

Controlling graphene-hBN rotational alignment

A critical parameter for graphene-based
moiré heterostructures

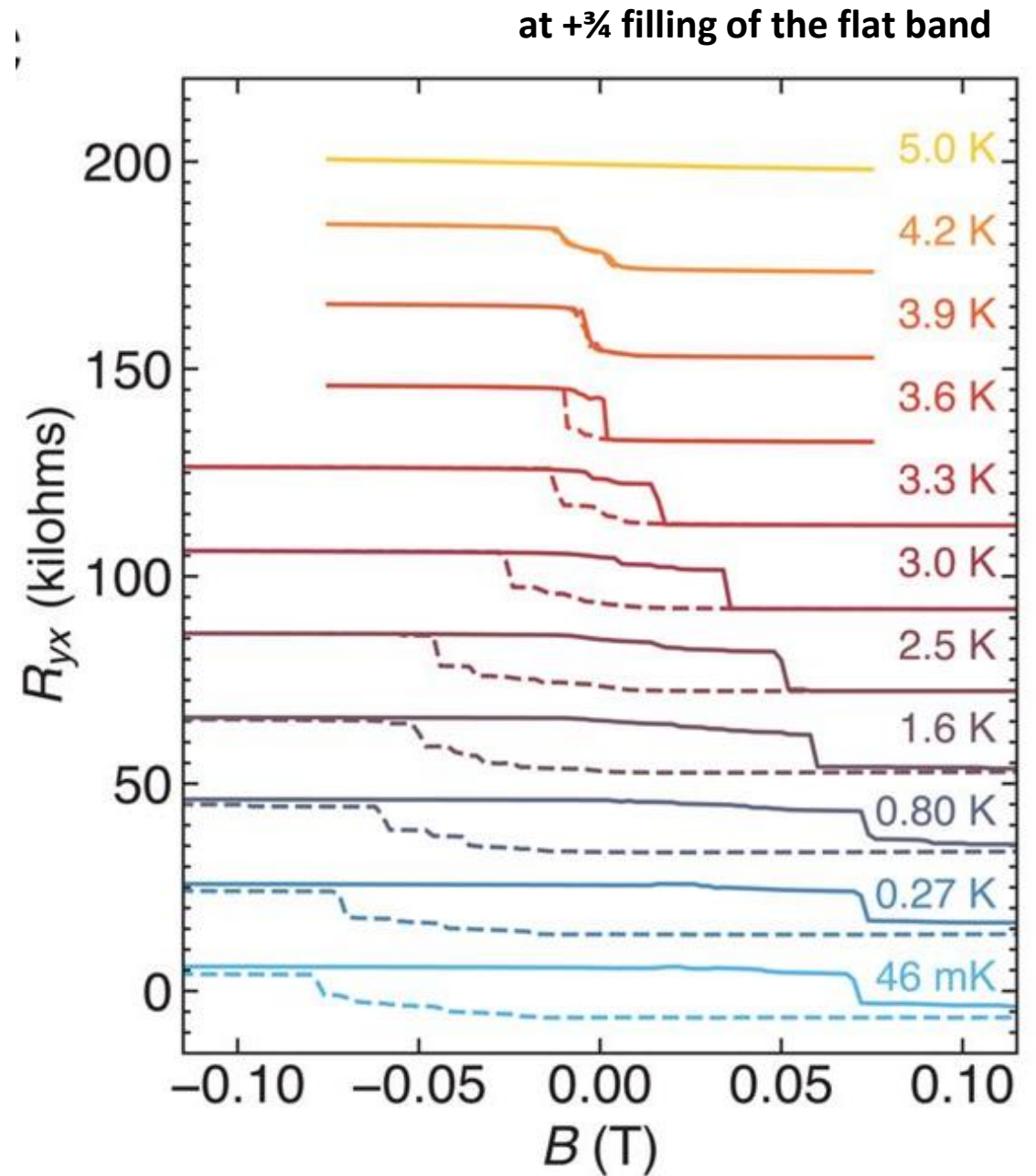
Rupini Kamat

Goldhaber-Gordon Group
Stanford Physics Department
Stanford Institute of Materials and Energy Sciences

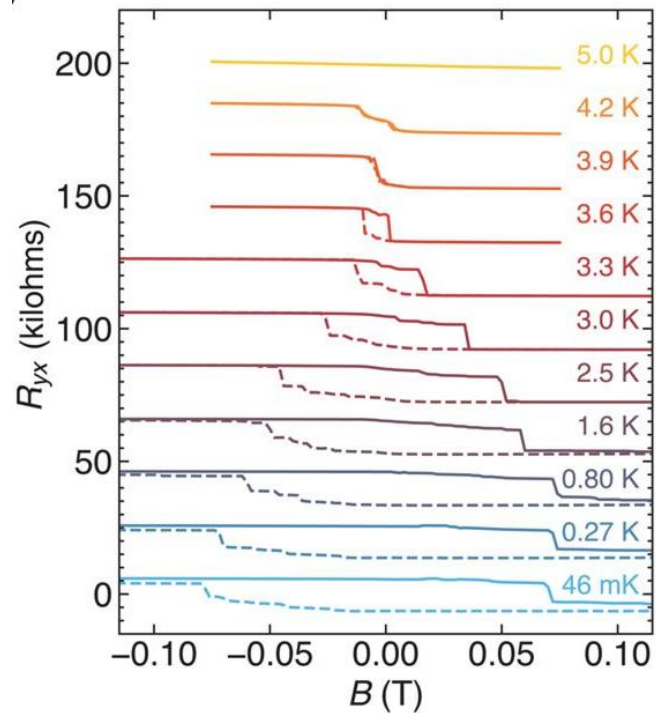
A very special magnet

[1] A. Sharpe, et al. *Science* 365.6453 (2019): 605-608

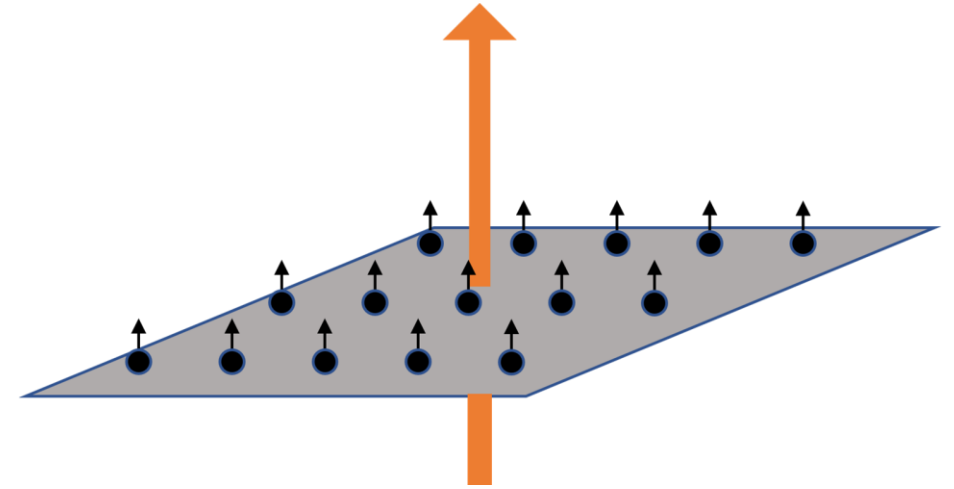
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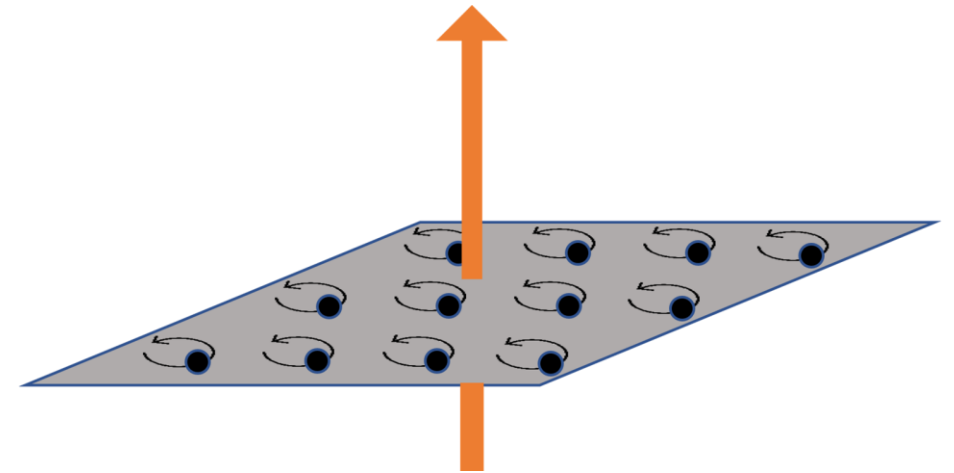
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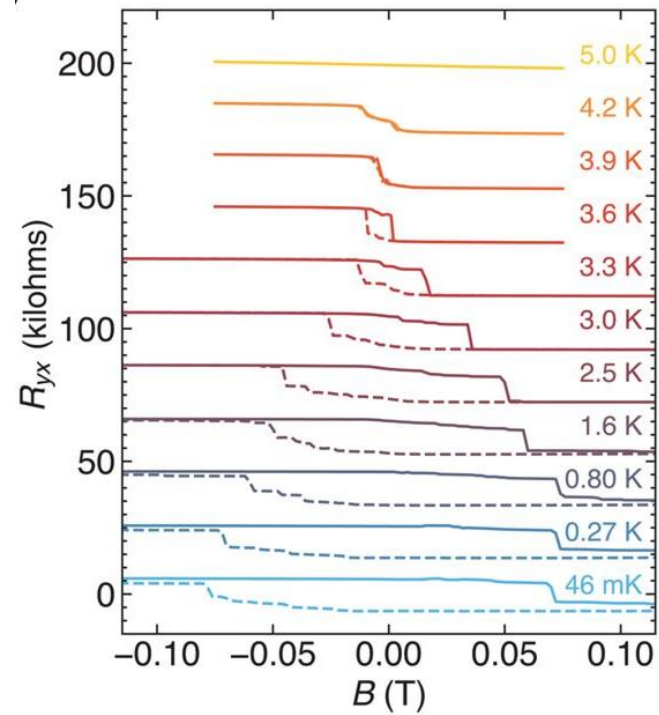


