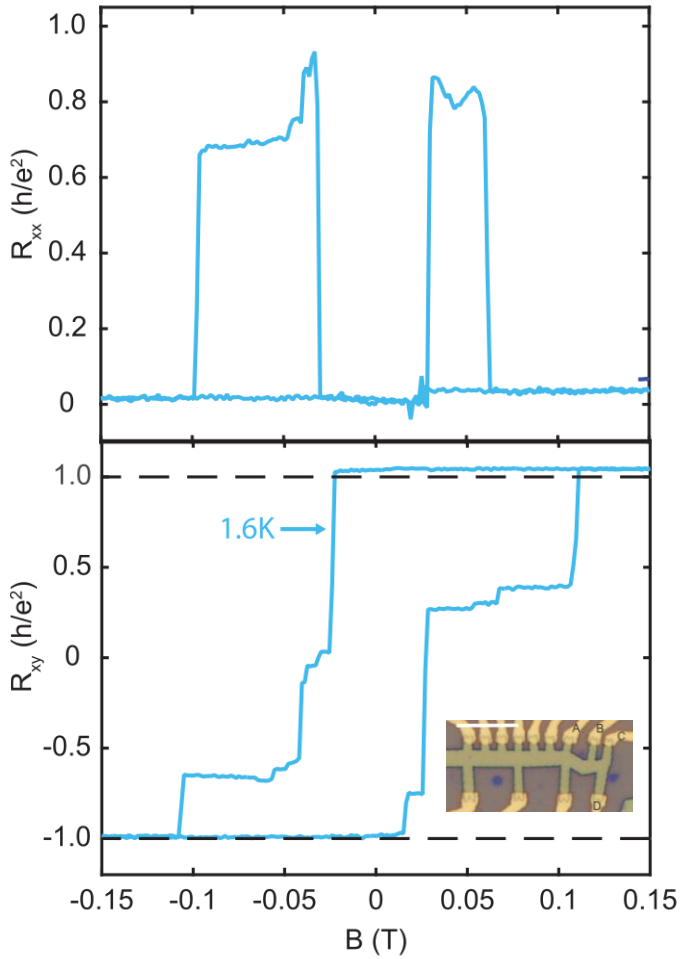

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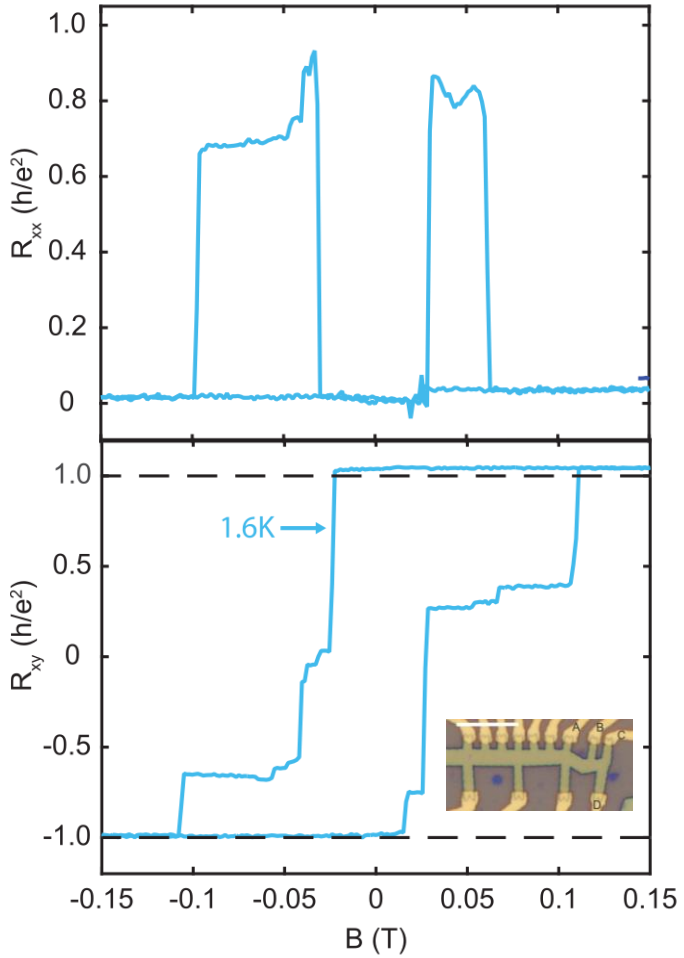

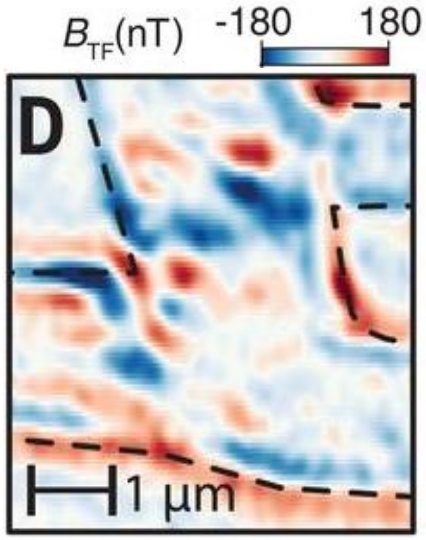

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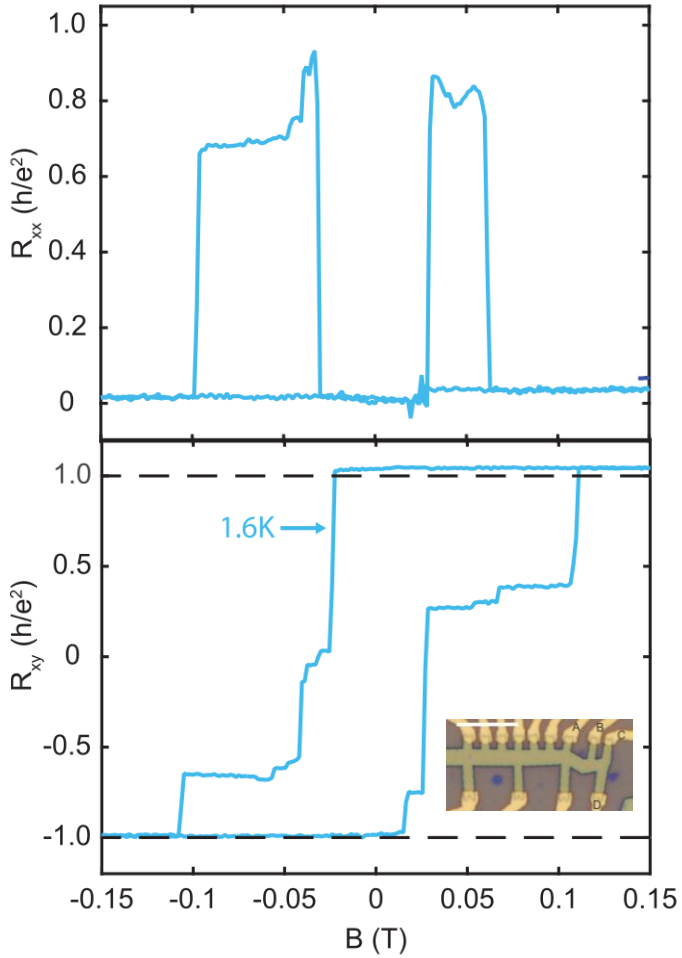

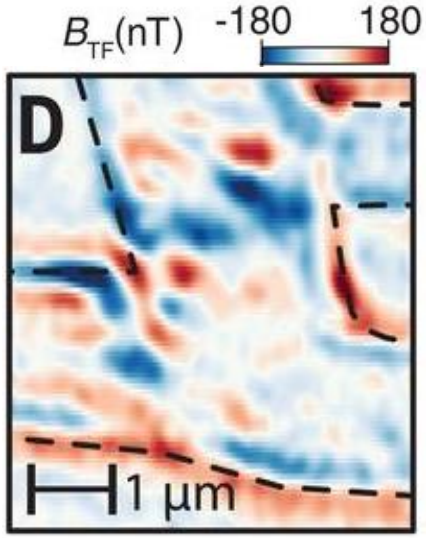

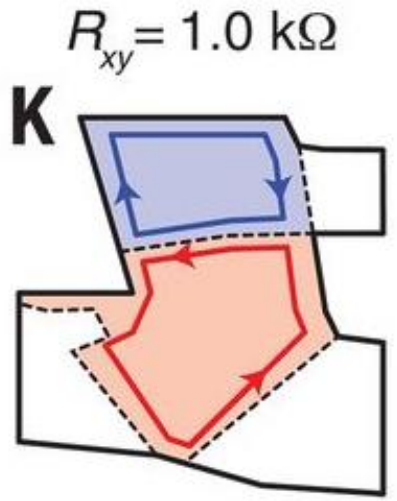

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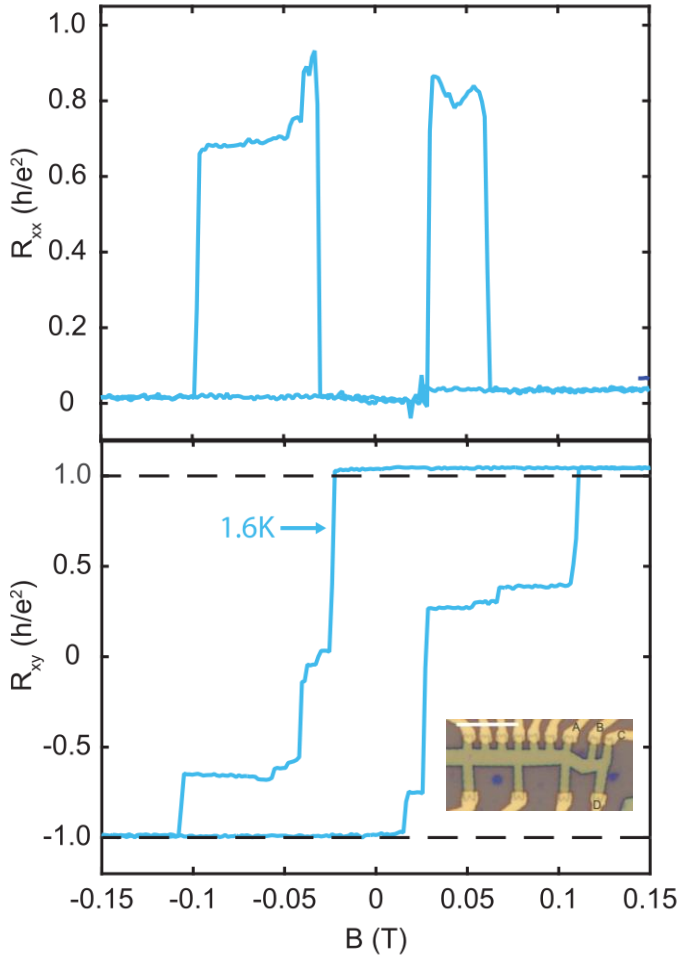

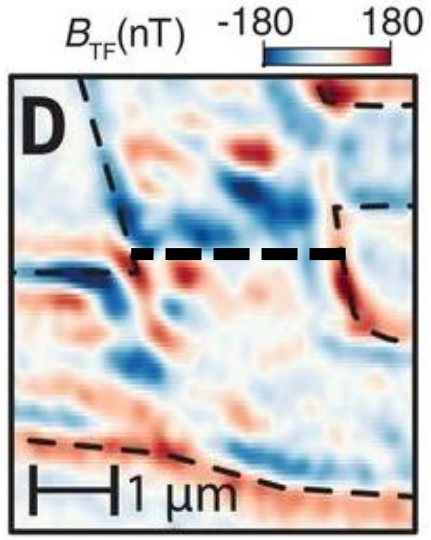

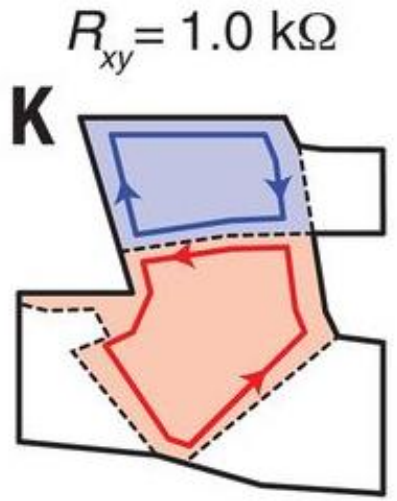

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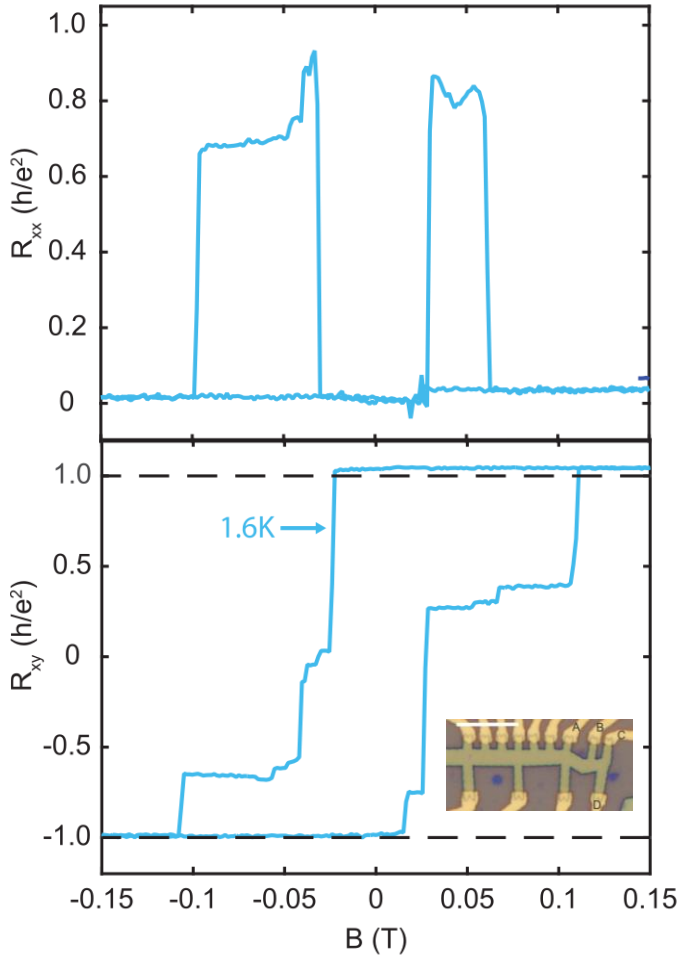

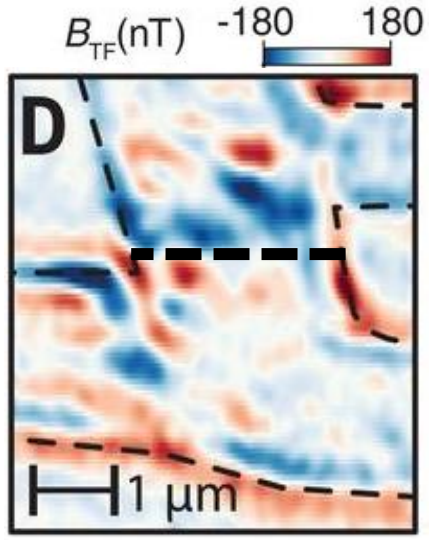

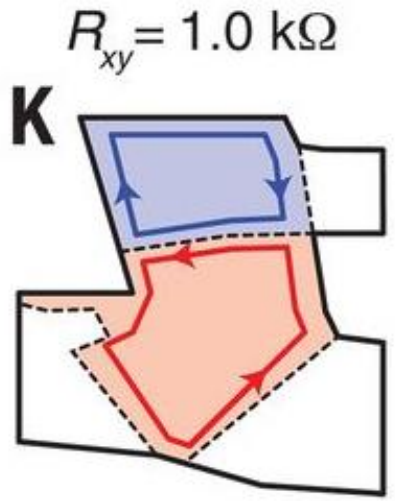

Splotchy magnet yet quantized?

Serlin, et al. Science (2020)
Tschirhart, et al. Science (2021)

(Q)AH is quite common in moirés!

MATBG aligned/commensurate with hBN:
A. Sharpe, et al. Science (2019)
M. Serlin, et al. Science (2020)
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P. Stepanov, et al. PRL (2021)
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Rhombohedral graphene
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Twisted bilayer MoTe2 (FCI)
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AB-stacked MoTe2/WSe2:
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Twisted double bilayer graphene:
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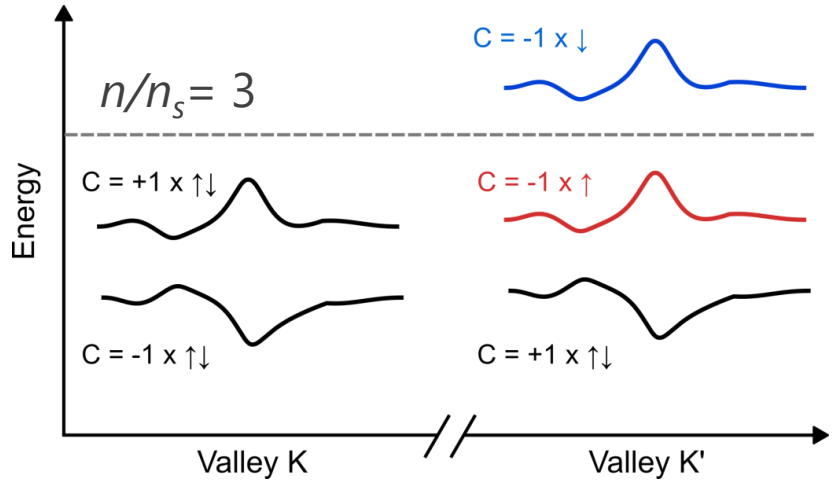
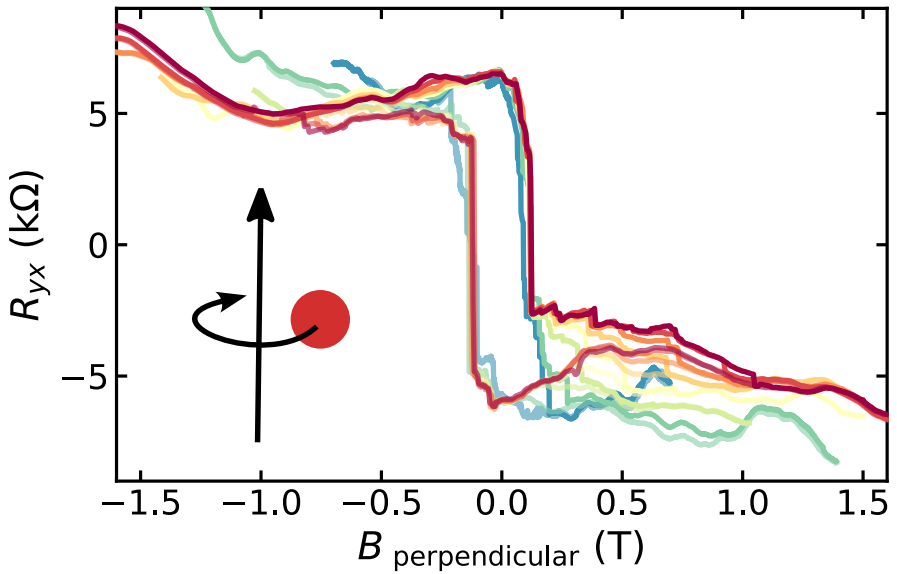
Summary

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 is likely crucial

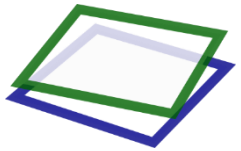
Why is this orbital magnet so hard to reproduce in TBG?
- Magnetism appears splotchy
- (Q)AH appears to be quite ubiquitous in moirés
- Shouldn't the hBN/graphene form a moiré?
- How should we model this?

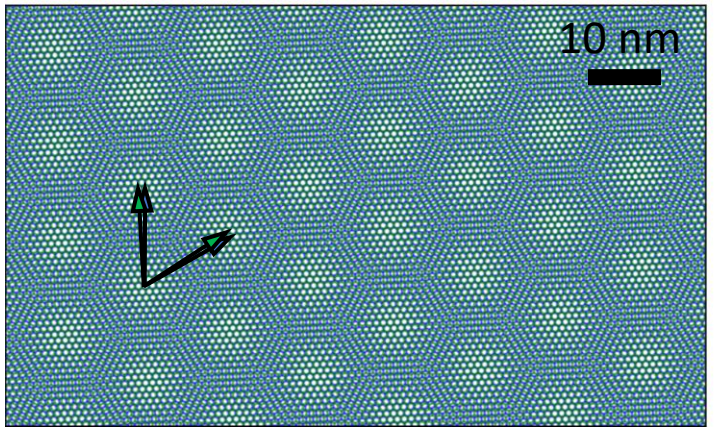




Sharpe, et al. Science (2019)
Sharpe, et al. Nano Lett (2021)

From moiré to multimoiré





Yang, et al. arXiv:2310.12961
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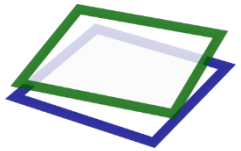

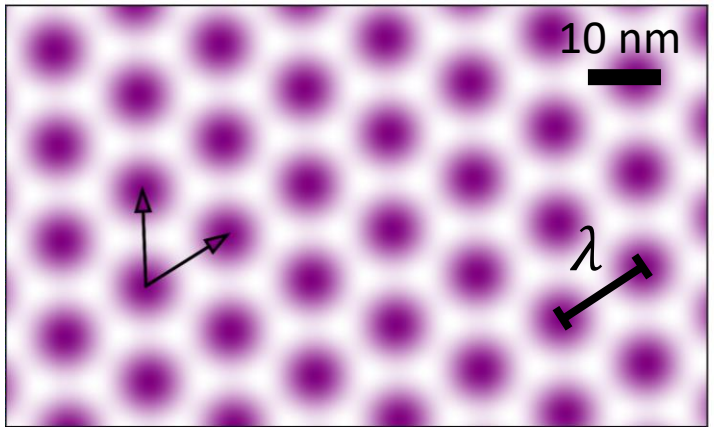

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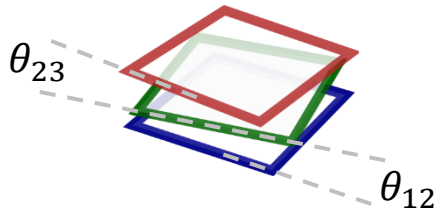

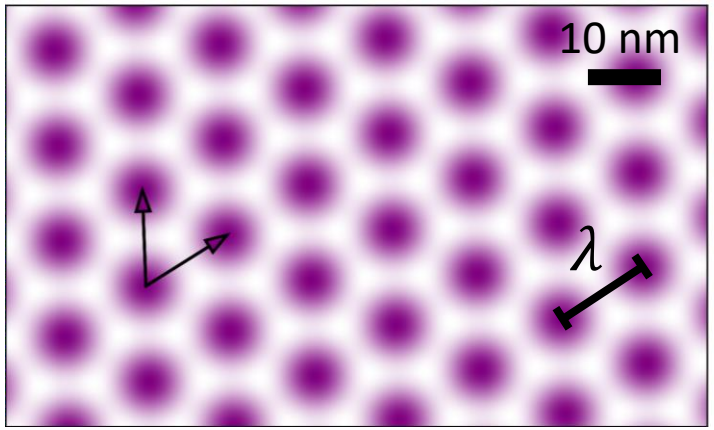

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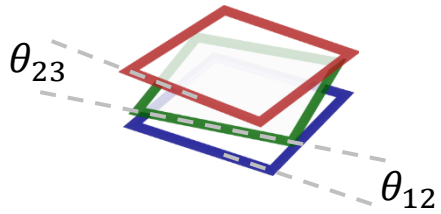

