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Long-range atomic ordering and variable interlayer interactions in two overlapping graphene lattices with stacking misorientations

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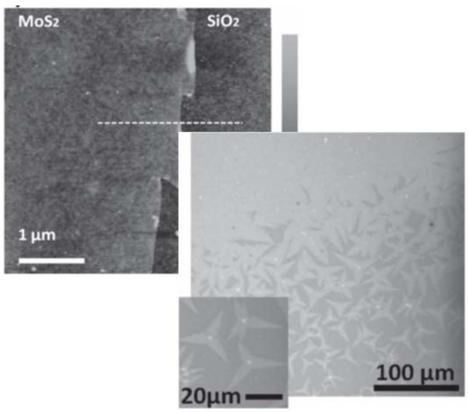
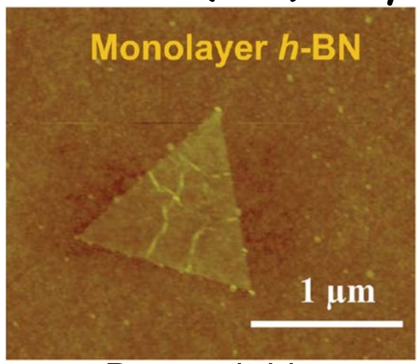
2D-crystals are becoming available

- Various two-dimensional (2D)-crystals



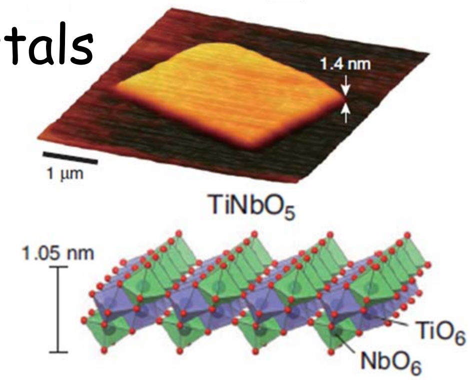
Molybdenum disulfide

Lee et al., Advanced Materials, 24, 2320 (2012)



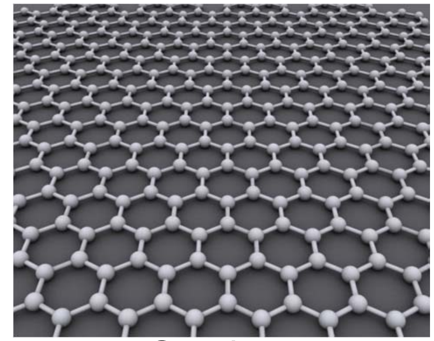
Boron nitride

Kim et al., Nano Lett., 12, 161 (2012)



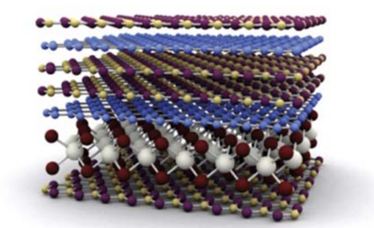
Titanium Niobate

Osada et al., Adv. Funct. Mater. 21, 3482 (2011)

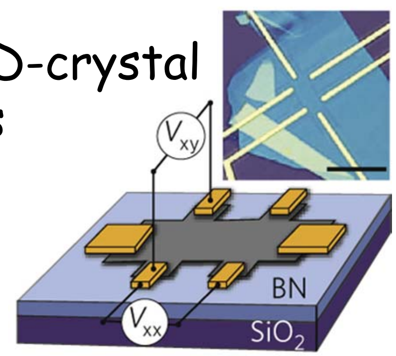

Graphene

http://en.wikipedia.org/wiki/Graphene

- Hybrid 2D-solids can be realized
  - Combining the property of each 2D-crystal
  - Exploring the emerging properties

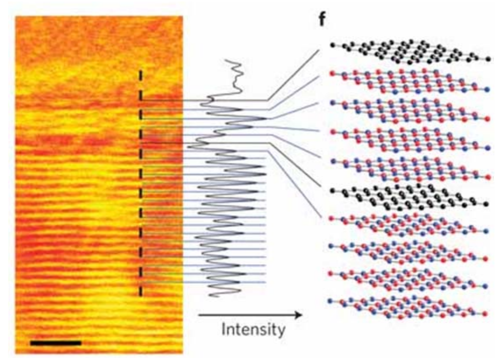

2D-based heterostructure

Novoselov et al., Nature 490, 192 (2012)