Spin Ferromagnet

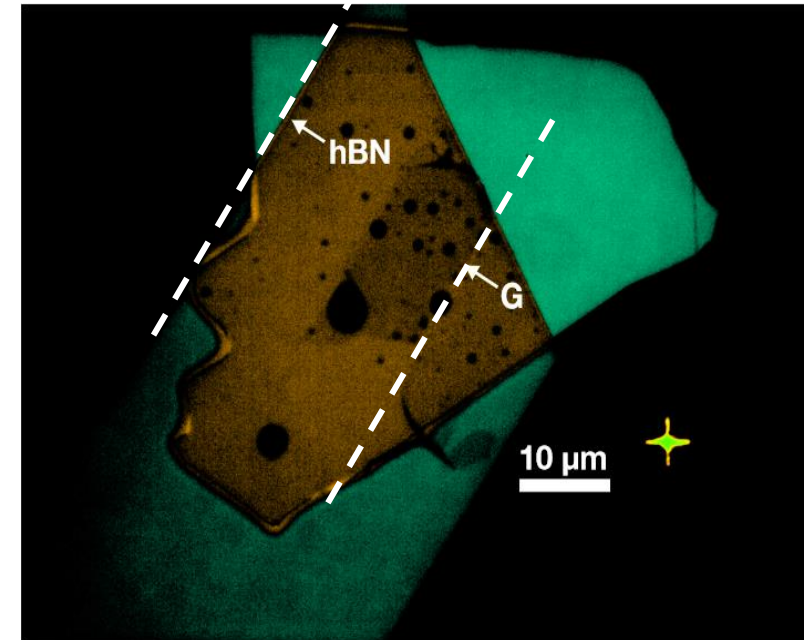
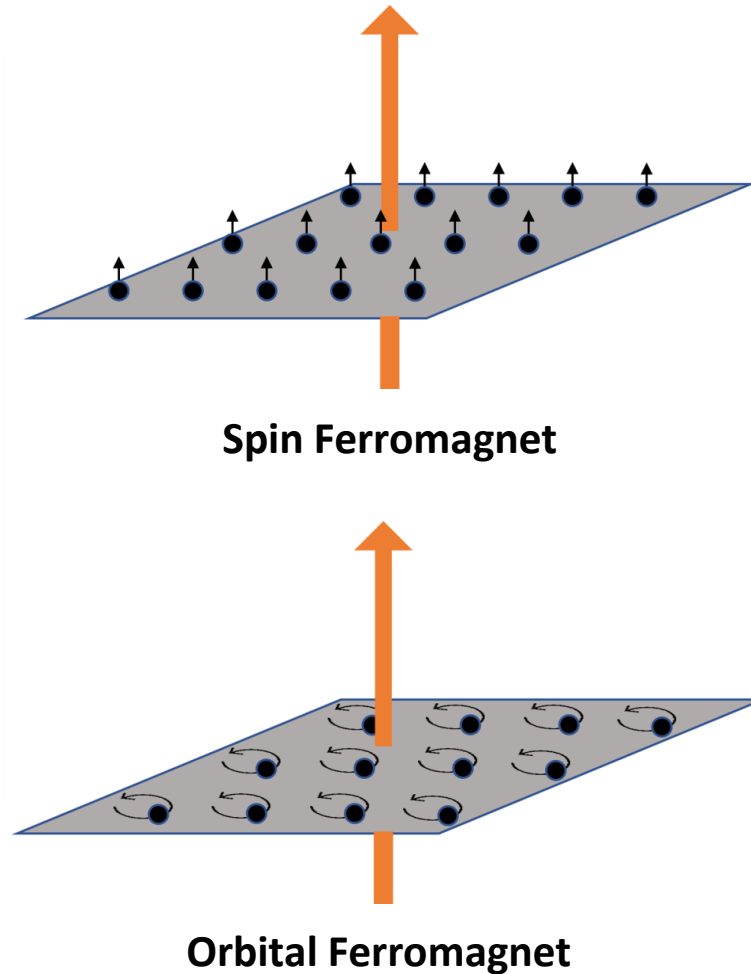


**Orbital
Ferromagnet**

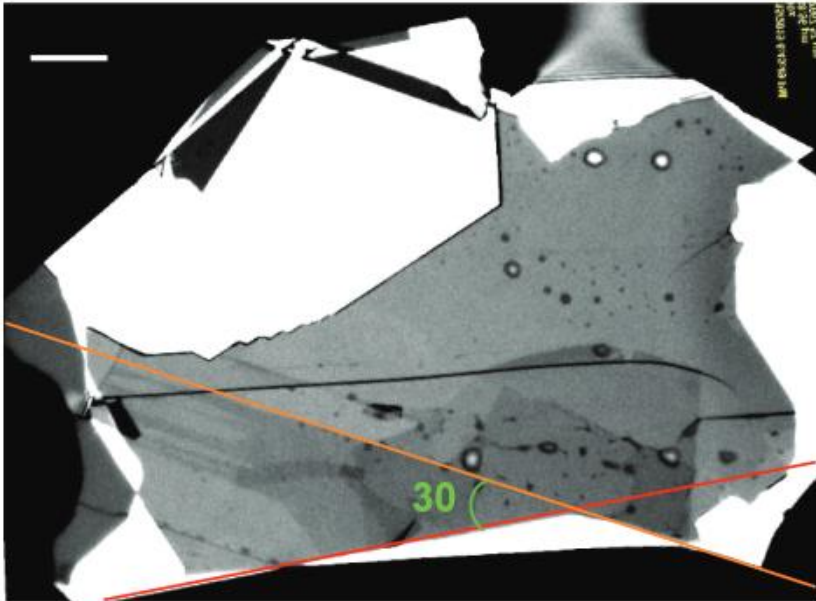
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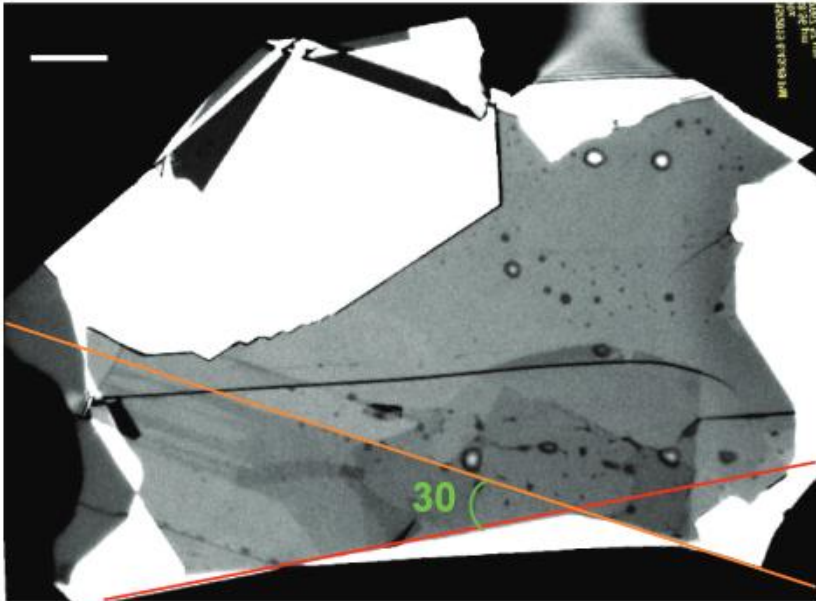


Another hBN-aligned TBG sample

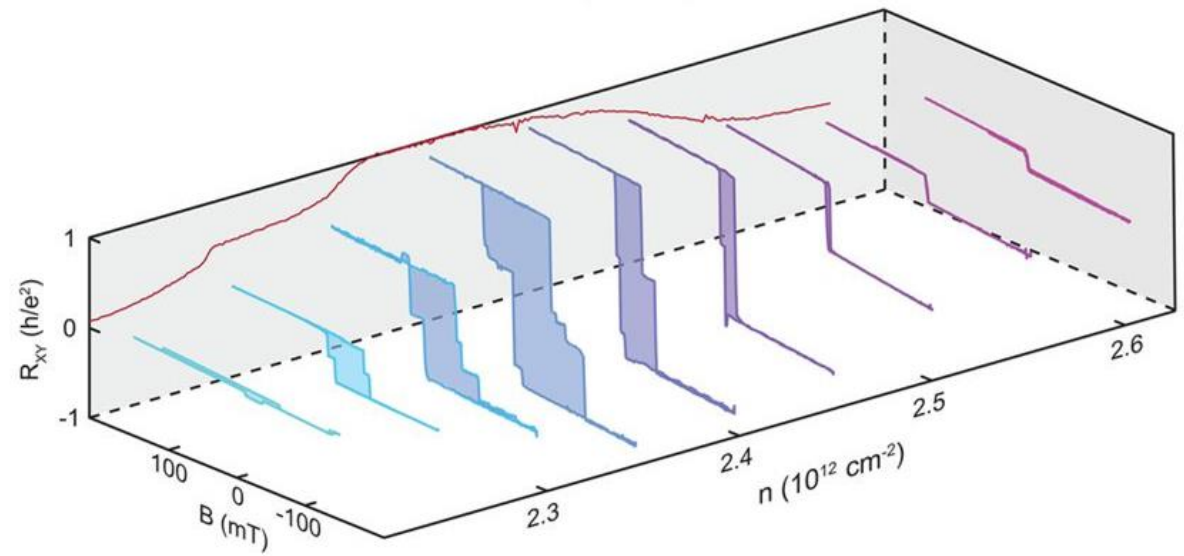


[3] M. Serlin, et al. *Science* 367.6480 (2020): 900-903
Young Group UCSB

Confirmation/extension!



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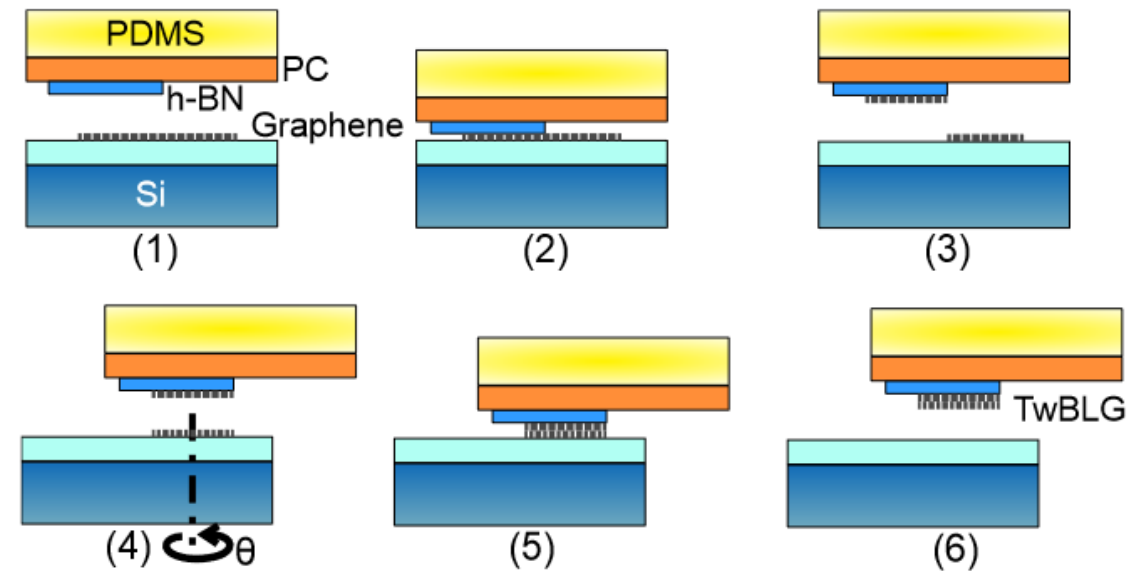


A problem

- ❖ 3 years and hundreds of MATBG samples later...
Still only 2 ferromagnetic TBG samples with this phase diagram
- ❖ Intentionally creating + characterizing a graphene-hBN “hetero moire” has significant challenges