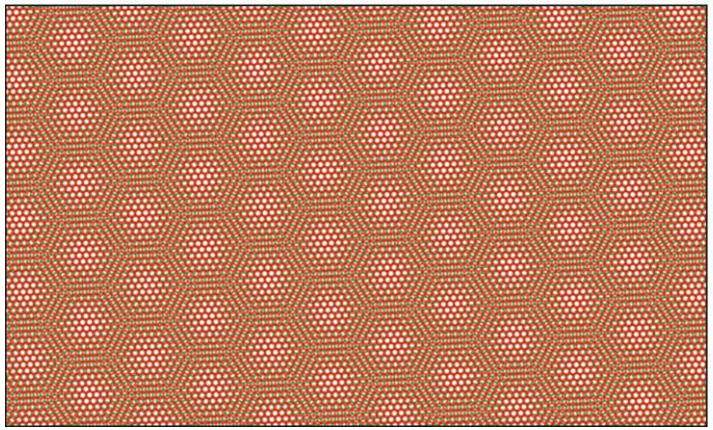

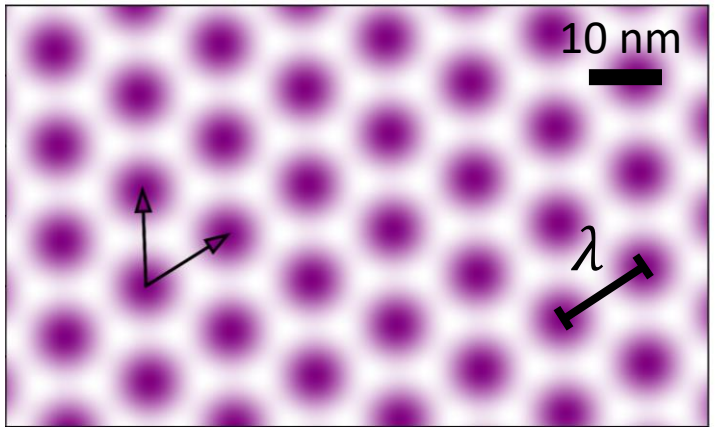

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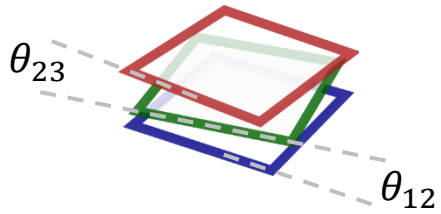

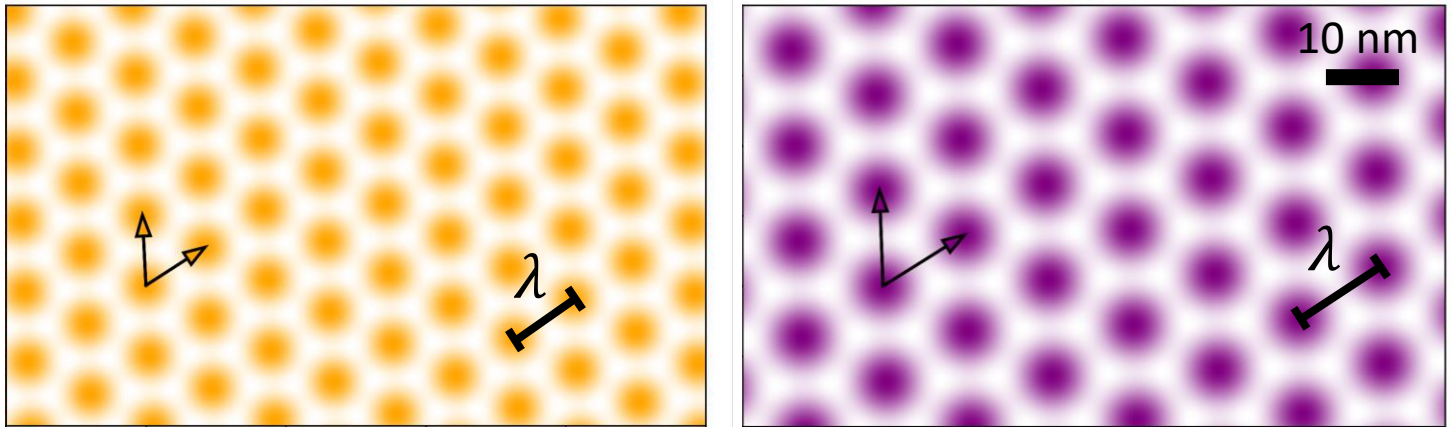

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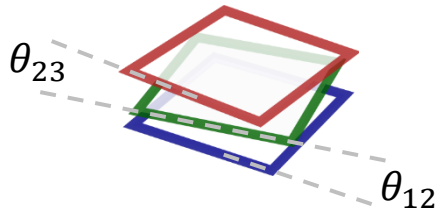

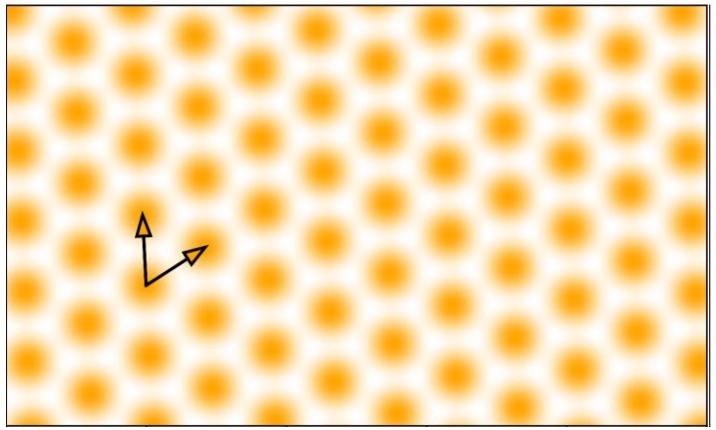

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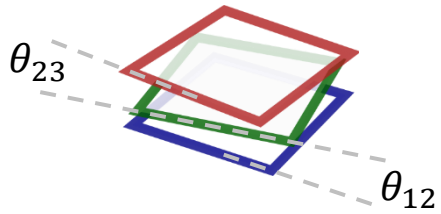

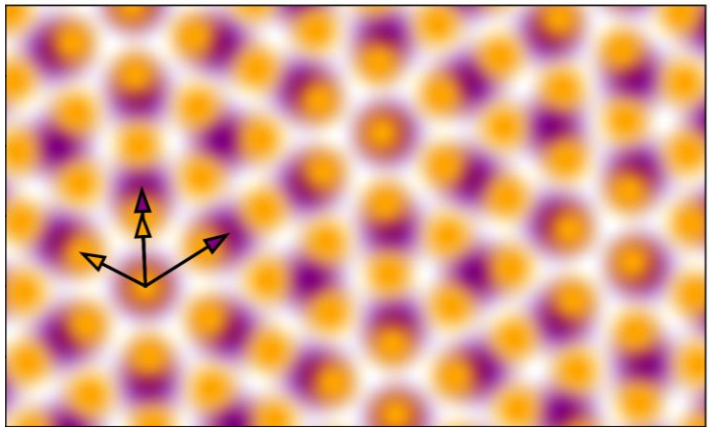

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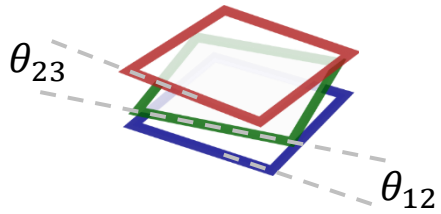

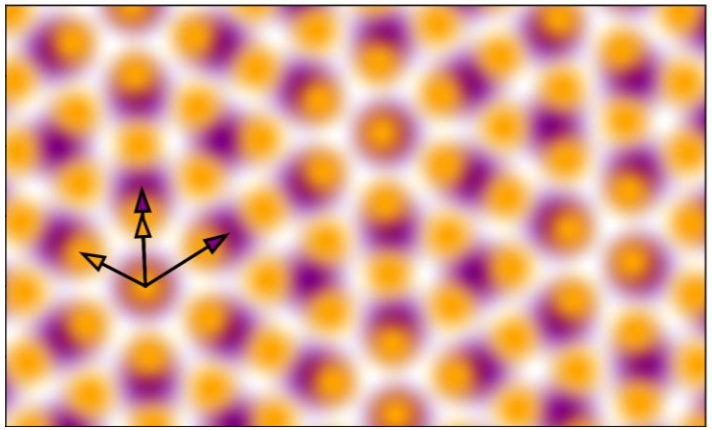

Moiré quasicrystal

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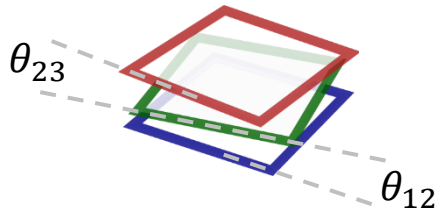

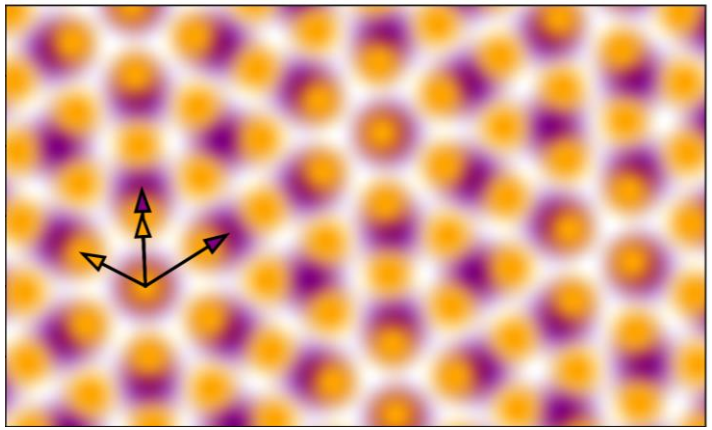

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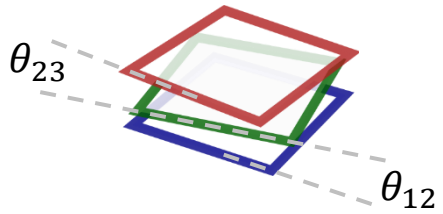

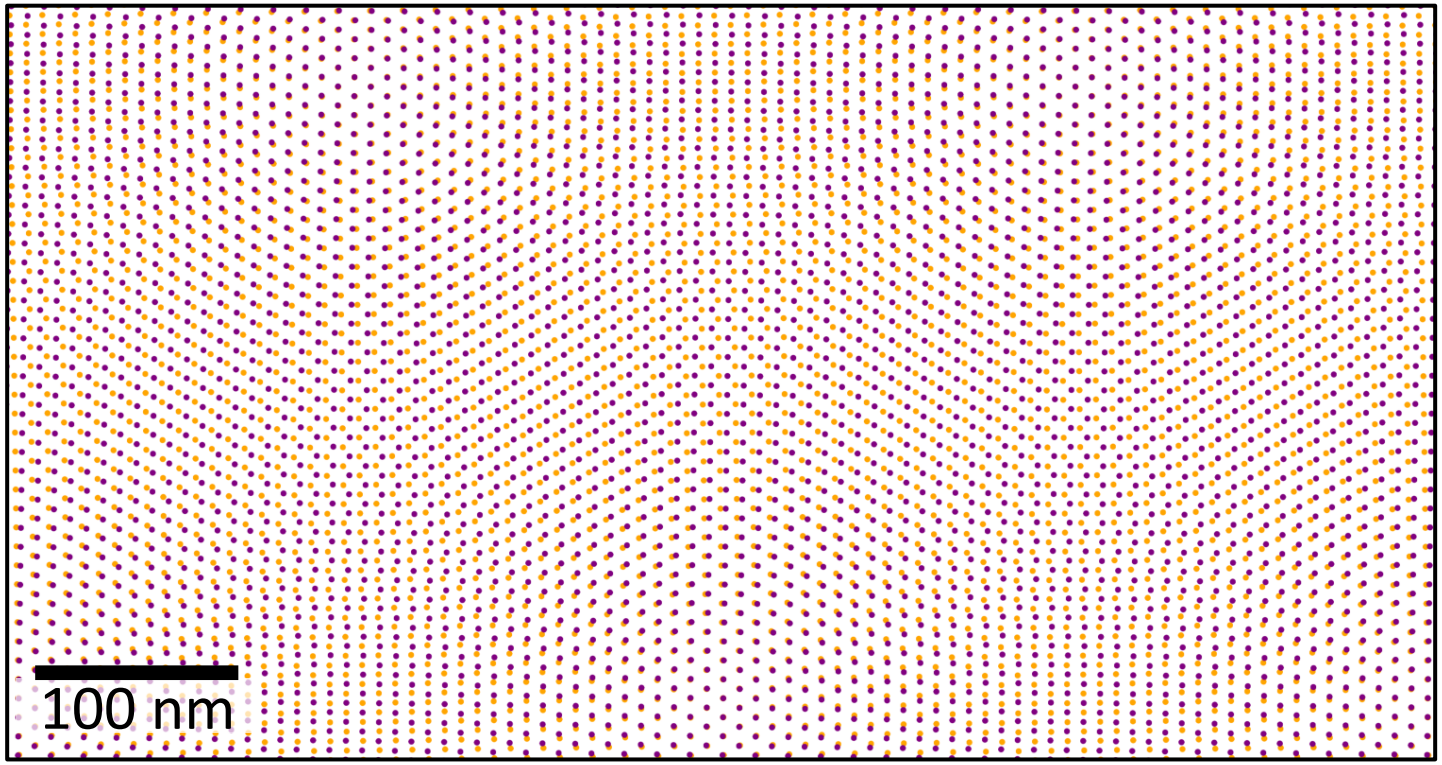

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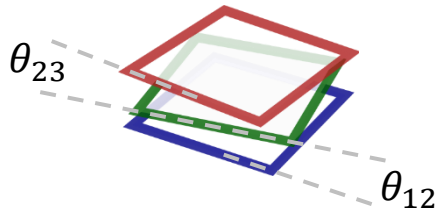
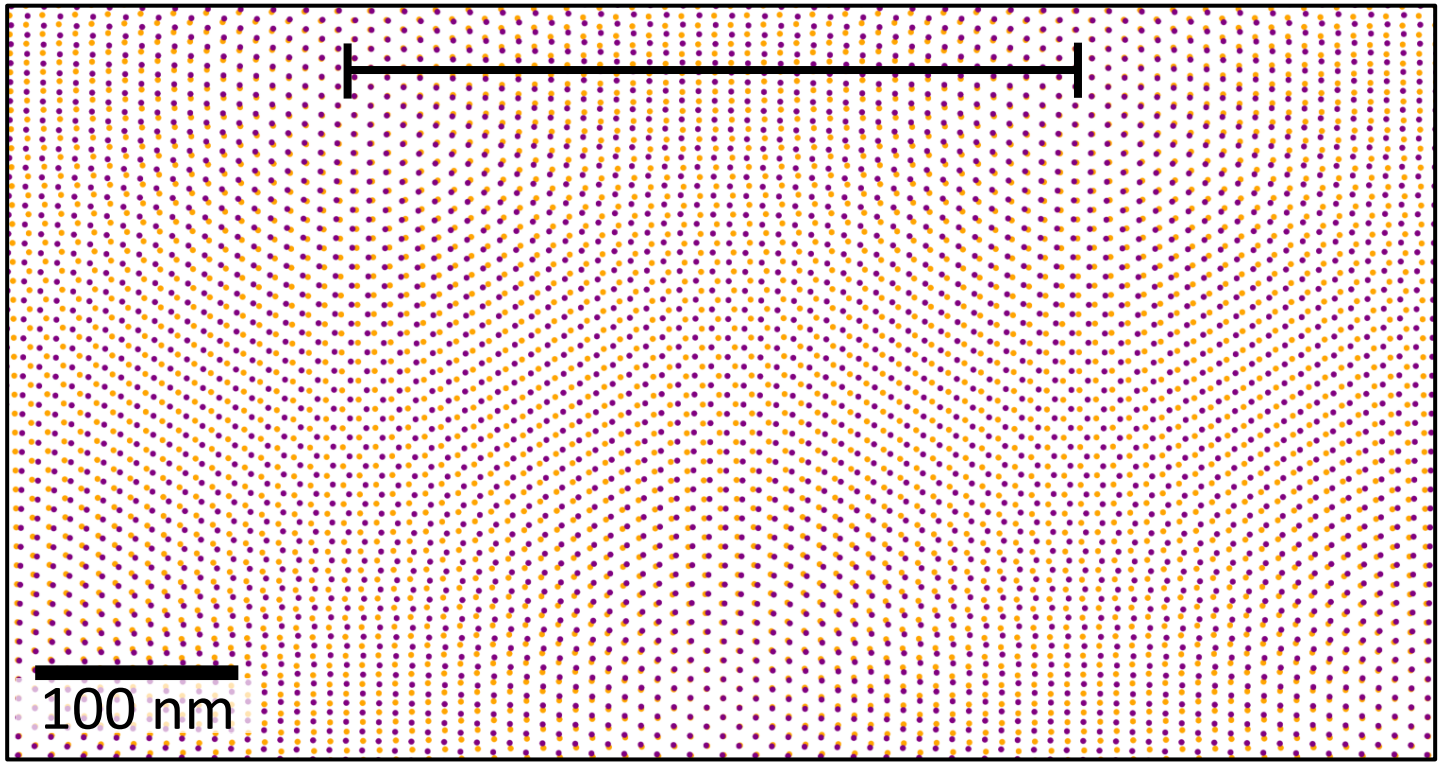


Supermoiré: $\lambda_{sm} = \frac{\lambda_m}{\theta} = \frac{a}{\theta^2}$



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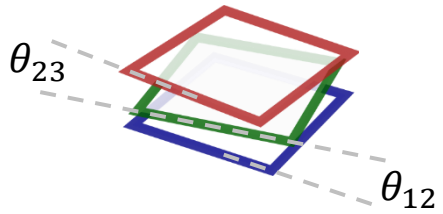

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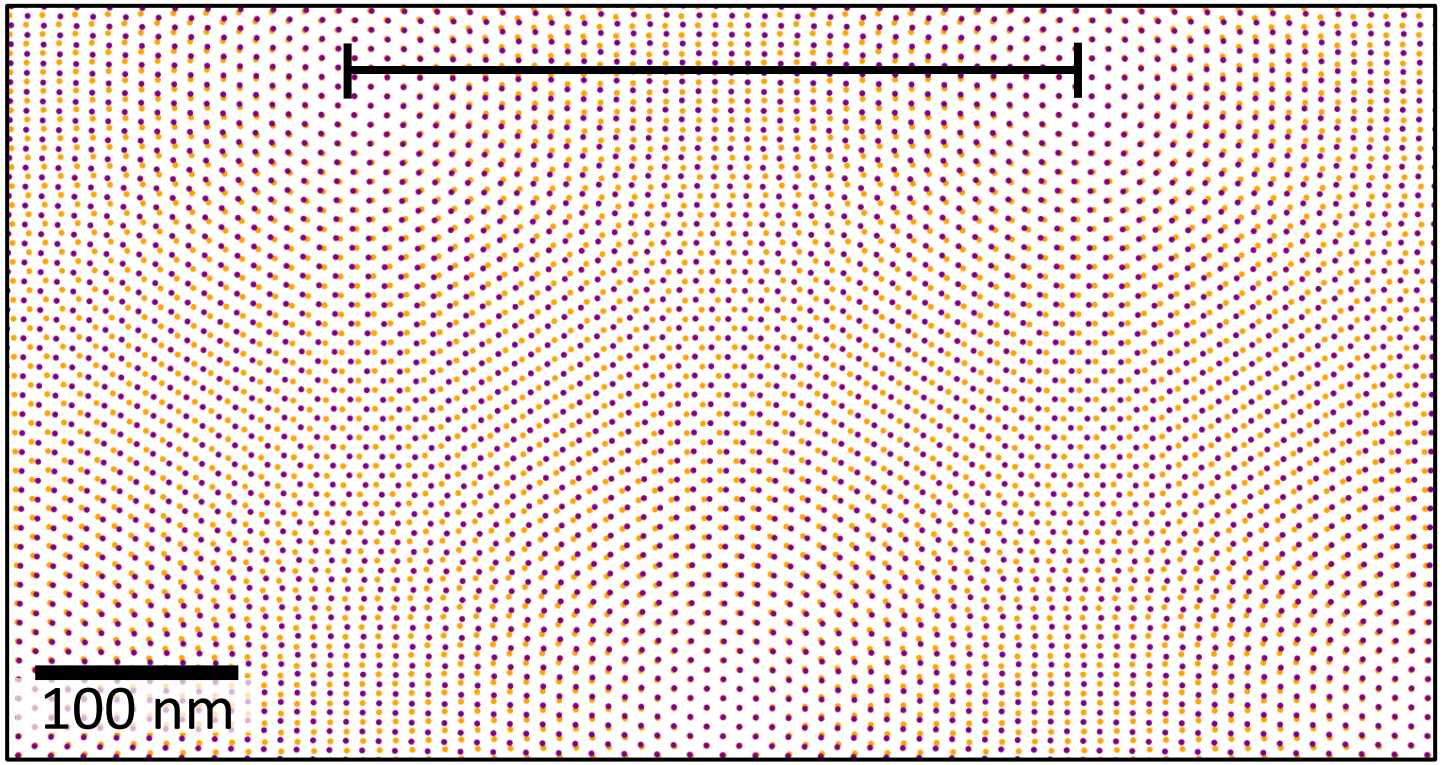

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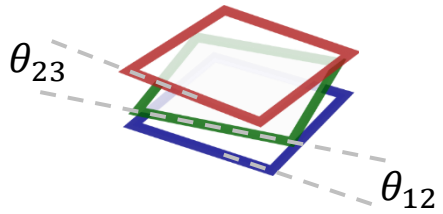

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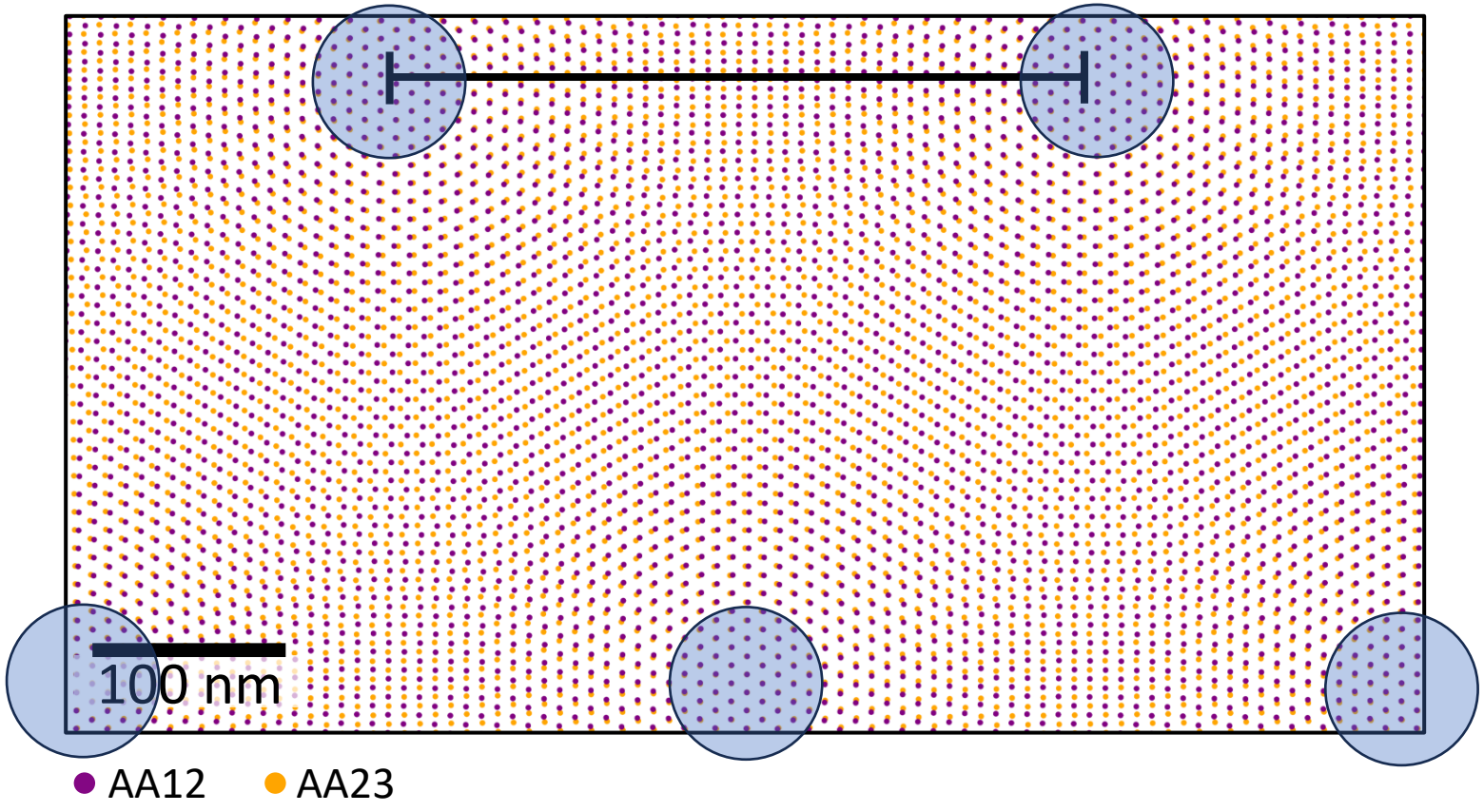

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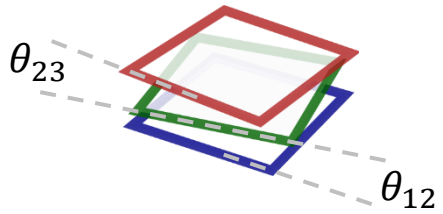