Graphene on BN

Dean et al., Nature Physics 7, 693 (2011)



Graphene/BN superlattice

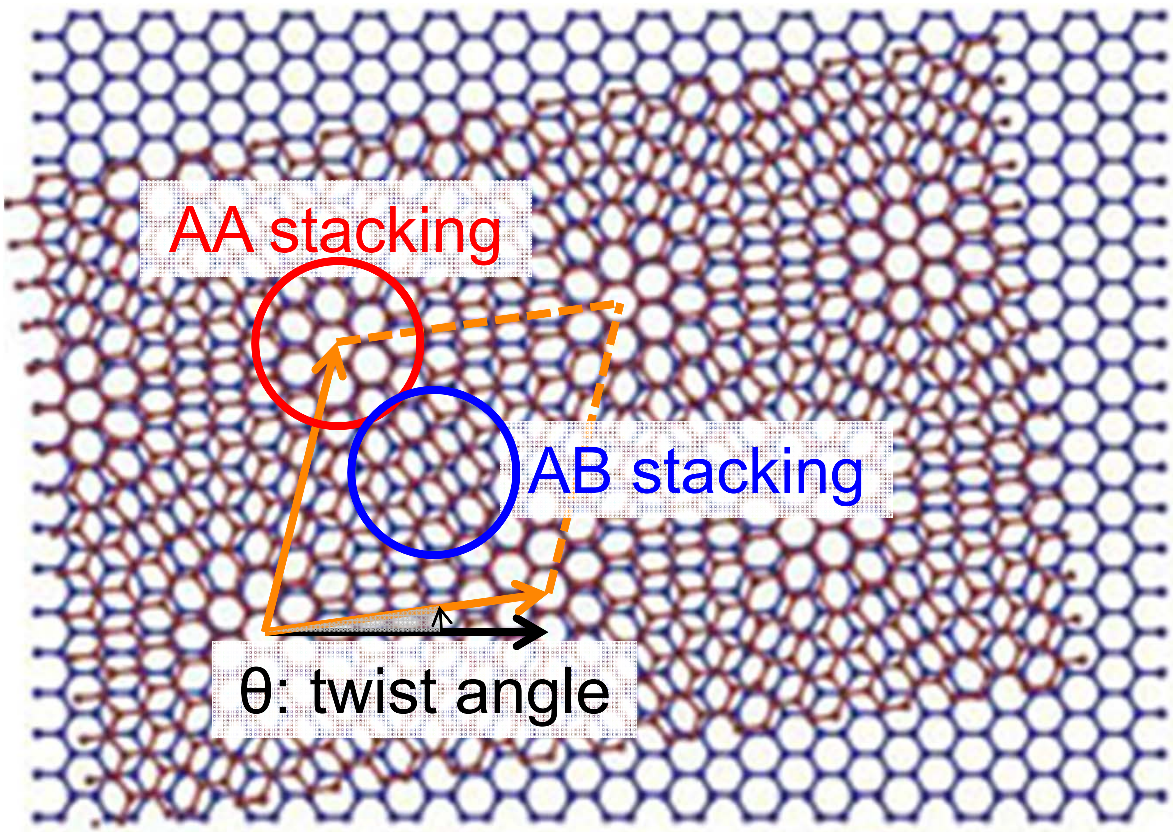
Haigh et al., Nature Materials 11, 764 (2012)

<u>How would 2D-crystals interact electronically with each other?</u>

- We examine this question using *twisted bilayer graphene*

What is twisted bilayer graphene (TBG)?

