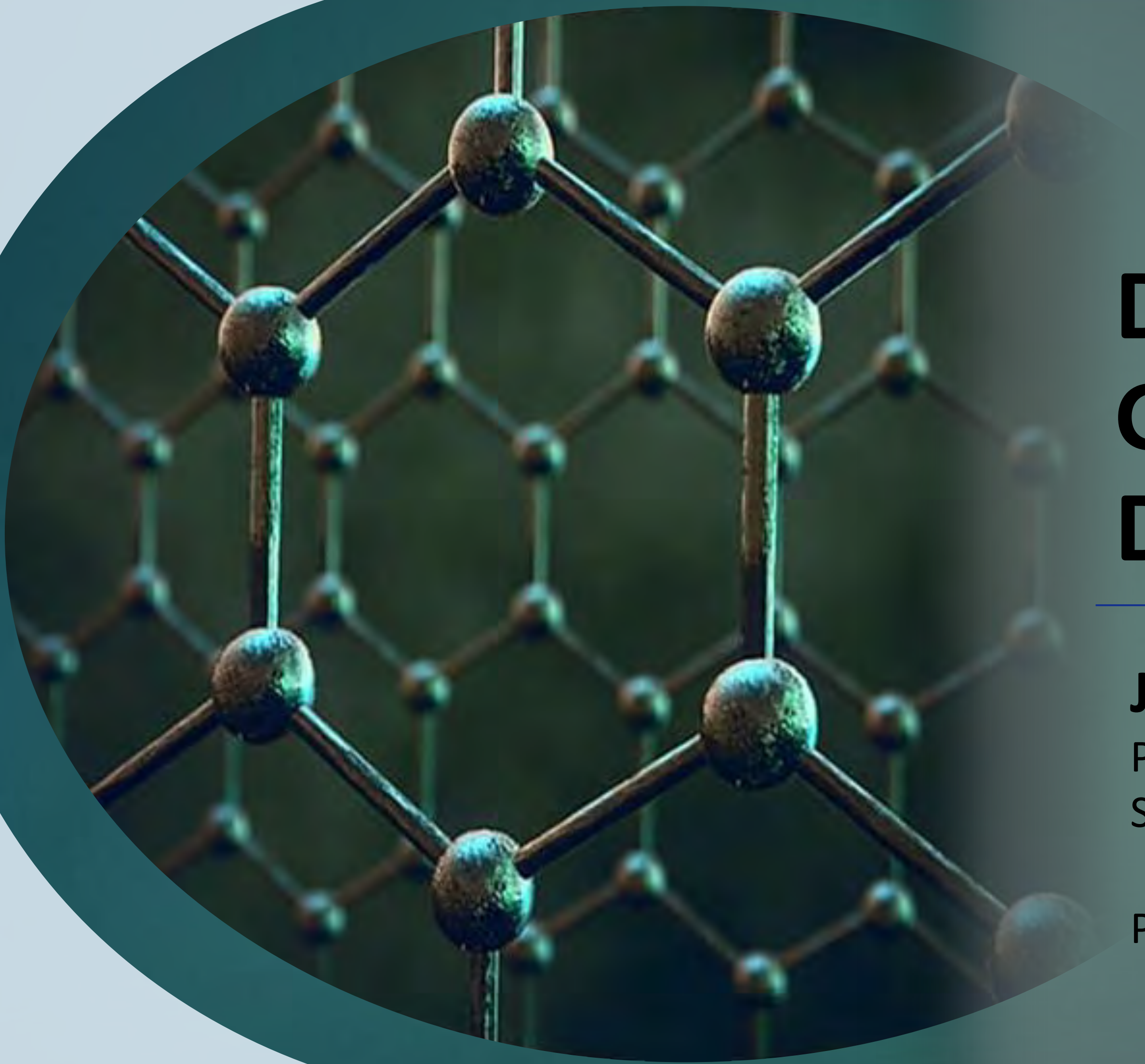


DESIGNING TWISTED GRAPHENE BASED DEVICES

JOVI K
P0181214
SEM 9 THESIS

PROJECT GUIDE: **DR. ROMAIN DANNEAU,**
Institut für Nanotechnologie,
Karlsruher Institut für Technologie

k.jovi@cbs.ac.in

ACKNOWLEDGEMENT

Image Location: INT-KIT, Karlsruhe

I give my deepest thanks to my supervisor
Dr. Romain Danneau,
and Prof. Ralph Krupke,
Group Head and my gratitude for all
members of Krupke Group and INT.

KIT
Karlsruher Institut für Technologie

CONTENTS

Image credit: GenerativeAI

SESSION 1

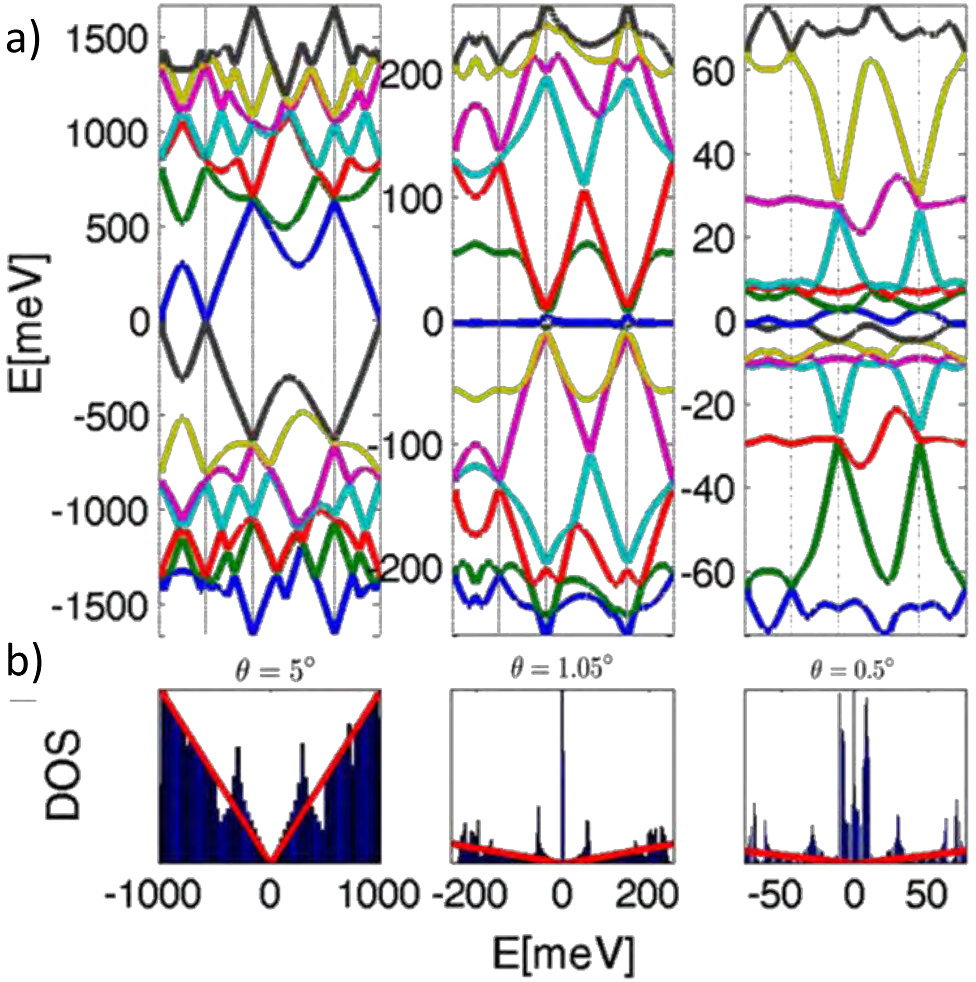
INTRODUCTION

Image credit: here

PREDICTION OF MAGIC ANGLE tBLG:

CONCLUSION

Emergence of strongly correlated system in Bi-Layer Graphene for twist $\sim 1.1^o, 0.5^o, \ldots$



GRAPH

Fig 2: Moiré bands: (a) Energy dispersion for the 14 bands closest to the Dirac point plotted along the k-space trajectory. (b) DOS.

Bistritzer, R. & MacDonald, A. H. Moiré bands in twisted double-layer graphene. Proceedings of the National Academy of Sciences 108, 12233–12237 (2011).

TEAR AND STACK PROCEDURE FOR BLG:

IMAGE

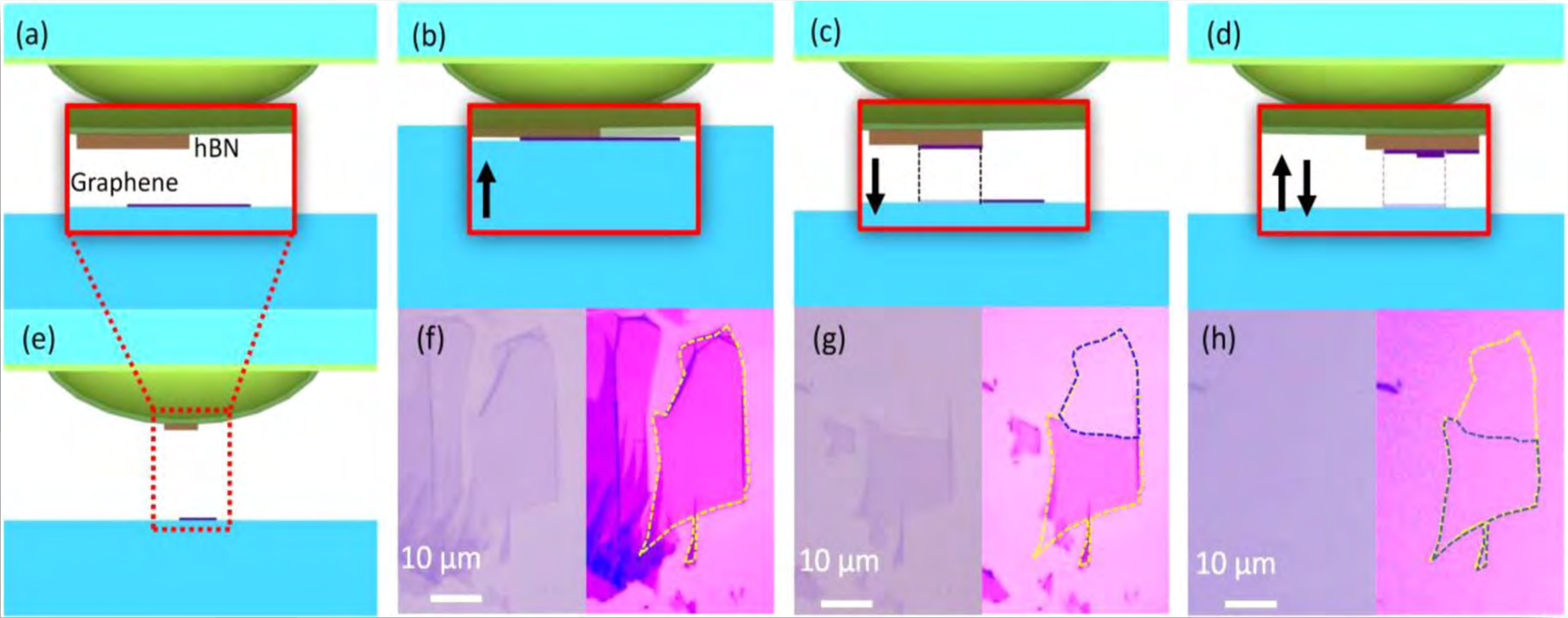


Fig 4: Rotationally aligned graphene double layer realized by successive transfers from a monolayer graphene using a hemispherical handle substrate.

CAPTION

Kim, K. et al. Van derWaals Heterostructures with High Accuracy Rotational Alignment. Nano Letters 16, 1989–1995 (2016).
Cao, Y. et al. Superlattice-Induced Insulating States and Valley-Protected Orbits in Twisted Bilayer Graphene. Physical Review Letters 117, 116804 (2016).

CONFIRMATION OF MAtBLG:

IMAGE

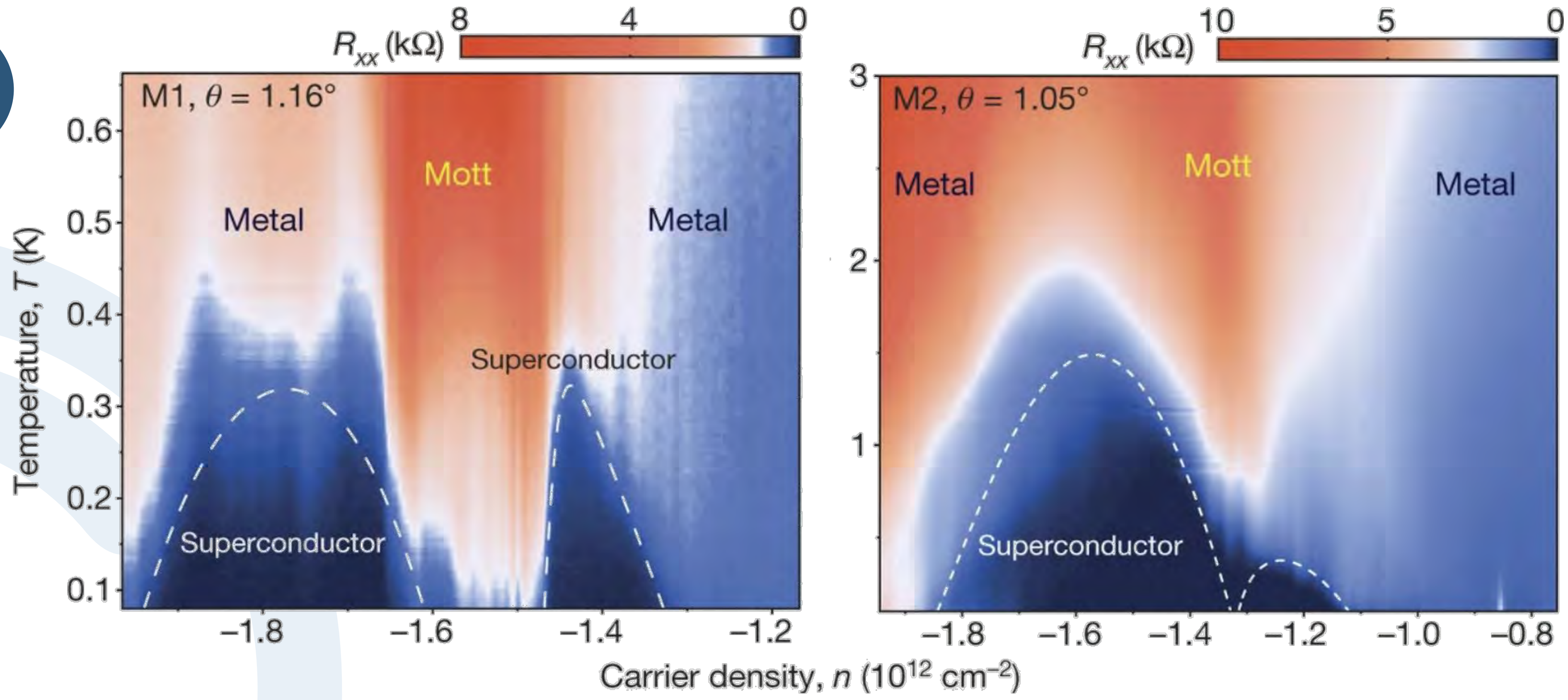


Fig 5: Unconventional Superconductivity in MAtBLG

CAPTION

Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

THESIS OBJECTIVE

STACKING PROCEDURE FOR tBLG

Standardising the **TEAR**, TWIST and **STACKING** in tBLG

- Exploring Efficient ways of tearing a graphene
- Standardising the procedure
- Conclude with fabricating MAtBLG

SESSION 2

Reviewing Previous Stacking Procedure

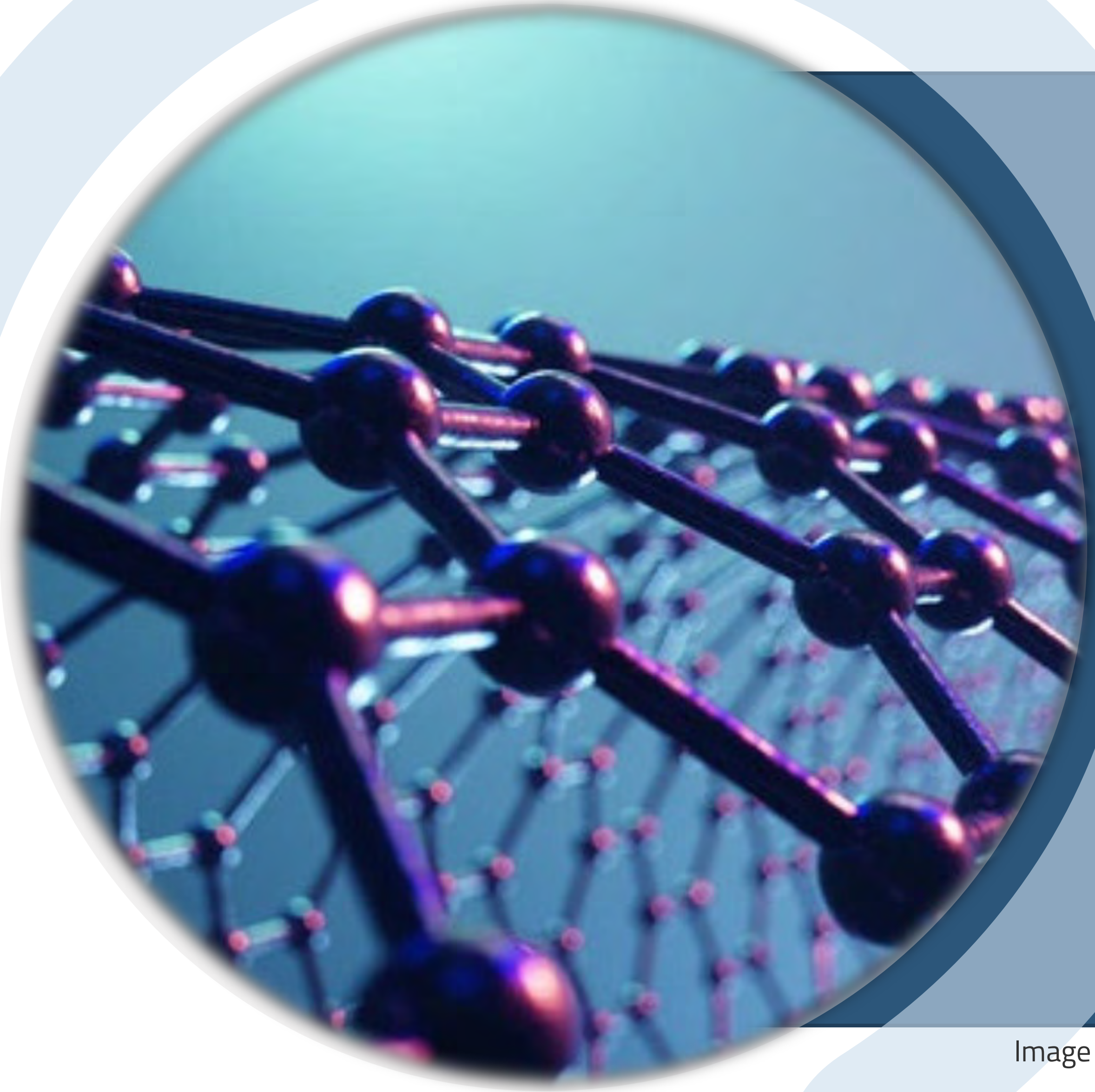


Image credit: Shutterstock

DRY PROCEDURE :

IMAGE

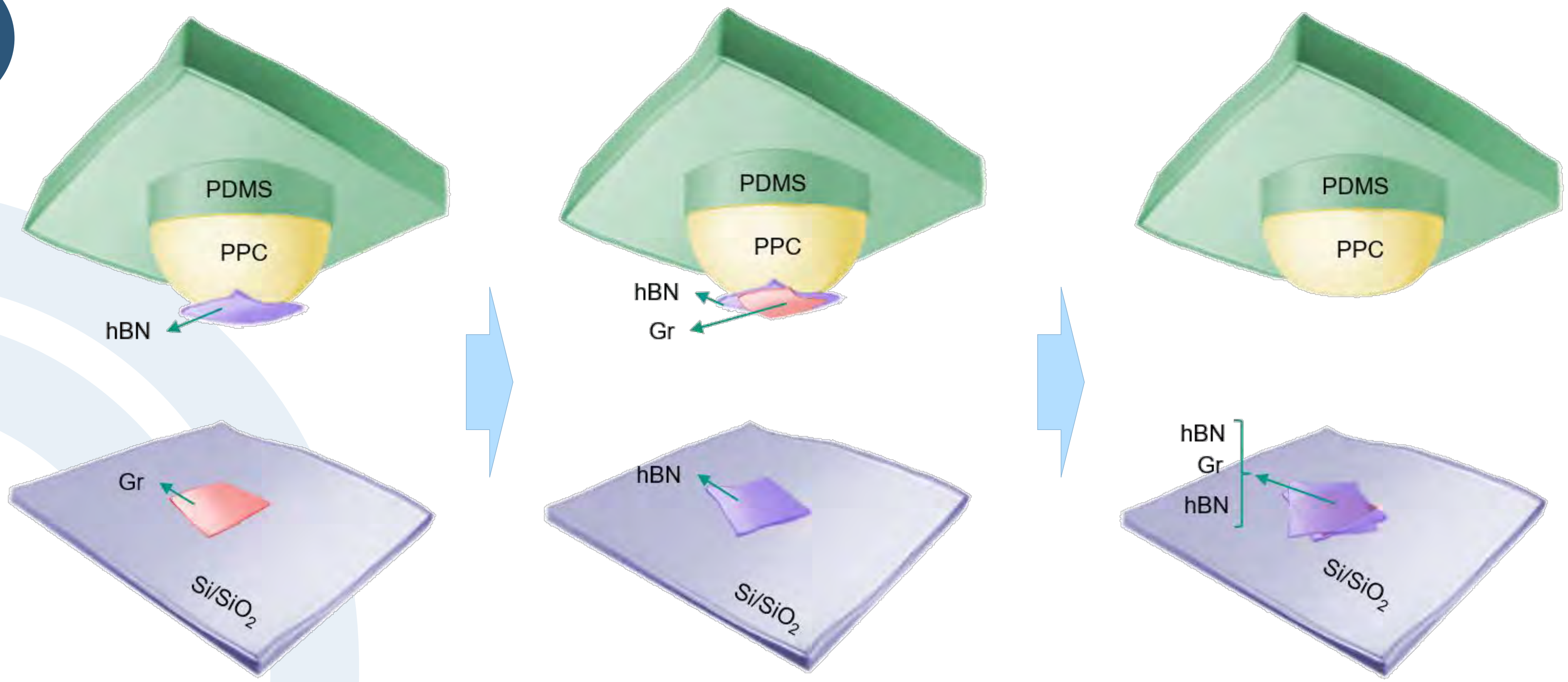


Fig 6: Illustration of van der Waals heterostructure assembly process as done in Danneau Group.