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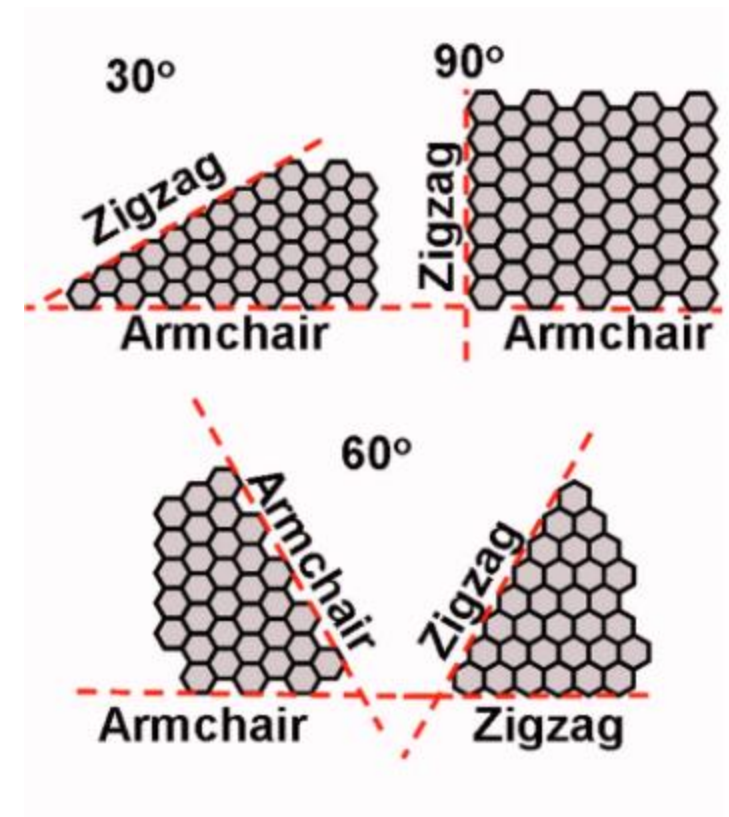
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[4] Y. You, et al. *Applied Physics Letters* 93.16 (2008): 163112.
Shen ZeXiang Group, NTU

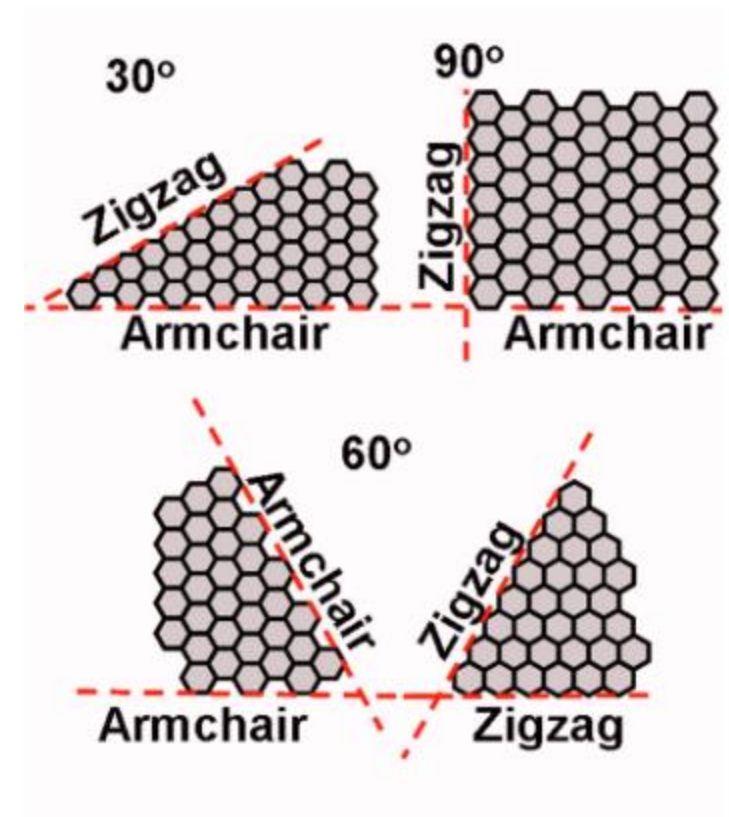


	ZZ	AC
ZZ	✓	✗
AC	✗	✓

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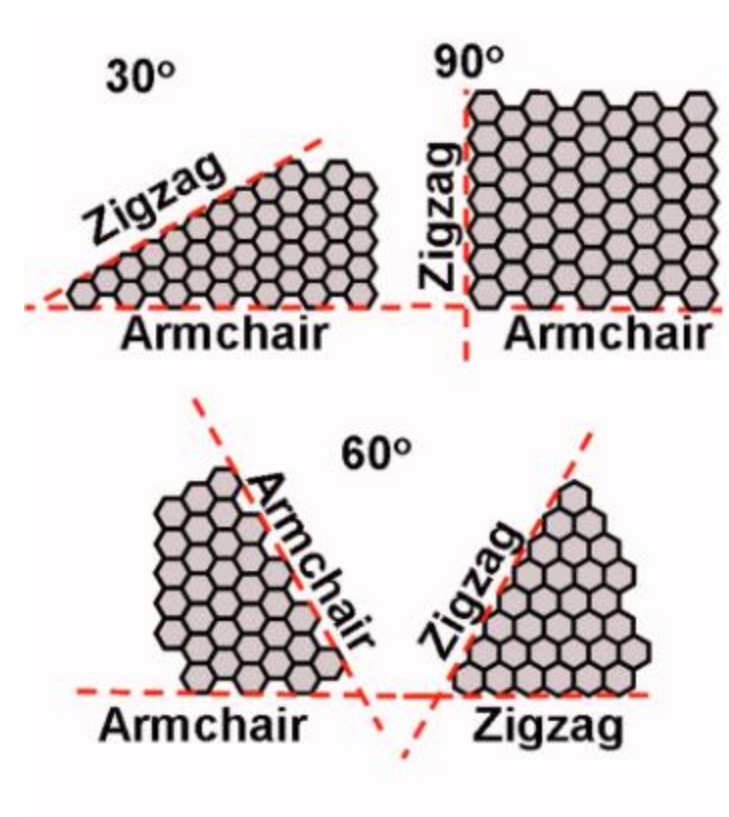
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Need greater control.

Strategy: 1) Characterize straight edges in flakes as ZZ or AC prior to stacking
2) Characterize moiré period after stacking.

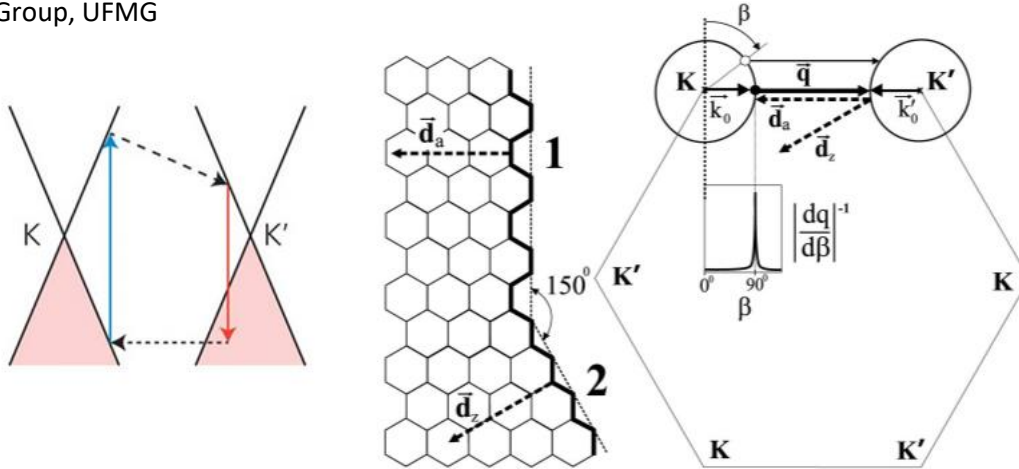
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Graphene Orientation: Polarized Raman

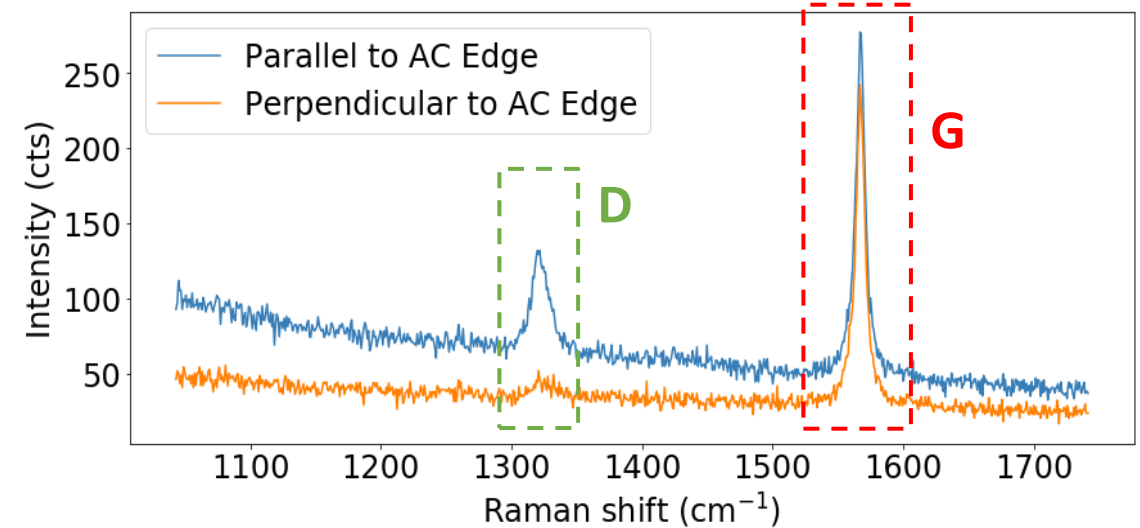
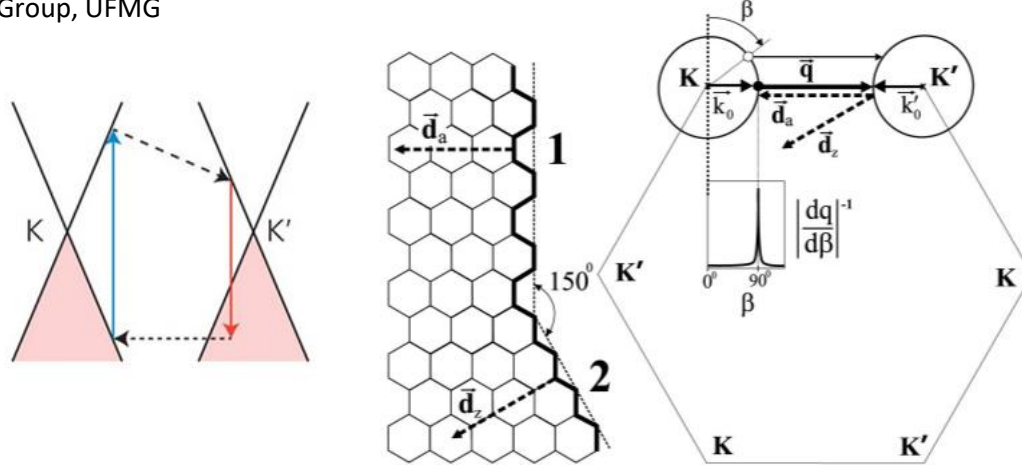
[5] L. G. Cancado, et al. *Physical review letters* 93.24 (2004): 247401.
Jorio Group, UFMG