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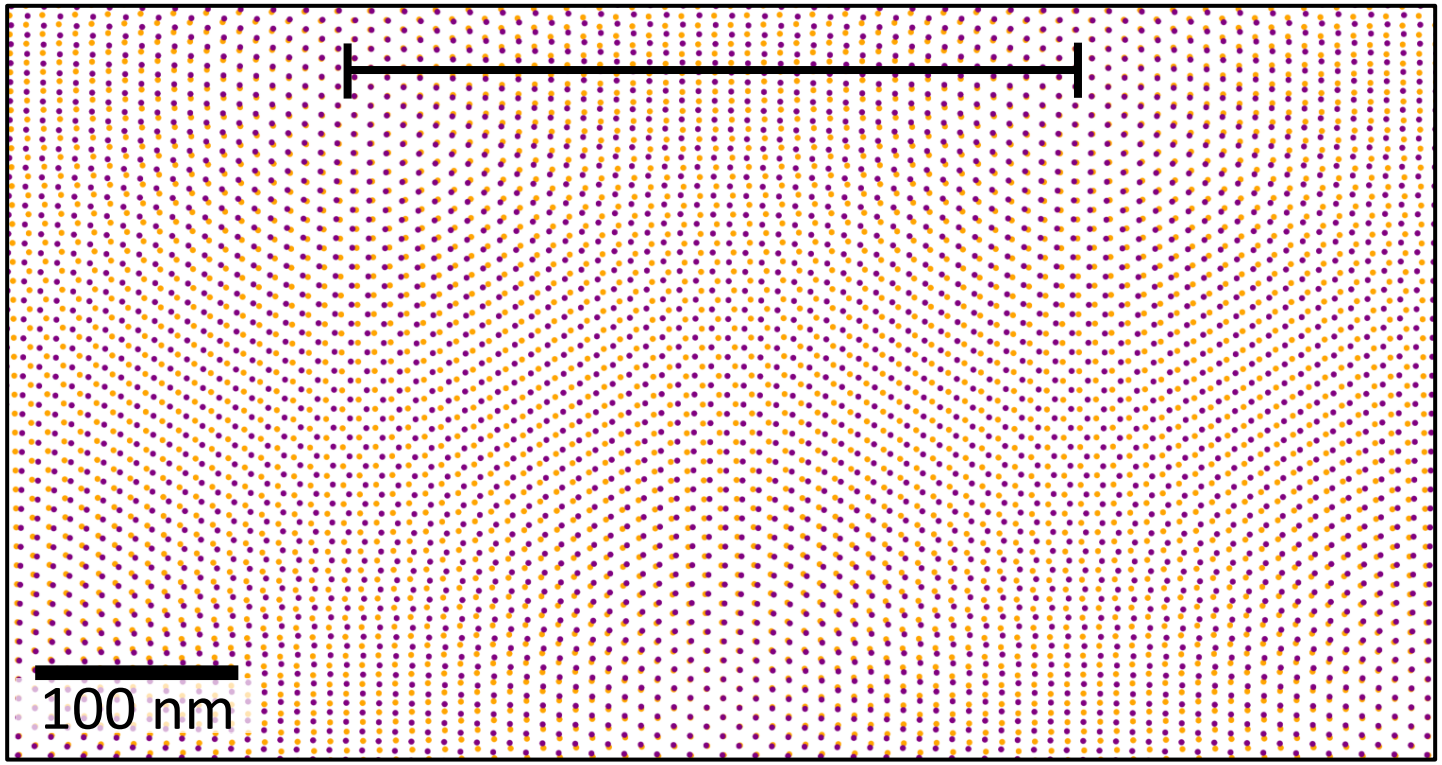

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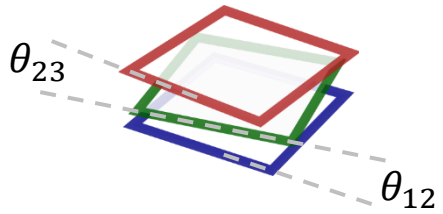

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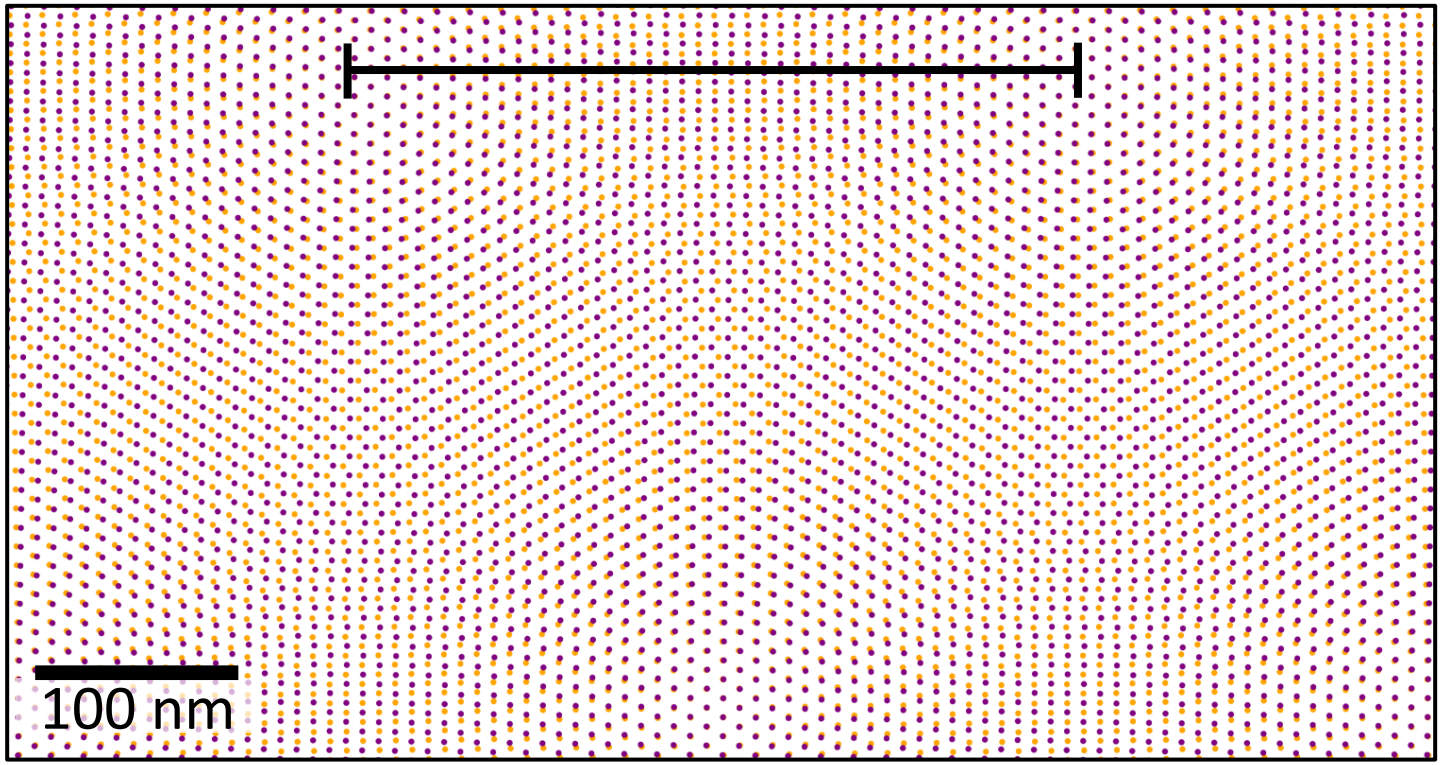

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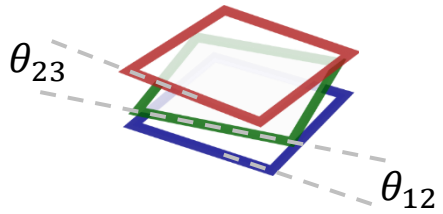

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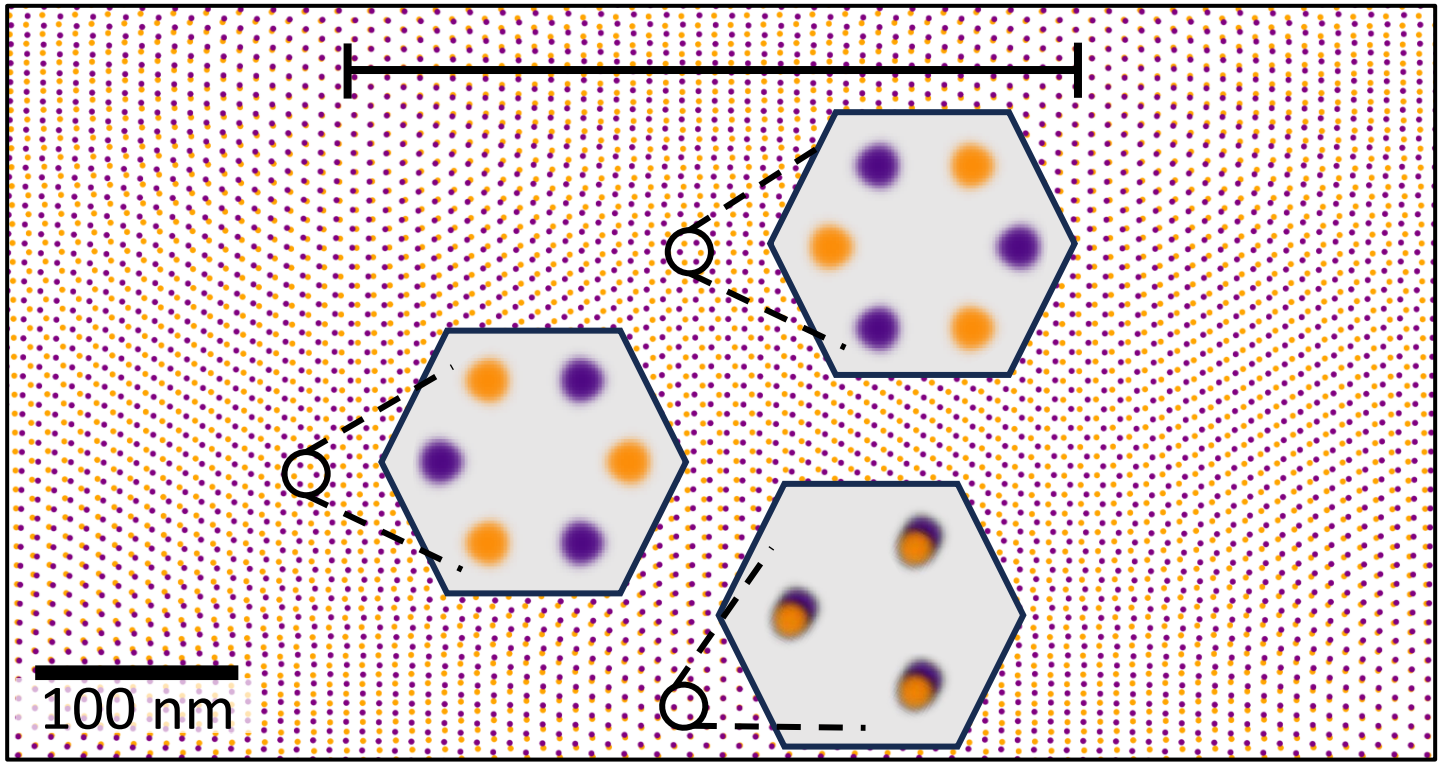

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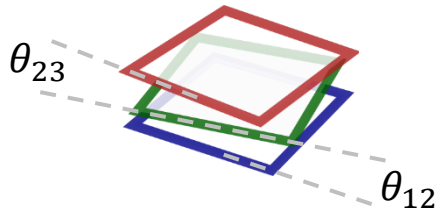

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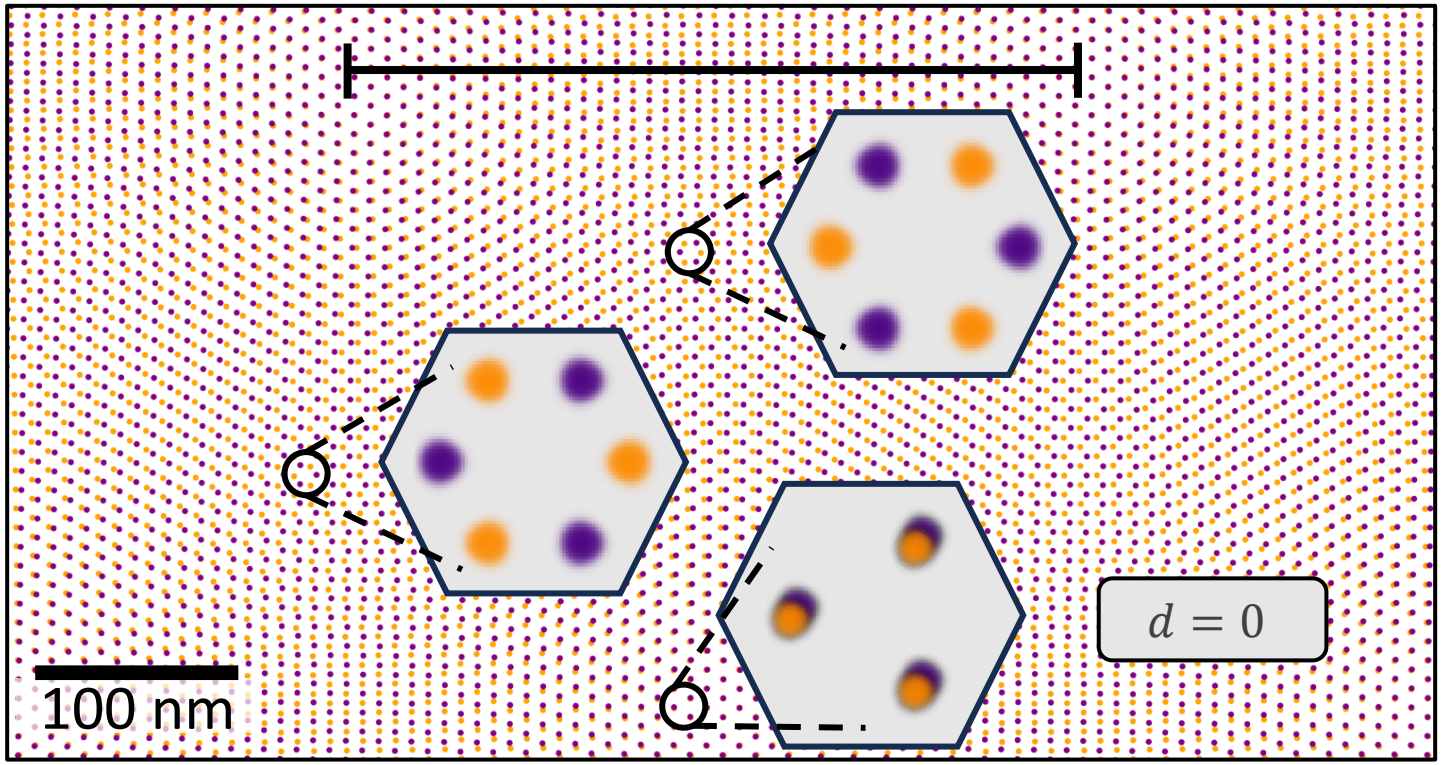

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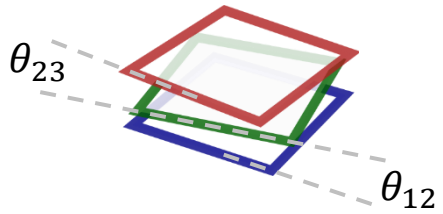

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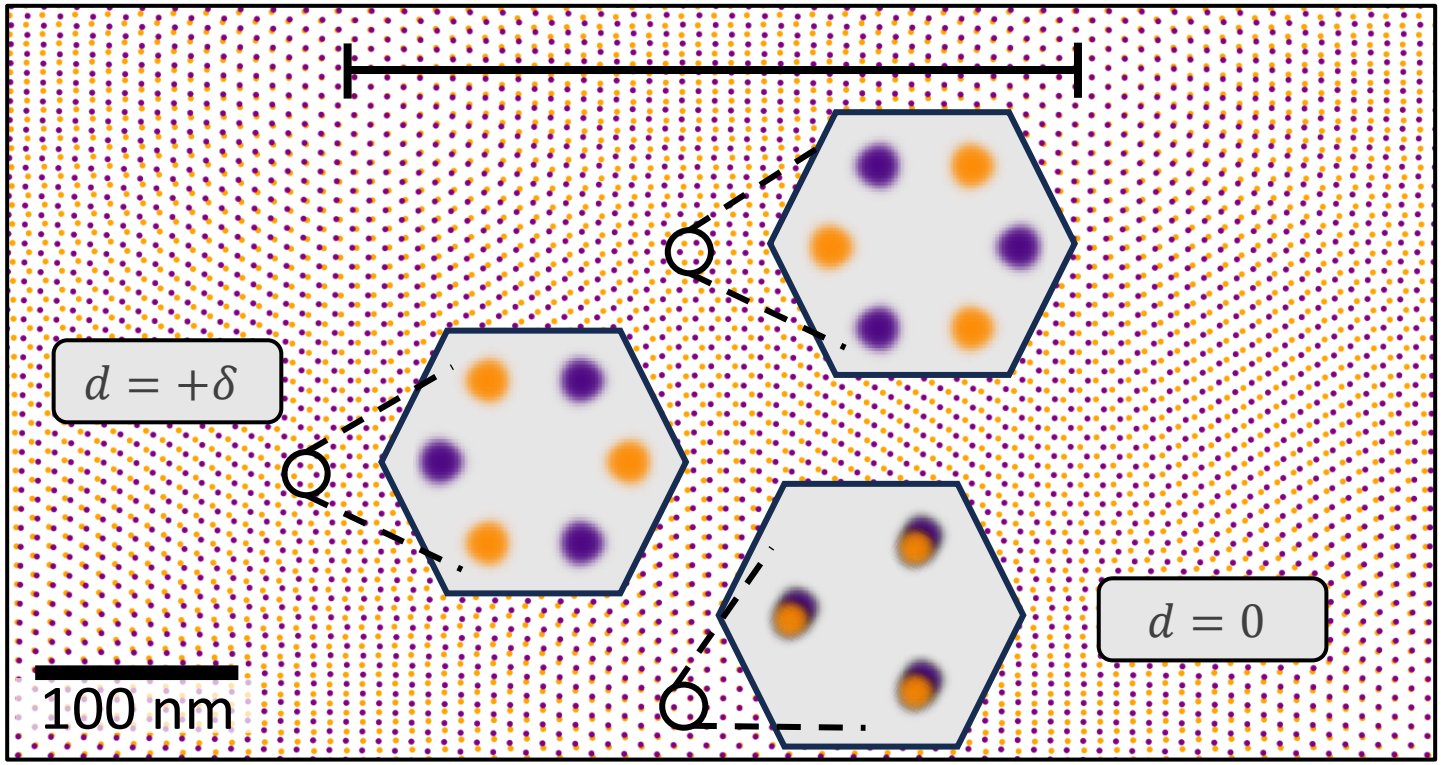

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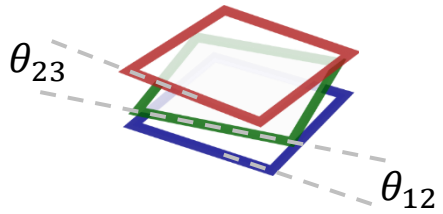

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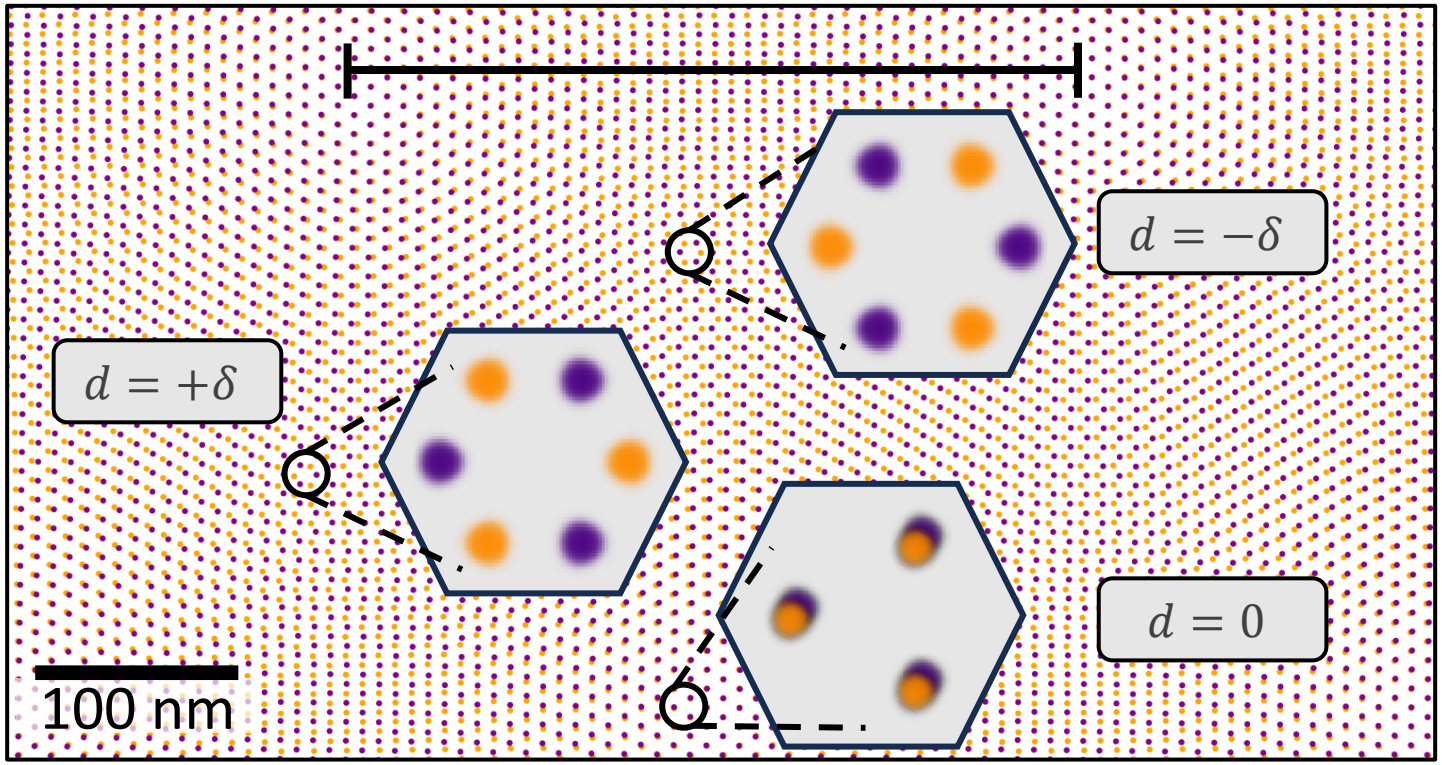