CAPTION

Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

DRAWBACKS FOR TBLG:

TEARING PROBLEM

Tear a graphene along the edge of hBN flake

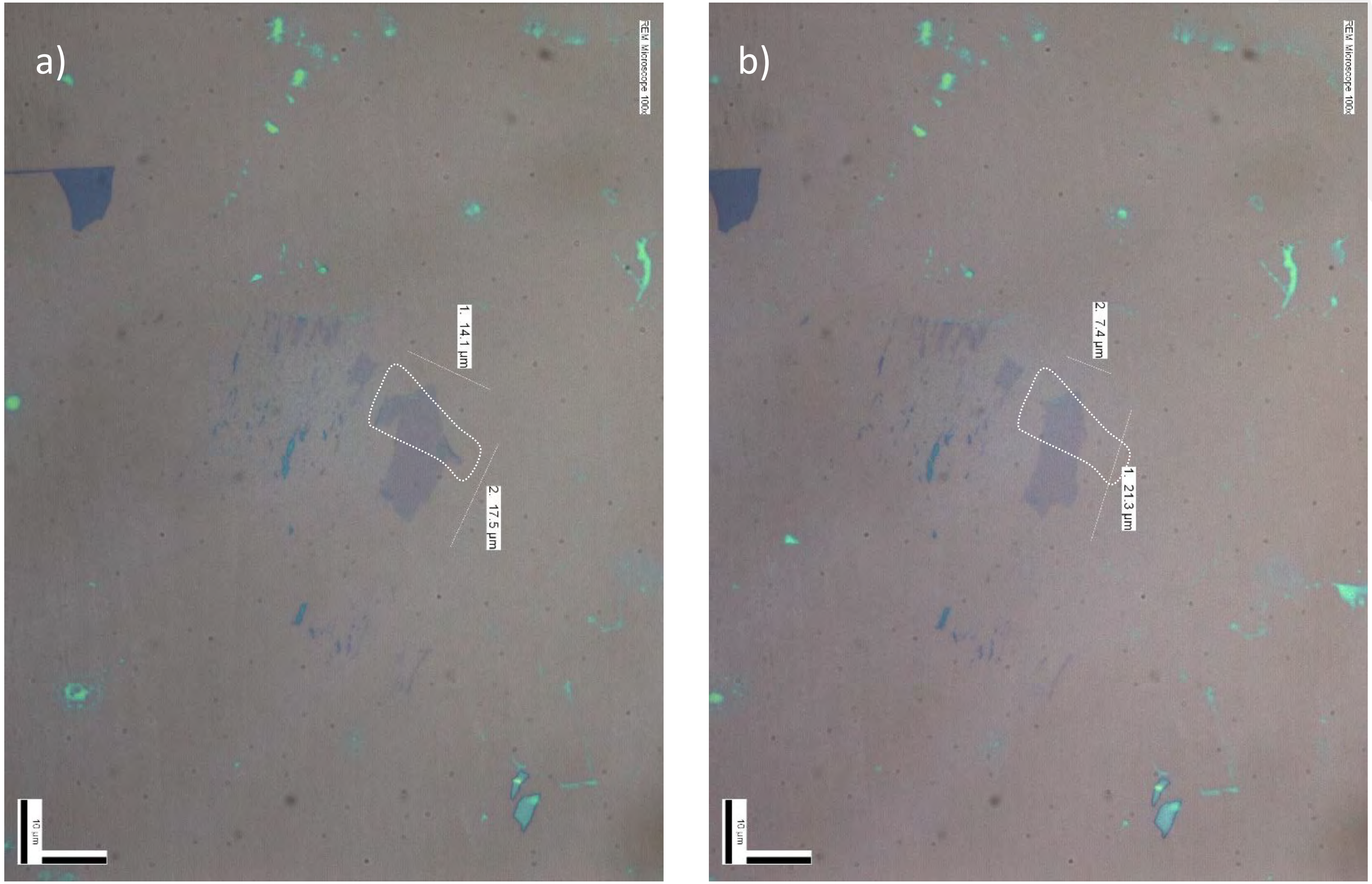
Avoid PPC stamp contamination on other half of graphene

CLEAN INTERFACE PROBLEM

SESSION 3

Modified Dry Stacking Procedure

TEAR PROBLEM



IMAGE

- Fig 7: a) A graphene flake before pickup. b) Same flake after pickup. Faint outline is the contact region of hBN

INFERENCE

- No trouble with Pickup of Graphene
- Tear only through previous cracks

TEARING A PAPER:

TEAR ALONG AN EDGE

RIPPING APART

TEARING A PAPER - RIPPING APART:

D. L. Mathieson, *The Tensile Strength of Paper*, Phys Teach **29**, 412 (1998).

TEAR ALONG AN EDGE - DRY STACK PROCEDURE :

IMAGE

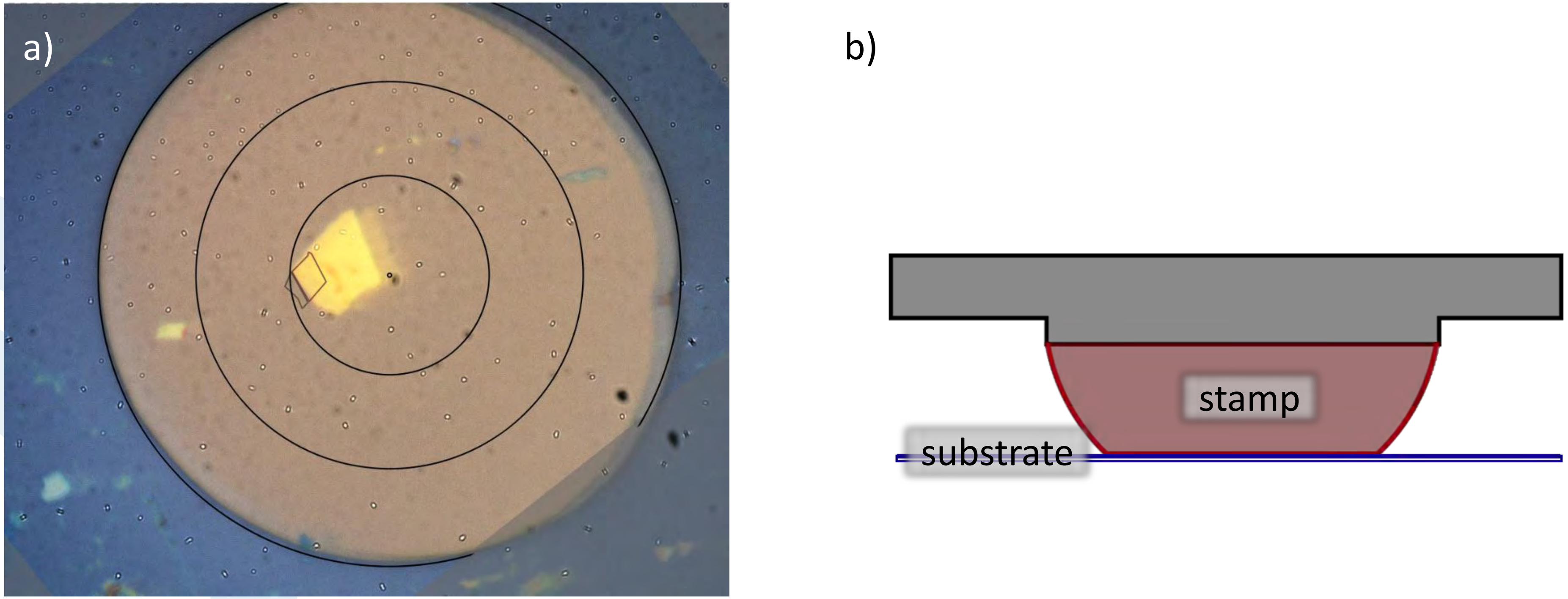


Fig 9: (a) Optical image during stacking. Concentric circles and tear outlines are overlayed and (b) cross sectional schematic.

CAPTION

Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

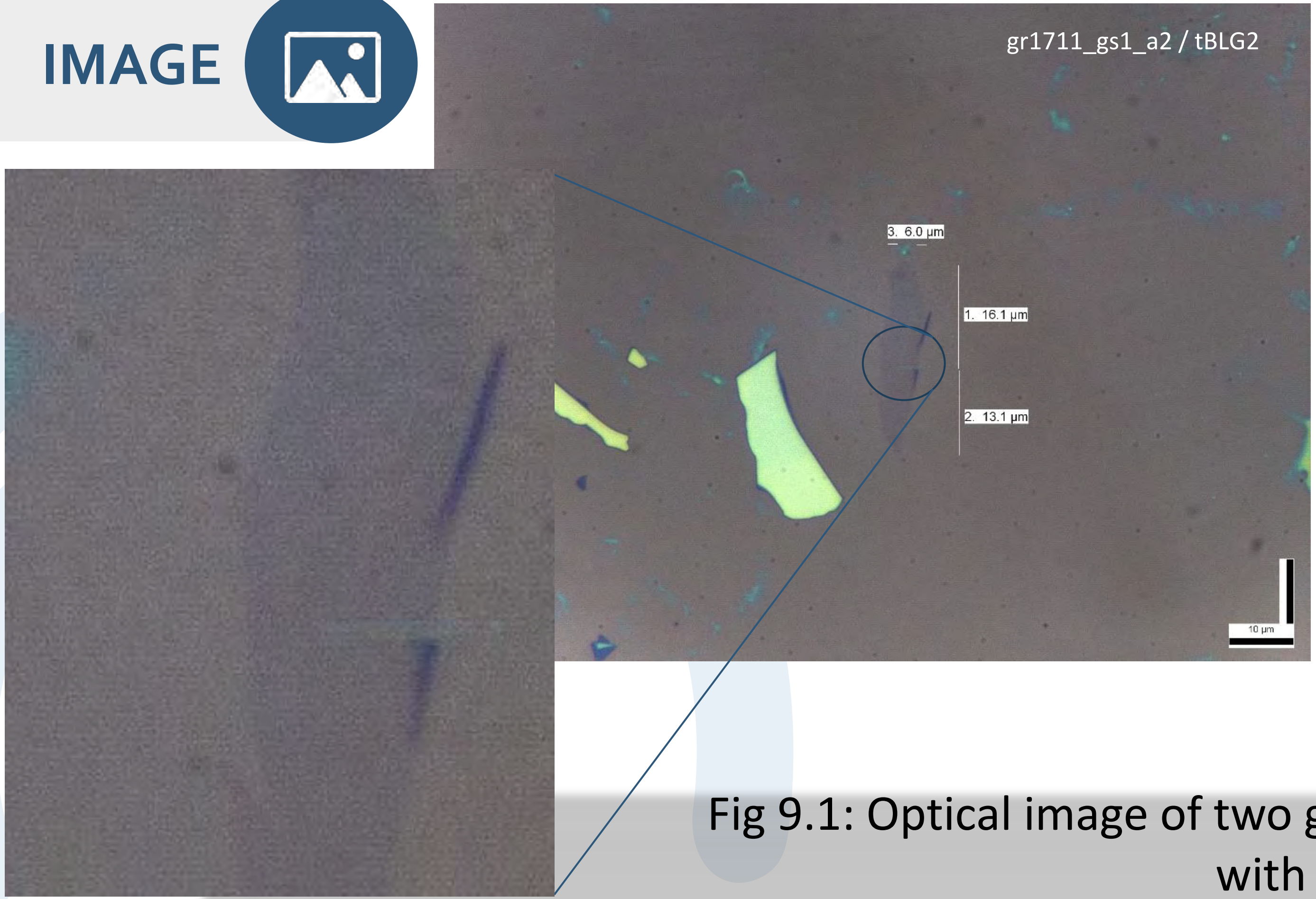
TEARING A PAPER:

TEAR ALONG AN EDGE

USING KNIFE

AFM TEAR - USING KNIFE:

IMAGE



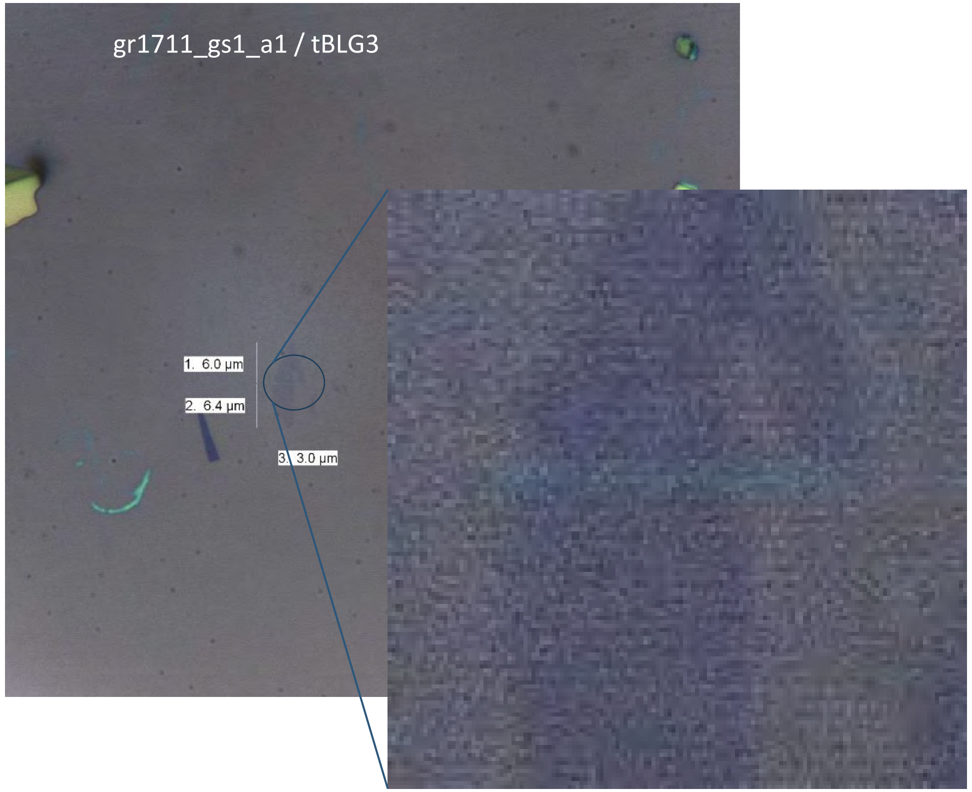


Fig 9.1: Optical image of two graphene flakes with a tear partition

CAPTION

Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

DRAWBACKS FOR TBLG:

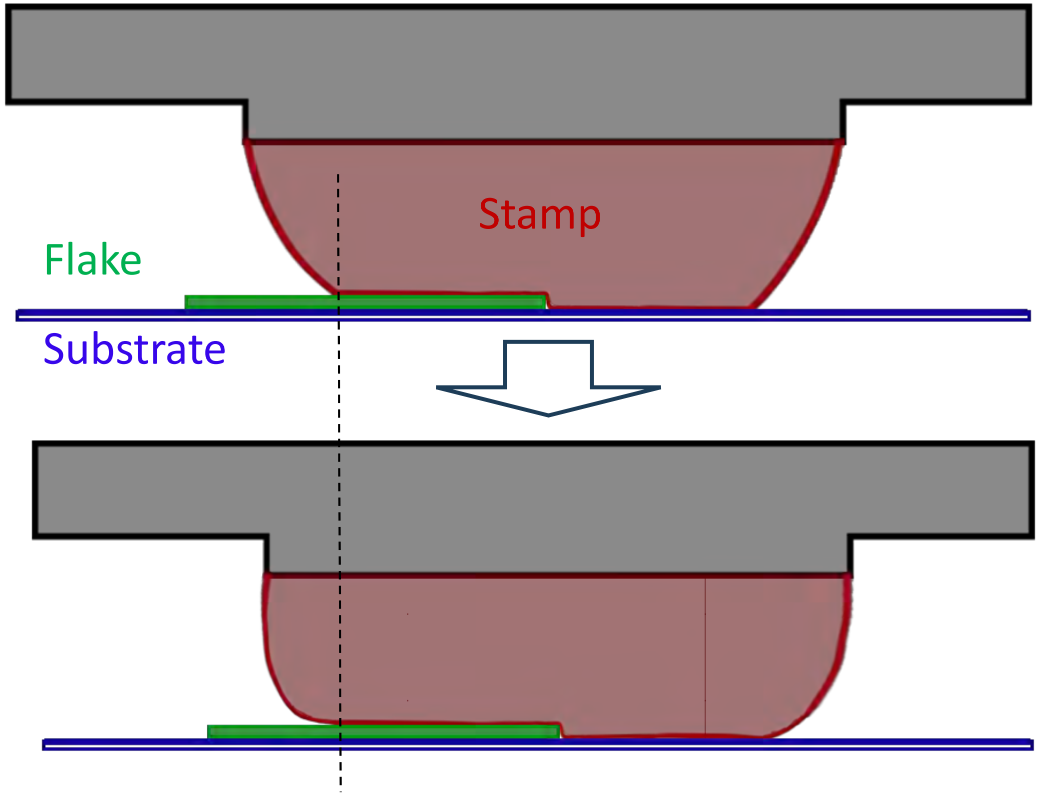
TEARING PROBLEM

Tear a graphene along the edge
of hBN flake

Avoid PPC stamp contamination
on other half of graphene

CLEAN INTERFACE PROBLEM

CLEAN INTERFACE PROBLEM



IMAGE

Fig 10: cross sectional schematic of the effect of heat on hemispherical stamp

CLEAN INTERFACE PROBLEM

CONVEX stamp

- ✓ Easy to detach from substrate after contact
- ✗ Contact edge expansion CANNOT be controlled

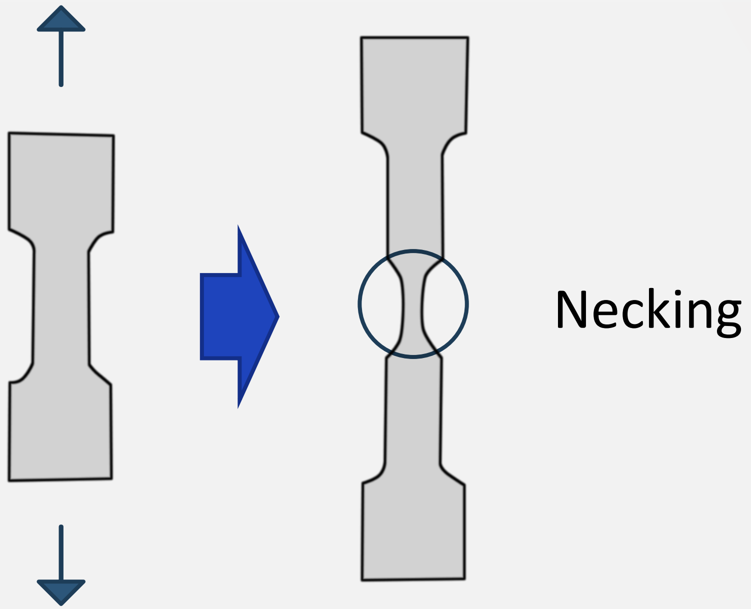


Fig 13: Formation and neck and eventual breakage.