- ❖ Armchair edges can be quickly identified by Raman spectroscopy
- ❖ Scattering off armchair edges gives rise to otherwise forbidden D peak in pristine graphene

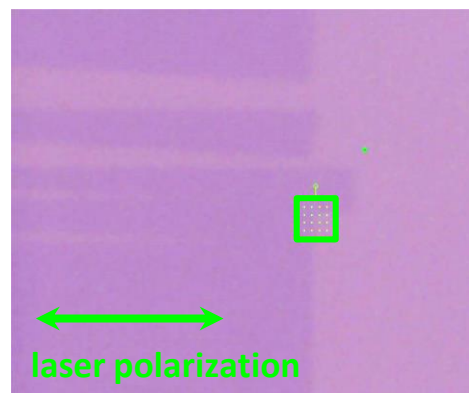
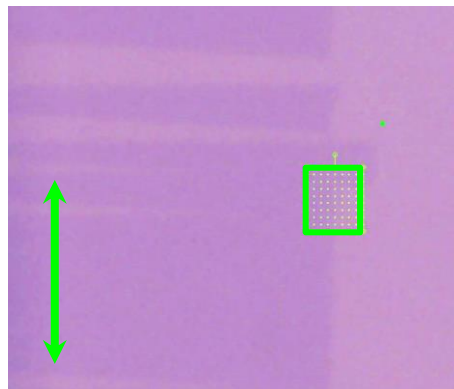
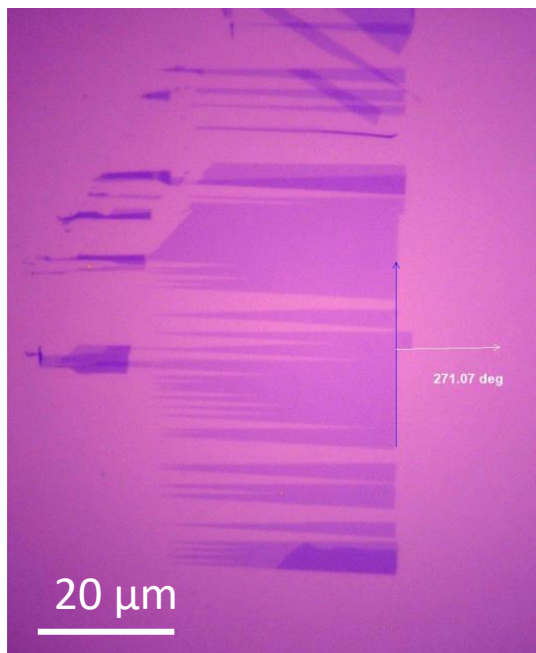
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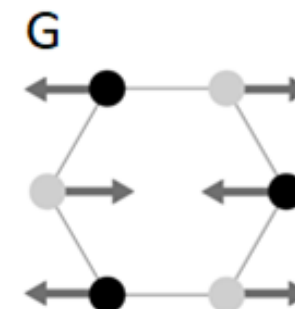
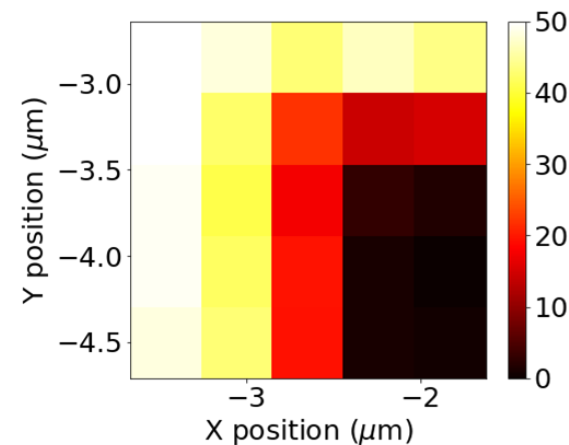
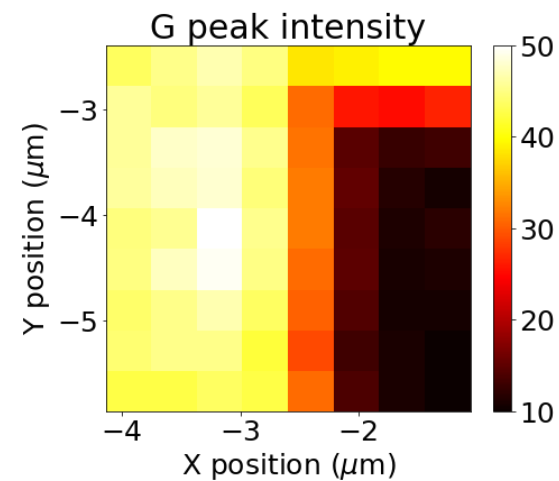
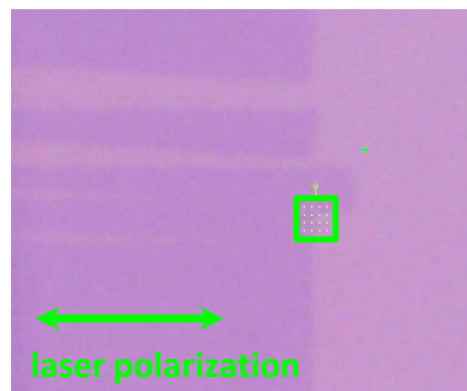
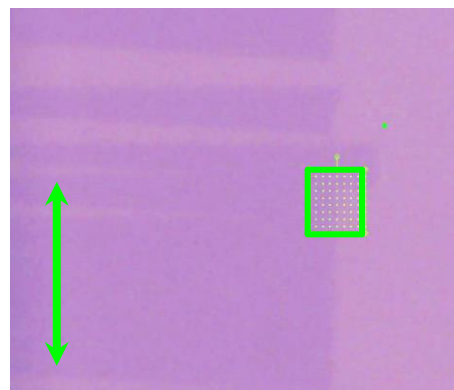
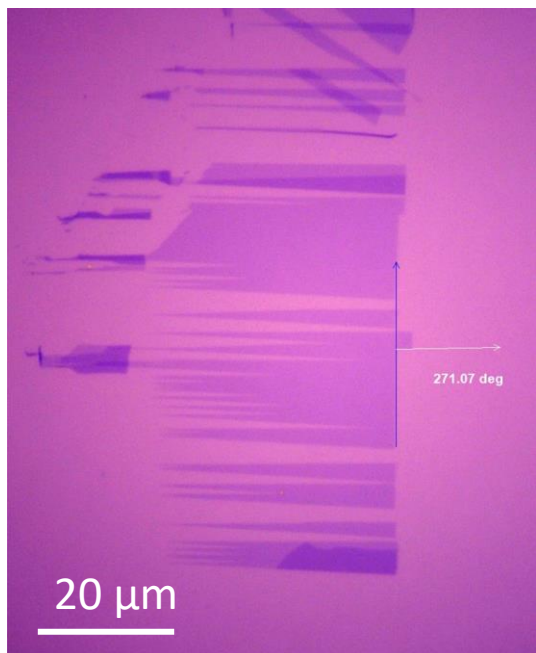


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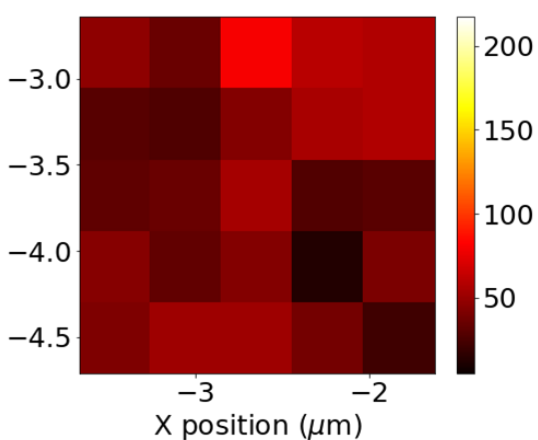
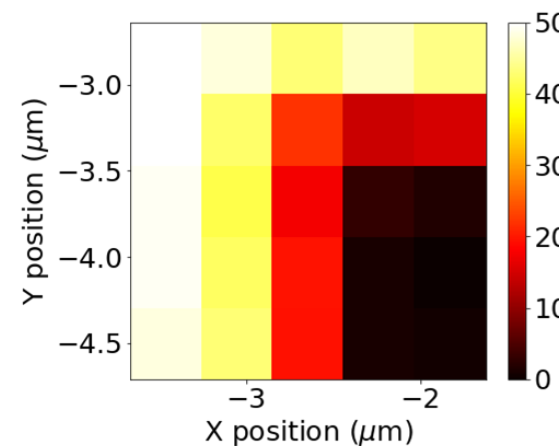
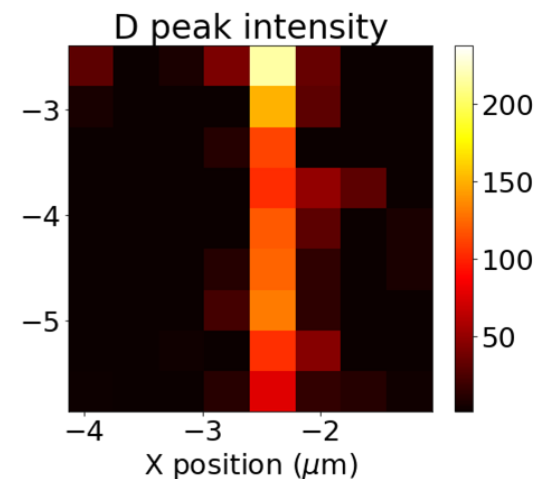
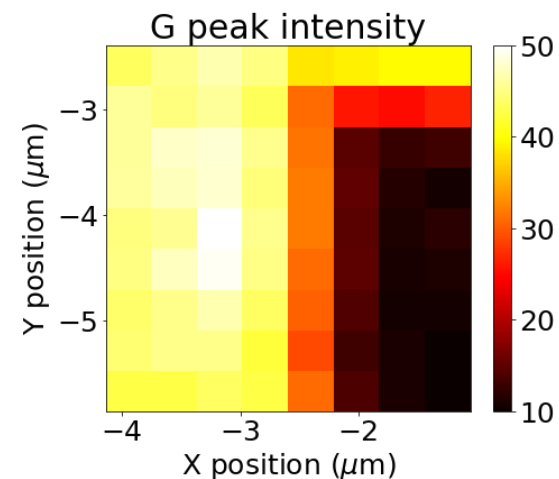
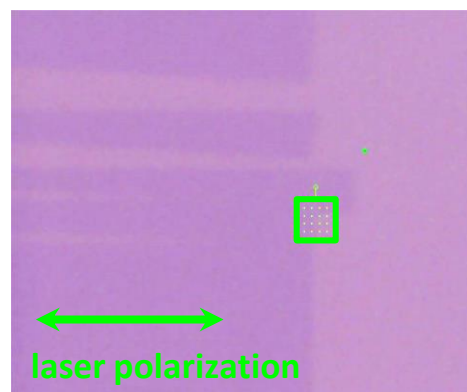
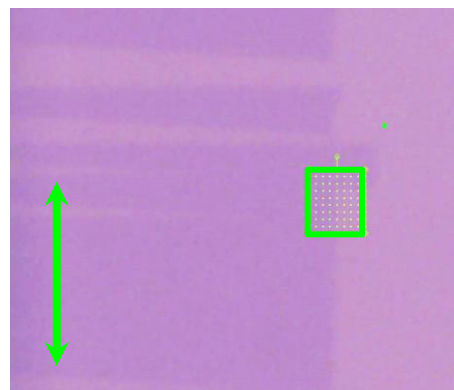
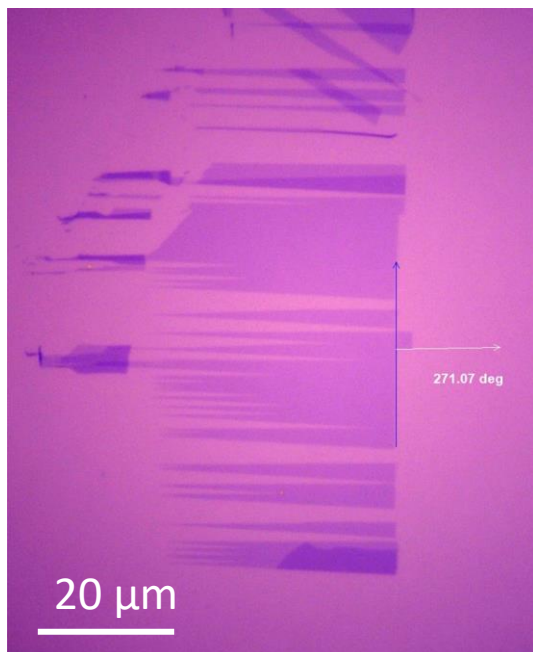
Raman Characterization of Graphene Orientation: An Example