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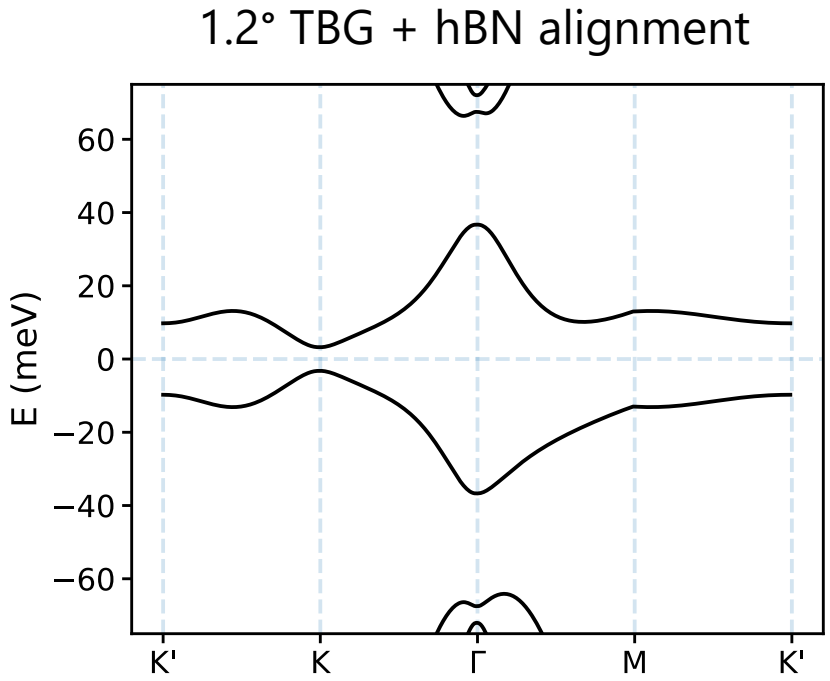
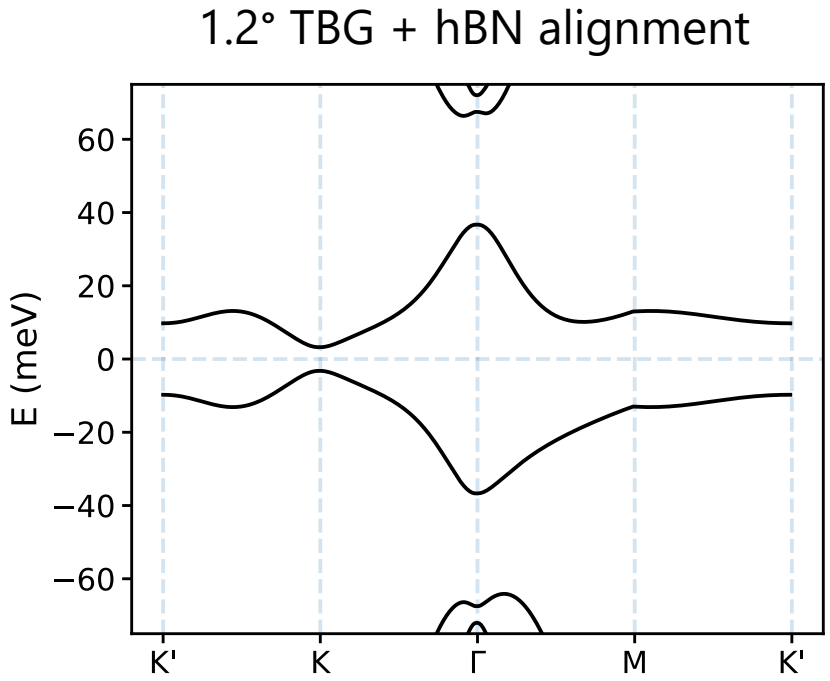
TBG/hBN multimoiré



https://git.sr.ht/~spxtr/bm_model
Shi et al. PRB (2021)

TBG/hBN multimoiré

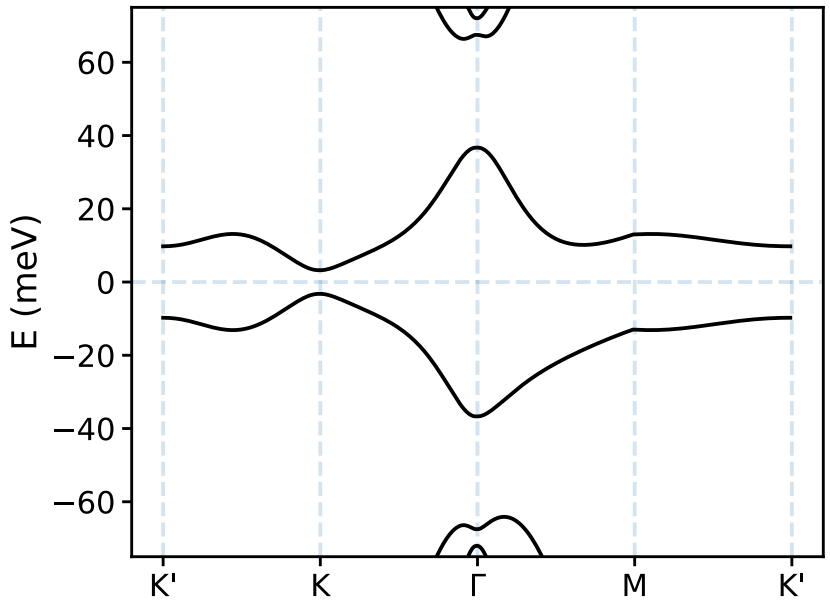


$$V_{BN} = \sum_{\mathbf{k}} \psi^{\dagger}_{1\mathbf{k}} (m_0 \sigma^z) \psi_{1\mathbf{k}}$$

https://git.sr.ht/~spxtr/bm_model
Shi et al. PRB (2021)

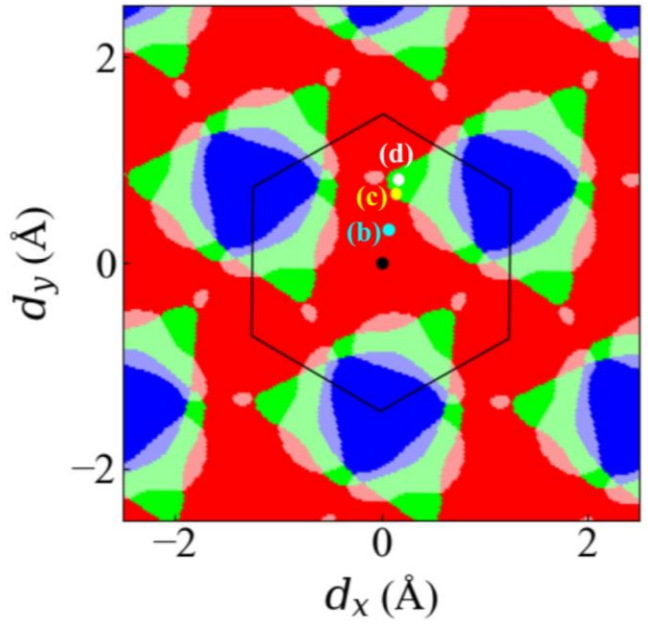
TBG/hBN multimoiré

1.2° TBG + hBN alignment



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

120° commensurate, $U = 0$



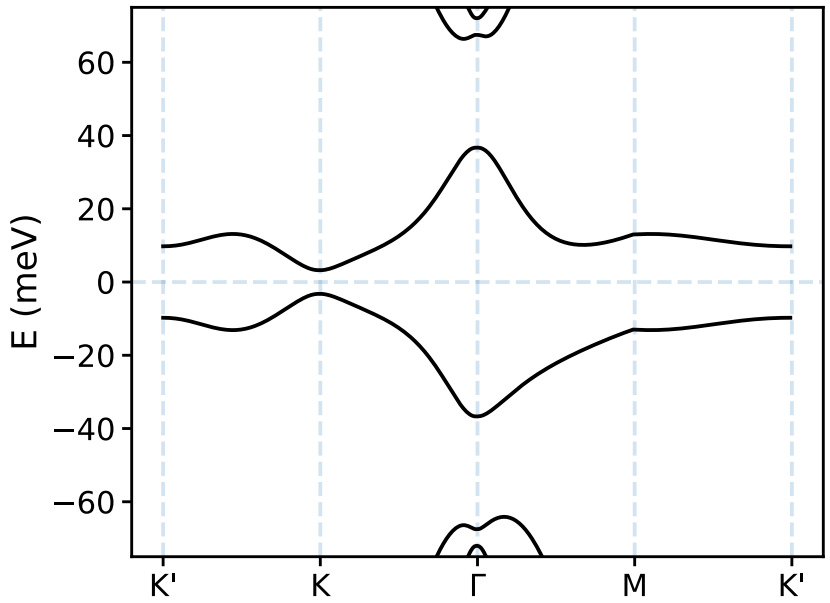
$C = +1$ $C = 0$ $C = -1$

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

https://git.sr.ht/~spxtr/bm_model
Shi et al. PRB (2021)

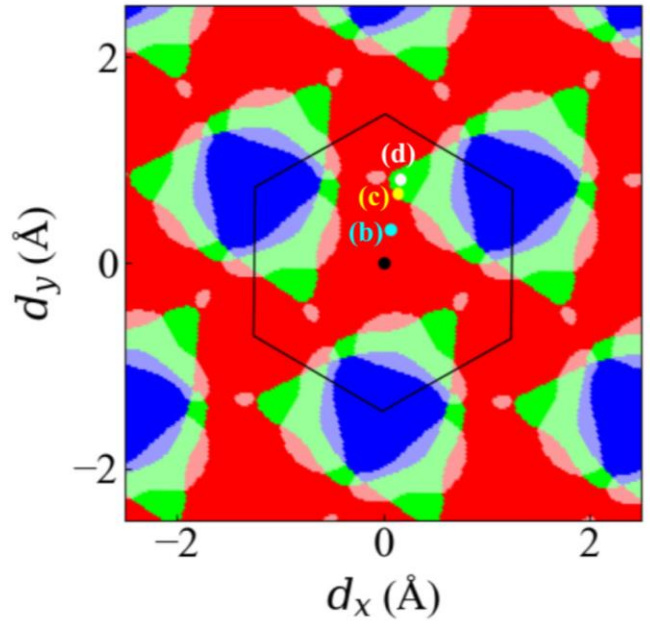
TBG/hBN multimoiré

1.2° TBG + hBN alignment



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

120° commensurate, $U = 0$



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

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

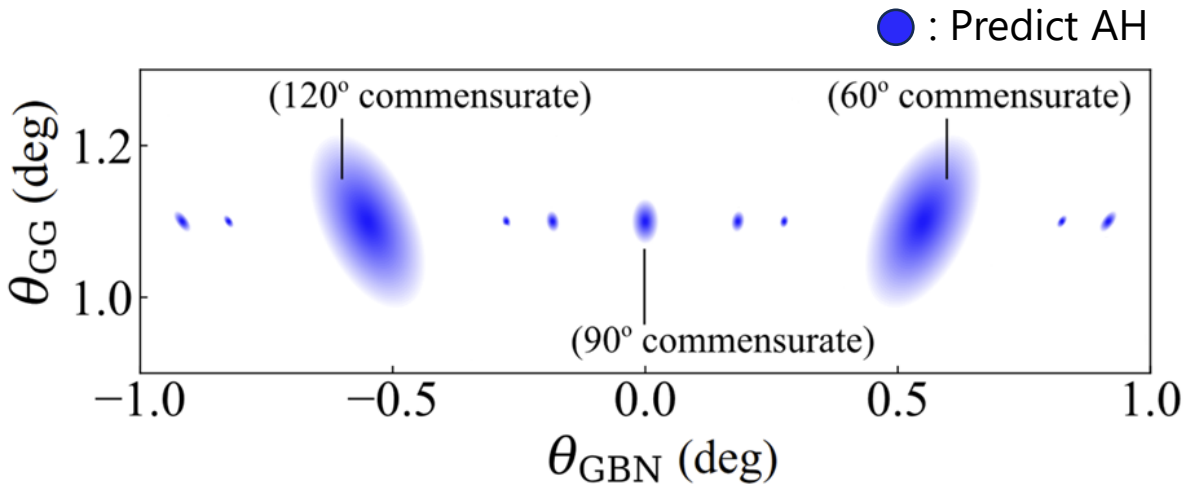
We must take the mesoscopics seriously!
Previous treatment too simplistic

https://git.sr.ht/~spxtr/bm_model
Shi et al. PRB (2021)

TBG/hBN multimoiré

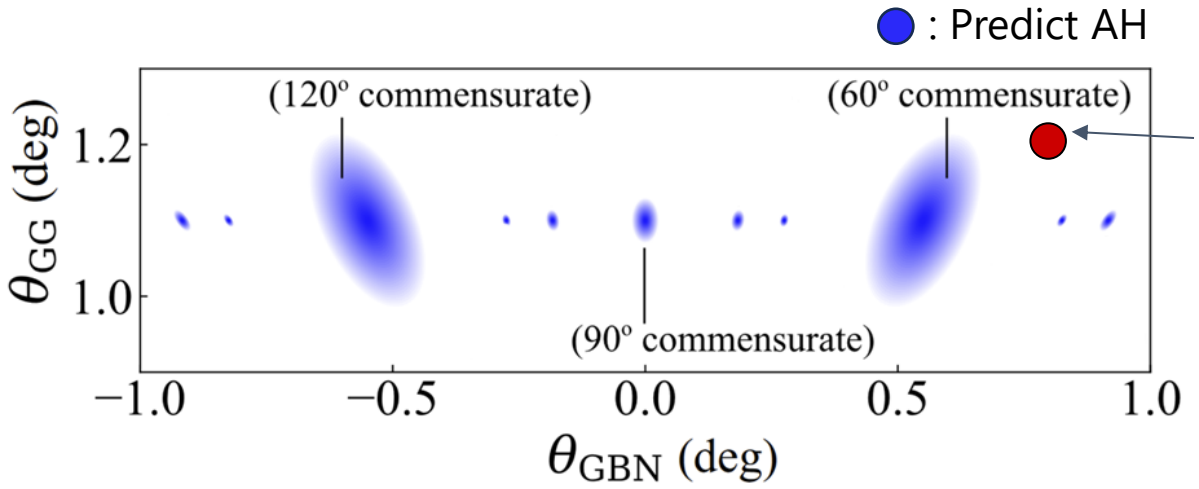
Shi et al. PRB (2021)

TBG/hBN multimoiré



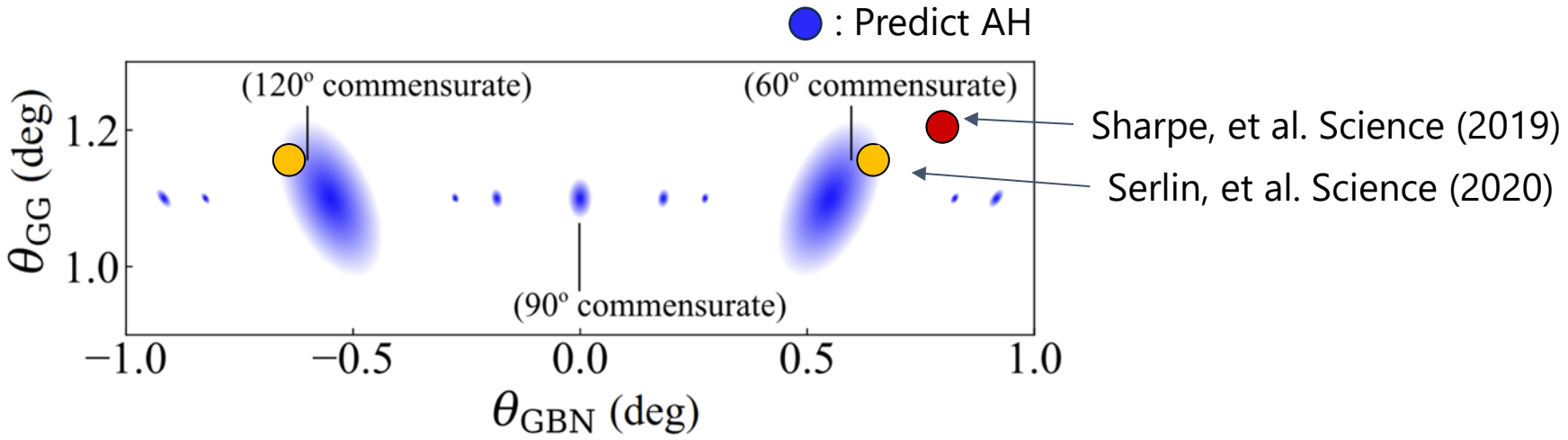
Shi et al. PRB (2021)

TBG/hBN multimoiré



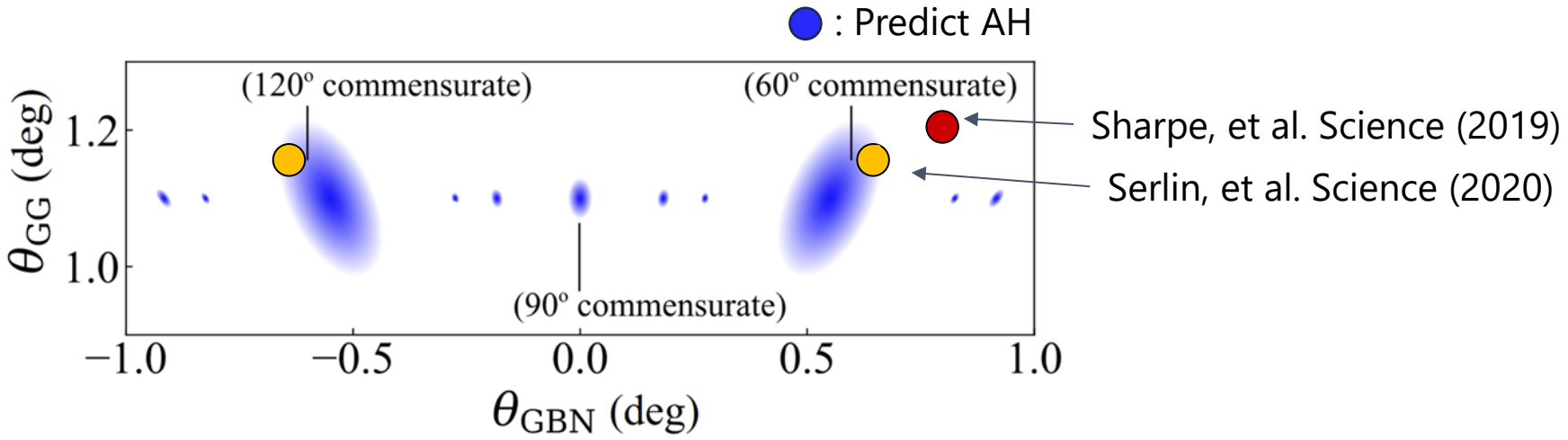
Shi et al. PRB (2021)

TBG/hBN multimoiré



Shi et al. PRB (2021)

TBG/hBN multimoiré



Shi et al. PRB (2021)

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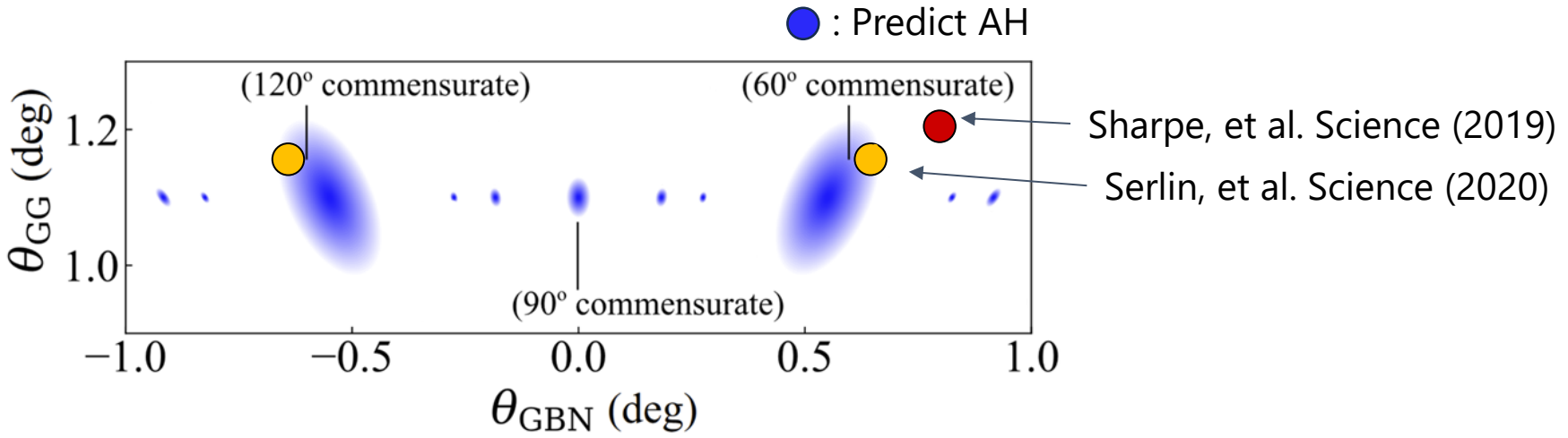
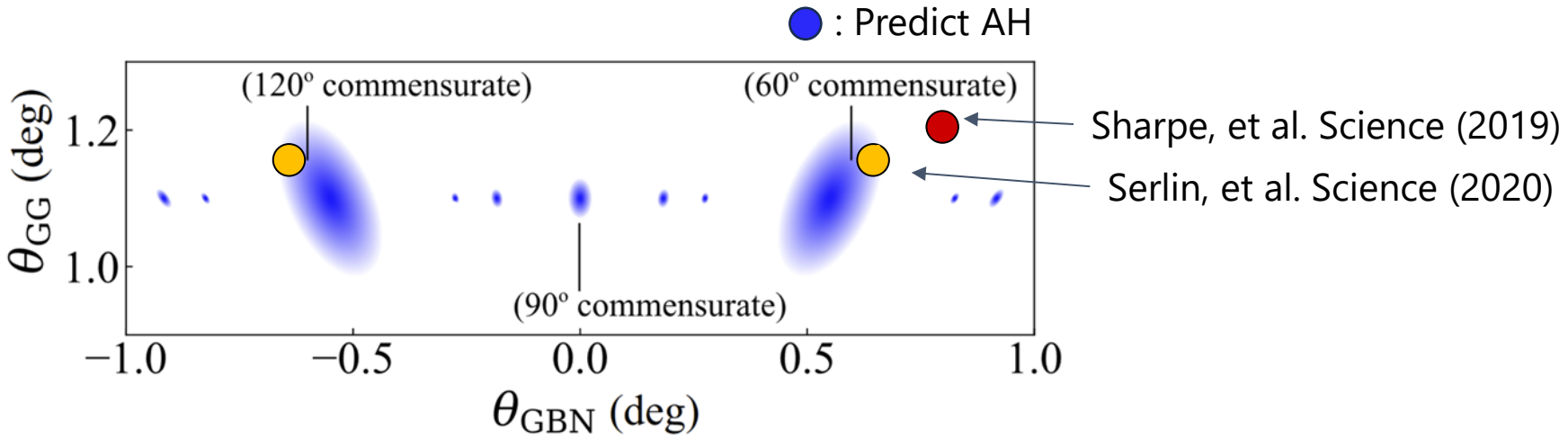


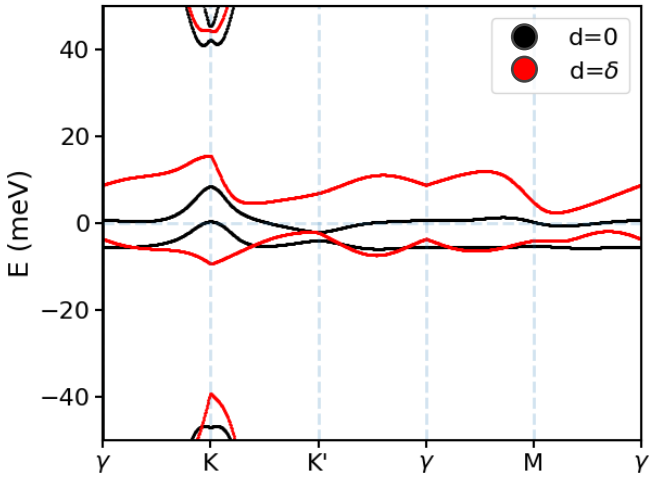
60 commensurate

Shi et al. PRB (2021)

TBG/hBN multimoiré

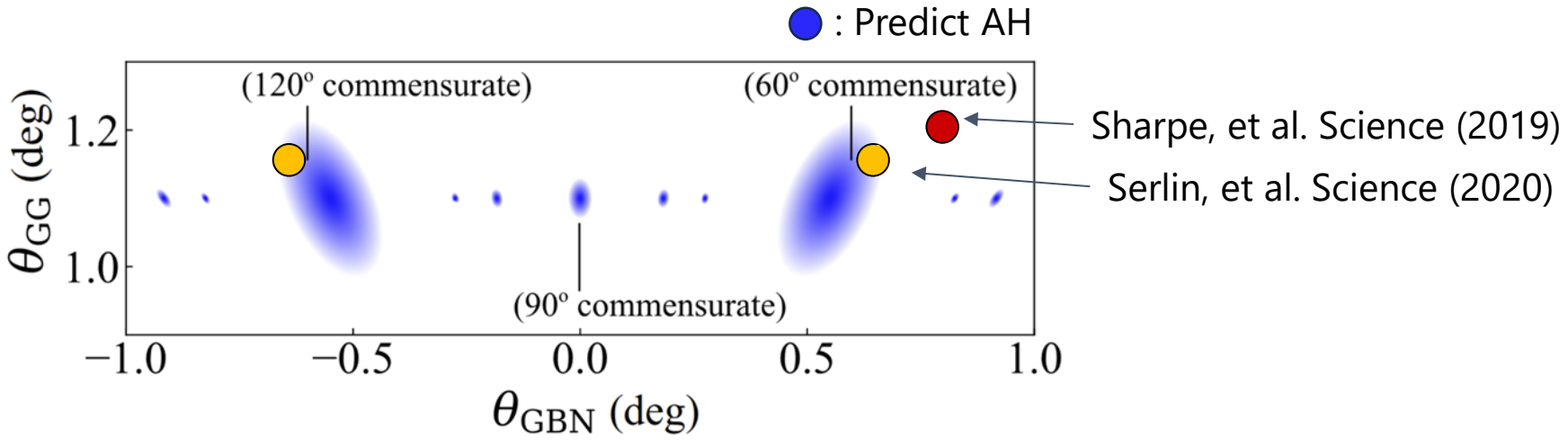


60 commensurate



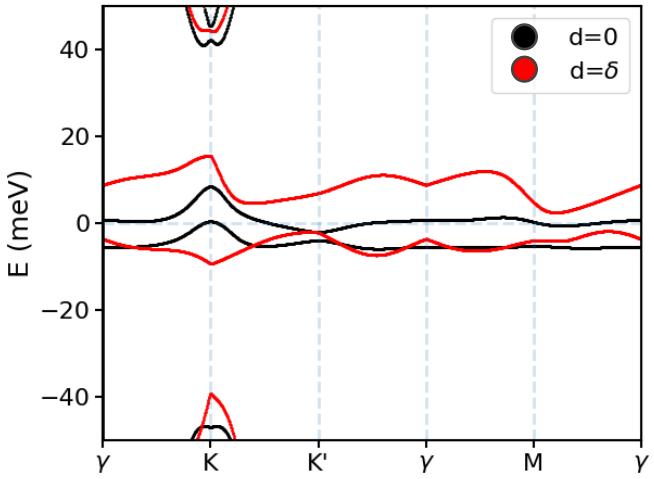
Shi et al. PRB (2021)