CONCAVE stamp

- ✗ NECK FORMATION and breakage
- ✓ Contact edge expansion CAN be controlled

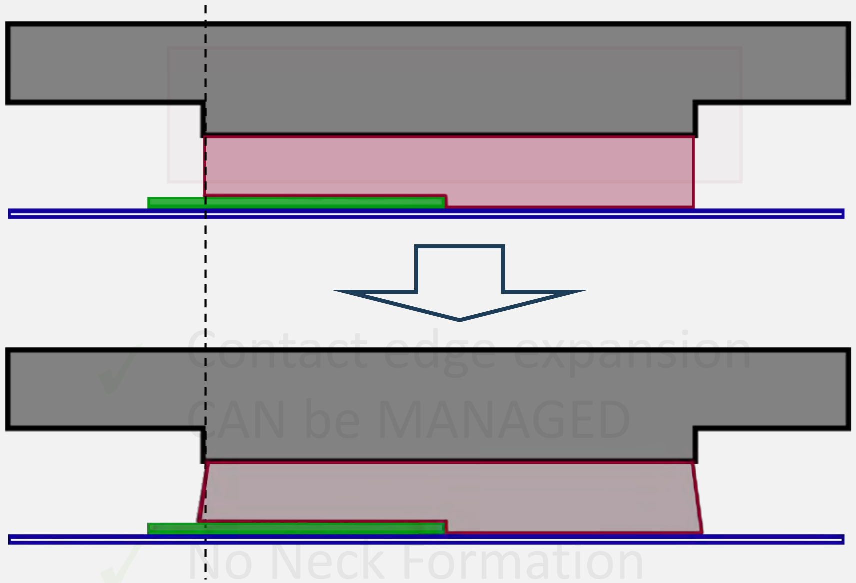
CLEAN INTERFACE PROBLEM

CONVEX stamp

✓ Easy to detach from substrate after contact

✗ Contact edge expansion CANNOT be controlled

CYLINDRICAL stamp



✓ Contact edge expansion CAN be MANAGED

✓ No Neck Formation

✗? **Easy to detach from substrate after contact**

CONCAVE stamp

✗ NECK FORMATION and breakage

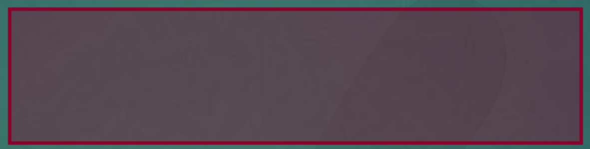
✓ Contact edge expansion CAN be controlled

CLEAN INTERFACE PROBLEM

CONVEX stamp

- ✓ Easy to detach from substrate after contact
- ✗ Contact edge expansion CANNOT be controlled

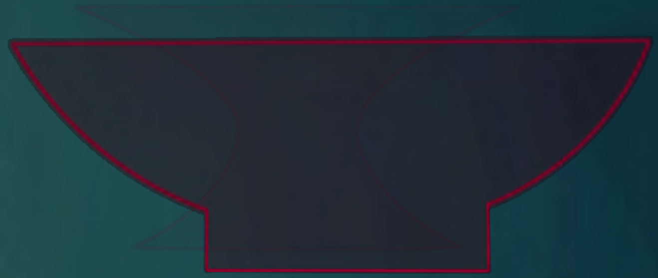
CYLINDRICAL stamp



- ✓ Contact edge expansion CAN be MANAGED
- ✓ No Neck Formation
- ✗ Easy to detach from substrate after contact

Convexo-Cylindrical stamp

CONCAVE stamp

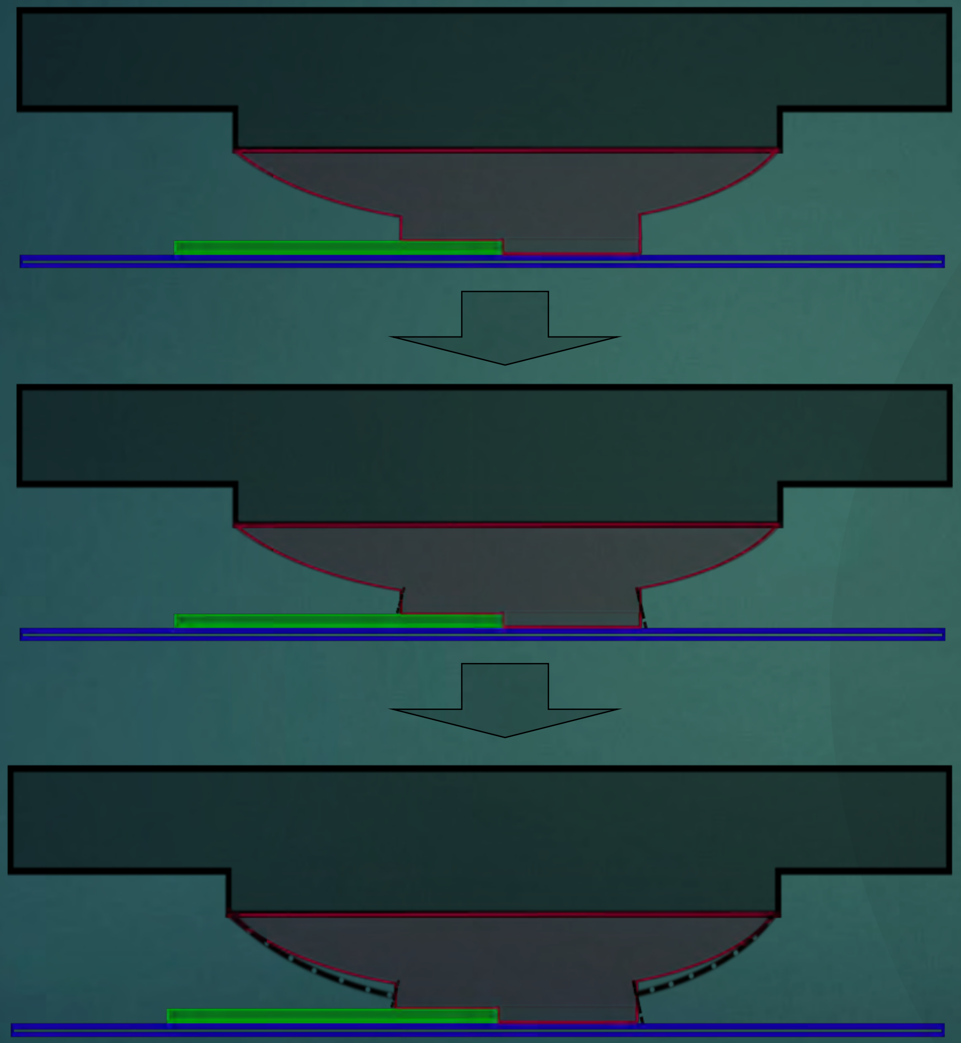


- ✓? Contact edge expansion CAN be MANAGED
- ✓? No Neck Formation
- ? Easy to detach from substrate after contact

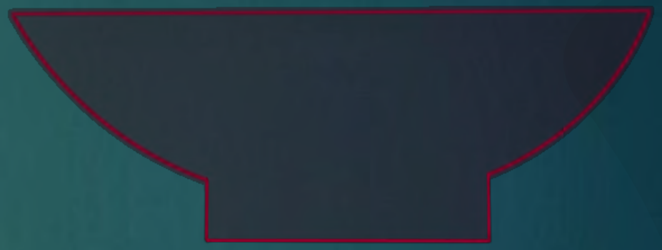
✗ NECK FORMATION and breakage

✓ Contact edge expansion CAN be controlled

CLEAN INTERFACE PROBLEM



<u>**Convexo-Cylindrical stamp**</u>



- ✓ Contact edge expansion CAN be MANAGED
- ✓ No Neck Formation
- ? Easy to detach from substrate after contact

SESSION 3

CONCLUSION

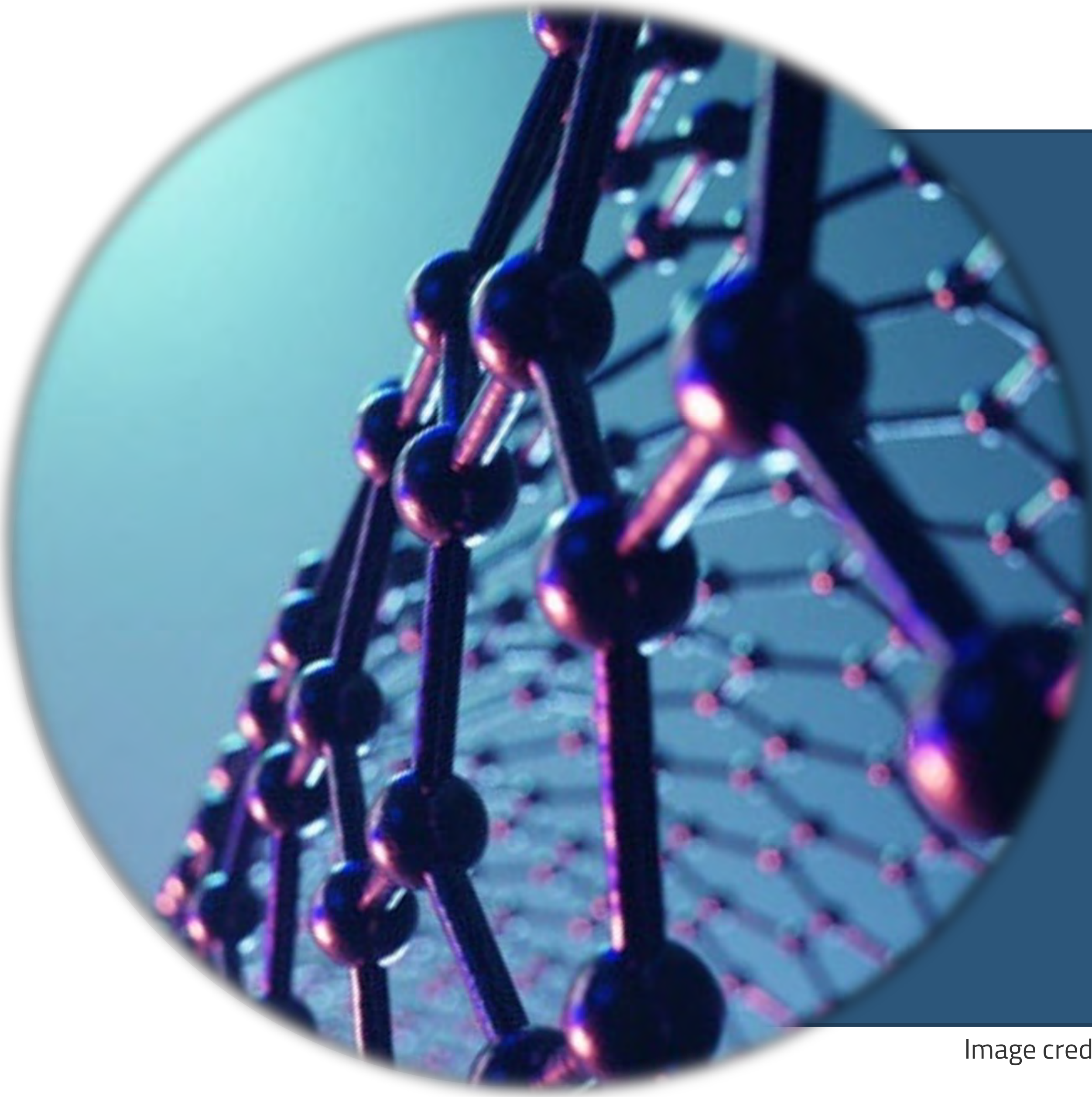
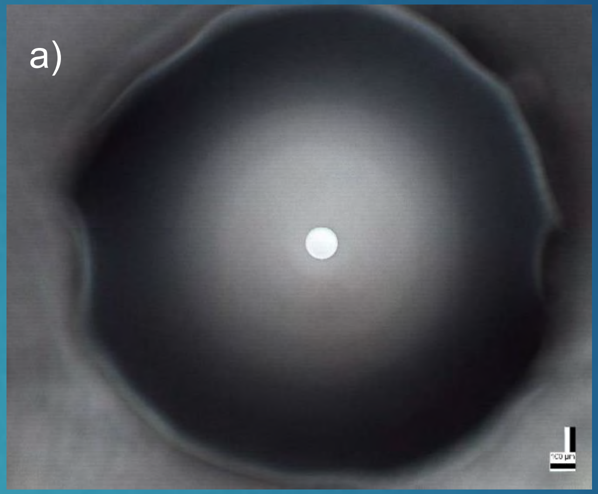


Image credit: Shutterstock

PROCEDURE FOR TBLG -

1. Exfoliation of hBN and graphene flakes
2. **Pickup of hBN**



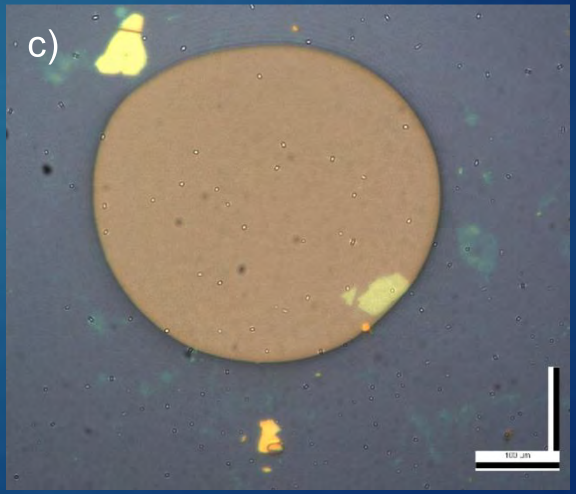


Figure 4.13: Convexo-rect. stamp. a) Optical image of hemispherical stamp with a cylindrical plateau at the tip and b) corresponding cross-section. c) is the optical image of (a) picking up an hBN flake.

PROCEDURE FOR TBLG -

1. Exfoliation of hBN and graphene flakes
2. **Pickup of hBN**
3. **Tear Graphene**
4. **Pickup of 1st half of graphene**

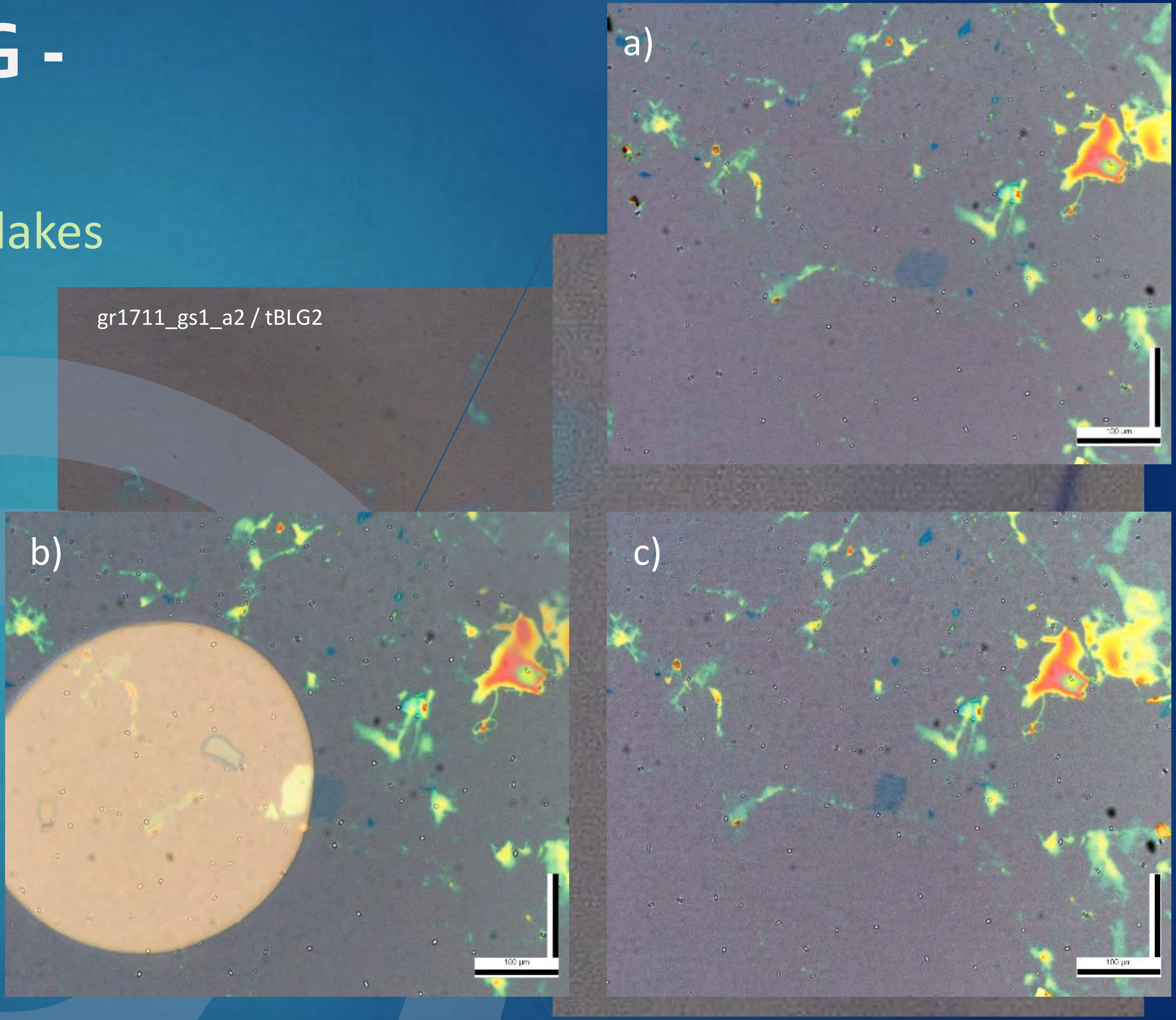


Fig 15: Optical image of a) graphene flake before tear-pickup, b) stamp with hBN in contact with half gr and c) other half of graphene left on substrate after tear.

Fig 14: AFM tear of a graphene flake and the pixel zoomed in view

PROCEDURE FOR TBLG -

2. Pickup of hBN
3. Tear Graphene
4. Pickup of 1st half of graphene
5. Twist and pickup of other half

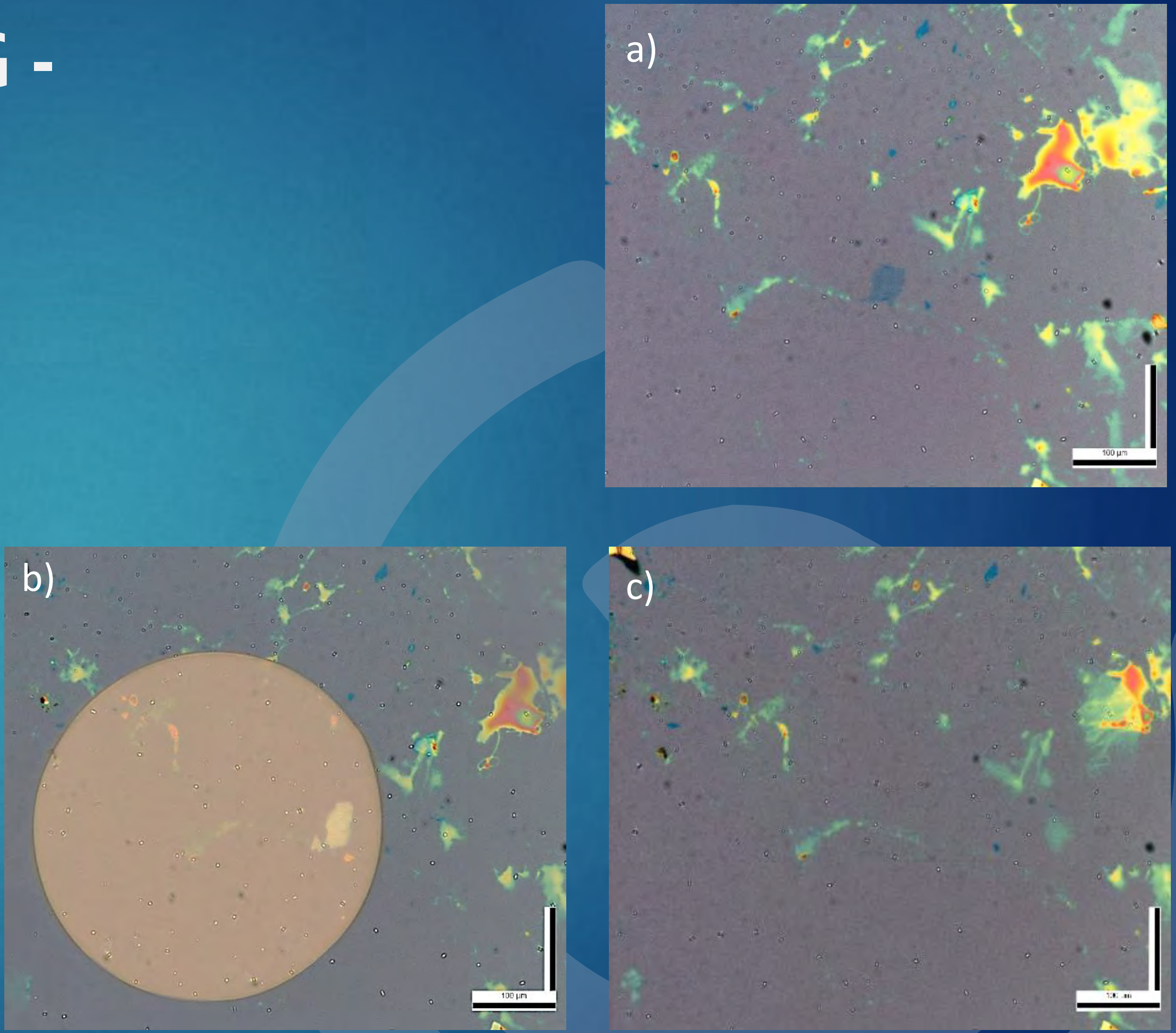
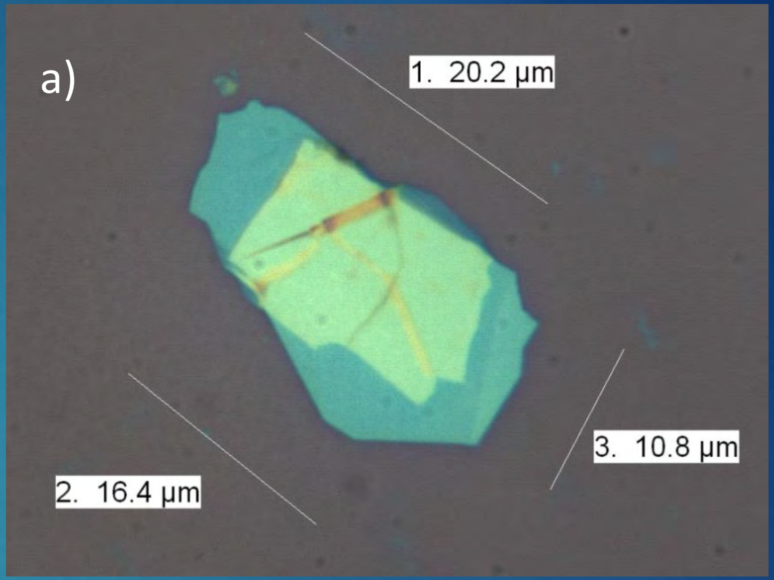


Fig 16: Optical image of a) graphene flake before final pickup, b) the twisted stamp containing hBN-gr in contact with the other half of gr and c) substrate after final pickup.