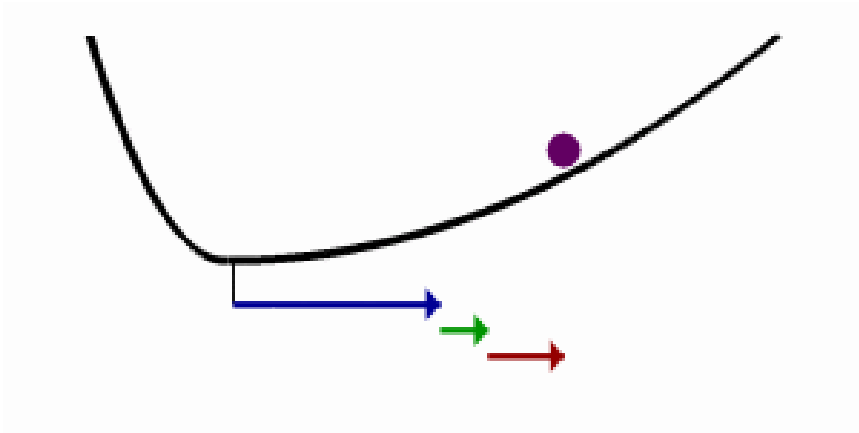
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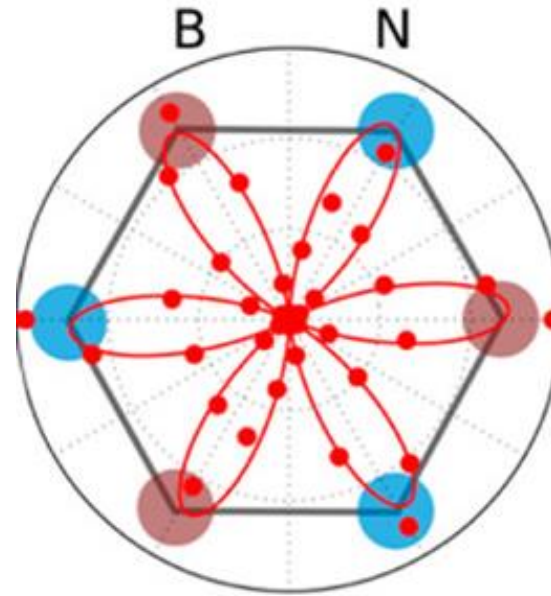
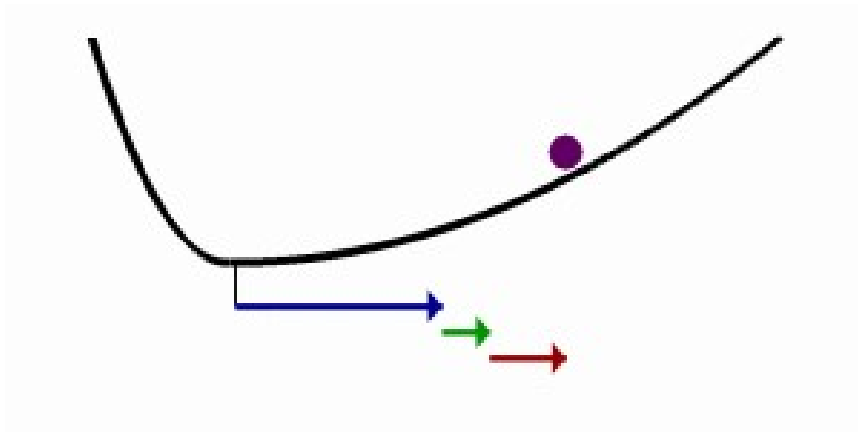


hBN Orientation: Second Harmonic Generation



- ❖ Non-centrosymmetric materials pumped by laser light at frequency f emit a second harmonic signal at $2f$

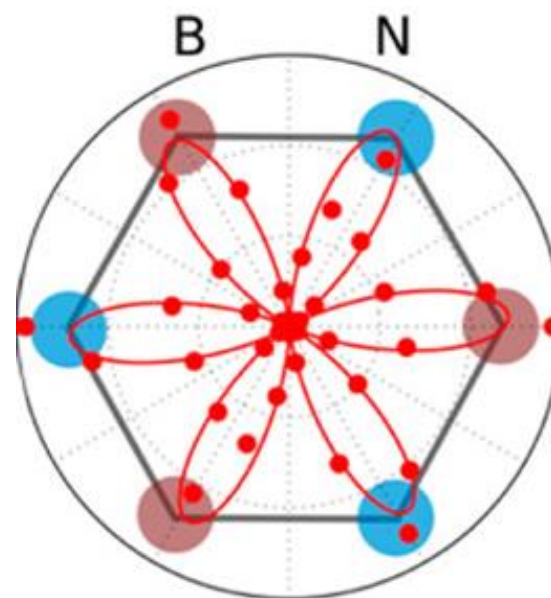
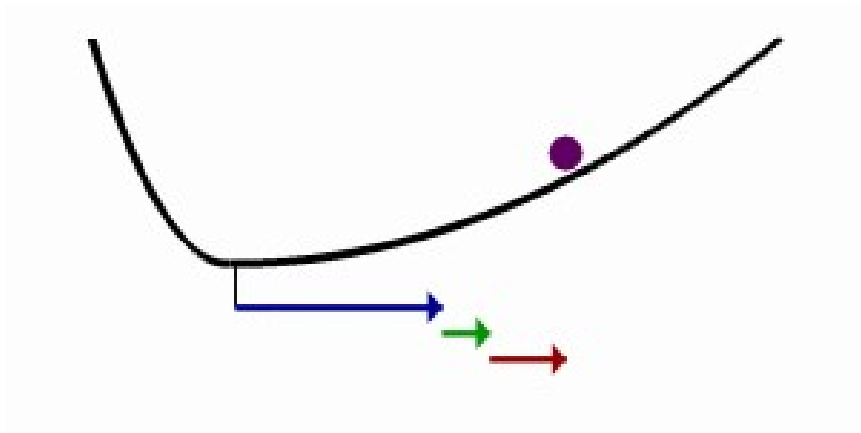
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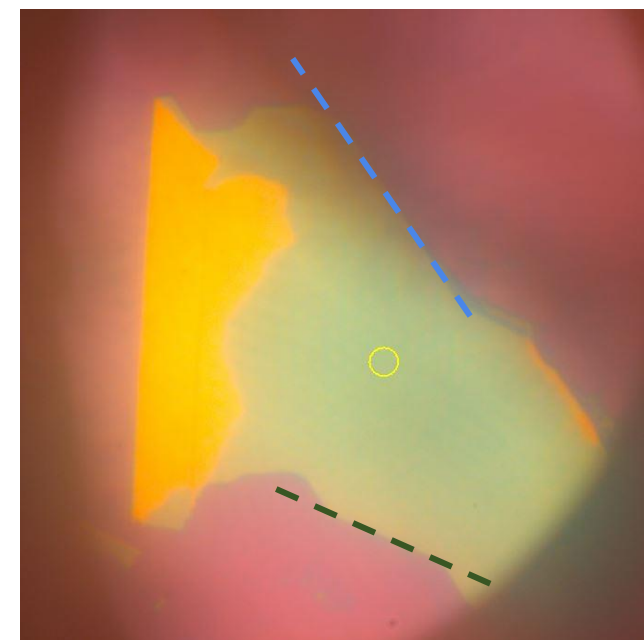
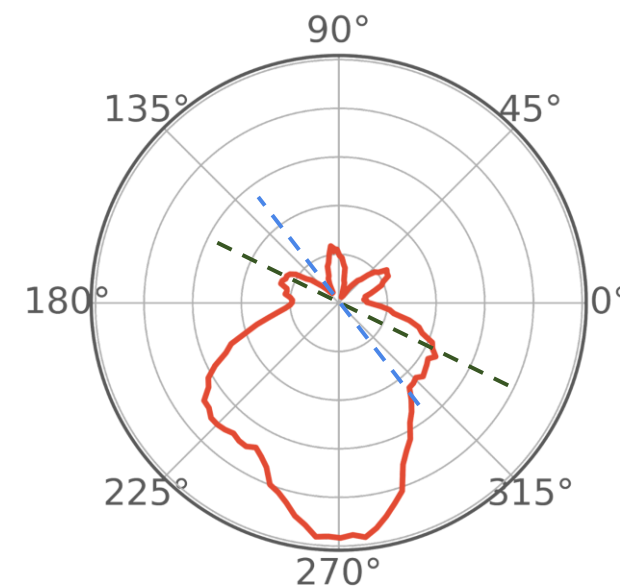
[6] Y. Li, et al. *Nano letters* 13.7 (2013): 3329-3333.
Heinz Group, Stanford

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- ❖ “Petals” of polarization-resolved SHG shows orientation of B-N bonds