TBG/hBN multimoiré



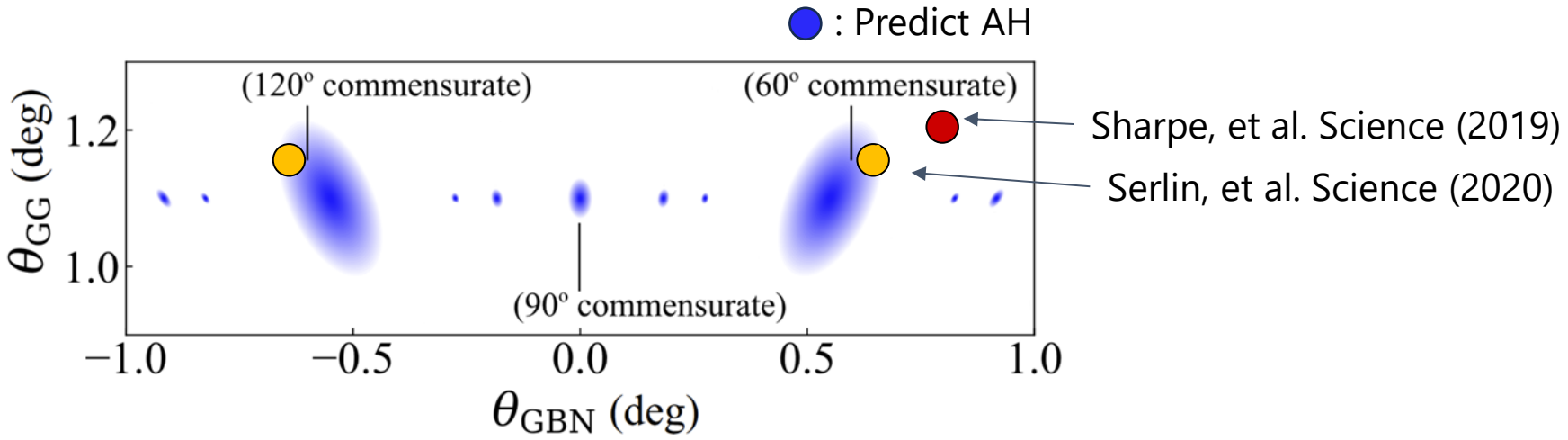
120 commensurate

60 commensurate

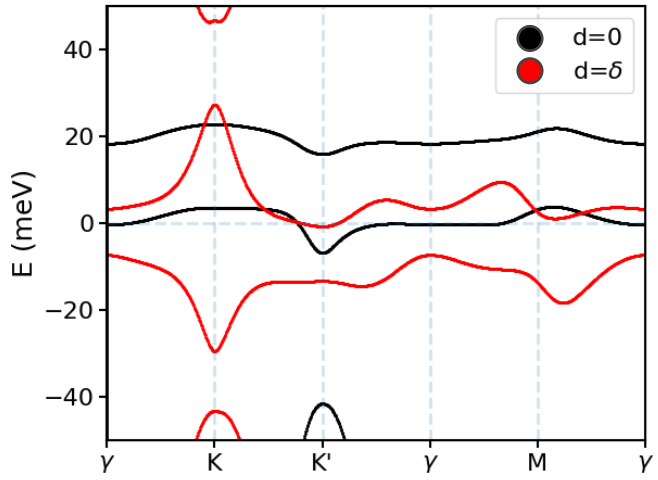


Shi et al. PRB (2021)

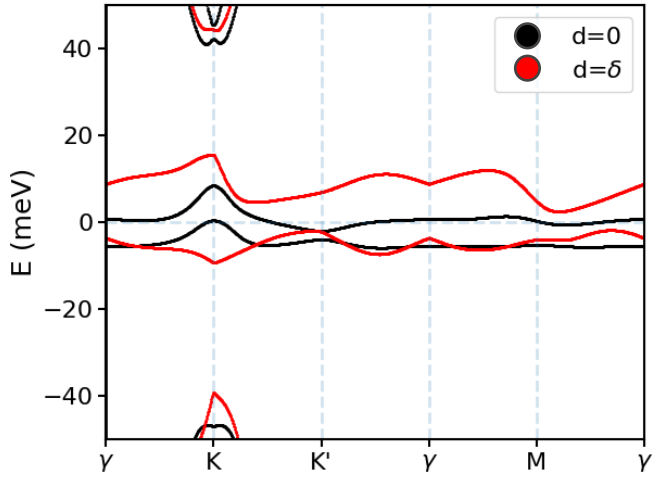
TBG/hBN multimoiré



120 commensurate

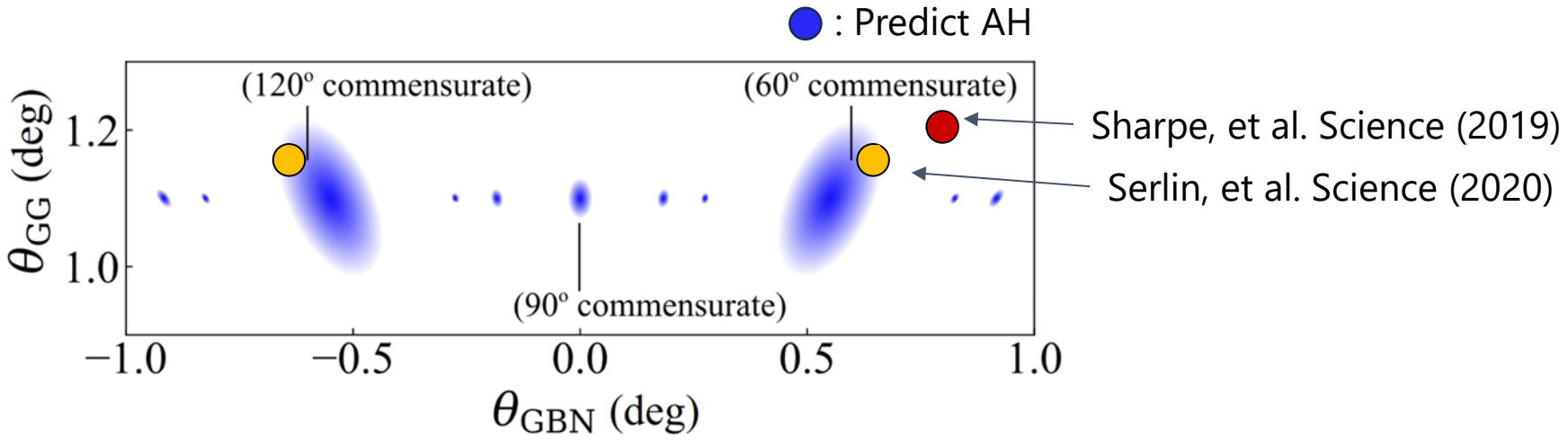


60 commensurate



Shi et al. PRB (2021)

TBG/hBN multimoiré



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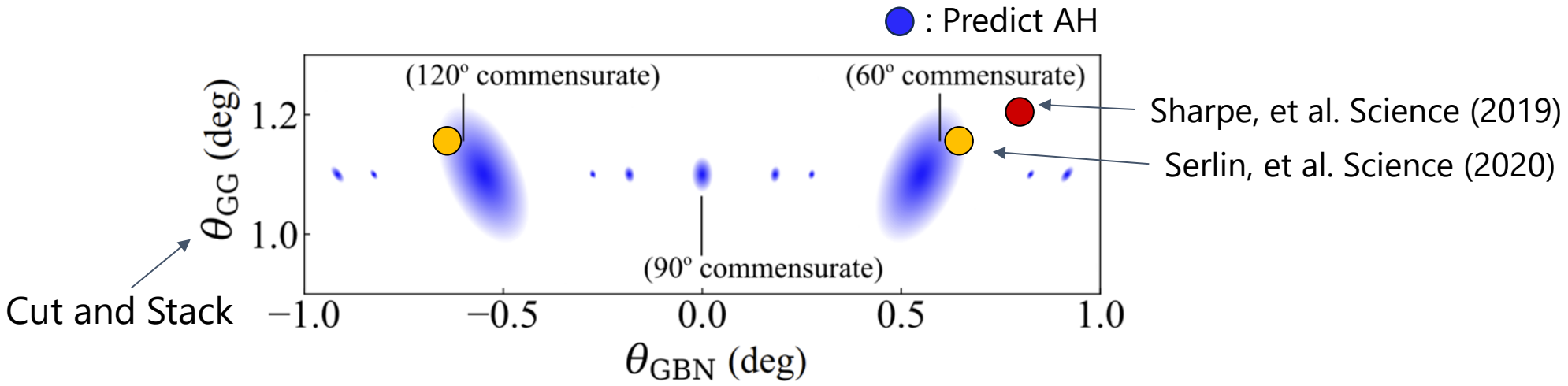

Shi et al. PRB (2021)

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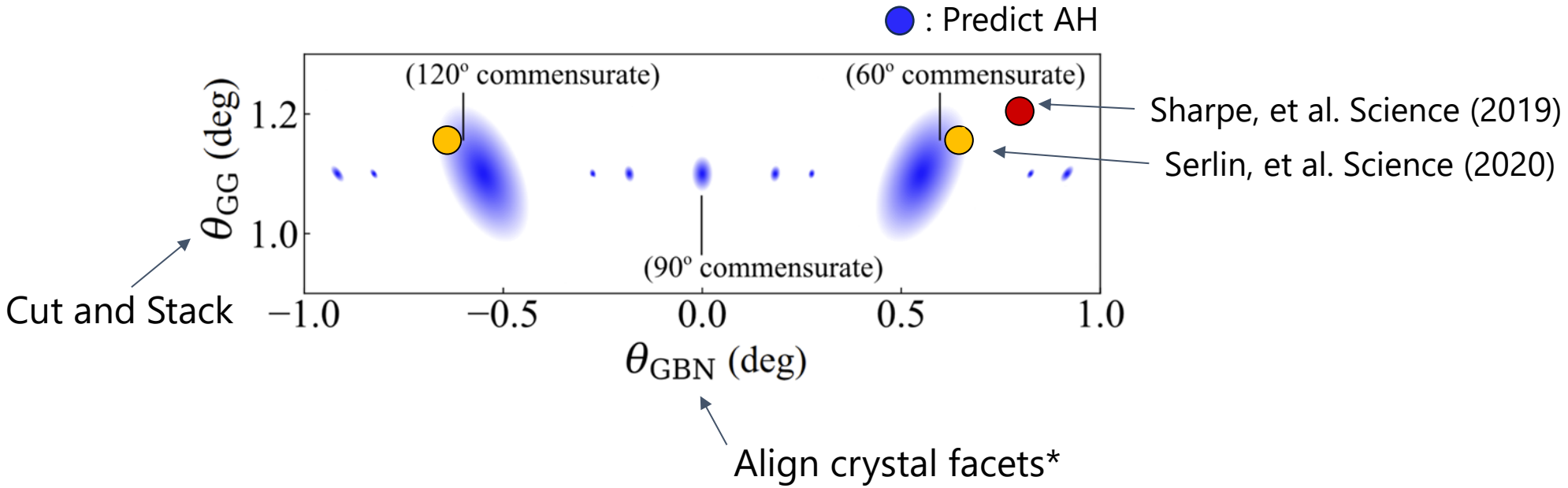

Shi et al. PRB (2021)

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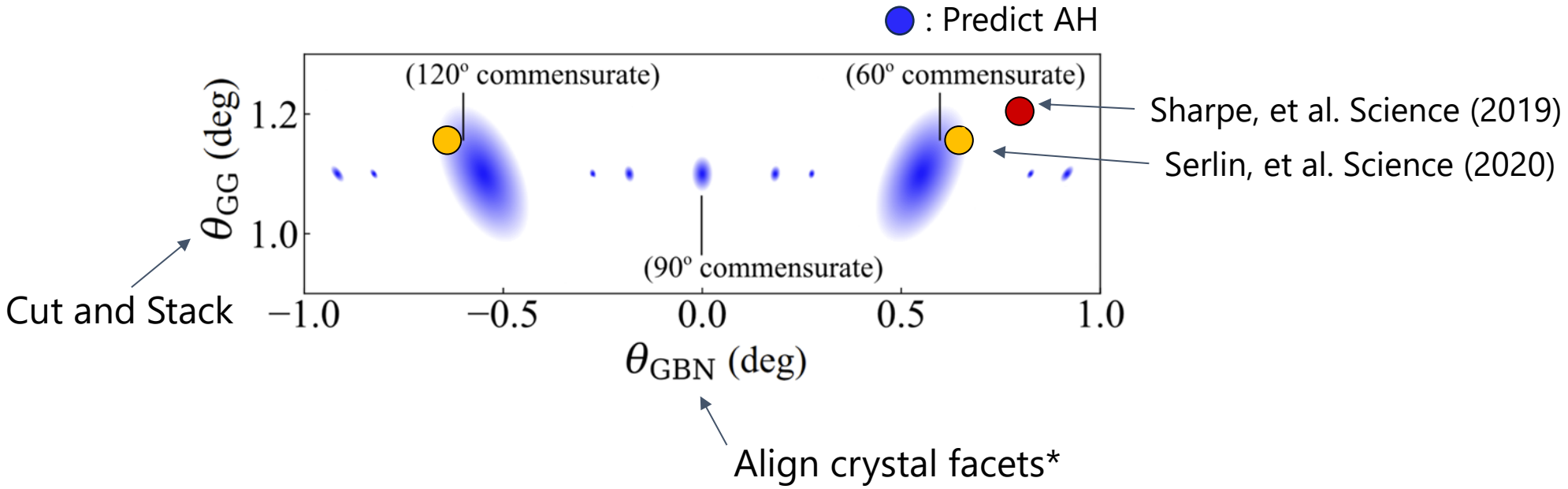
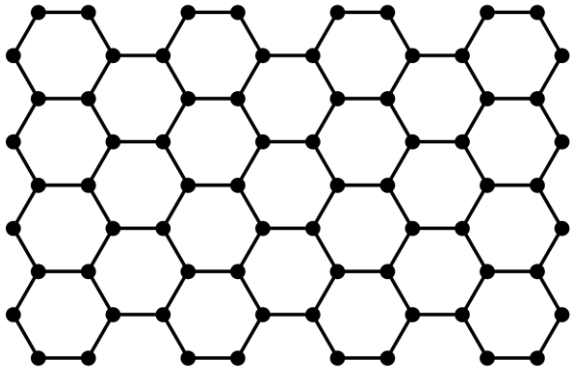




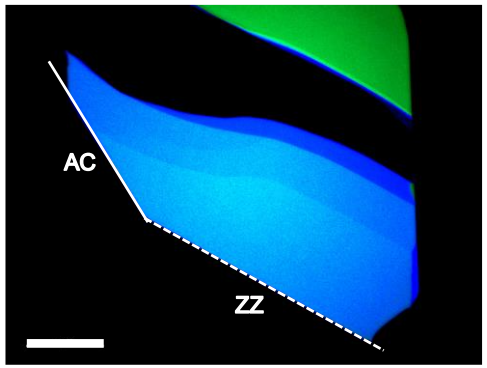
Zig-Zag

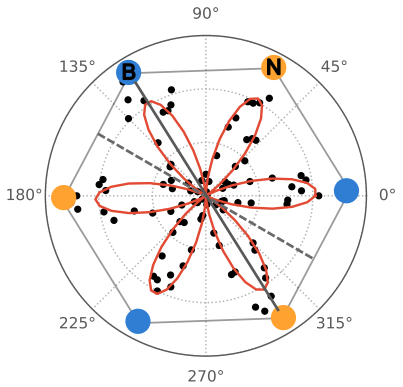
Armchair

Shi et al. PRB (2021)

Deterministic fab with rapid feedback

Second harmonic generation for hBN

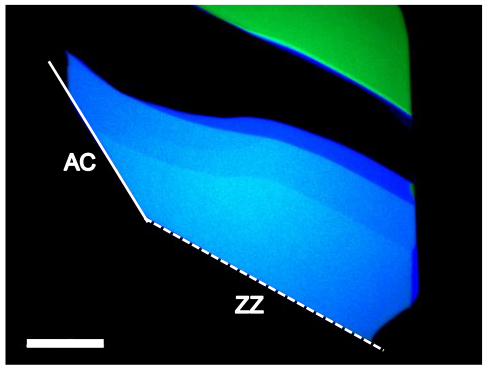


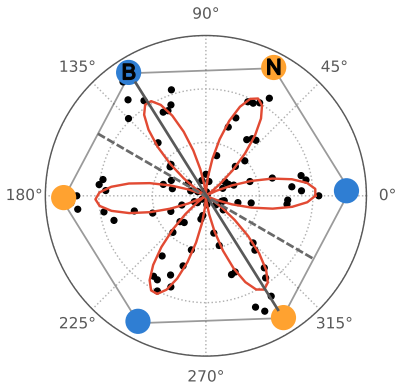


Kamat, Sharpe, et al. PNAS (2024)
Pendharkar, et al. PNAS (2024)

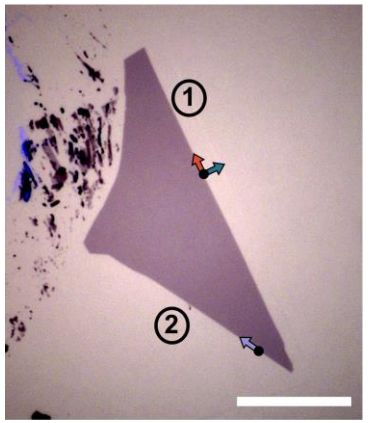
Deterministic fab with rapid feedback

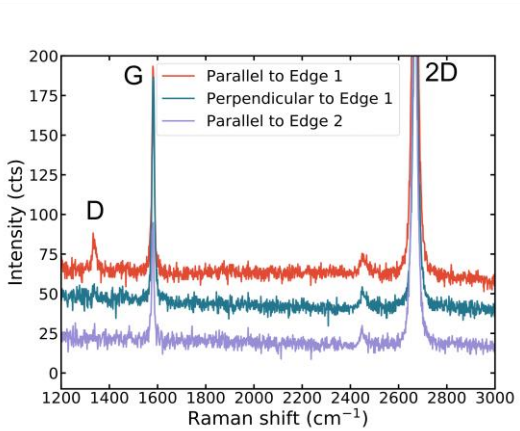
Second harmonic generation for hBN





Polarized Raman for graphene





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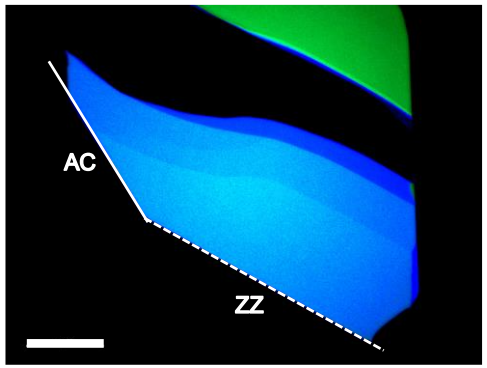

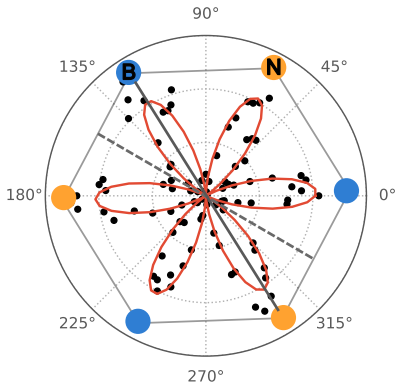

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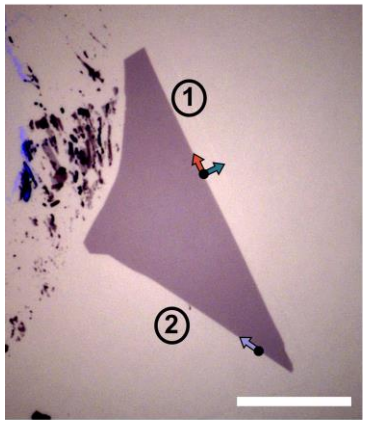
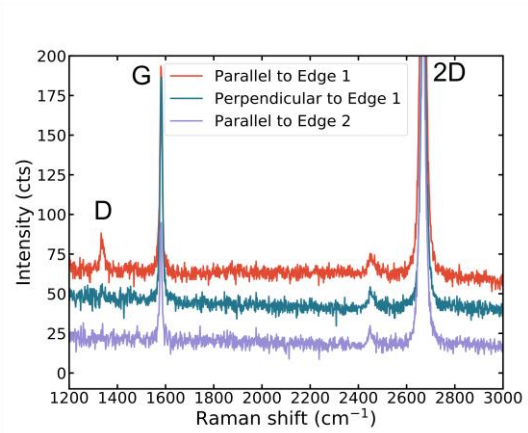
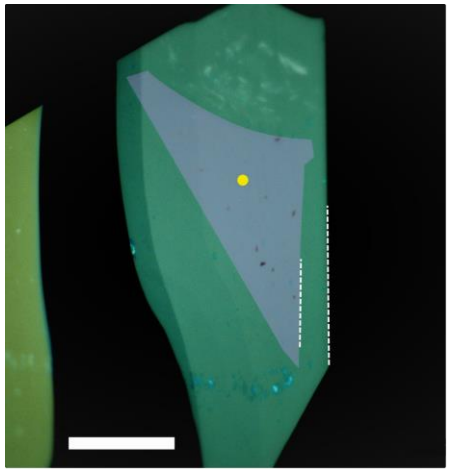
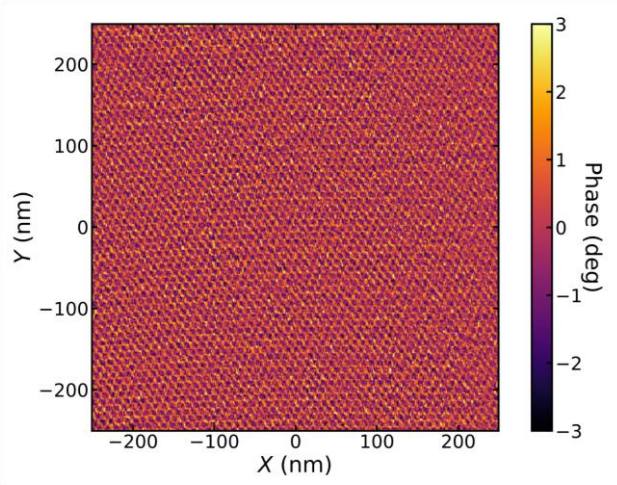