PROCEDURE FOR TBLG -

3. **Tear Graphene**
4. **Pickup of 1st half of graphene**
5. **Twist and pickup of other half**
6. hBN encapsulation and post processing



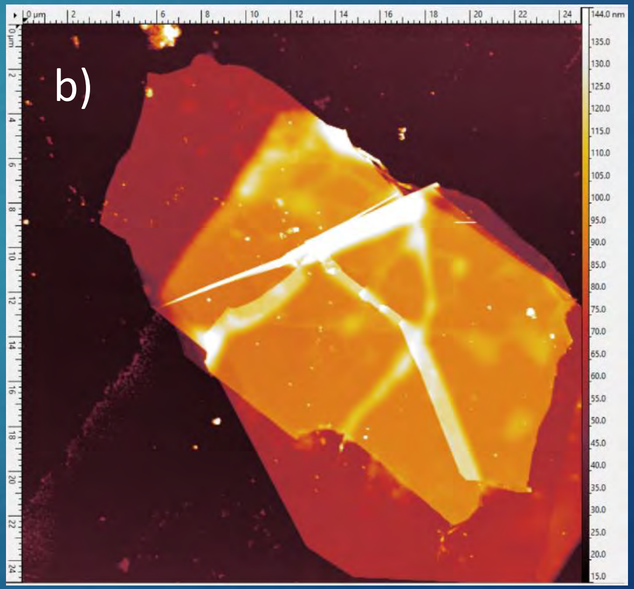
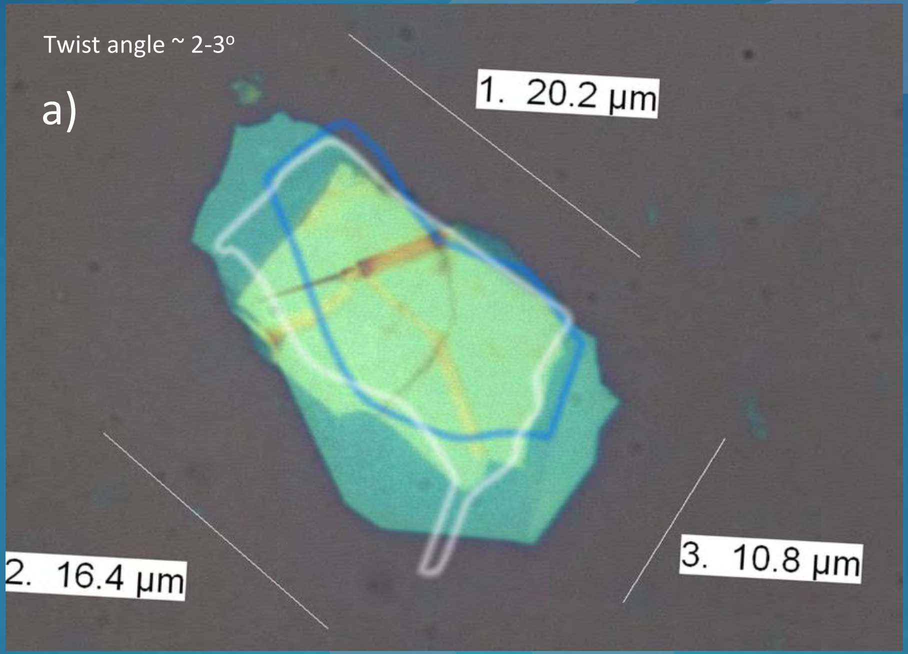
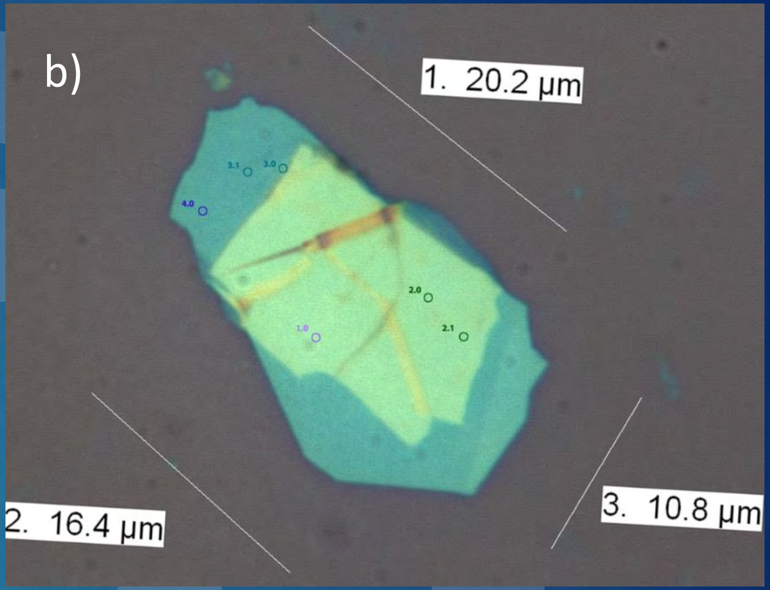


Fig 17: a) Optical Image of the final stack and b) AFM image of the same

PROCEDURE FOR TBLG -

7. Analysis





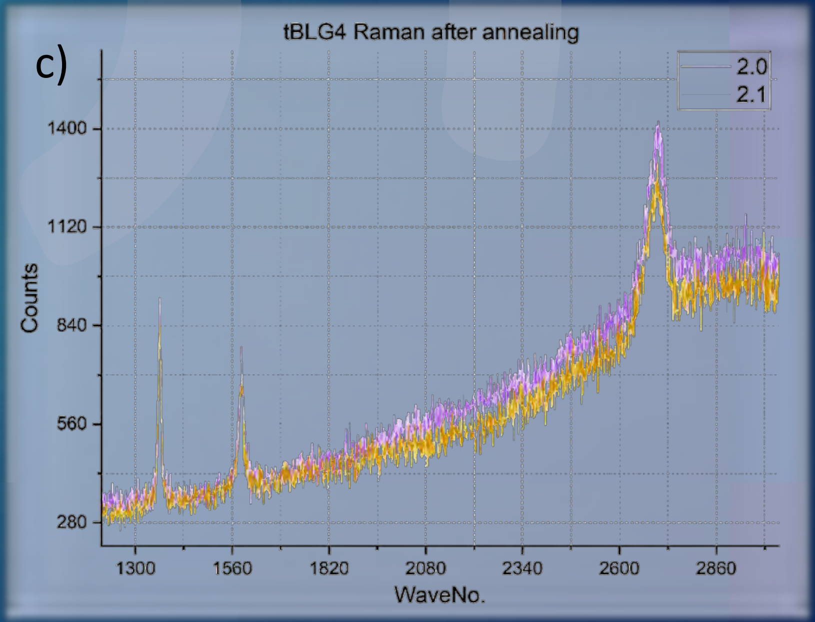


Fig 18: a) Optical Image with overlay of the gr flakes. The twist is estimated from the images b) Stack with markings on Raman spectrum sites c) Relevant Raman Spectrum.

SUMMARY OF MY WORK

- Clean tearing edge by use of AFM
- Innovative solution to the clean interface problem leads to ultra clean interface.
- Standardised the dry stacking procedure for tear and stack.
- Easily generalisable for a MAtBLG.

BIBIOGRAPHY

- **Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, *Unconventional Superconductivity in Magic-Angle Graphene Superlattices*, Nature 2018 556:7699 556, 43 (2018).**
- Y. Cao et al., *Correlated Insulator Behaviour at Half-Filling in Magic-Angle Graphene Superlattices*, Nature 2018 556:7699 556, 80 (2018).
- A. L. Sharpe, E. J. Fox, A. W. Barnard, J. Finney, K. Watanabe, T. Taniguchi, M. A. Kastner, and D. Goldhaber-Gordon, *Emergent Ferromagnetism near Three-Quarters Filling in Twisted Bilayer Graphene*, Science (1979) 365, 605 (2019).
- K. Kim, A. DaSilva, S. Huang, B. Fallahazad, S. Larentis, T. Taniguchi, K. Watanabe, B. J. LeRoy, A. H. MacDonald, and E. Tutuc, ***Tunable Moiré Bands and Strong Correlations in Small-Twist-Angle Bilayer Graphene***, Proc Natl Acad Sci U S A 114, 3364 (2017).
- R. Bistritzer and A. H. Macdonald, ***Moiré Bands in Twisted Double-Layer Graphene***, PNAS 108, 12233 (2011).

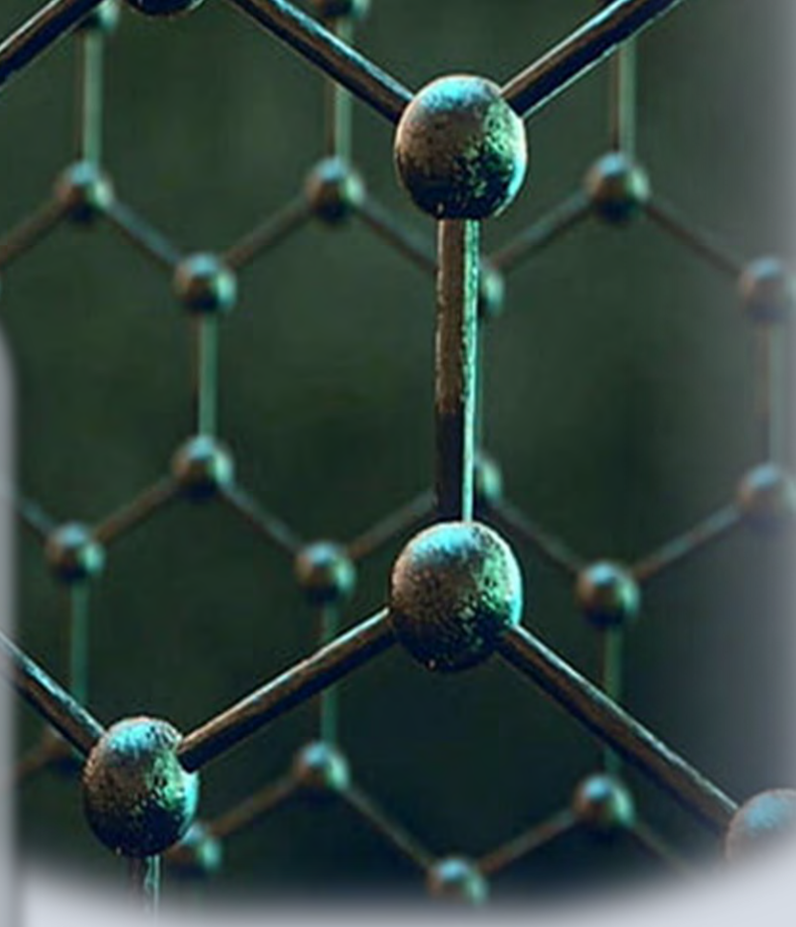
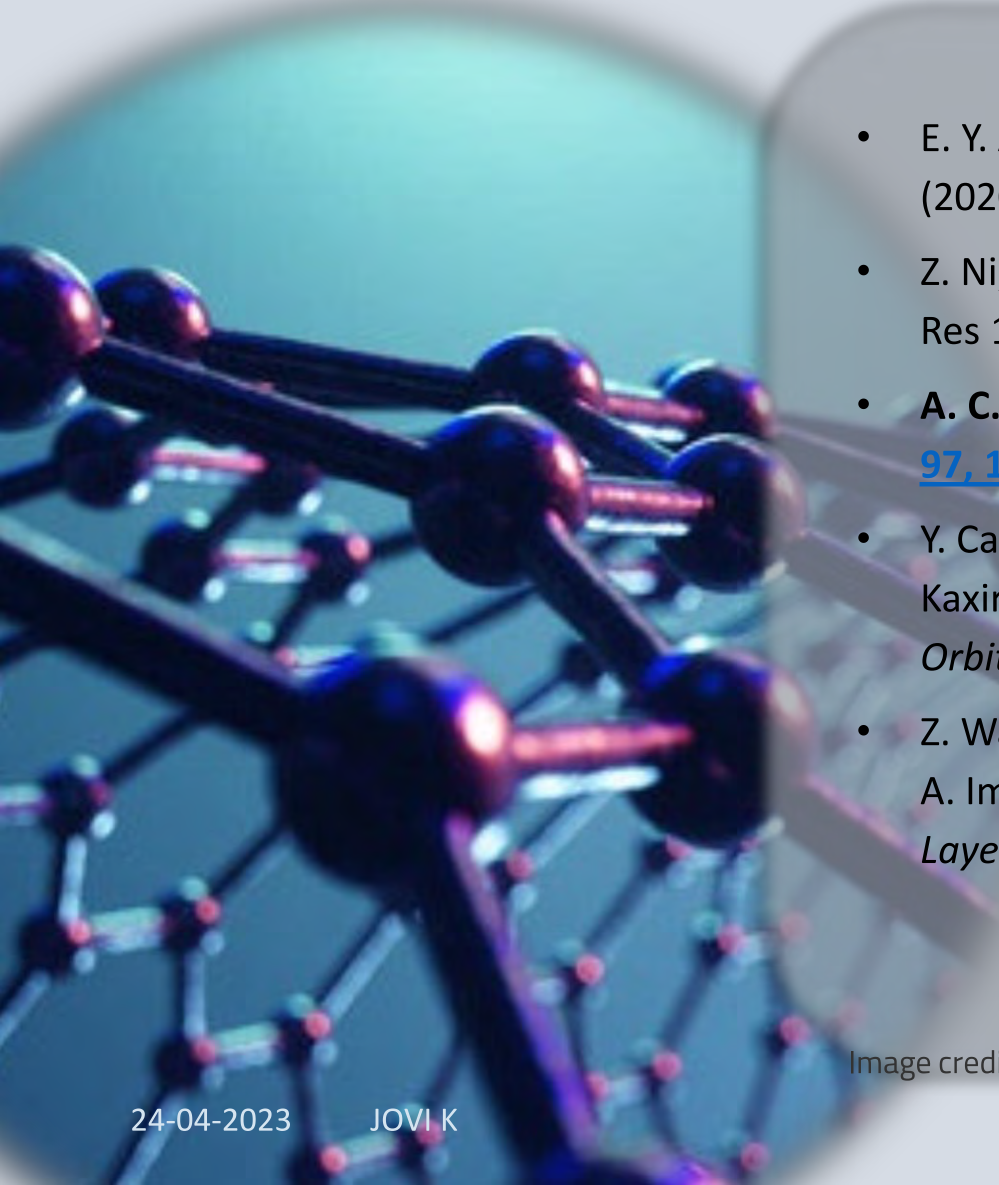


Image credit: SEP Hall Thruster, NASA



BIBIOGRAPHY

- E. Y. Andrei and A. H. MacDonald, *Graphene Bilayers with a Twist*, Nat Mater 19, 1265 (2020).
- Z. Ni, Y. Wang, T. Yu, and Z. Shen, *Raman Spectroscopy and Imaging of Graphene*, Nano Res 1, 273 (2008).
- **A. C. Ferrari et al., *Raman Spectrum of Graphene and Graphene Layers*, Phys Rev Lett 97, 187401 (2006).**
- Y. Cao, J. Y. Luo, V. Fatemi, S. Fang, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, *Superlattice-Induced Insulating States and Valley-Protected Orbits in Twisted Bilayer Graphene*, Phys Rev Lett 117, 116804 (2016).
- Z. Wang, I. Gutiérrez-Lezama, N. Ubrig, M. Kroner, M. Gibertini, T. Taniguchi, K. Watanabe, A. Imamoğlu, E. Giannini, and A. F. Morpurgo, *Very Large Tunneling Magnetoresistance in Layered Magnetic Semiconductor CrI3*, Nature Communications 2018 9:1 9, 1 (2018).

Image credit: here

THANK YOU

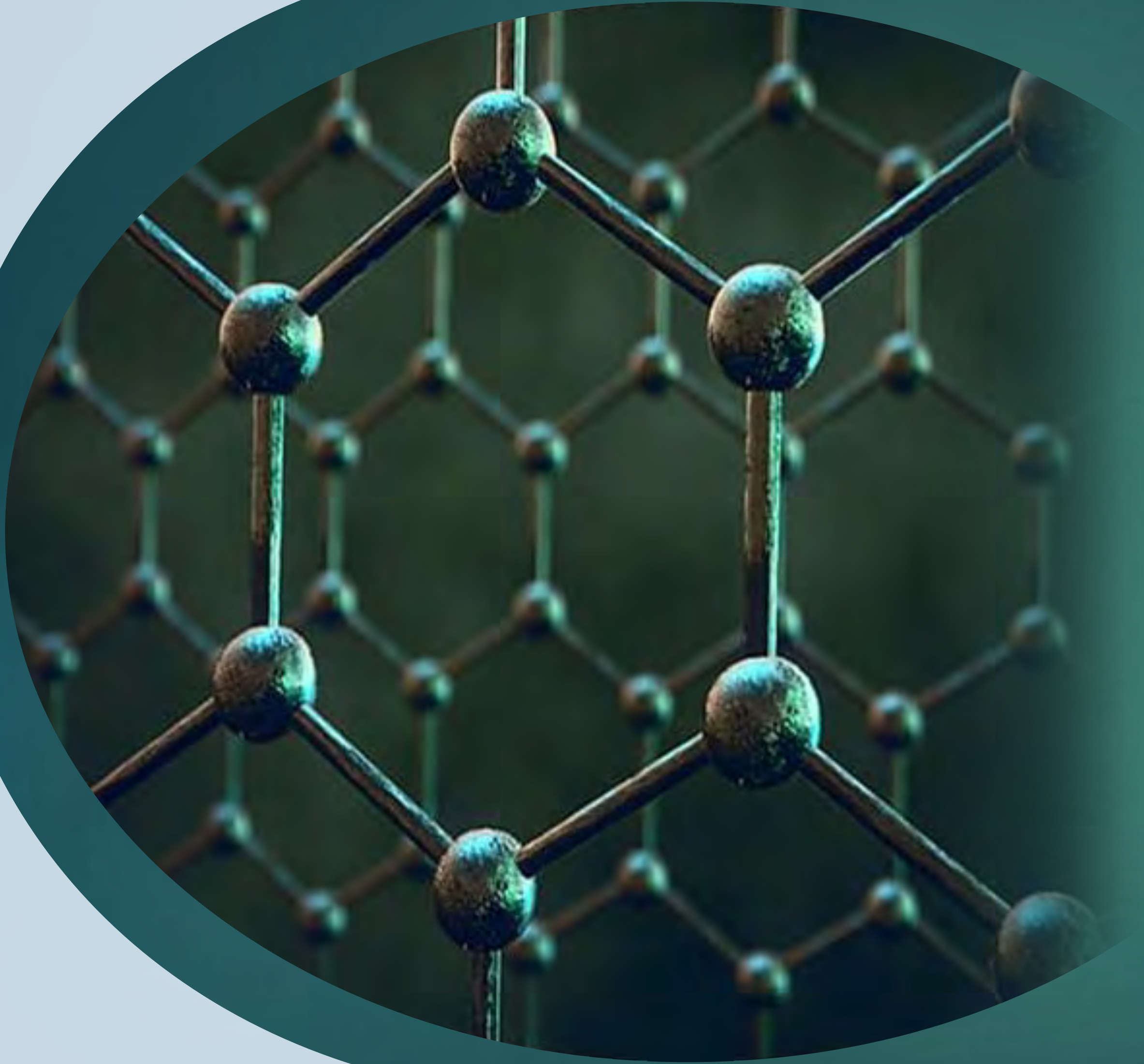
k.jovi@cbs.ac.in

Jovi K,

P0181214,

UM DAE CEBS

THE END



APPENDIX

SLG – MOIRE SUPERLATTICE:

IMAGE

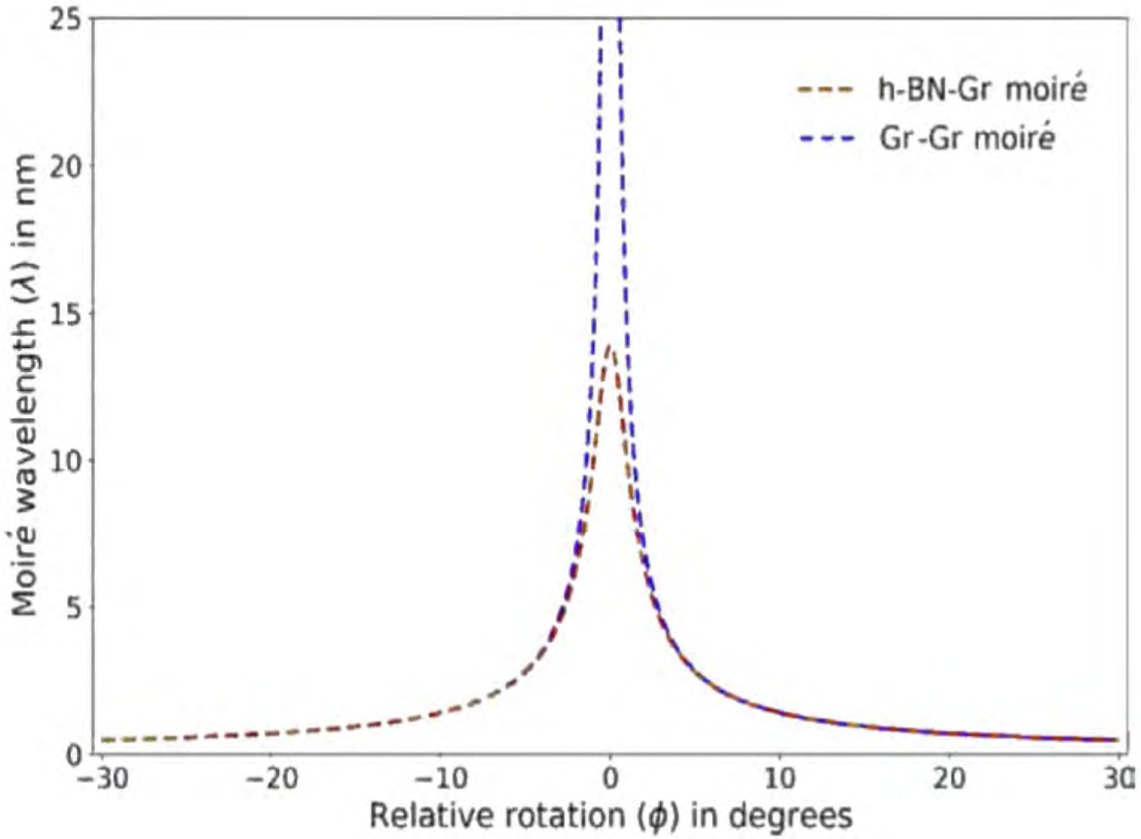


Figure 2.2: Energy Band in Graphene: a) Plot of Energy band in reciprocal space. b) is the corresponding Reciprocal space.