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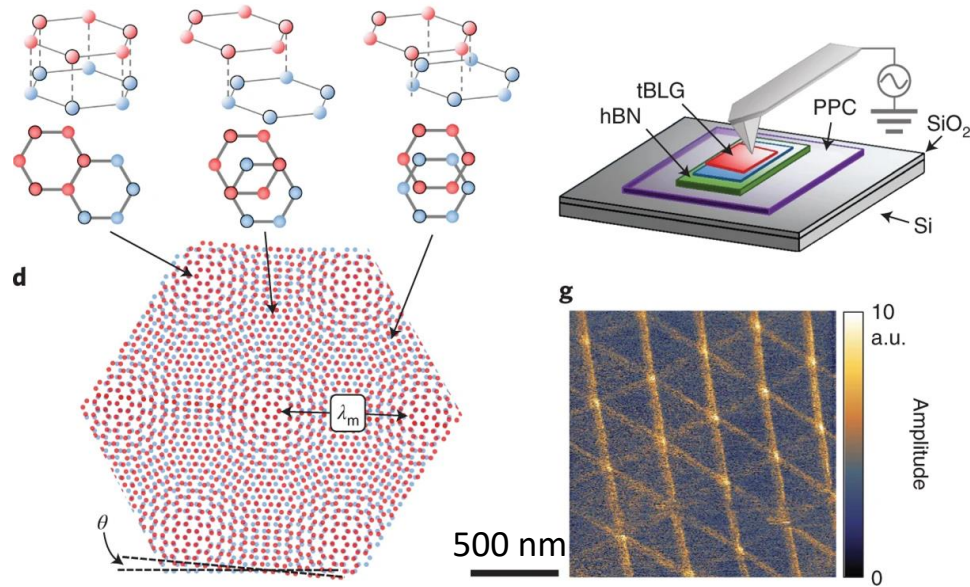
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Post-Stack Characterization: Piezoresponse Force Microscopy (PFM)

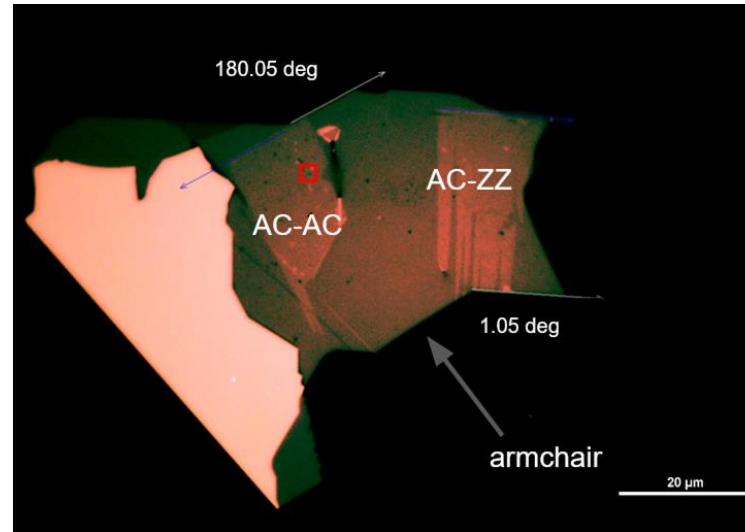
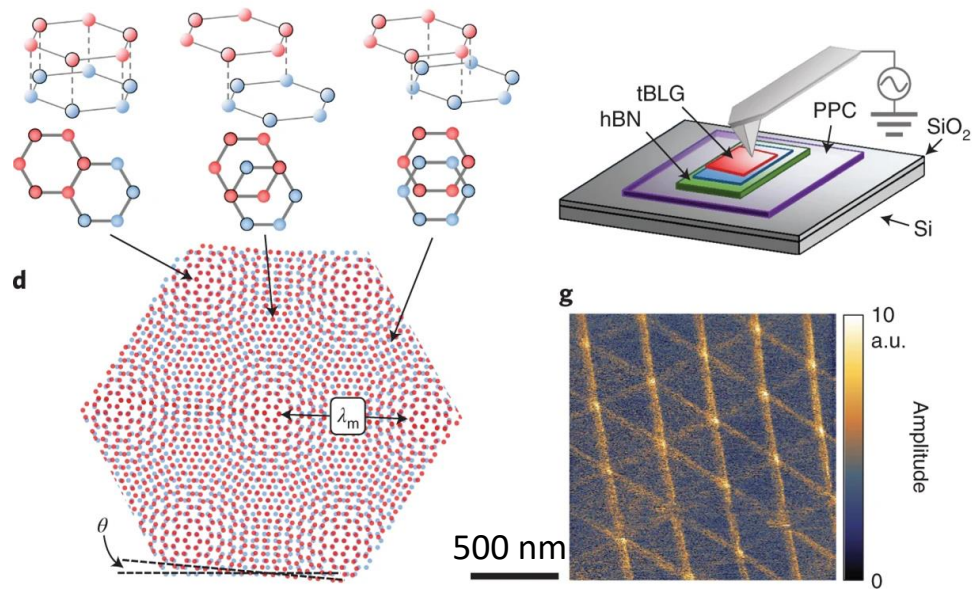
[7] L. McGilly, et al. *Nature Nanotechnology* 15.7 (2020): 580-584.
Dean Lab, Columbia



Rapid verification of alignment + direct measurement of moiré done via PFM, which measures local electromechanical response of material

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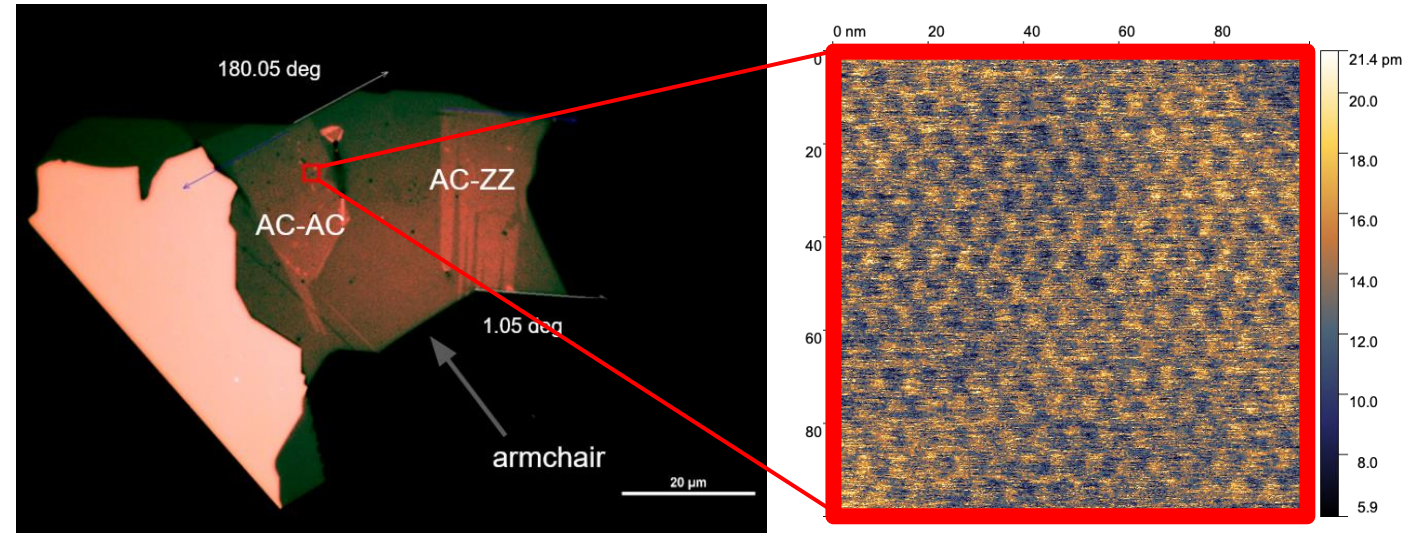
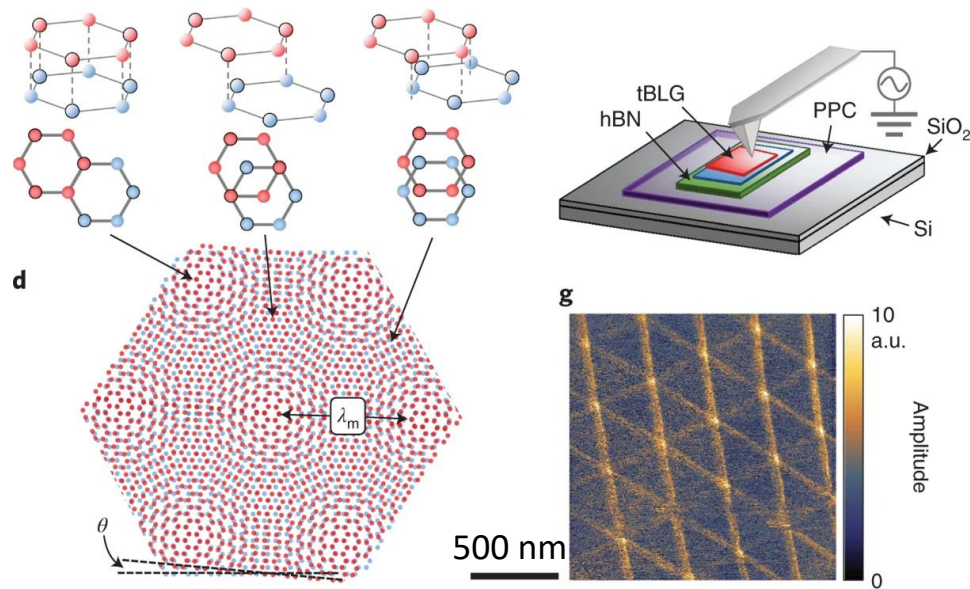
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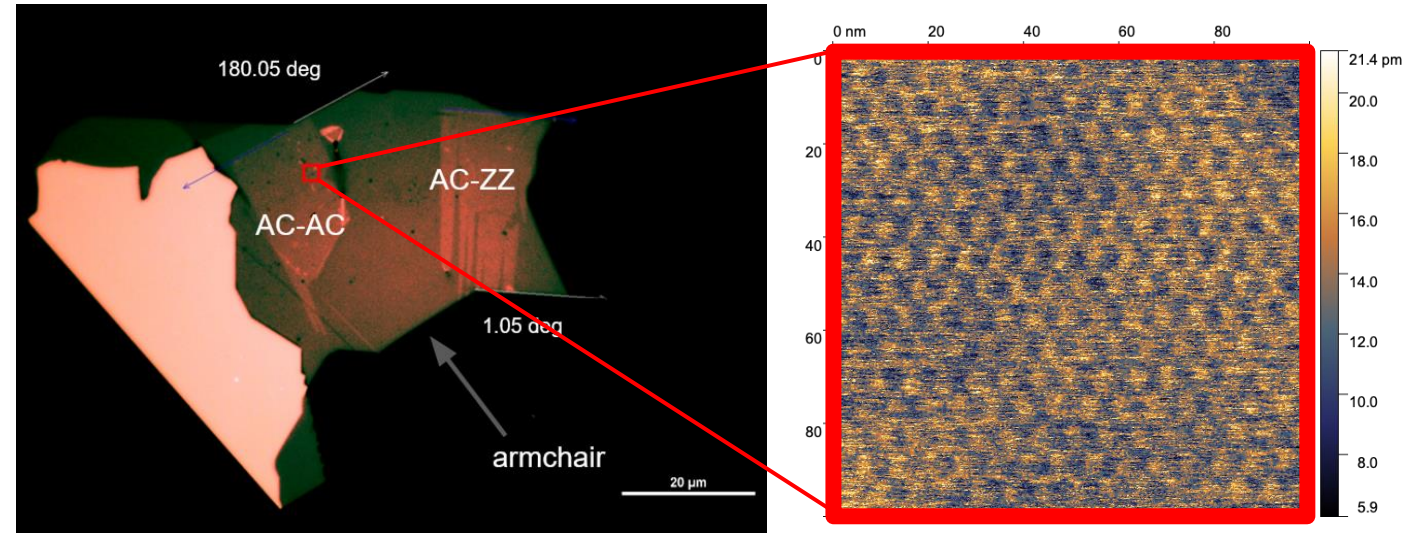
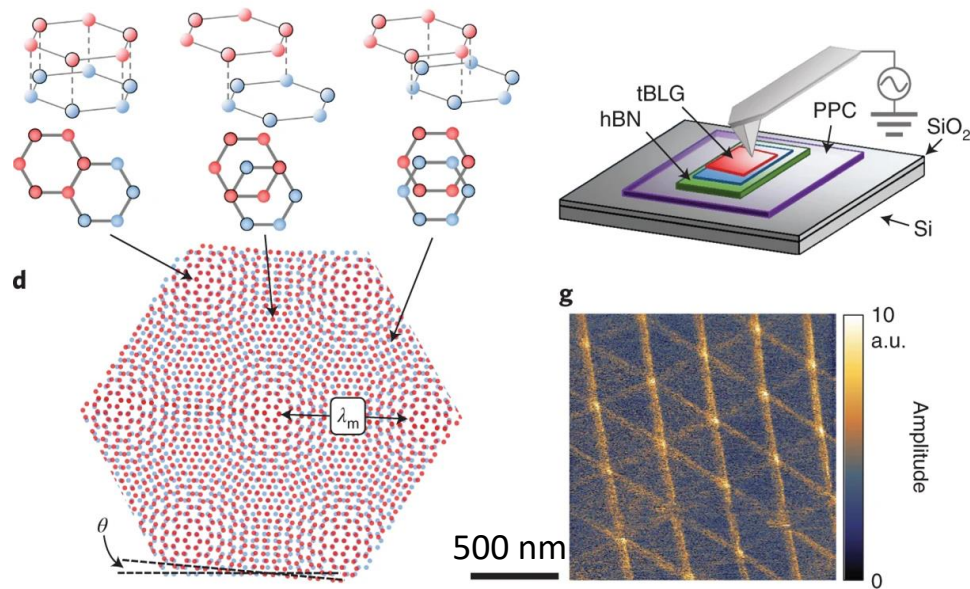