Torsional force microscopy for moirés





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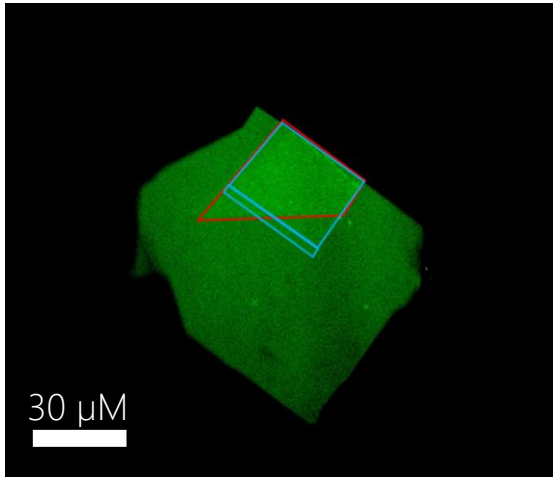
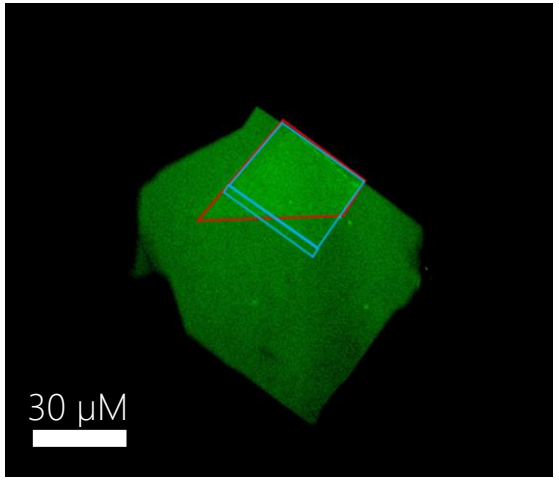
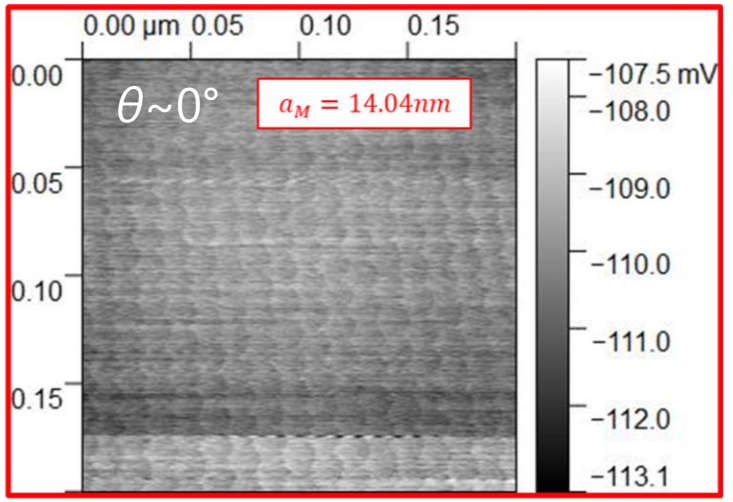
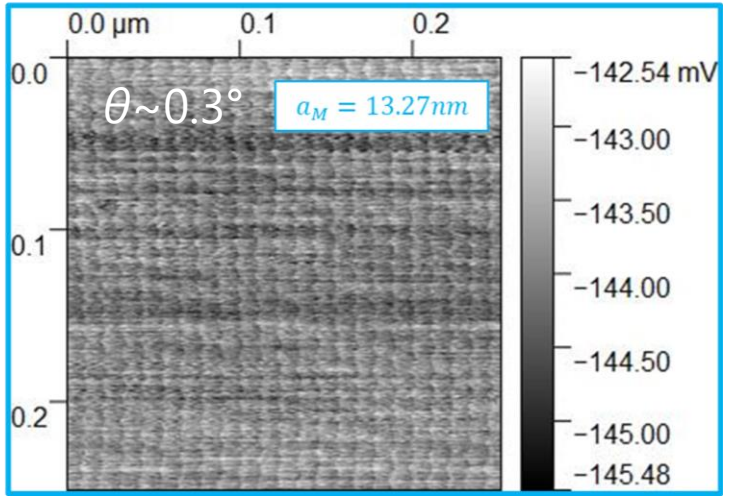
Deterministic fab with rapid feedback



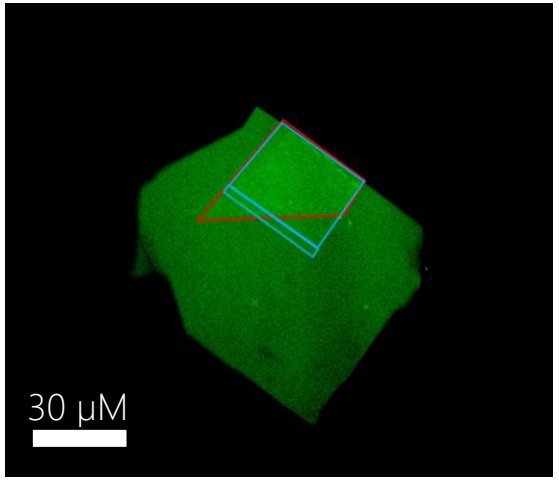
Deterministic fab with rapid feedback

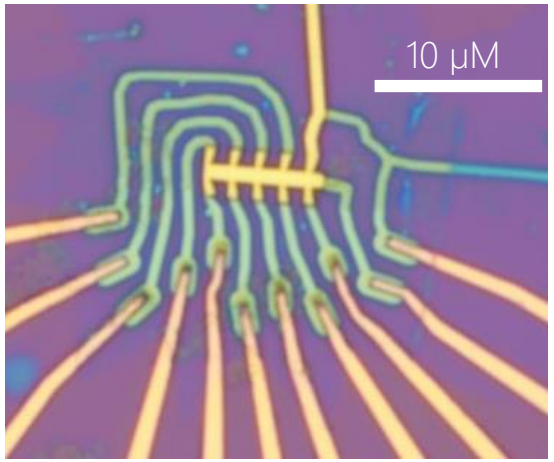


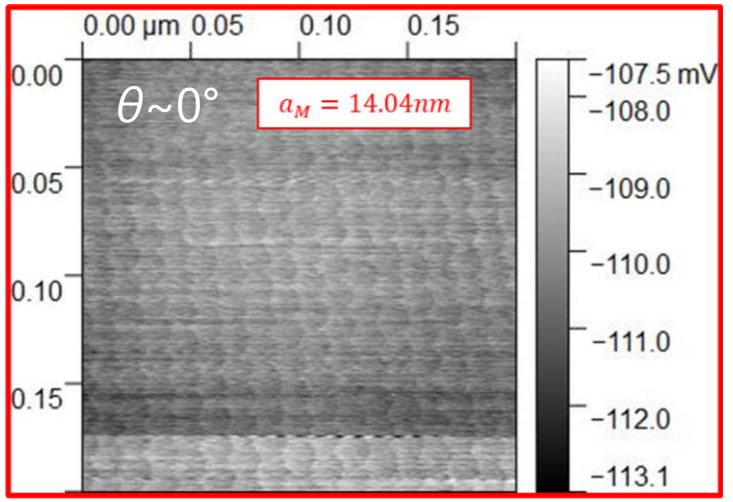


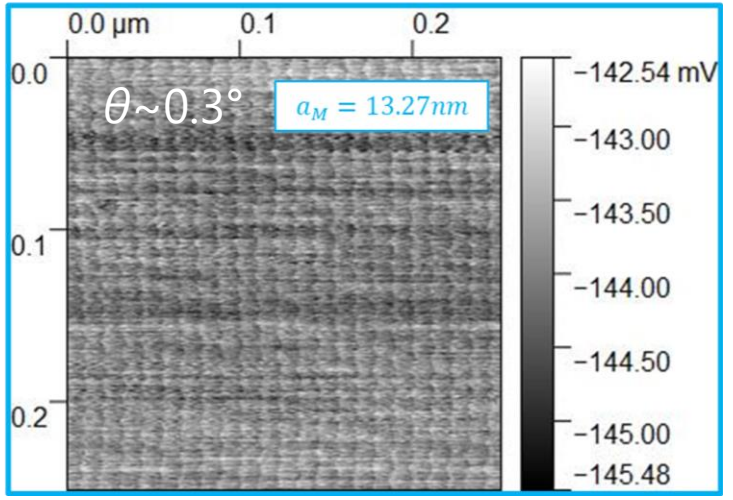


Deterministic fab with rapid feedback

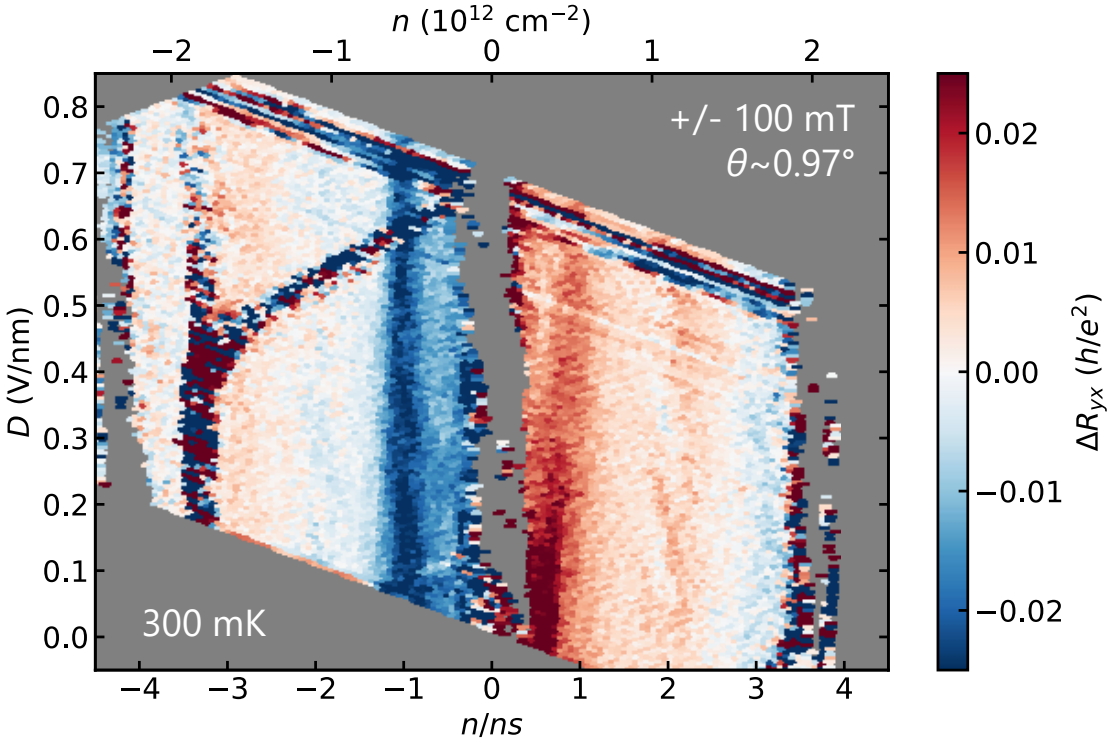




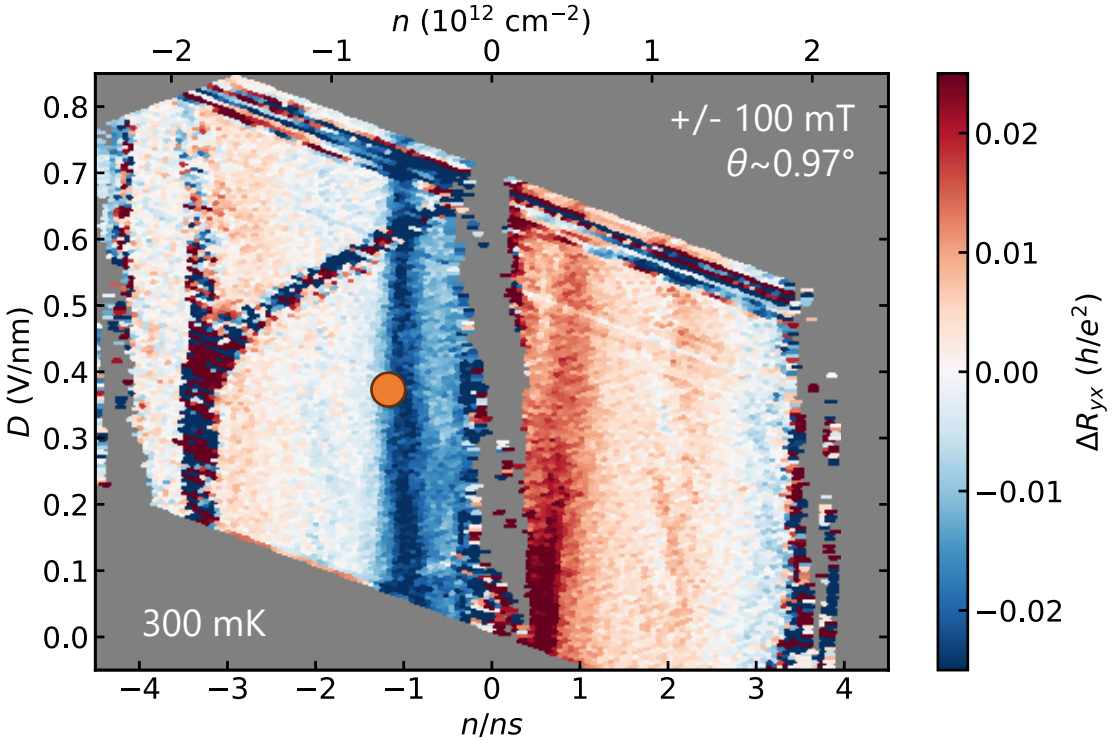


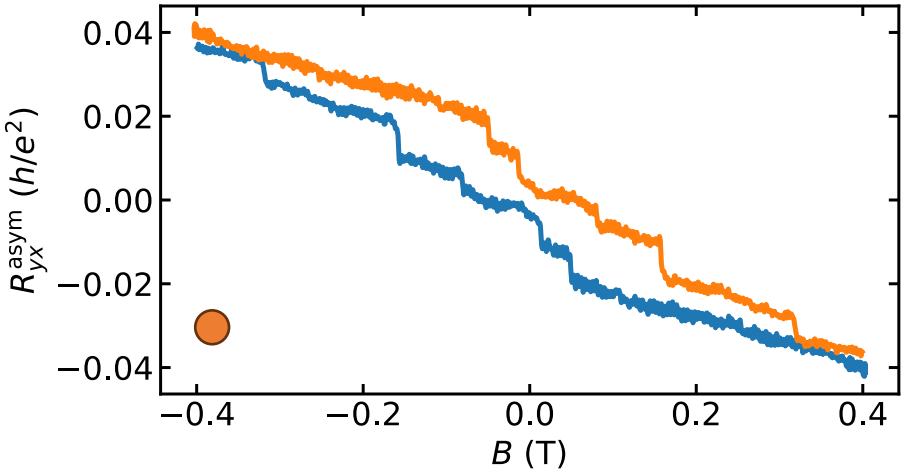


Anomalous Hall!



Anomalous Hall!





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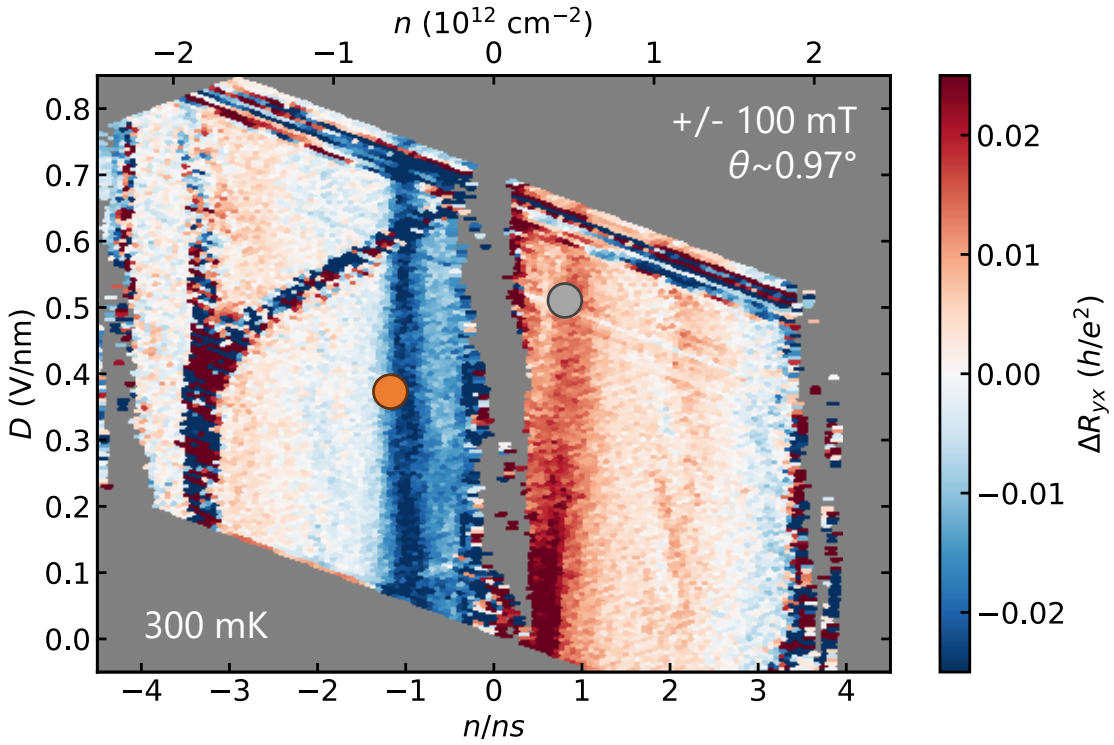

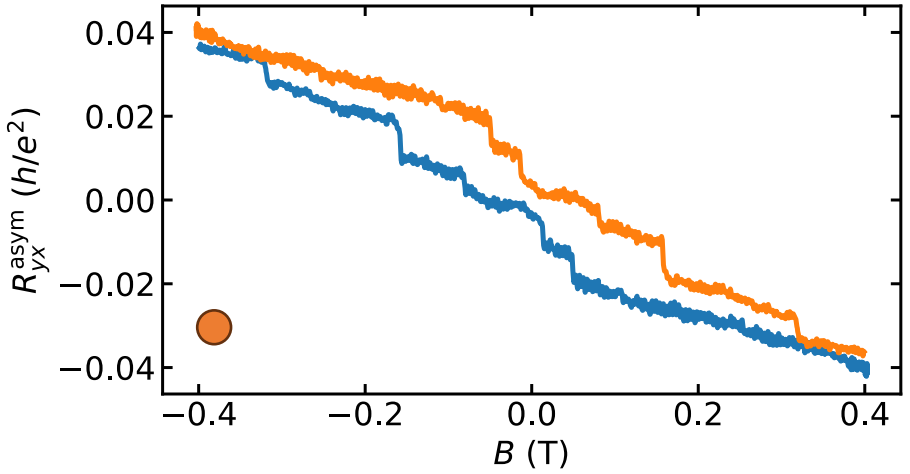

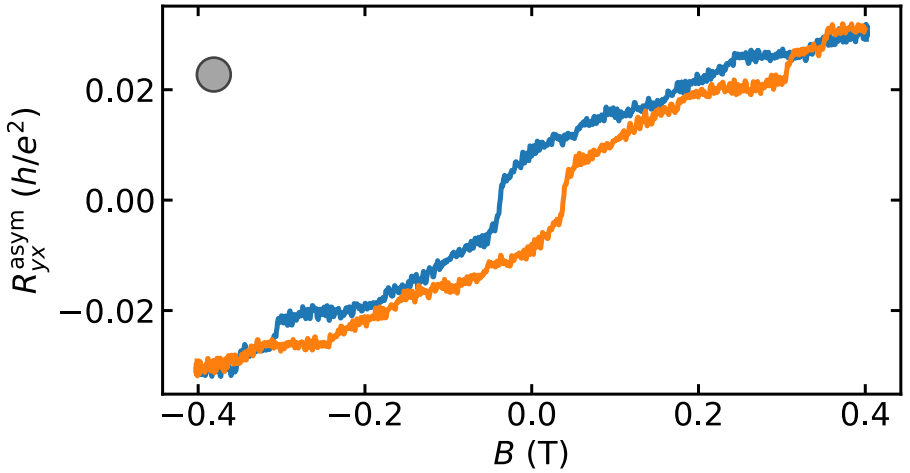

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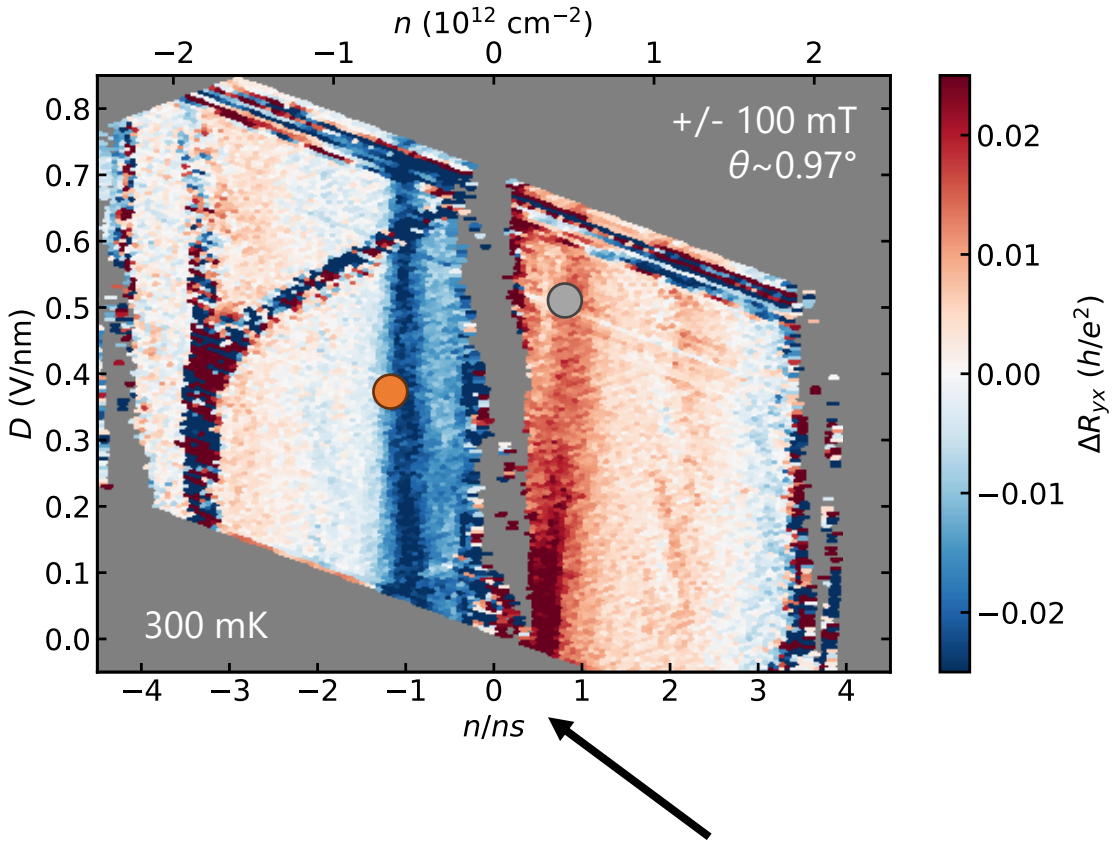
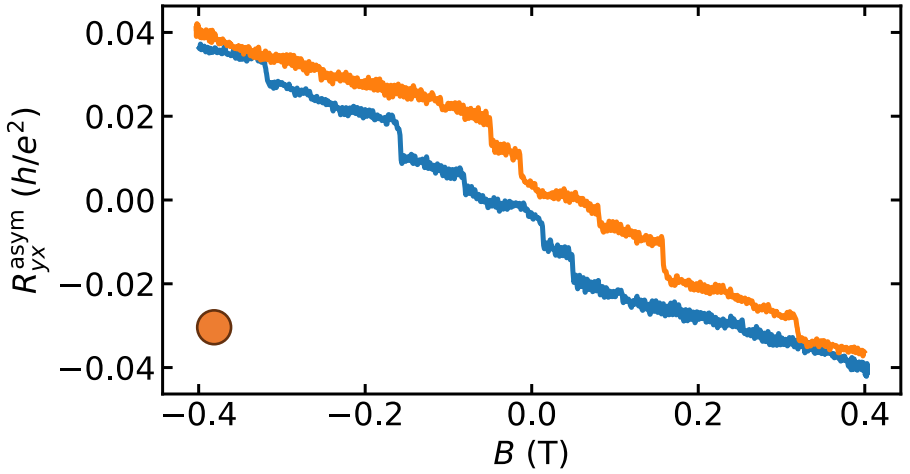
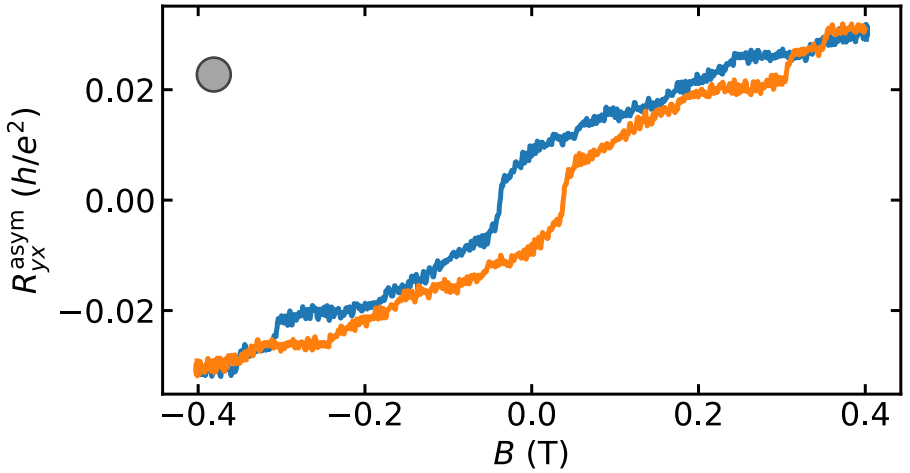
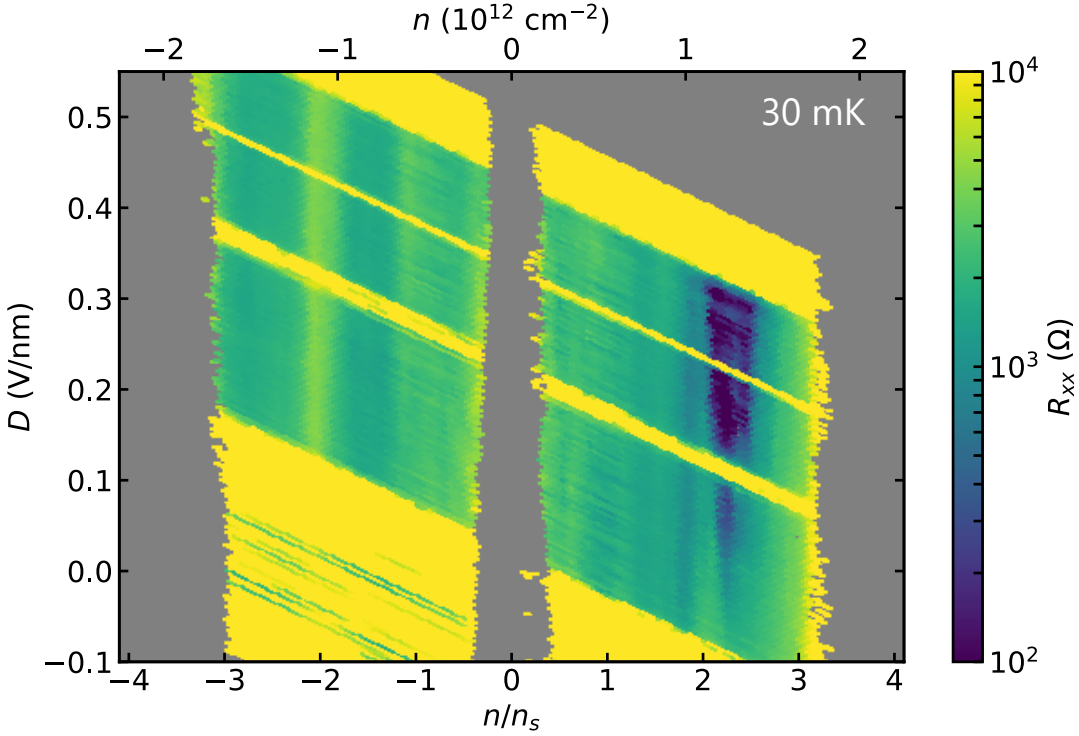