CAPTION

Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

FABRICATION MAGIC ANGLE TBLG:

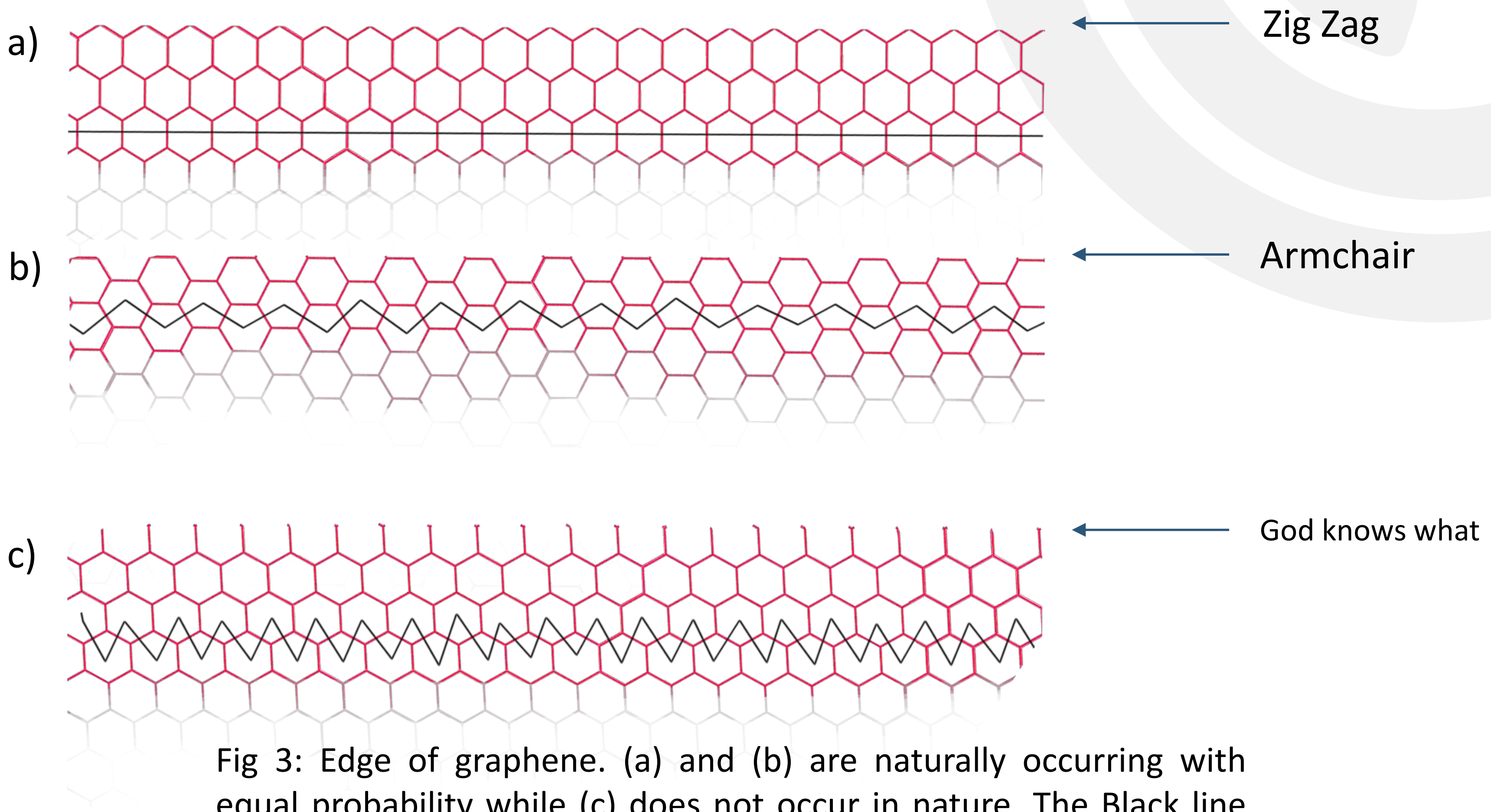


Fig 3: Edge of graphene. (a) and (b) are naturally occurring with equal probability while (c) does not occur in nature. The Black line represent the bond cu

SLG – DIRAC CONES:

IMAGE

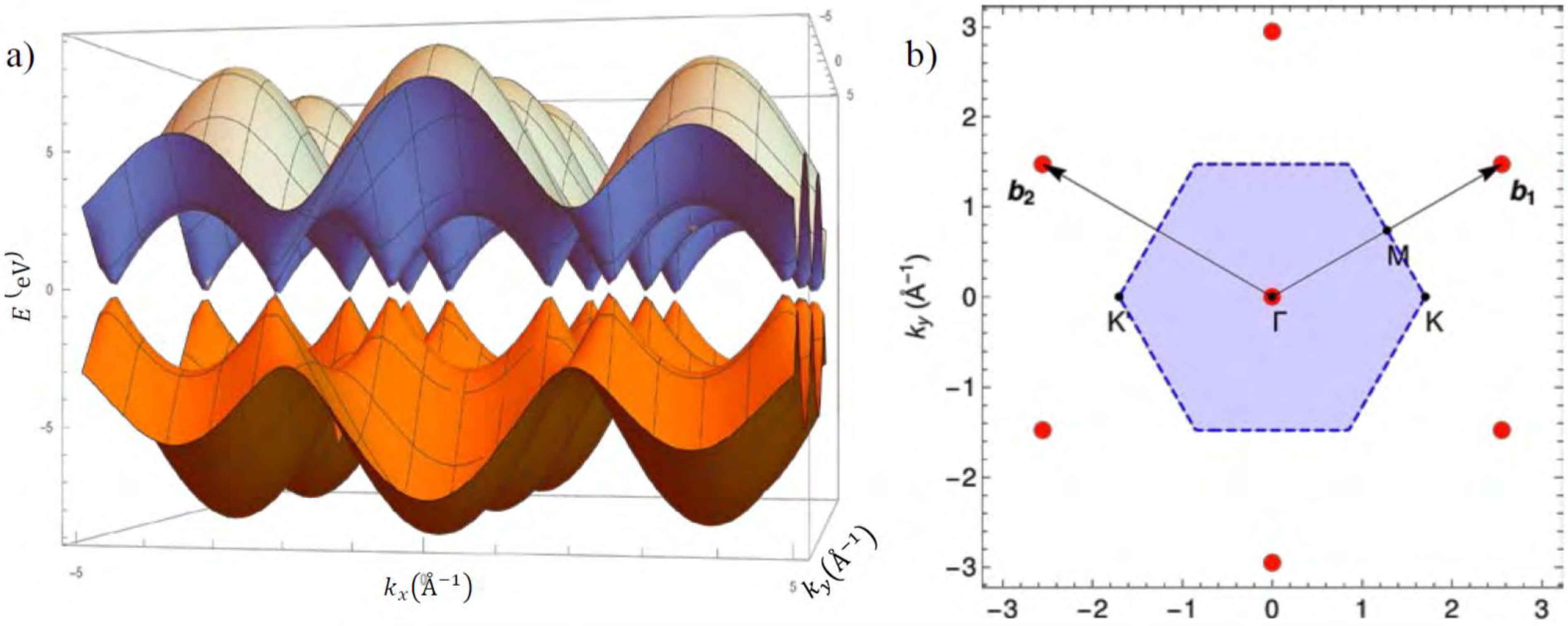
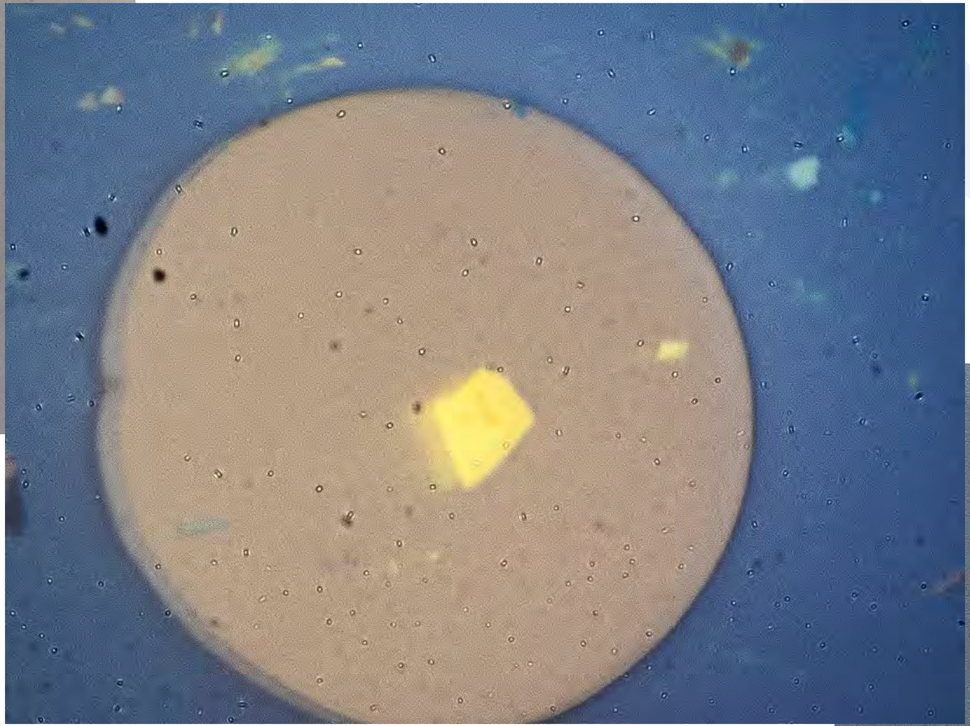


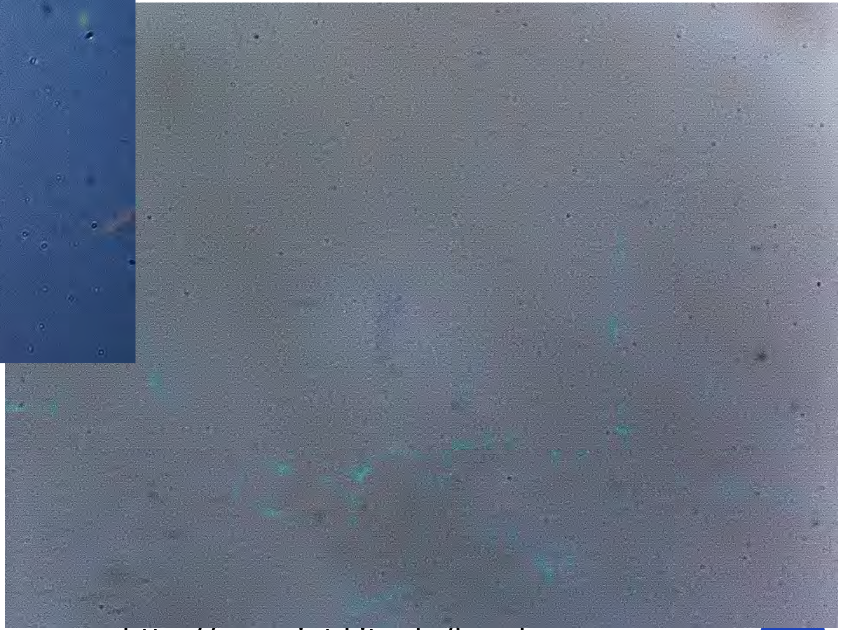
Figure 2.2: E[illegible]
reciproca[illegible]

$$E(\boldsymbol{k}) = \pm t\sqrt{4\cos\left(\frac{\sqrt{3}}{2}dk_x\right)\cos\left(\frac{3}{2}dk_y\right) + 2\cos(\sqrt{3}dk_x) + 3}$$

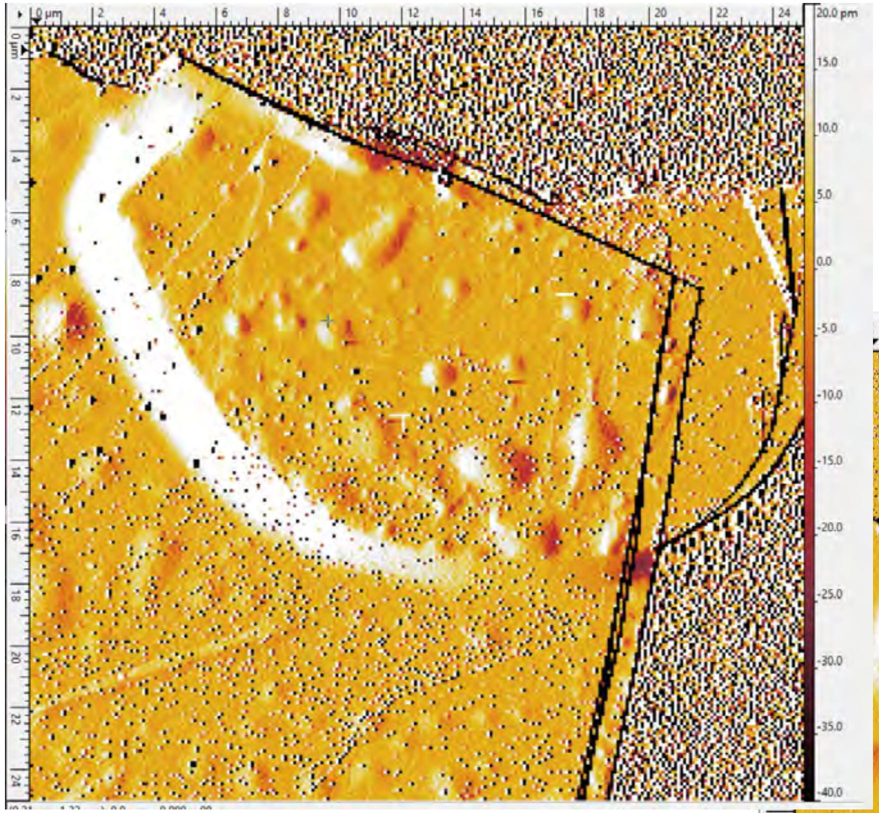
Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).

tBLG_1 Angle ~24°

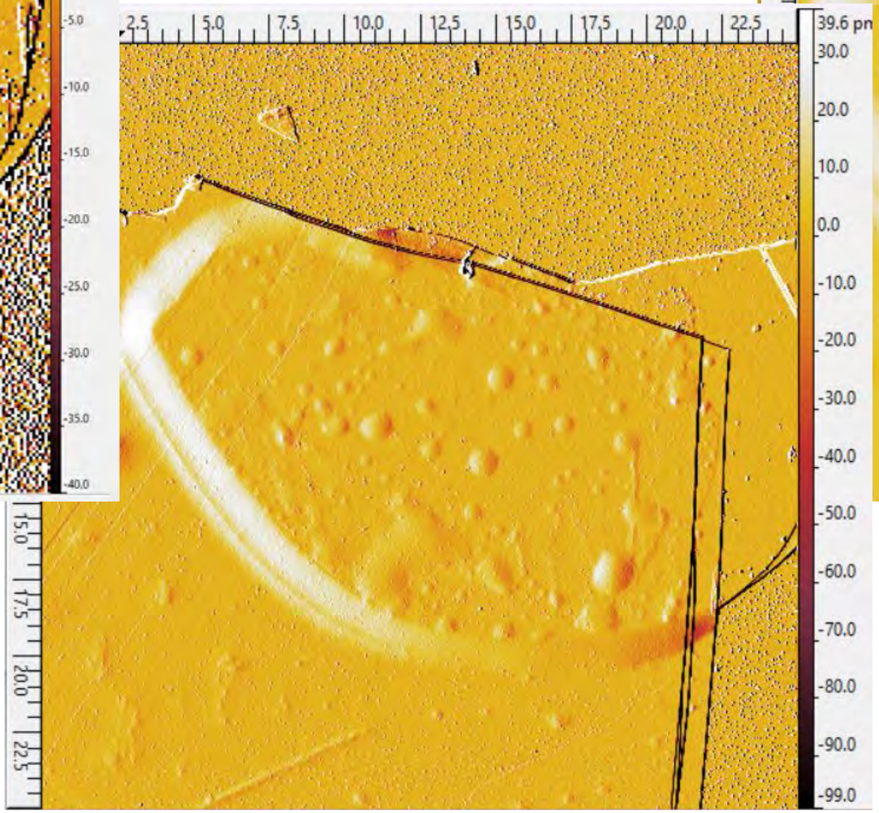


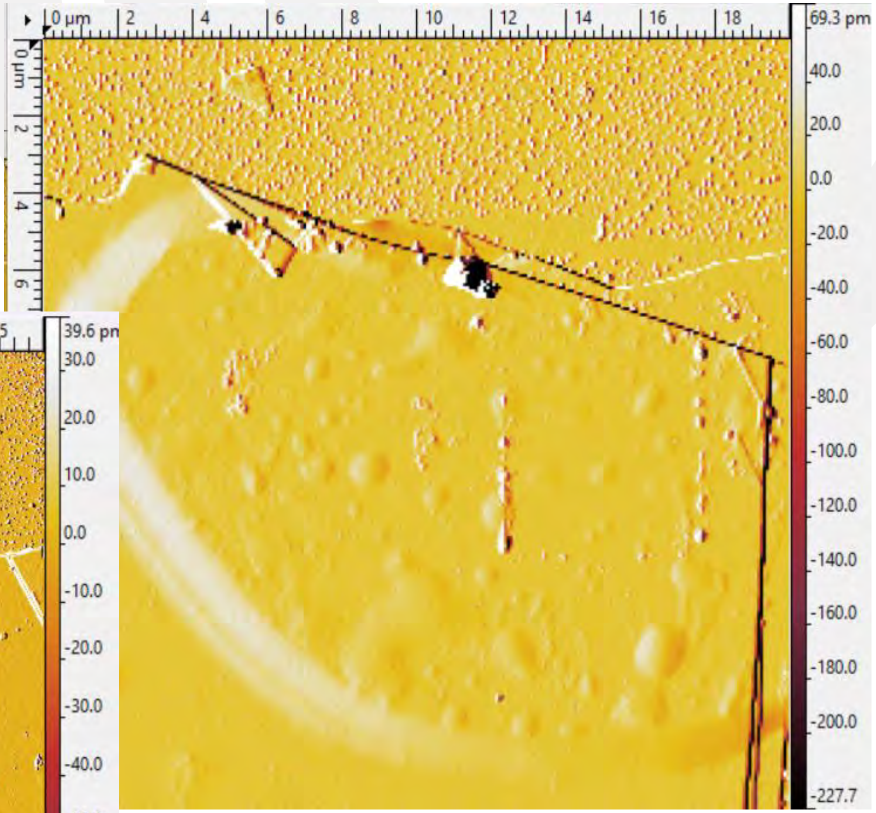


tBLG_1 Angle ~24^o



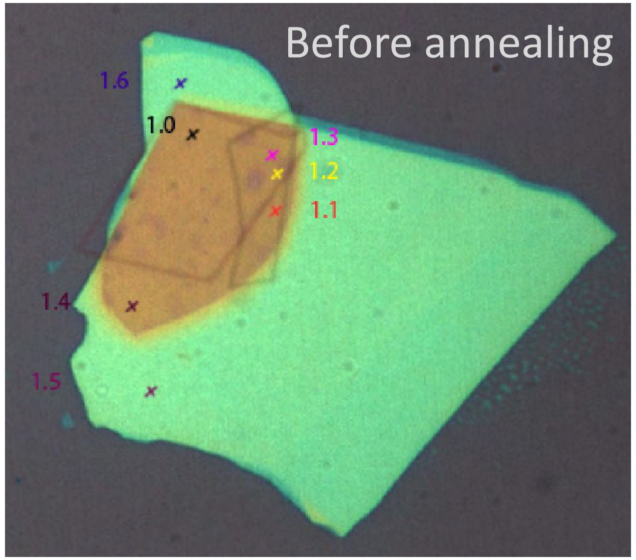
Annealing at
300^o for 3 hrs.