Early results confirm successful alignment using pre-stack characterization!

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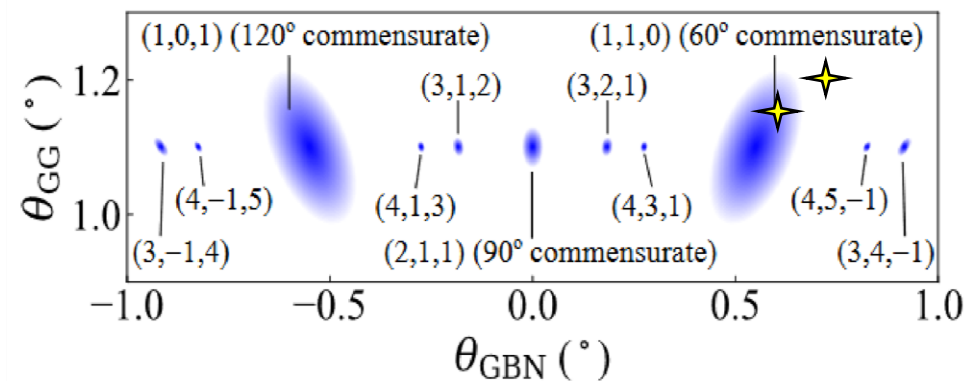


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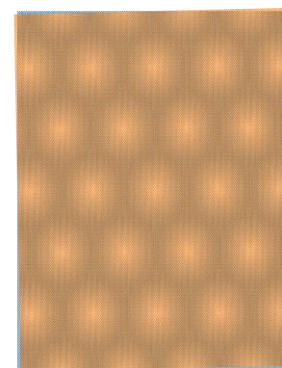
Early results confirm successful alignment using pre-stack characterization!

Precise alignment remains a challenge, moiré period ~ 7.2 nm moiré period $\rightarrow 1.69$ deg twist angle

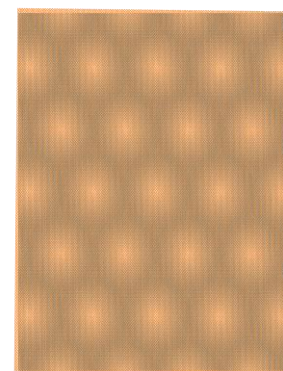
Outlook



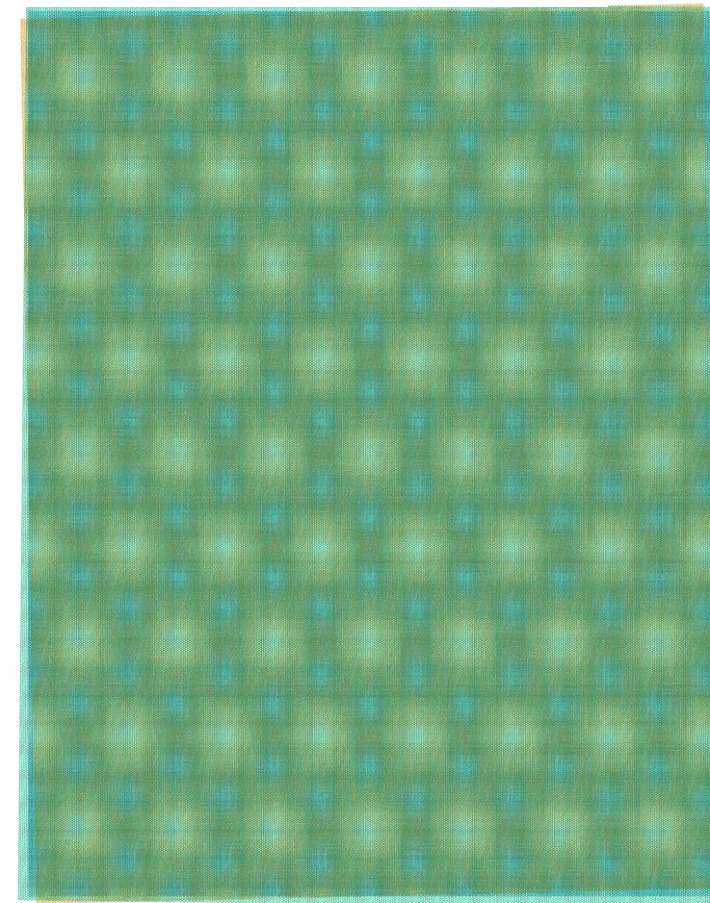
[8] J. Shi, et al. *Physical Review B* 103.7 (2021): 075122. MacDonald Group, UT Austin



$\theta_{\text{GG}} = 1.15^\circ$



$\theta_{\text{GBN}} = -0.6^\circ$

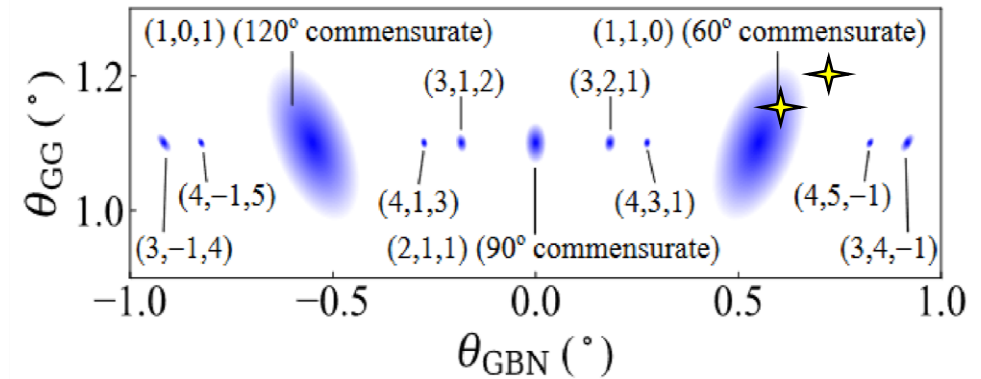


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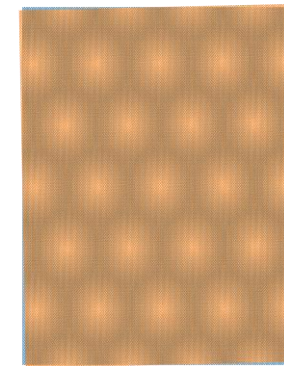
- ❖ Process flow of pre-stack characterization/stacking/post-stack characterization enables new degree of control AND verification of Gr-hBN moiré

Combining these tools can enable:

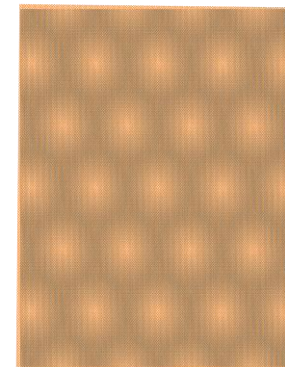
- 1) reliable reproduction of novel correlated electron states (like orbital ferromagnetism in TBG) that may depend on this supermoiré
- 1) systematic exploration of device behavior as a function of graphene-hBN alignment.