Large gap at CNP

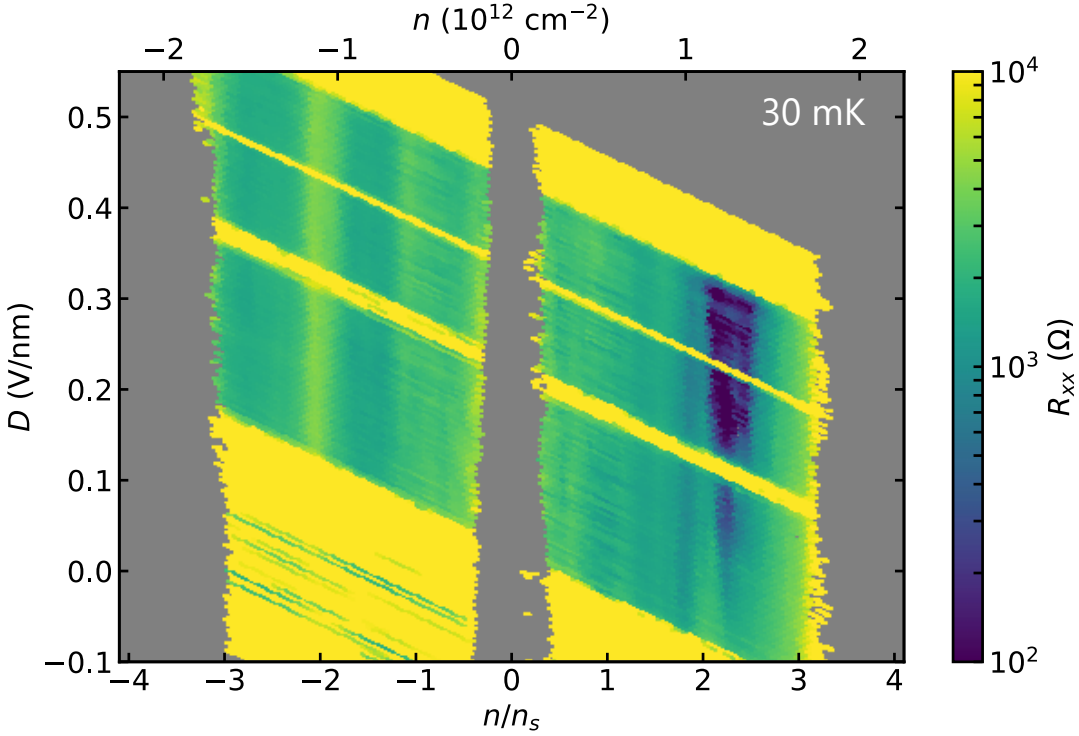




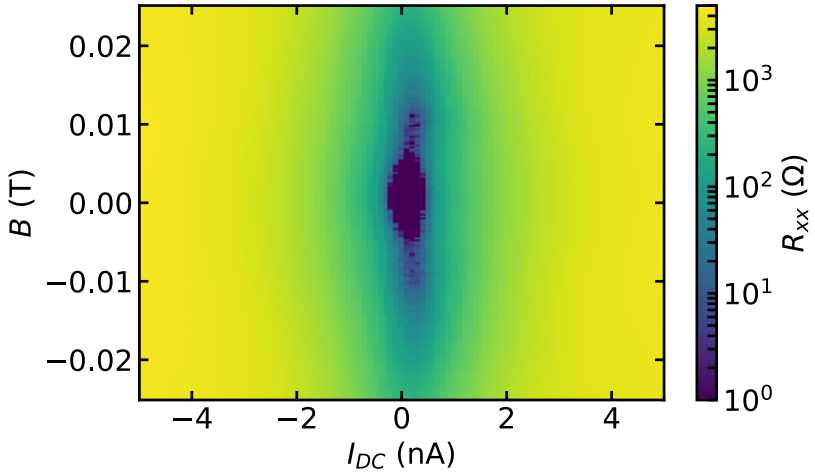
Cooling to 30 mK, superconductivity!?



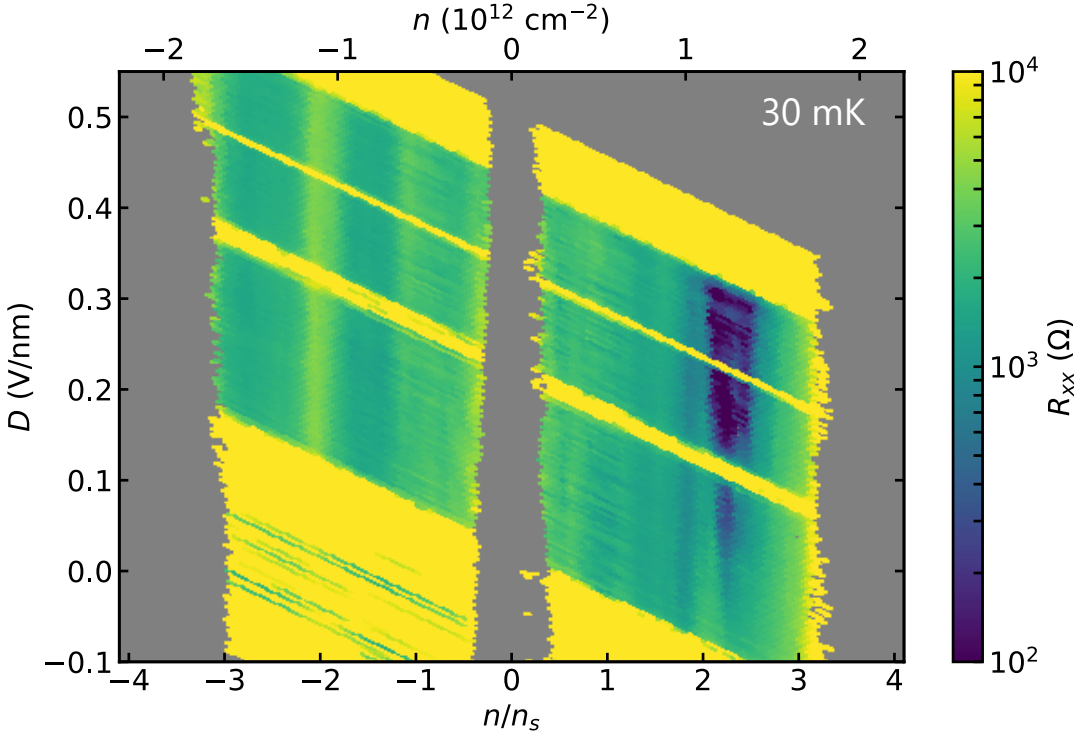
Cooling to 30 mK, superconductivity!?



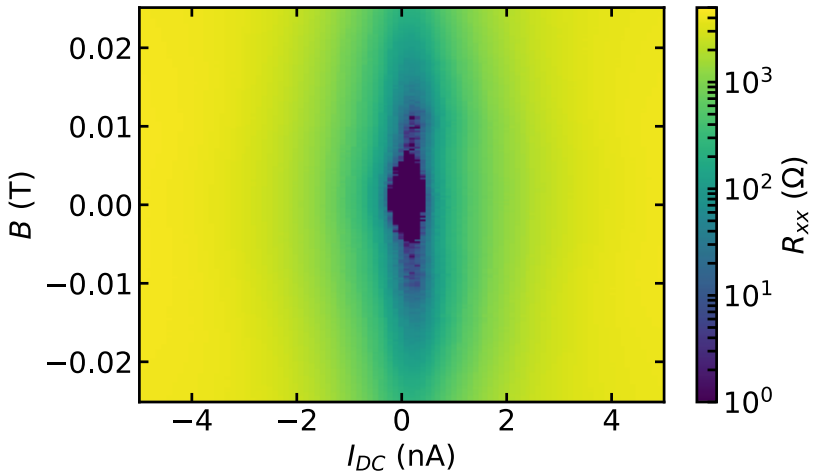
Near optimal doping



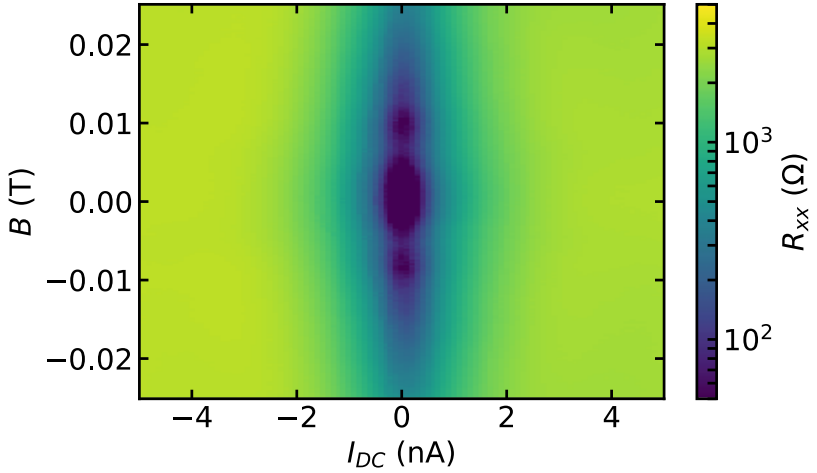
Cooling to 30 mK, superconductivity!?



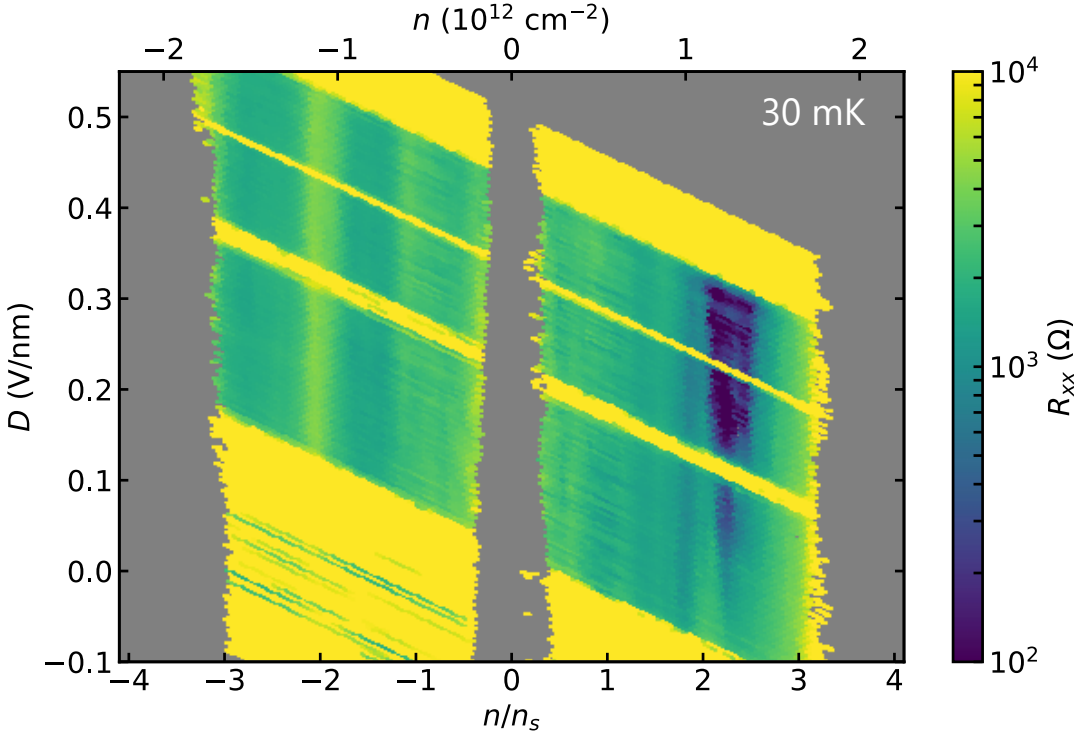
Near optimal doping



Edge of dome

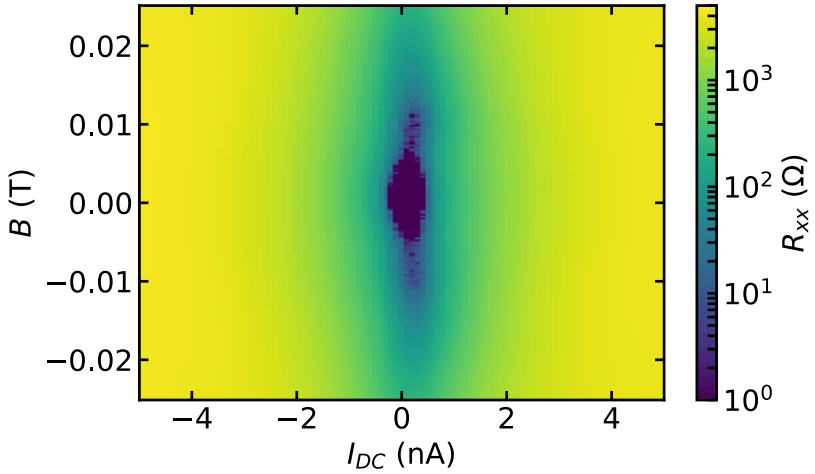


Cooling to 30 mK, superconductivity!?

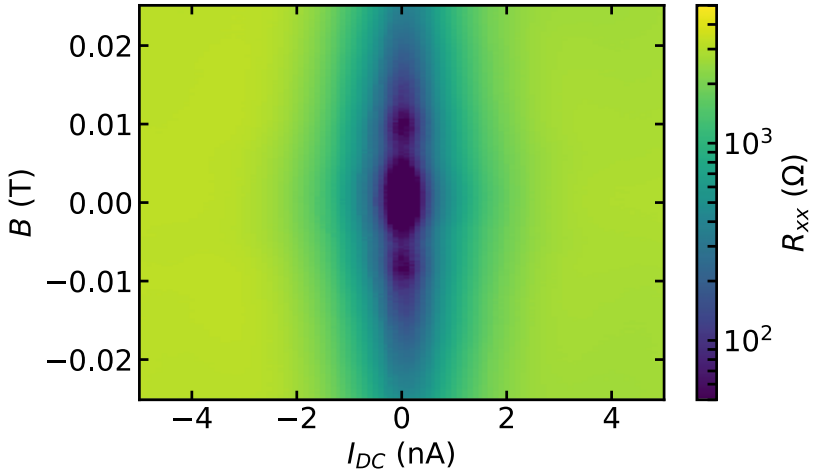


This device exhibits both magnetism and superconductivity!

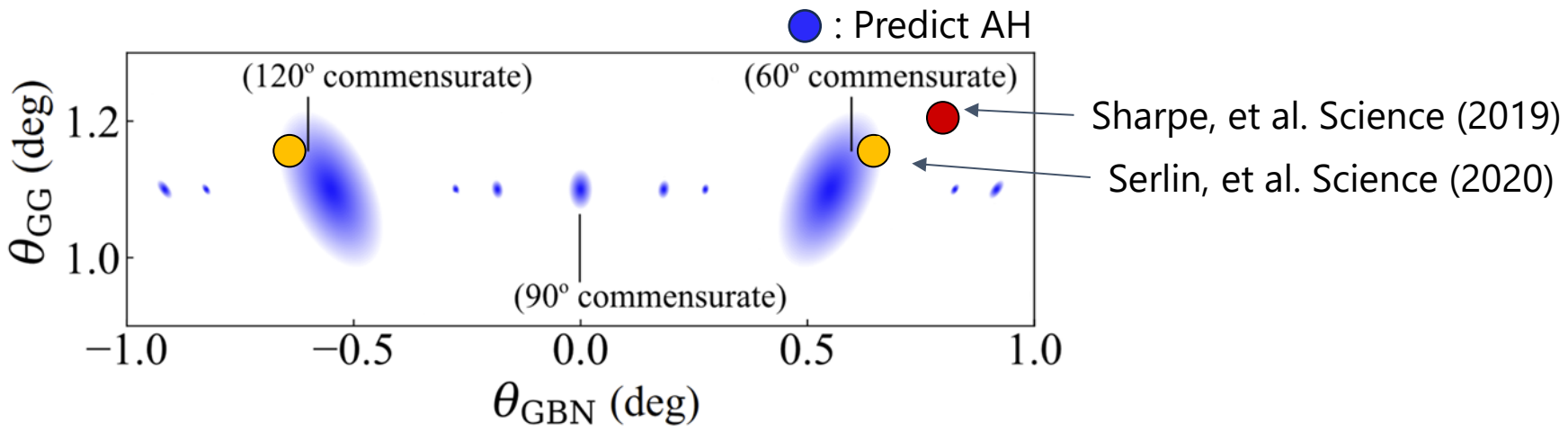
Near optimal doping



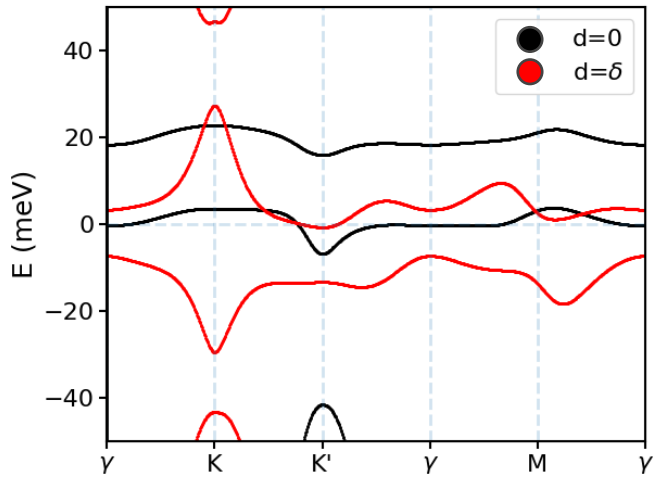
Edge of dome



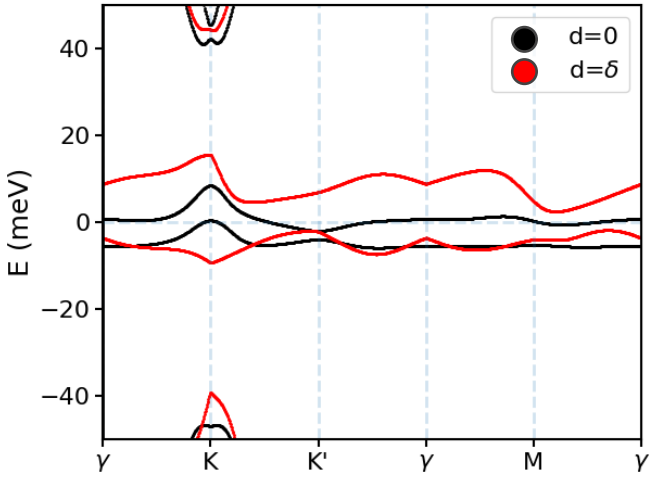
What is θ_{GBN}?



120 commensurate

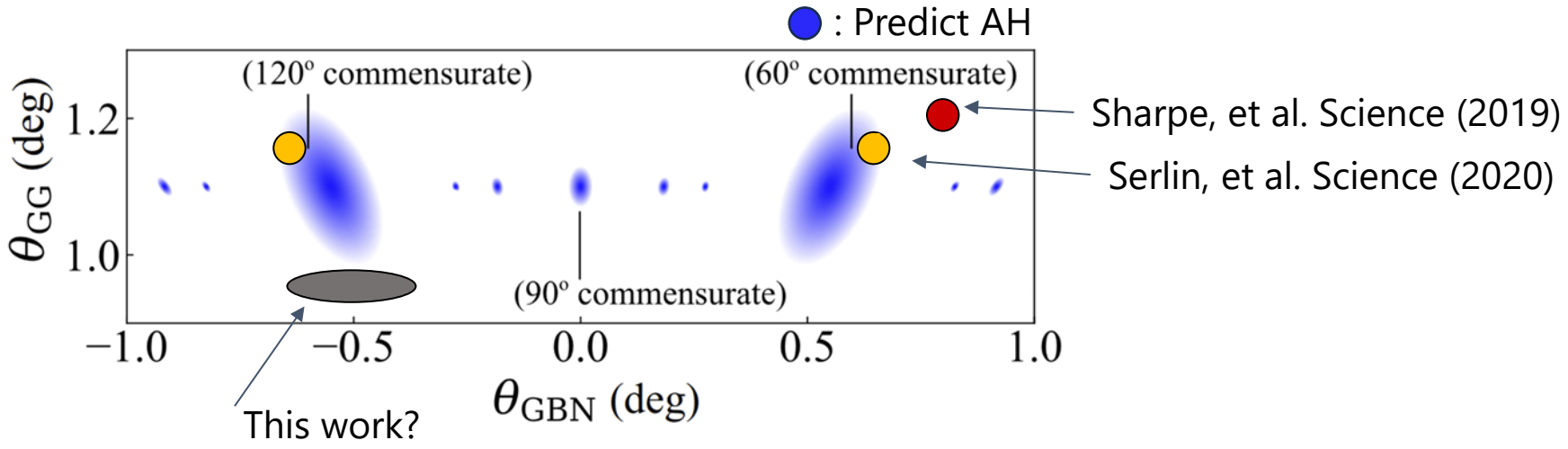


60 commensurate



Shi et al. PRB (2021)

What is θ_{GBN}?