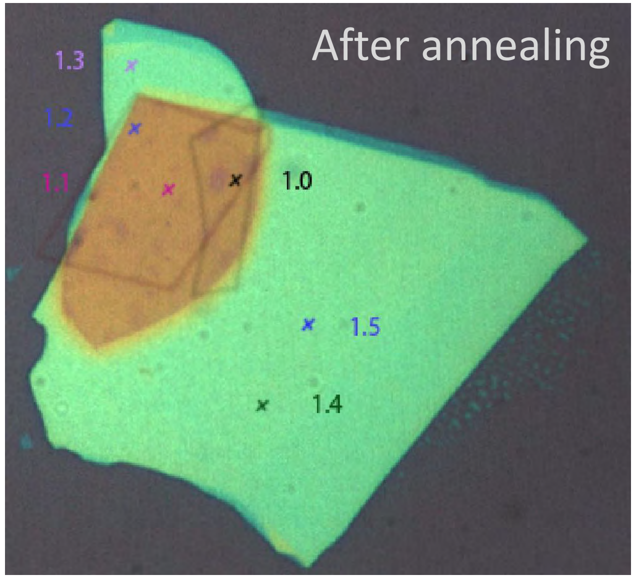


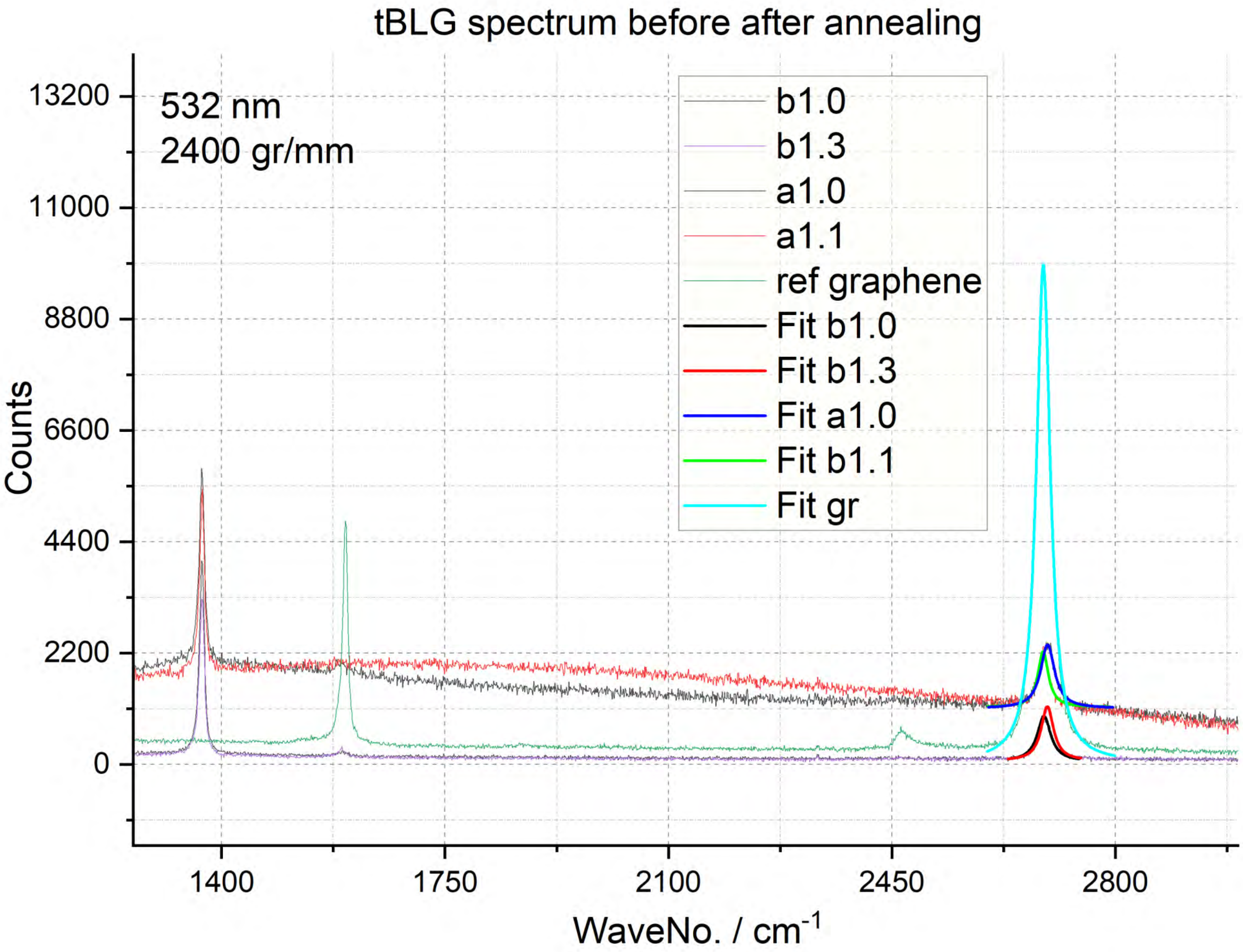


AFM Ironing

tBLG_1 Angle ~24°

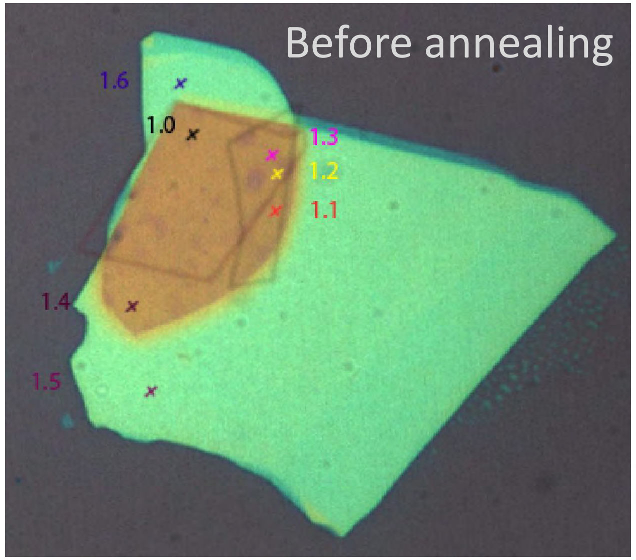


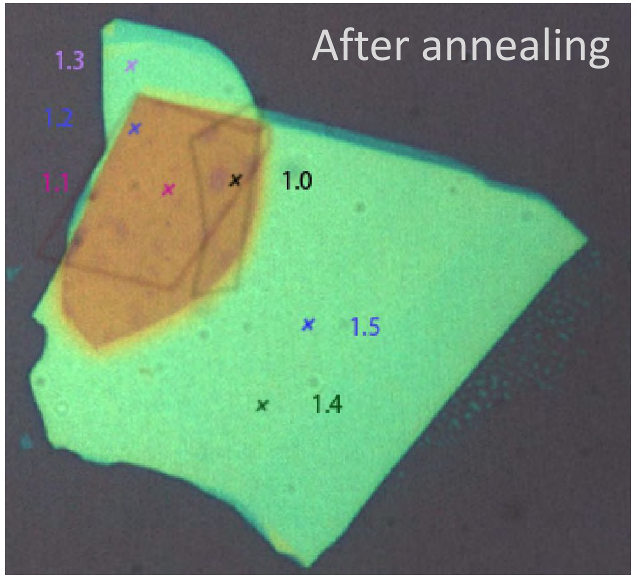


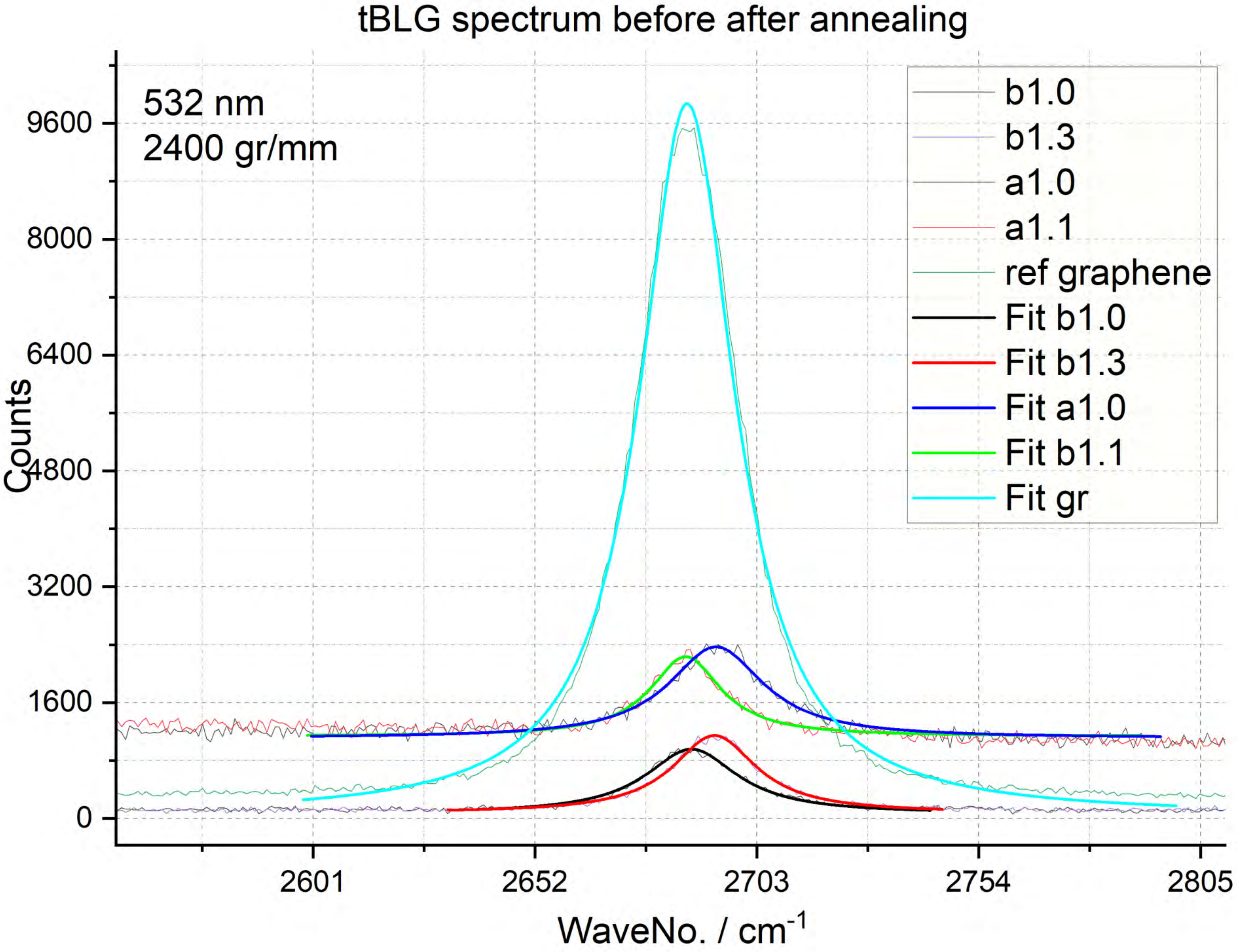


24-04-2023 JOVI K

tBLG_1 Angle ~24º

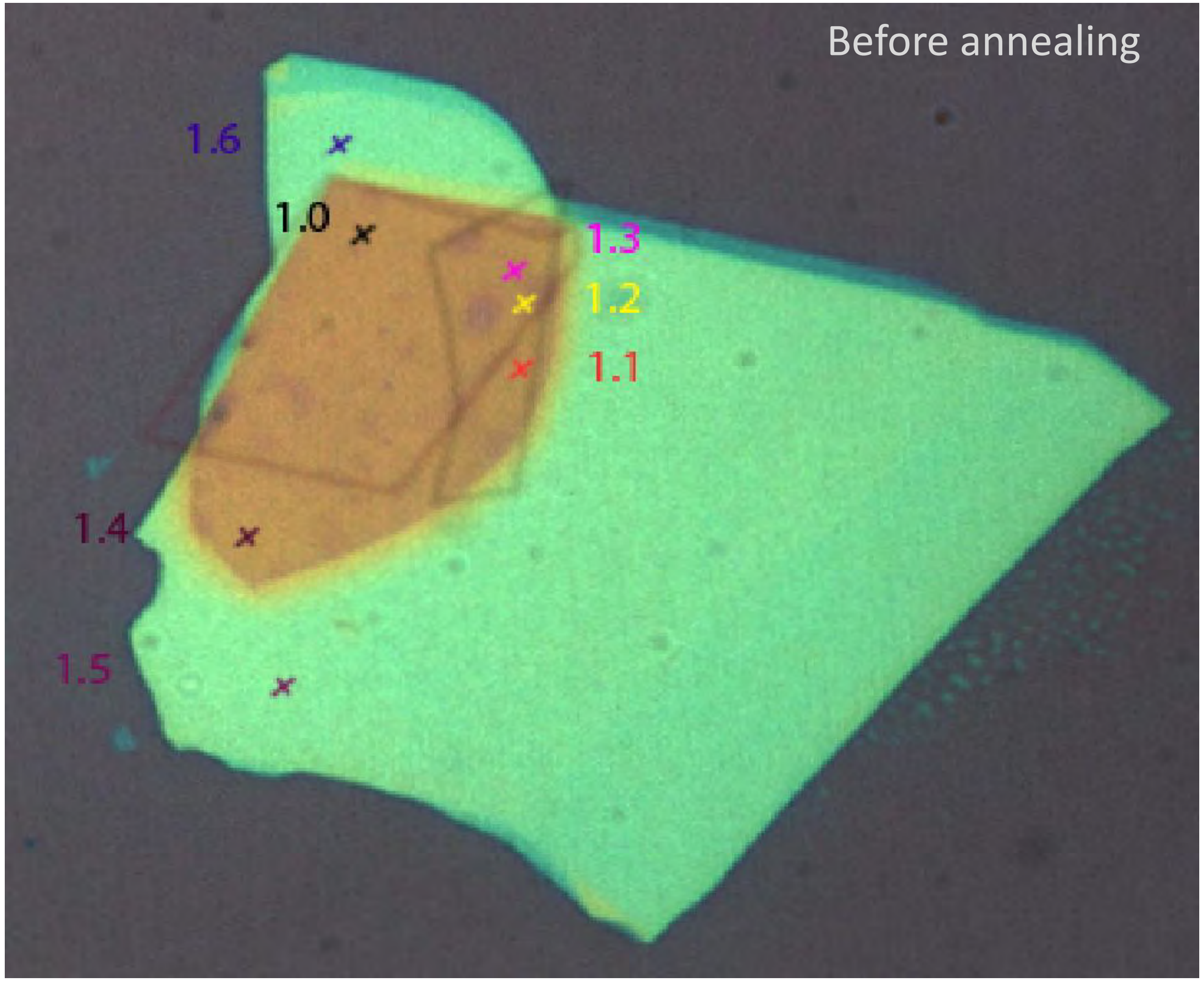


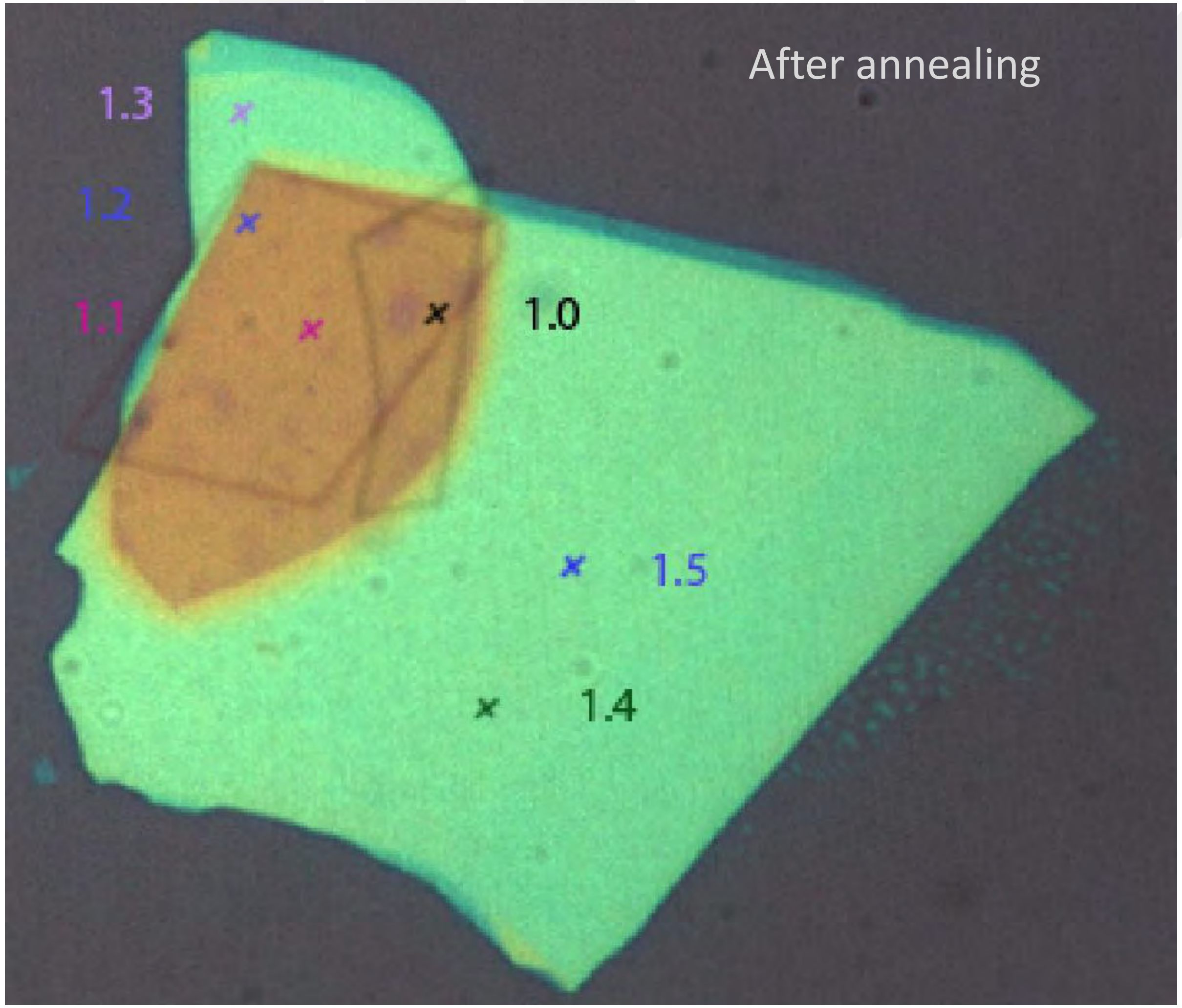




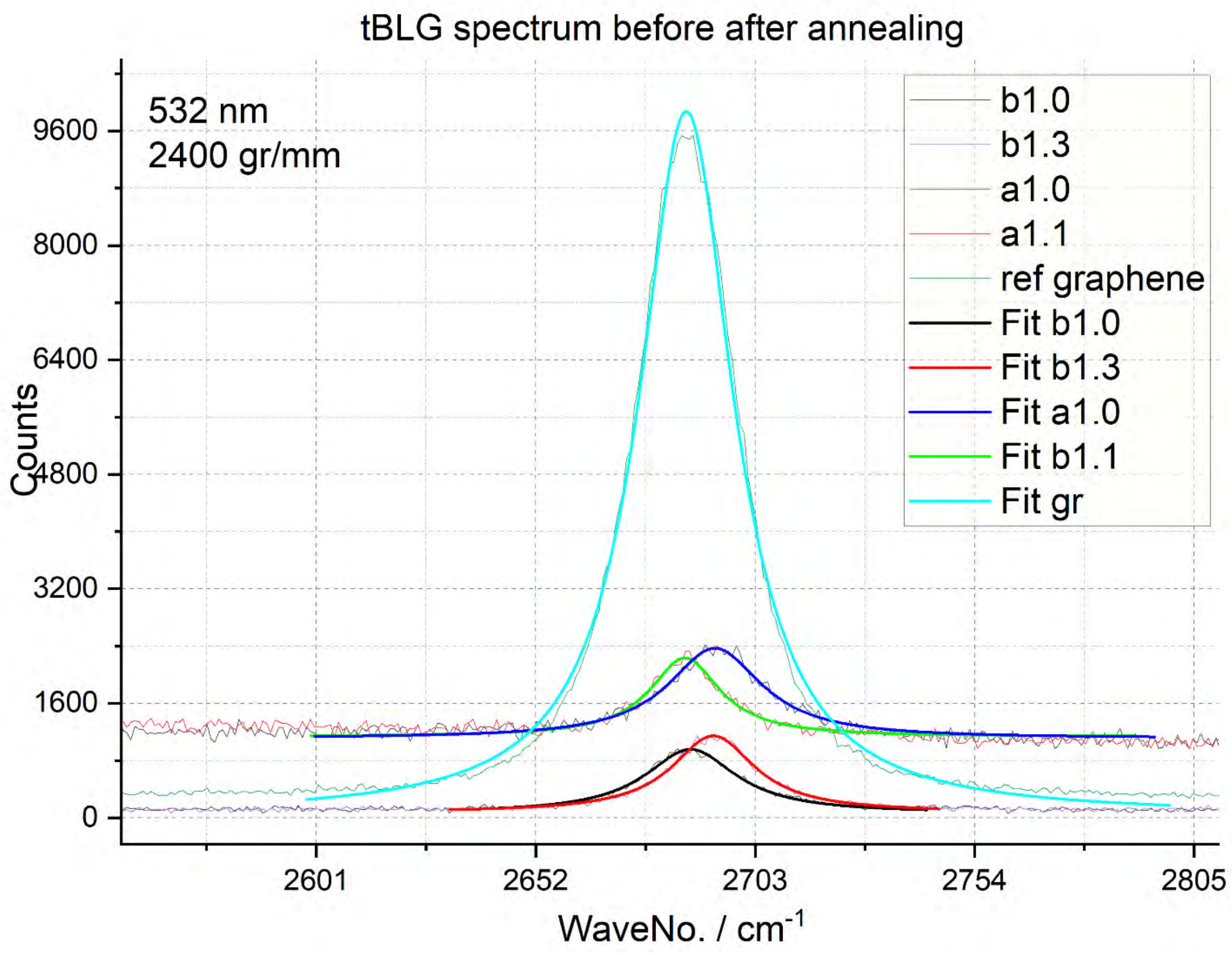
24-04-2023 JOVI K

5% Power


Before annealing
1.6
1.0
1.3
1.2
1.1
1.4
1.5


After annealing
1.3
1.2
1.1
1.0
1.5
1.4

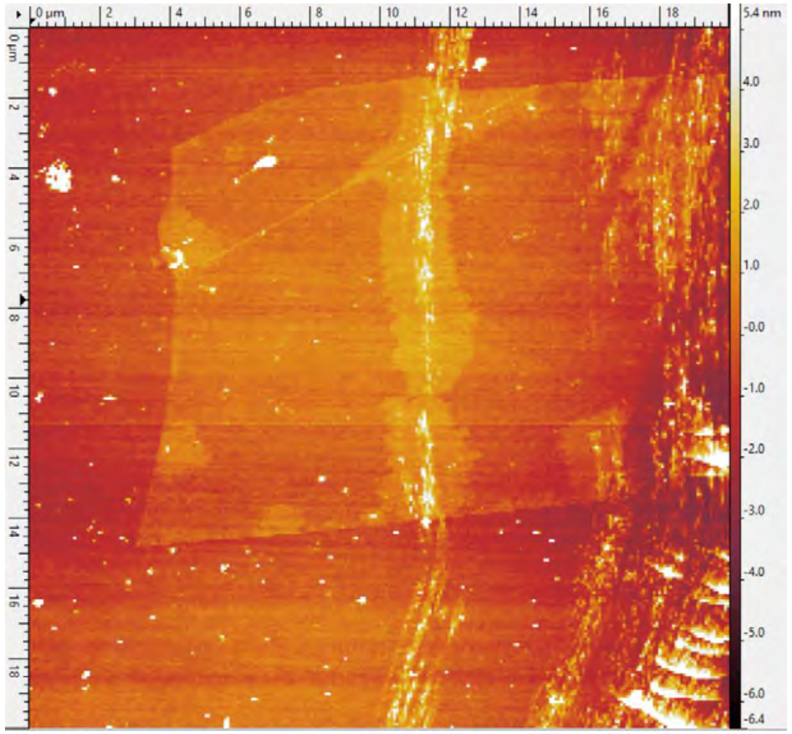
tBLG Angle ~24º