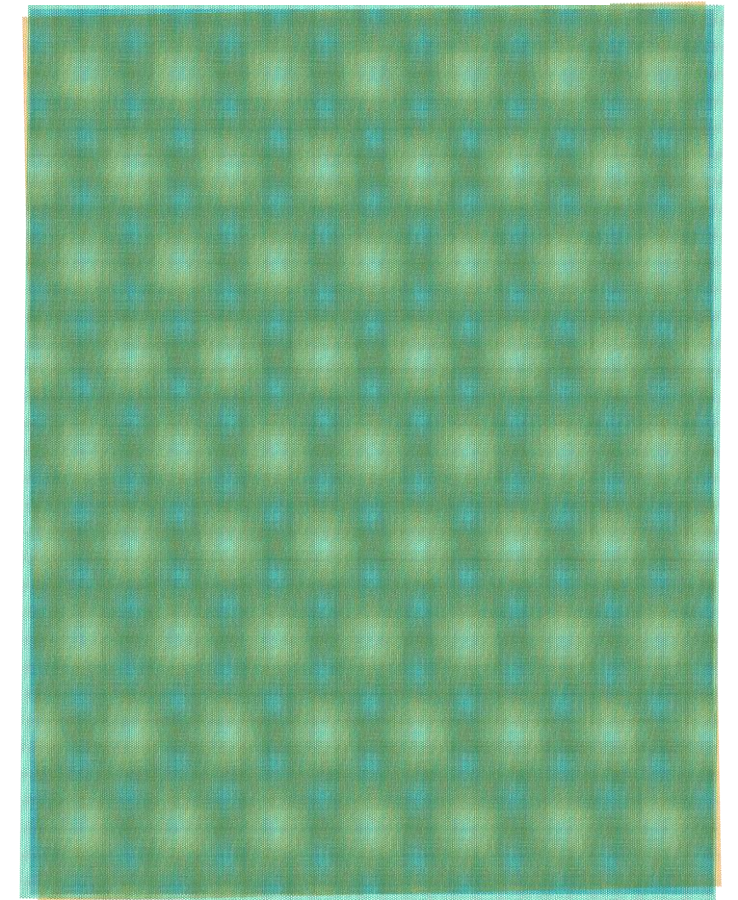
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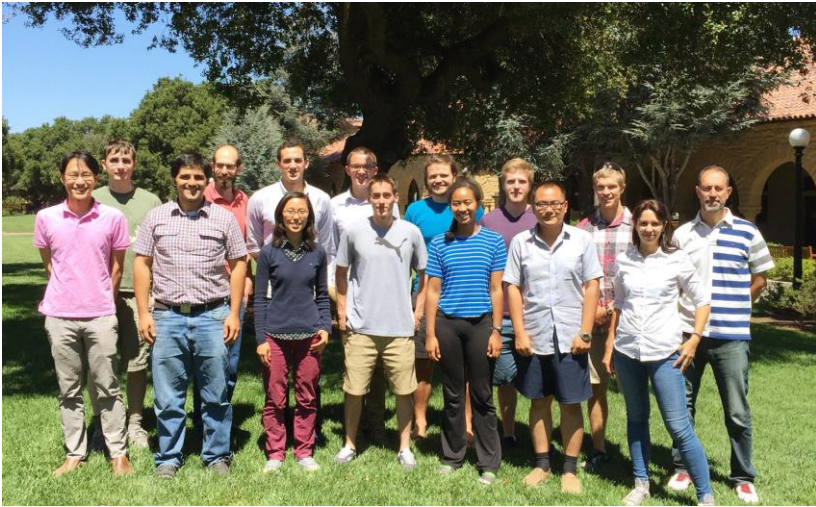
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Acknowledgments



Goldhaber-Gordon Group



Sandia National Labs:

Aaron Sharpe

Goldhaber-Gordon Group:

Greg Zaborski, Mihir Pendharkar,
Marc Kastner, David Goldhaber-
Gordon

Tony Heinz Group:

Jenny Hu, Tony Heinz

NIMS:

Takashi Taniguchi, Kenji

Watanabe

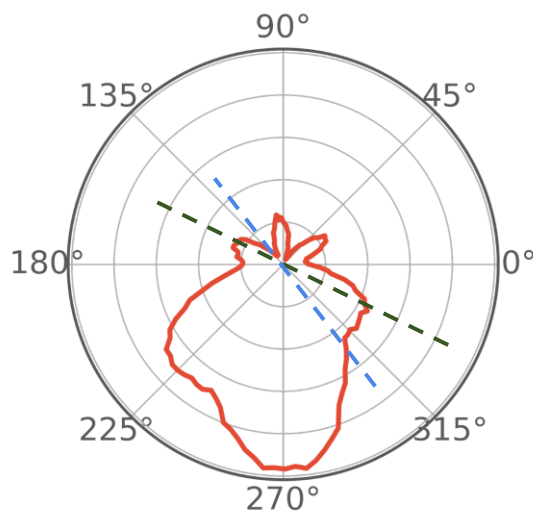
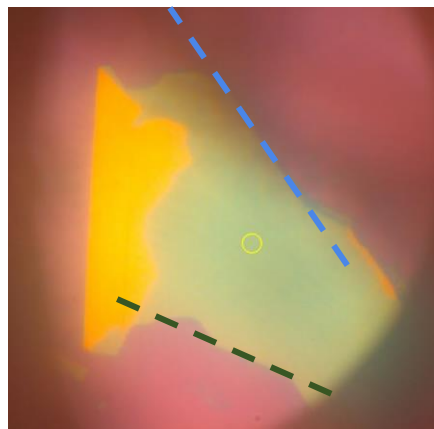
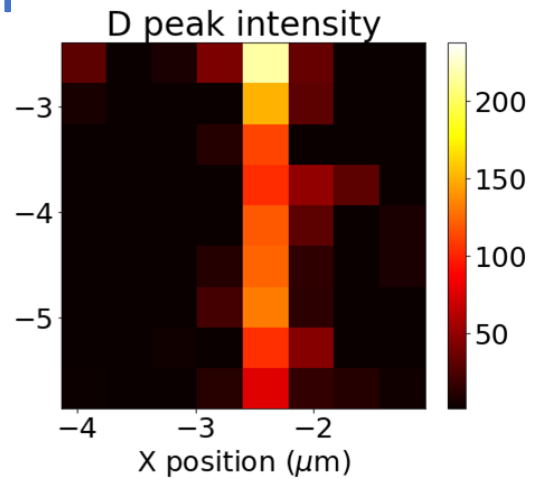
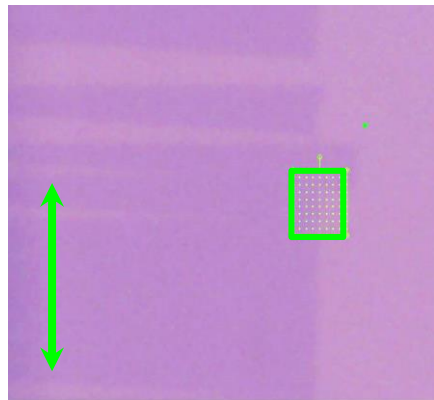
Funding acknowledgement:



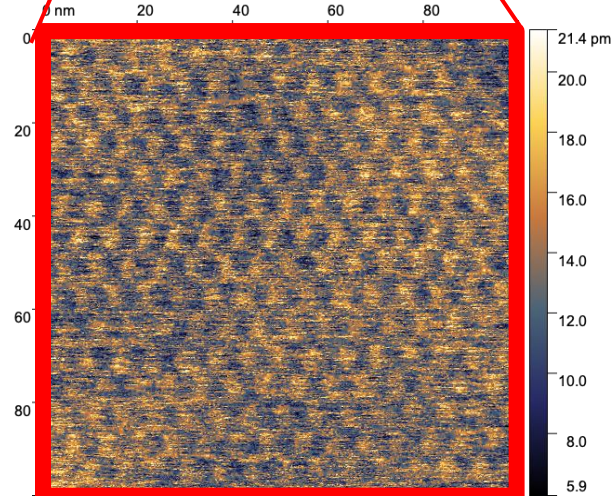
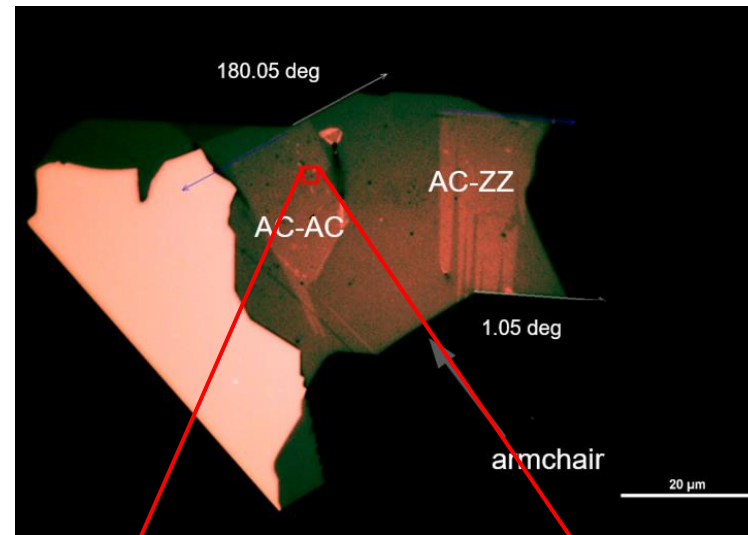
**Sandia
National
Laboratories**

Questions?

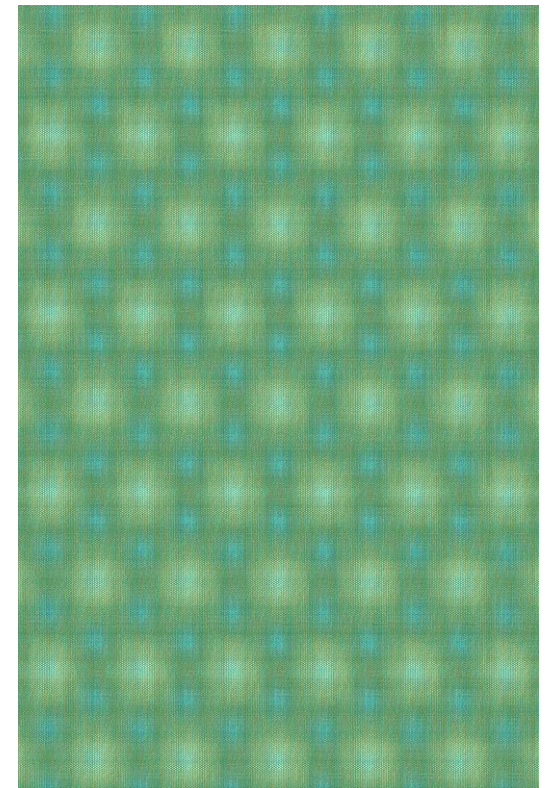
Pre-Characterization



PFM moiré Confirmation



'Super-moiré' of Interest



References

- [1] Sharpe, Aaron L., et al. "Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene." *Science* 365.6453 (2019): 605-608.
- [2] A. H. MacDonald, "Bilayer graphene's wicked, twisted road", *Physics* 12, 12 (2019).
- [3] Serlin, M., et al. "Intrinsic quantized anomalous Hall effect in a moiré heterostructure." *Science* 367.6480 (2020): 900-903.
- [4] You, YuMeng, et al. "Edge chirality determination of graphene by Raman spectroscopy." *Applied Physics Letters* 93.16 (2008): 163112.
- [5] Cancado, L. G., et al. "Influence of the atomic structure on the Raman spectra of graphite edges." *Physical review letters* 93.24 (2004): 247401.
- [6] Li, Yilei, et al. "Probing symmetry properties of few-layer MoS₂ and h-BN by optical second-harmonic generation." *Nano letters* 13.7 (2013): 3329-3333.
- [7] McGilly, Leo J., et al. "Visualization of moiré superlattices." *Nature Nanotechnology* 15.7 (2020): 580-584.
- [8] Shi, Jingtian, Jihang Zhu, and A. H. MacDonald. "Moiré commensurability and the quantum anomalous Hall effect in twisted bilayer graphene on hexagonal boron nitride." *Physical Review B* 103.7 (2021): 075122.