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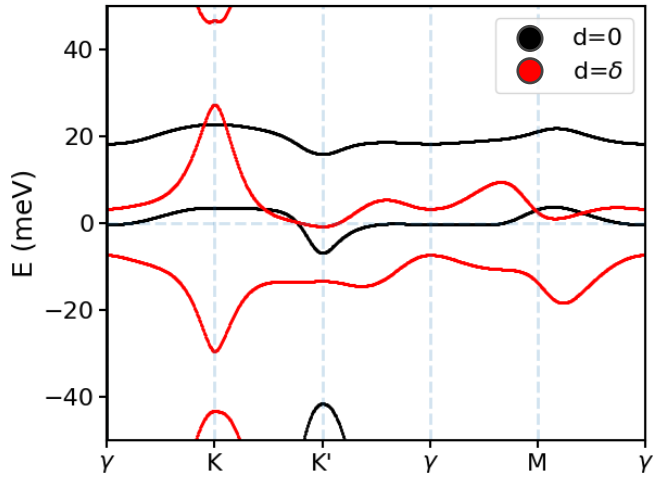
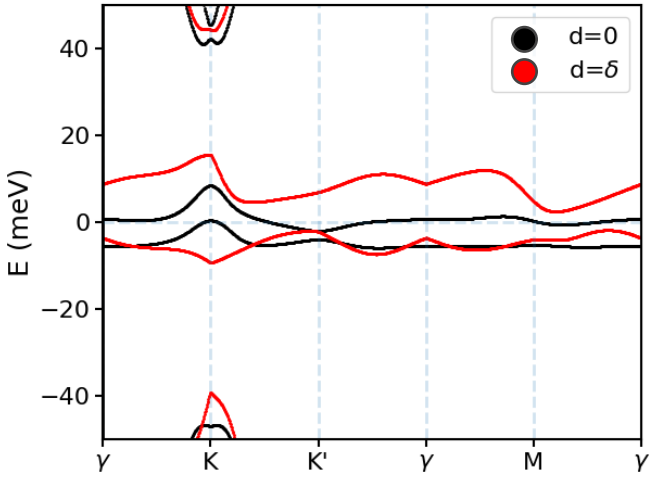
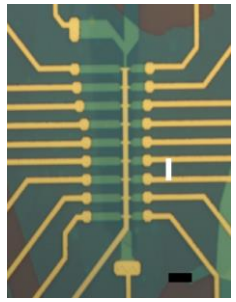


60 commensurate

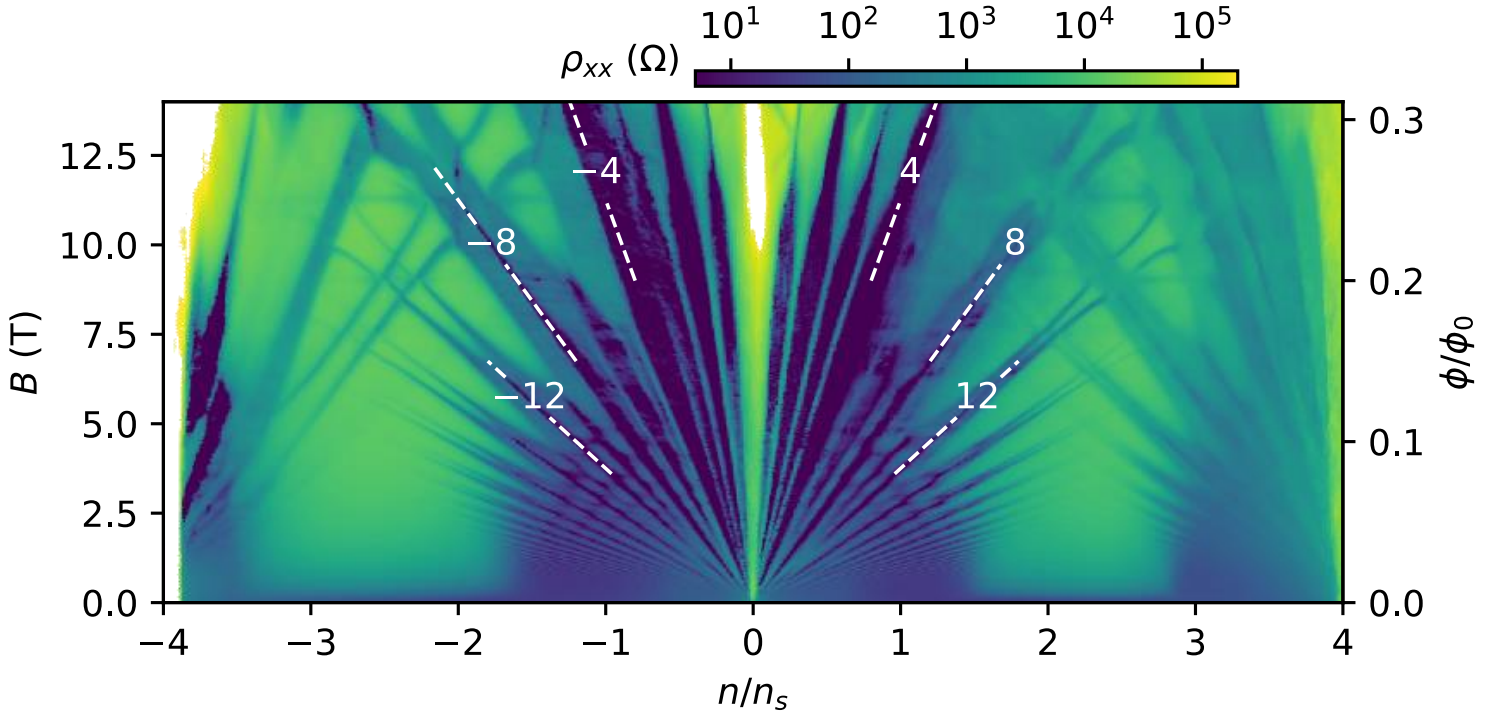


Shi et al. PRB (2021)

TBG is still hiding secrets

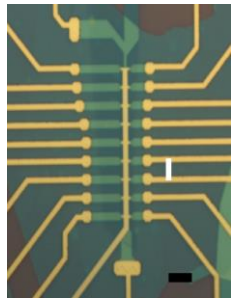


1.38° TBG

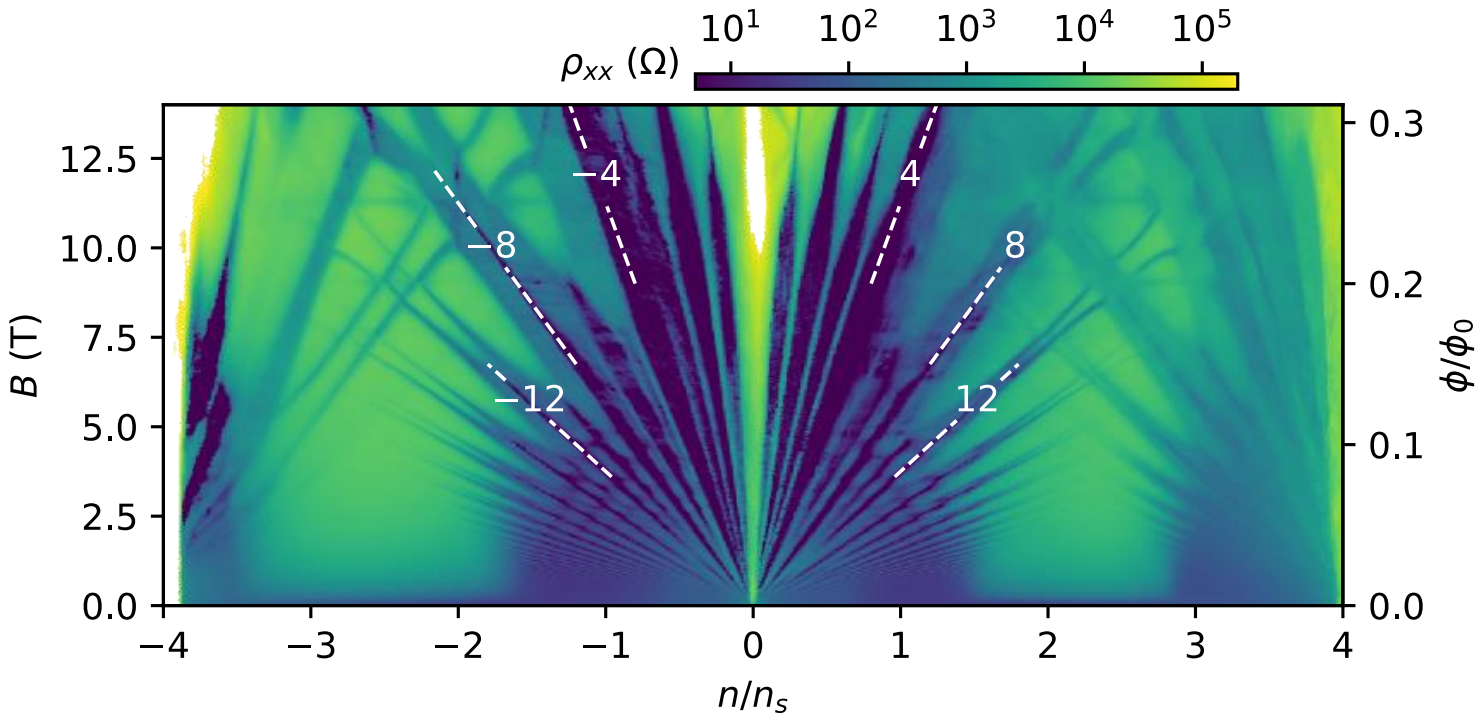


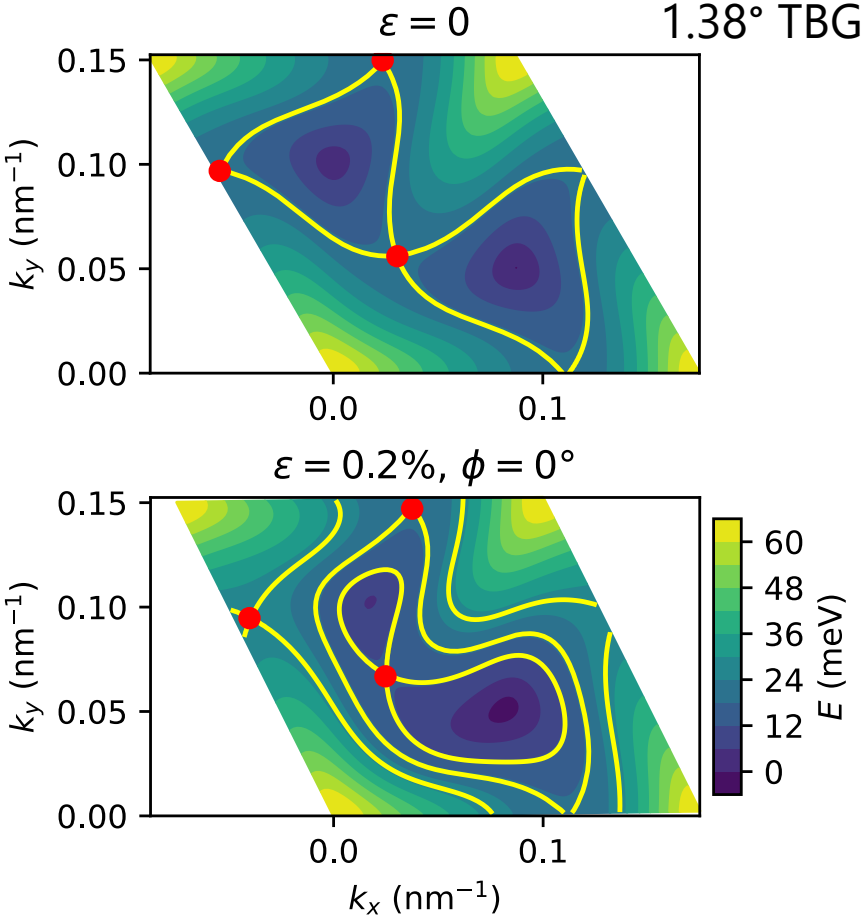
Wang, Finney, Sharpe, et al. PNAS (2023)
Finney, Sharpe, et al. PNAS (2022)

TBG is still hiding secrets



1.38° TBG





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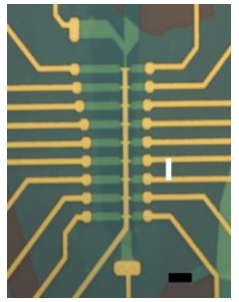
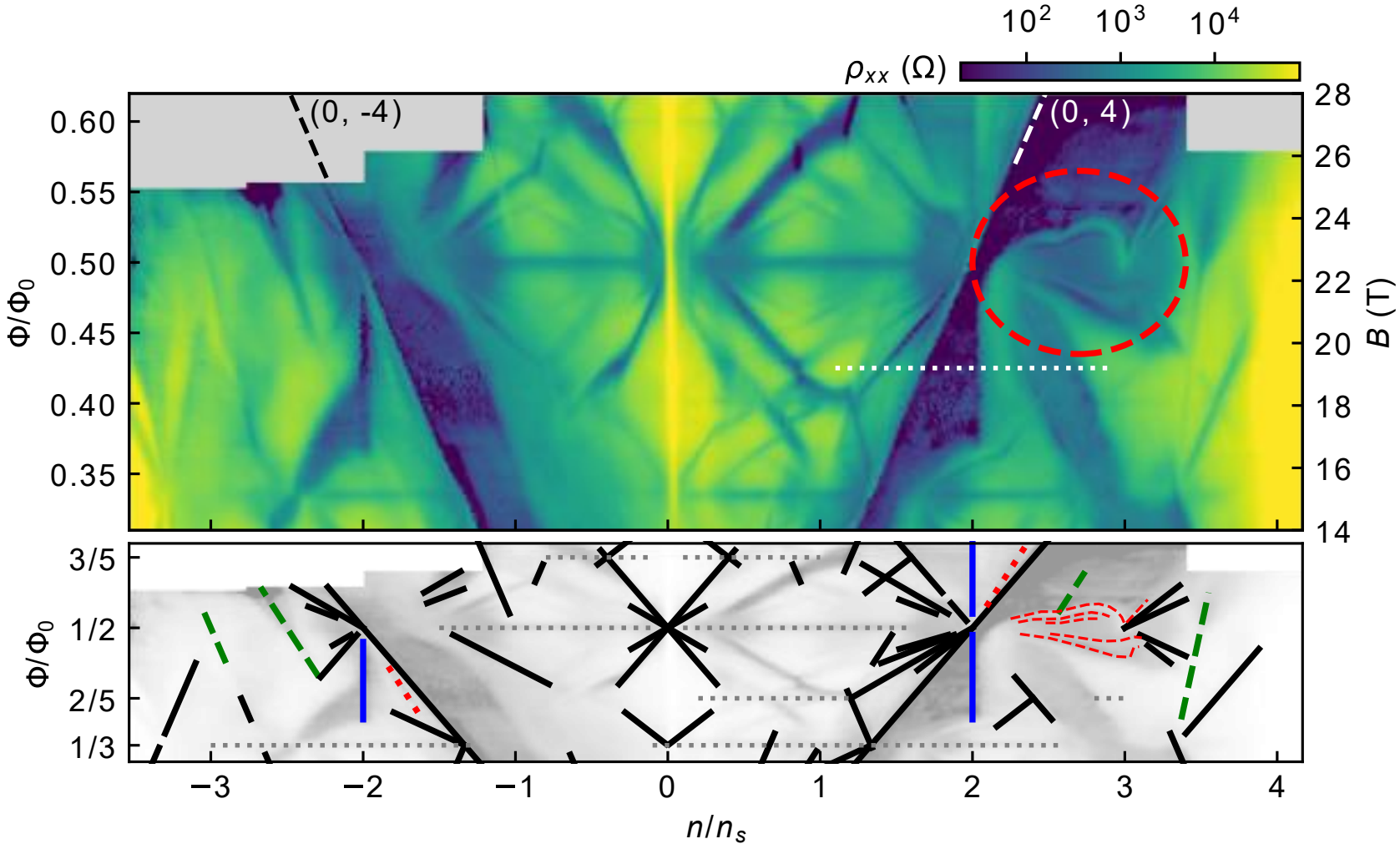
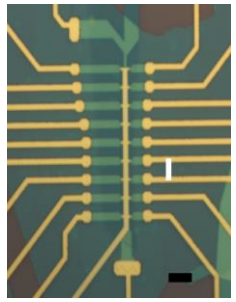


1.38° TBG



TBG is still hiding secrets



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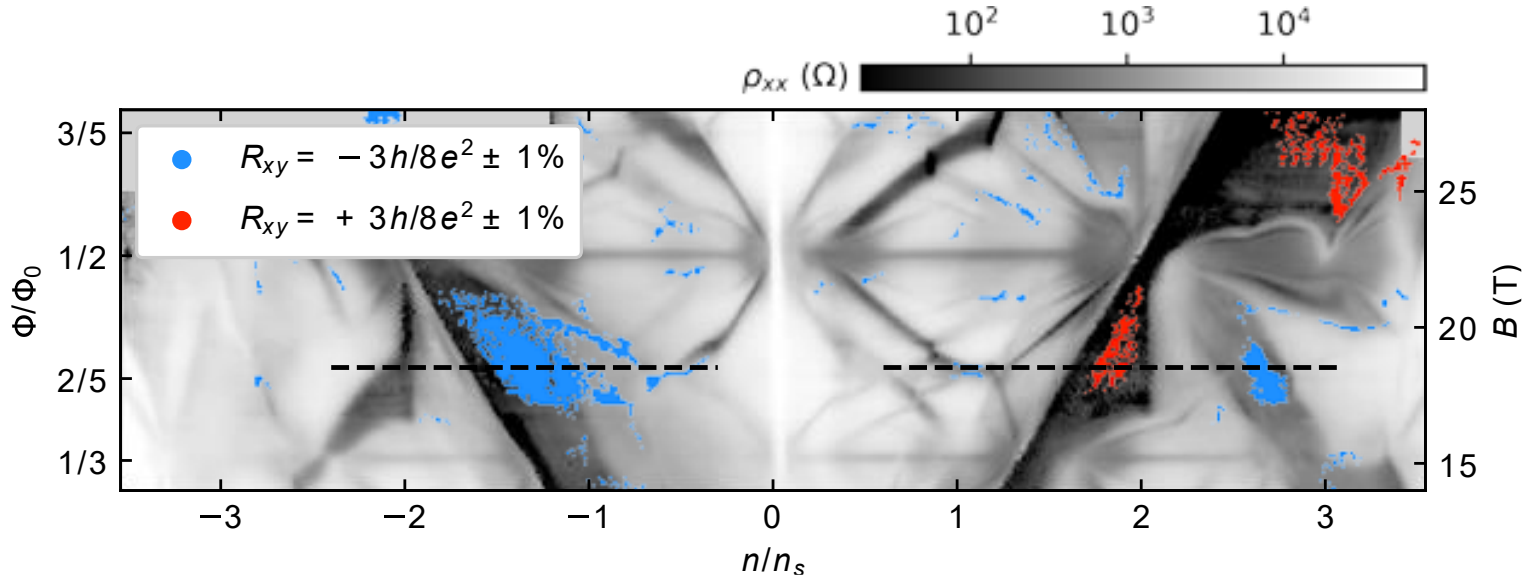
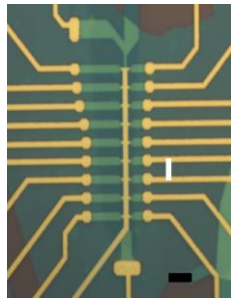


TBG is still hiding secrets



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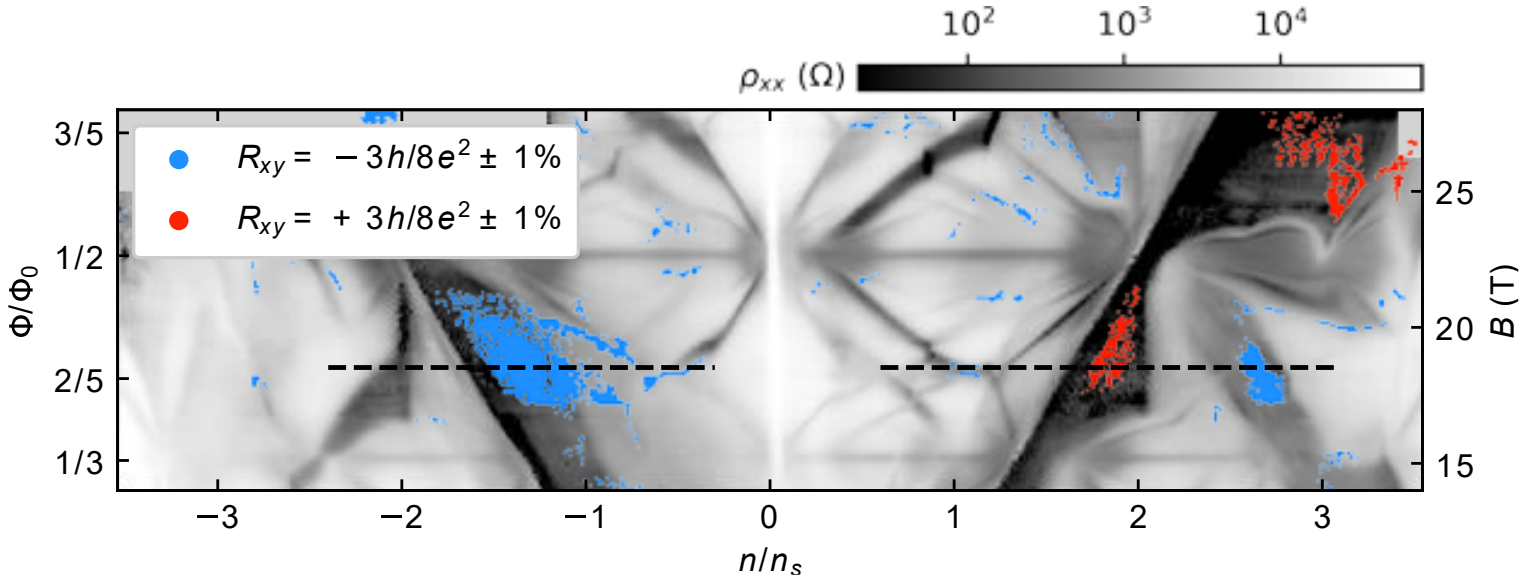
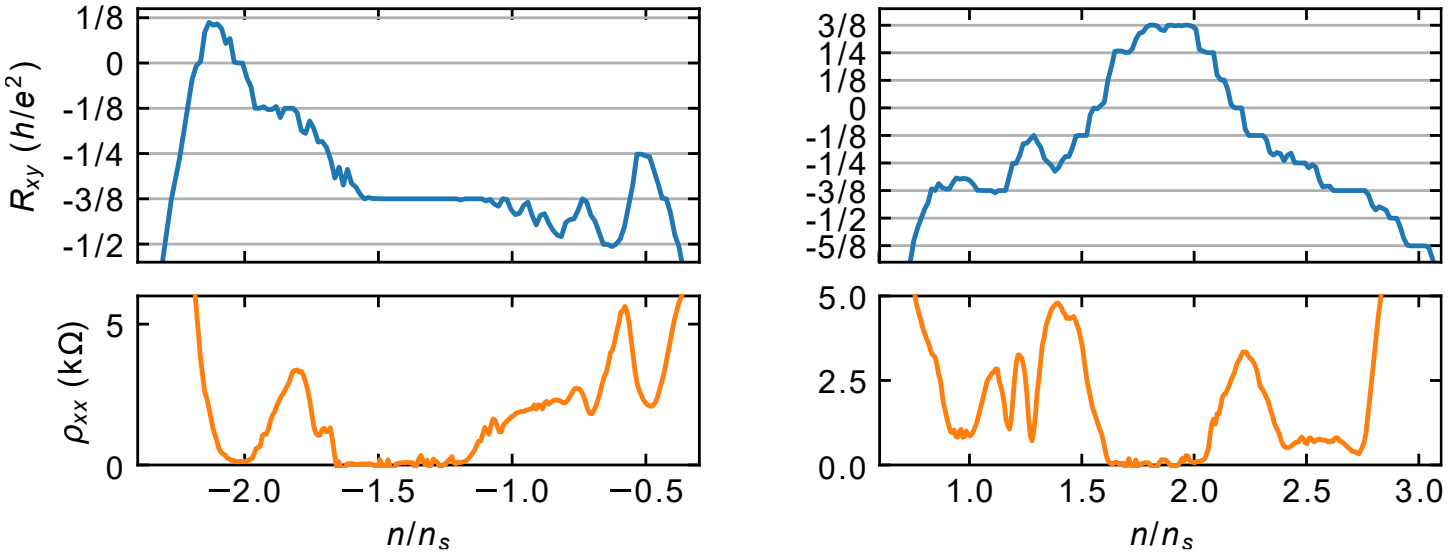
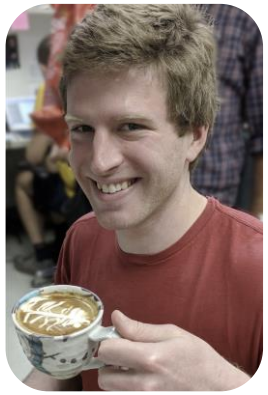




Acknowledgments



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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