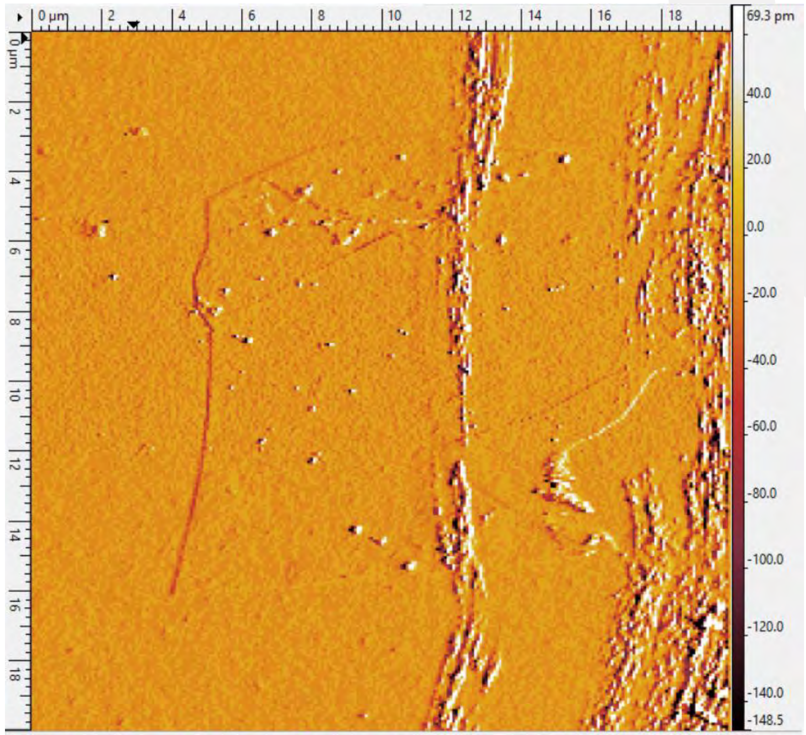
Site	Comment	Centre	FWHM	COD
ref	sLG	2686.97147	26.6696	0.99631
b1.0	Gr stack	2687.27108	25.9488	0.98766
b1.3	tBLG	2693.26407	23.5229	0.99223
a1.0	tBLG	2693.63354	24.6959	0.94786
a1.1	Gr stack	2686.63373	19.9056	0.91869

- tBL has blueshift ~6 cm-1
- Before after heat blueshift is same

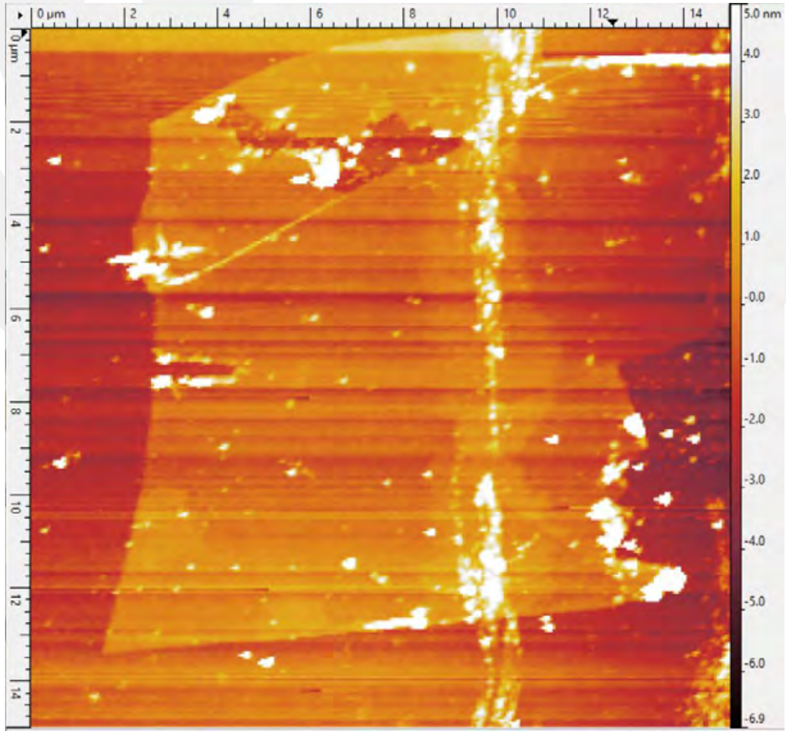
AFM contact Tear



Tap before tear
IG – 0.5
PG – 5
Target – 310mV
Setpoint – 270mV

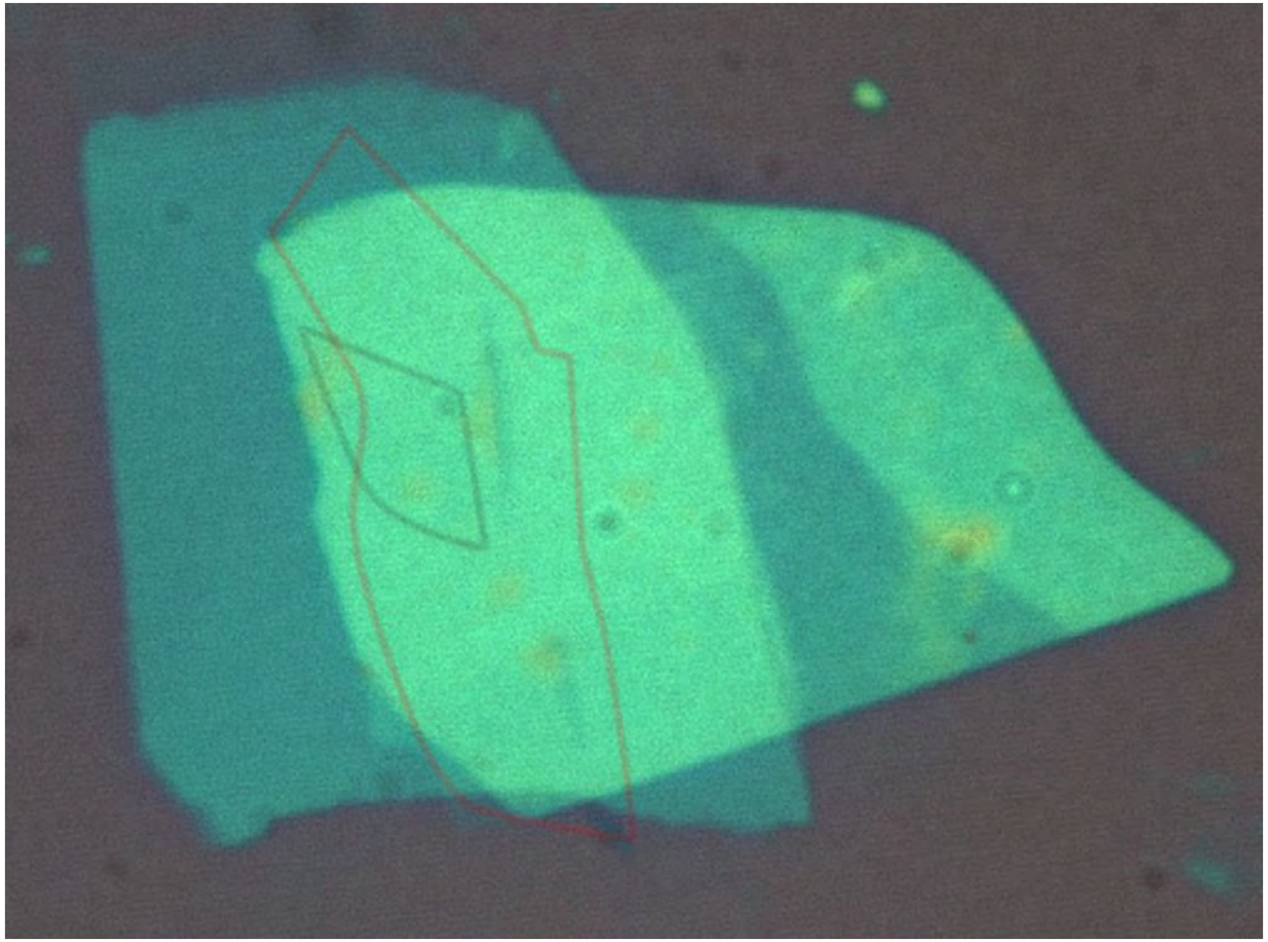


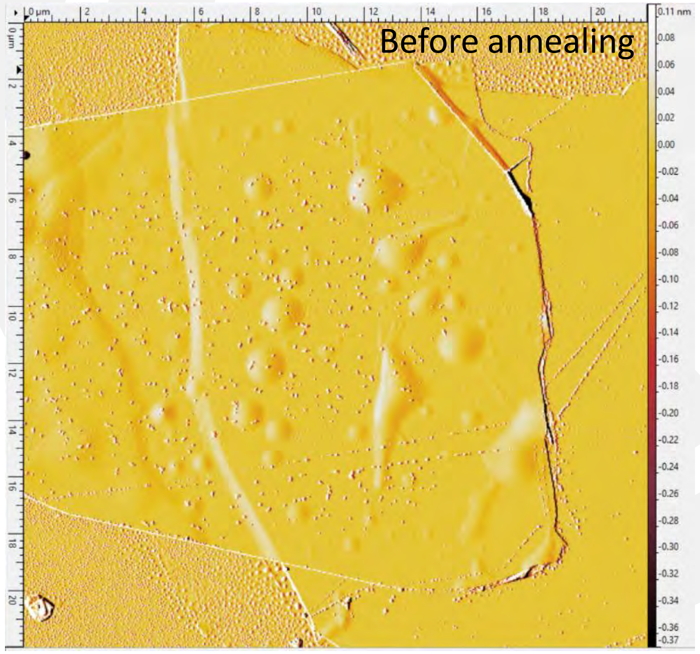
Contact before tear
IG – 5
PG – 10
Setpoint – 0.6 V

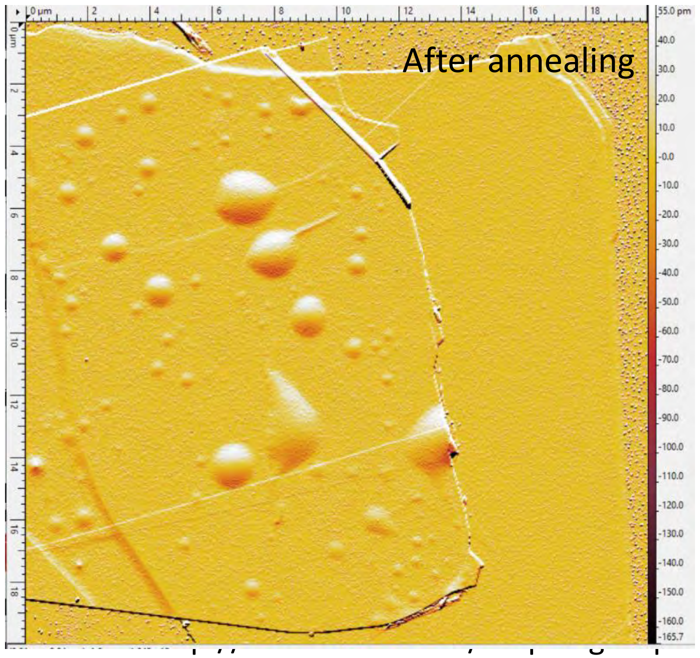


Tap after tear
IG – 0.5
PG – 5
Target – 310mV
Setpoint – 240mV

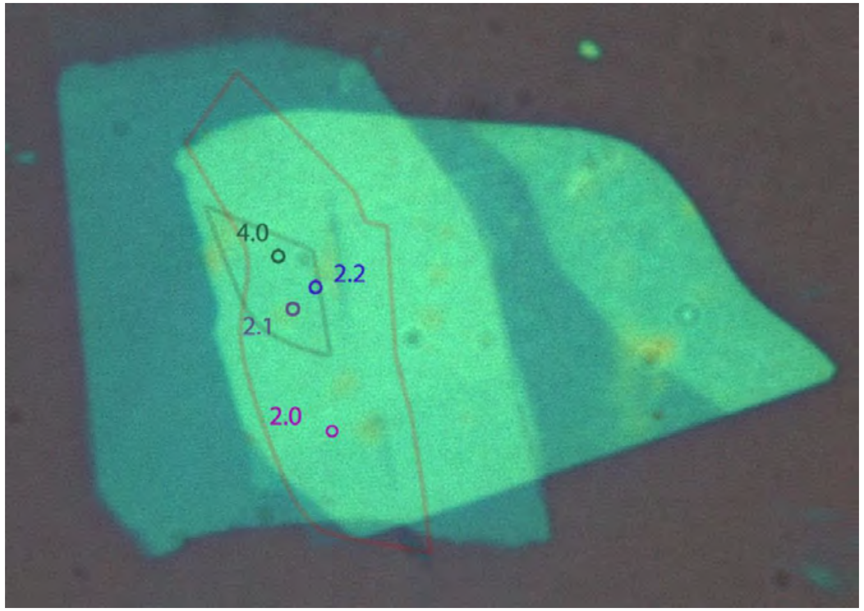
tBLG_2 Expected angle ~ 3^{o}

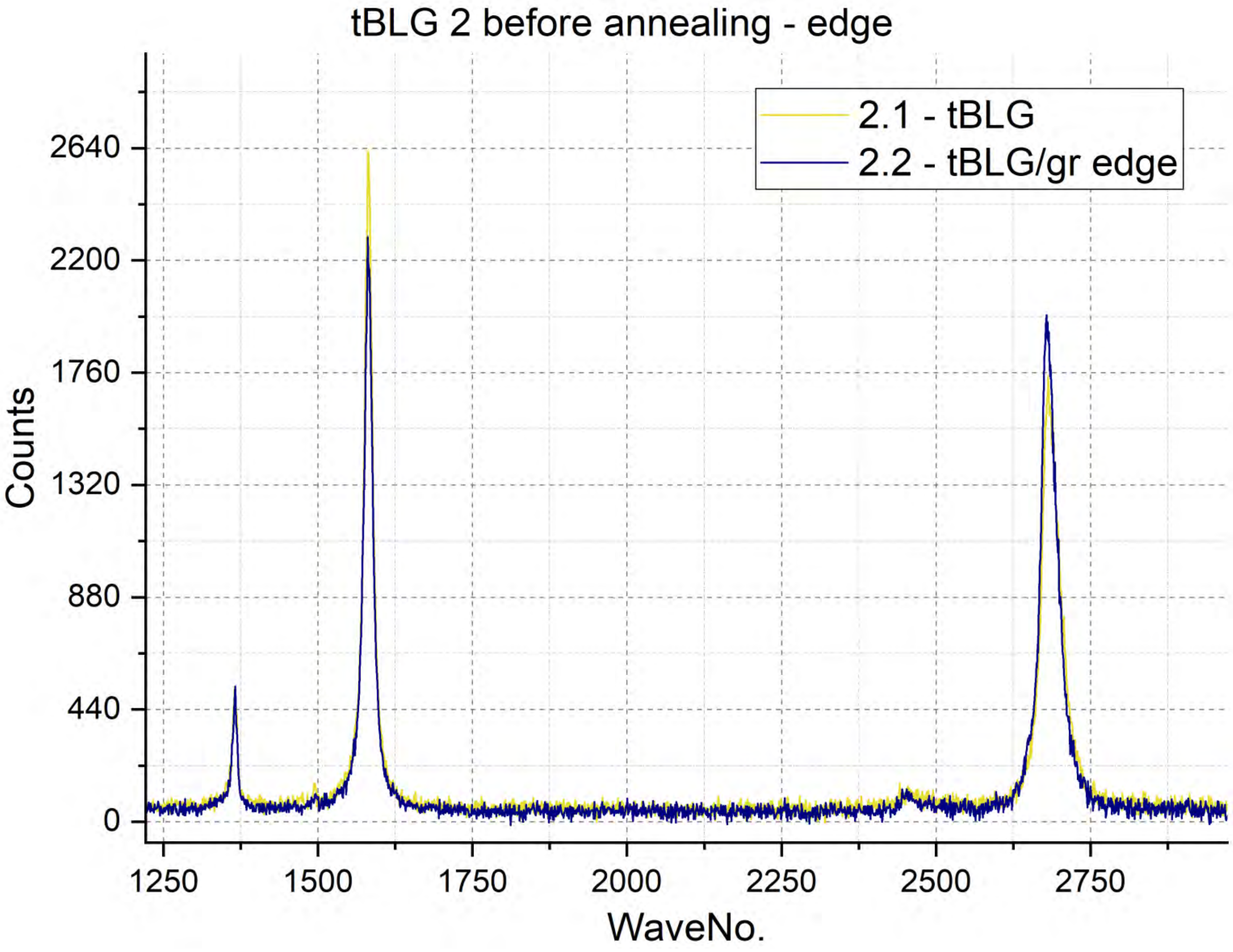




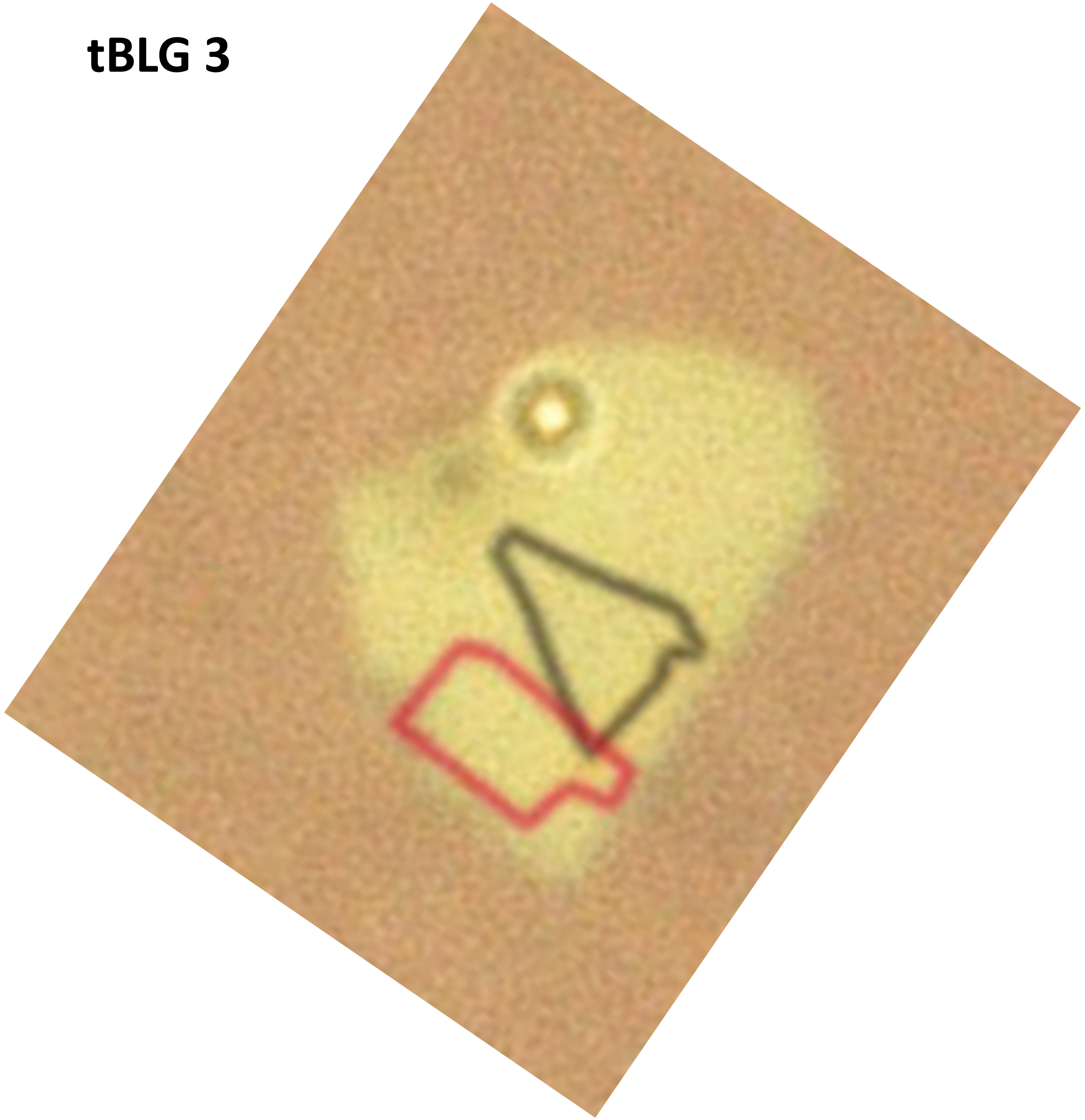


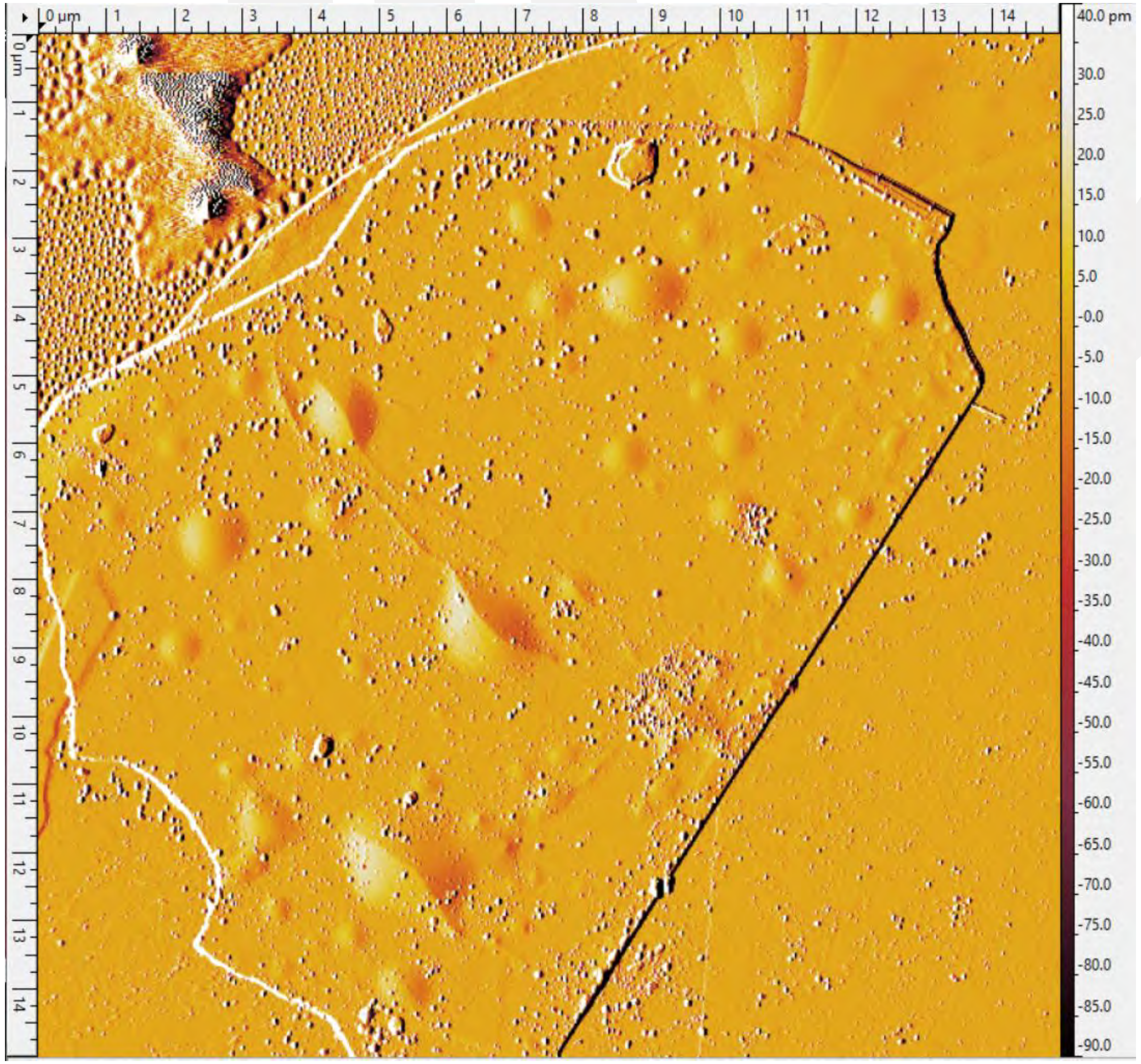
tBLG_2 Expected angle ~ 5°





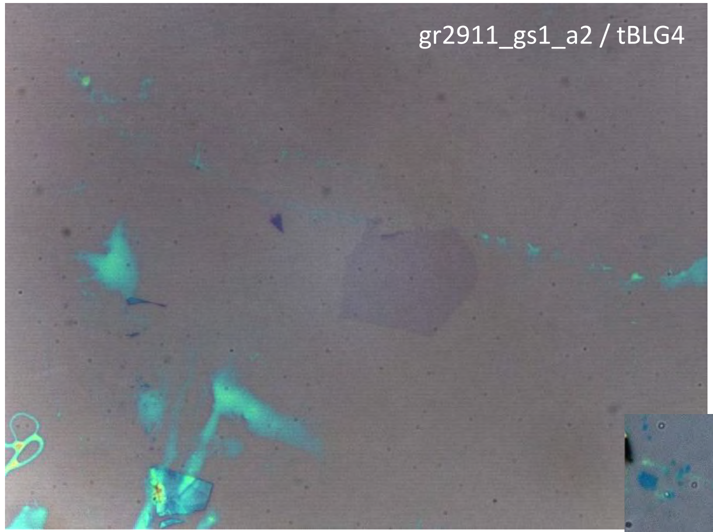
tBLG 3



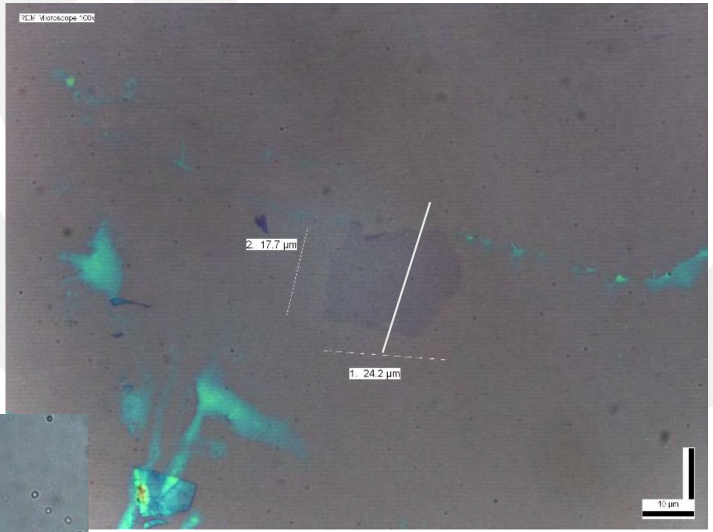


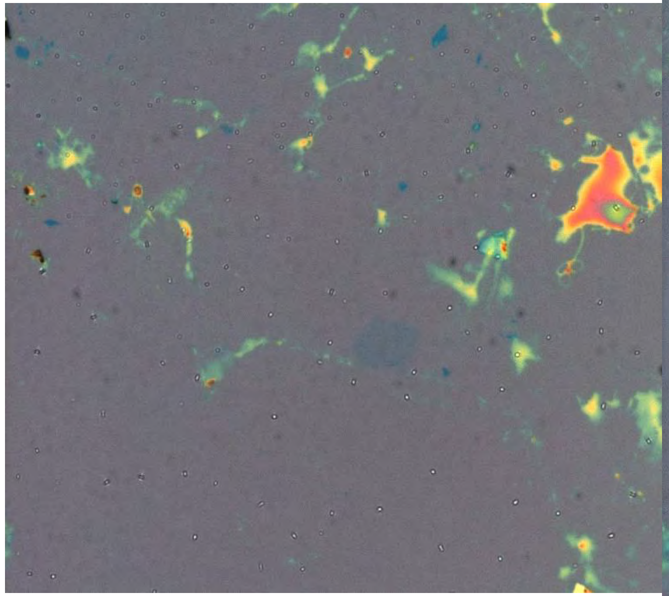
0 µm
1
2
3
4
5
6
7
8
9
10
11
12
13
14
40.0 pm
30.0
25.0
20.0
15.0
10.0
5.0
-0.0
-5.0
-10.0
-15.0
-20.0
-25.0
-30.0
-35.0
-40.0
-45.0
-50.0
-55.0
-60.0
-65.0
-70.0
-75.0
-80.0
-85.0
-90.0

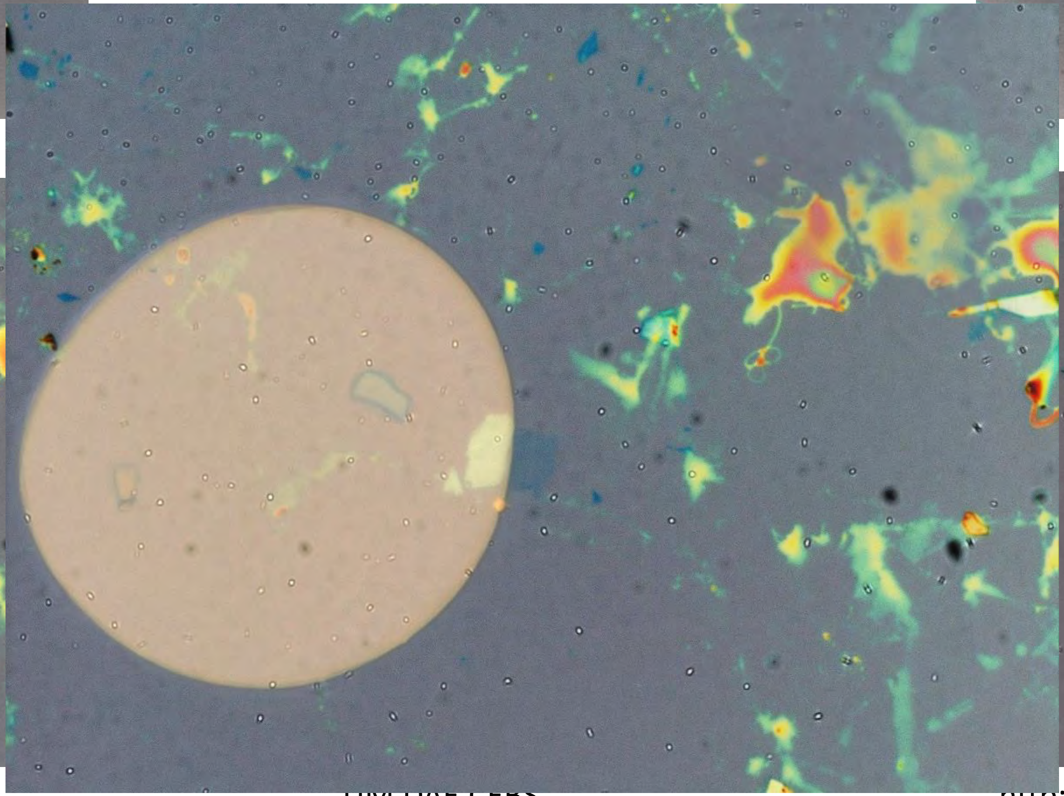
Clean Interface – tBLG4

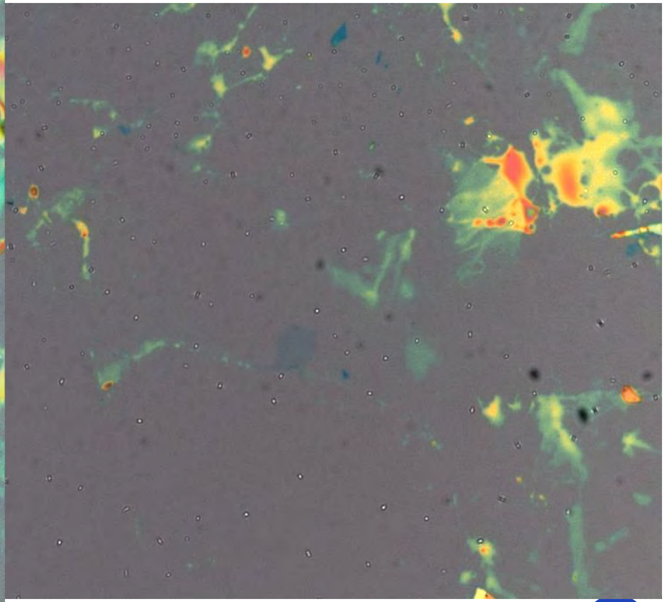


AFM tear

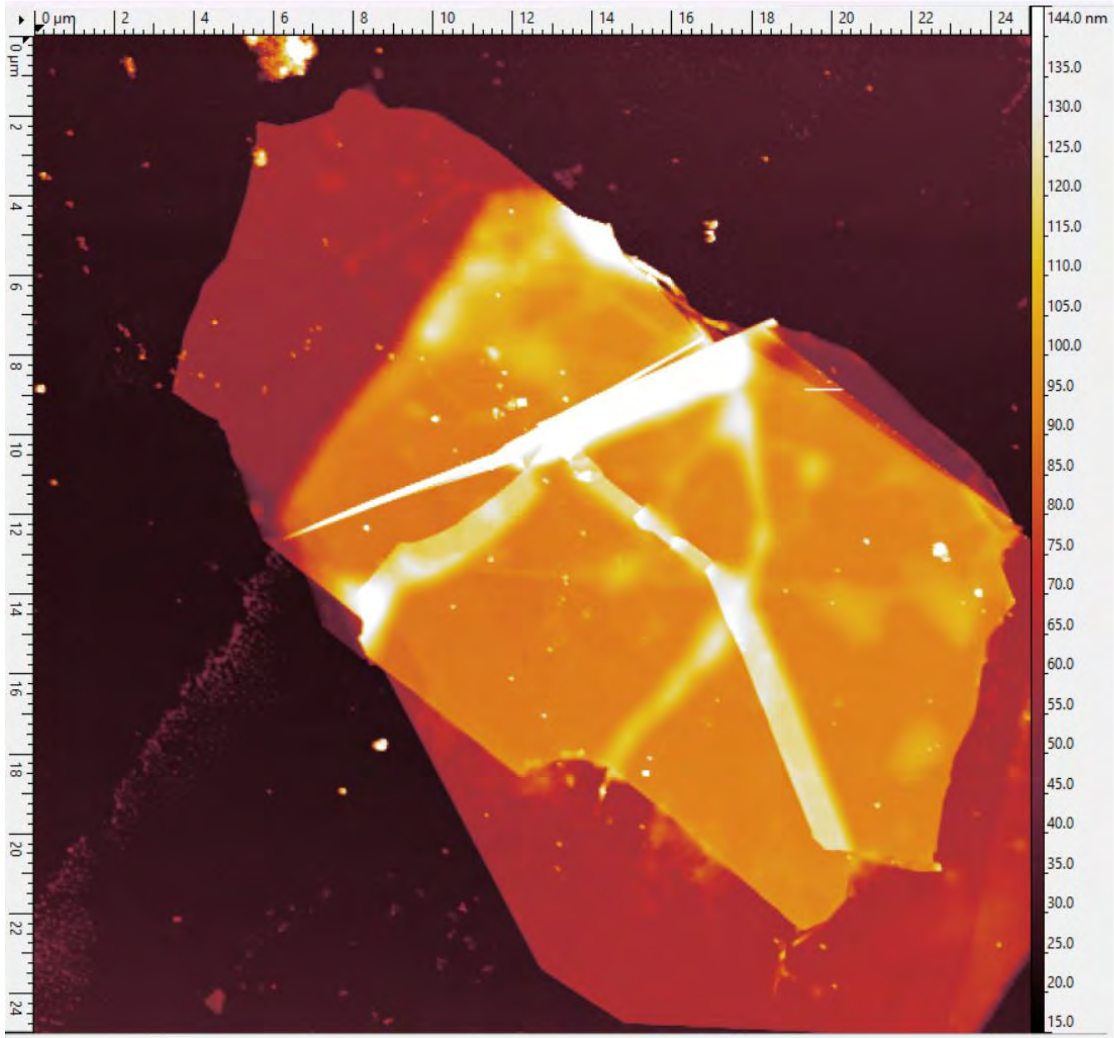


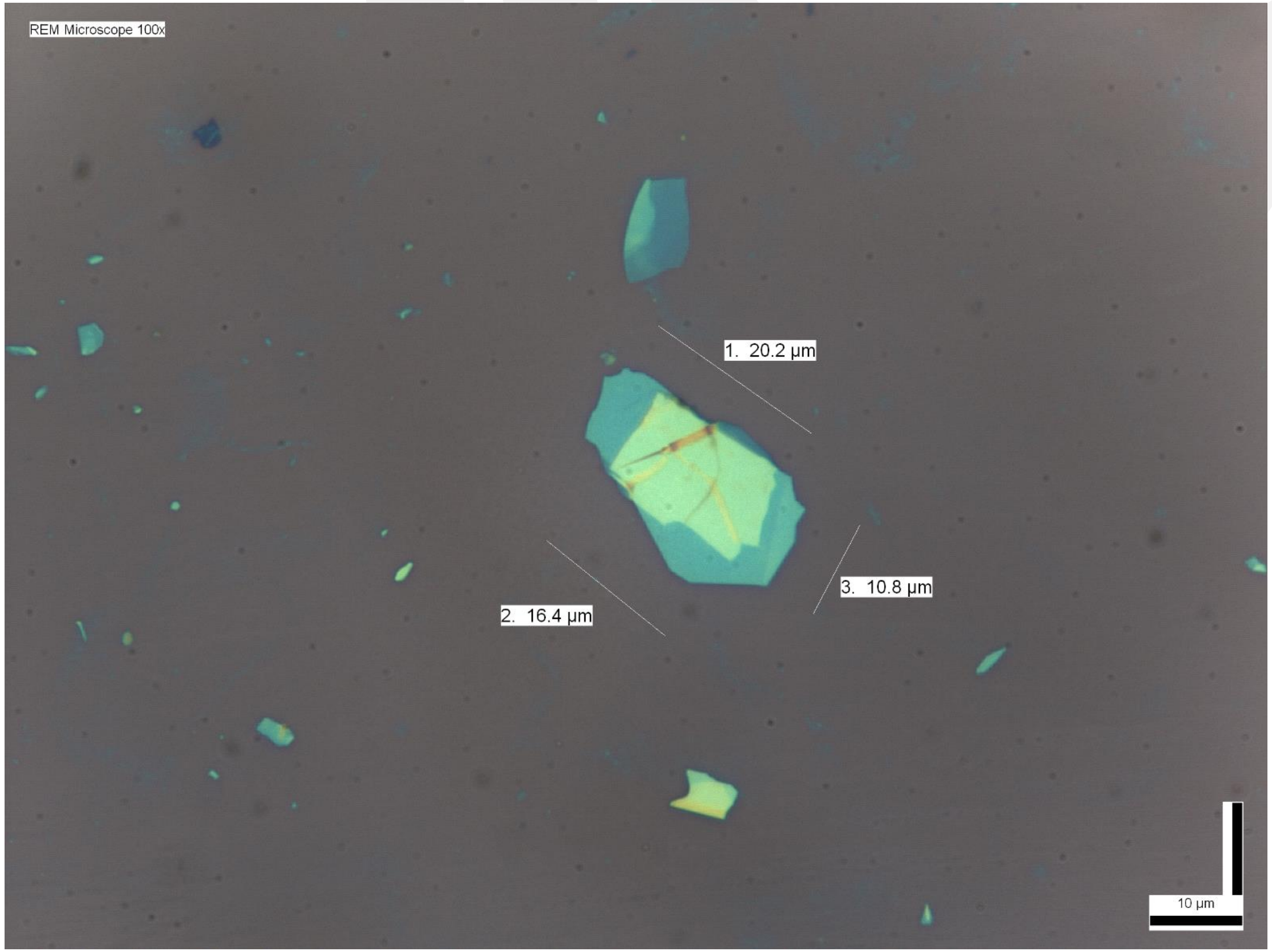






Clean Interface – tBLG4





Clean Interface – tBLG4