- Two layers of graphene stacked with a relative in-plane angular misorientation

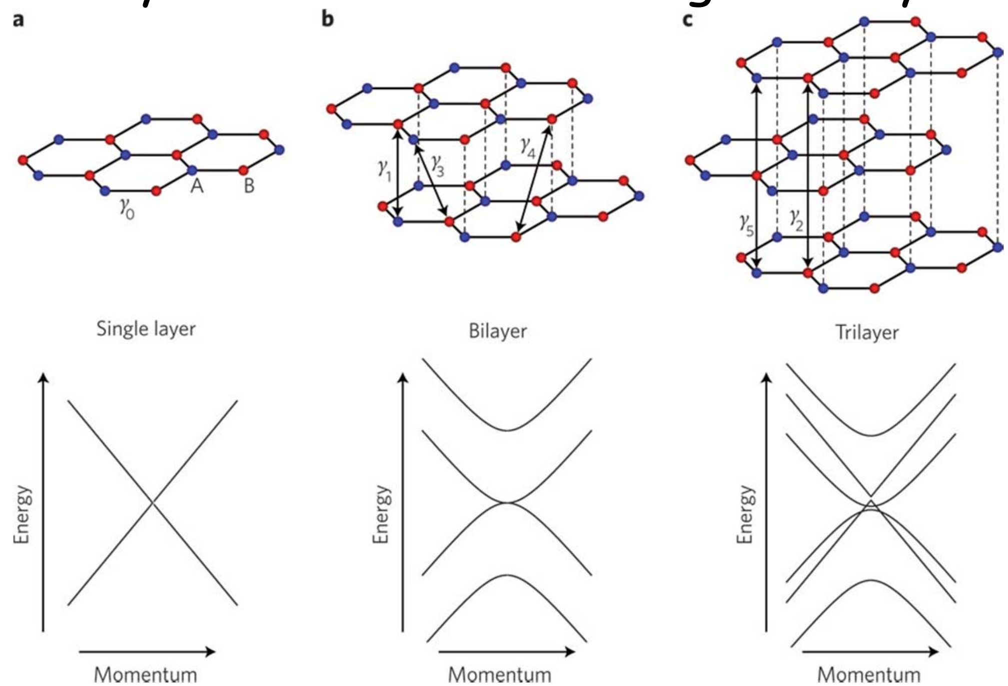


http://www.nist.gov/cnst/epg/sds_graphene.cfm

- Twisting of graphene is the origin of turbostatic disorder in graphite
- TBG is known to exist in various forms of graphene
  - CVD grown graphene on metals
  - Epitaxial graphene grown on Carbon-face of SiC

Do graphene sheets interact with misorientation?

- Bernal stacked bilayer is known for strong interlayer interaction

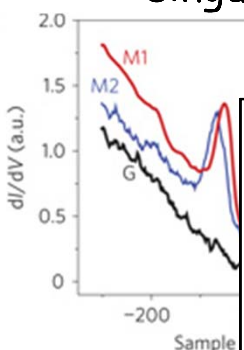


Freitag, Nature Physics 7 596 (2011)

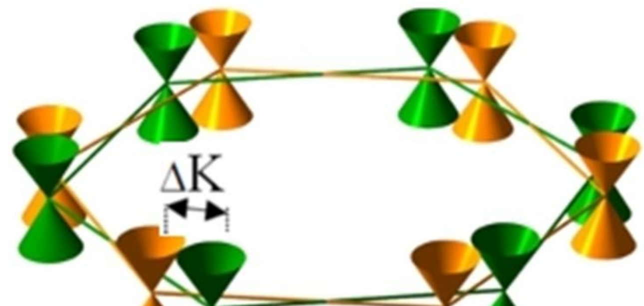
Do graphene sheets interact with misorientation?

- Bernal stacked bilayer is known for strong interlayer interaction

- What is known about twisted graphene?
  - STS indicates the existence of van Hove singularities (vHs) in CVD graphene

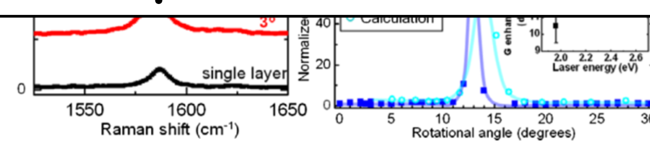


Li et al., Nature Ph

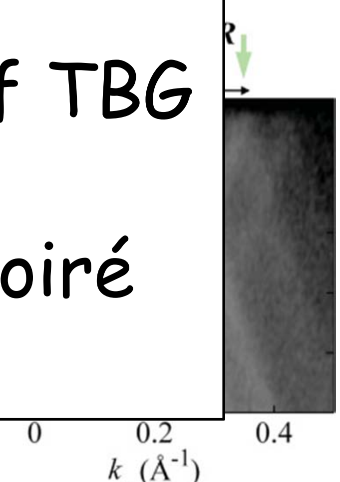


Are there vHs?

- Microscopic and atomic view of TBG
- Interacting Dirac cones
- Band renormalization due to moiré periodic potential



Kim et al., PRL 108, 246103 (2012)

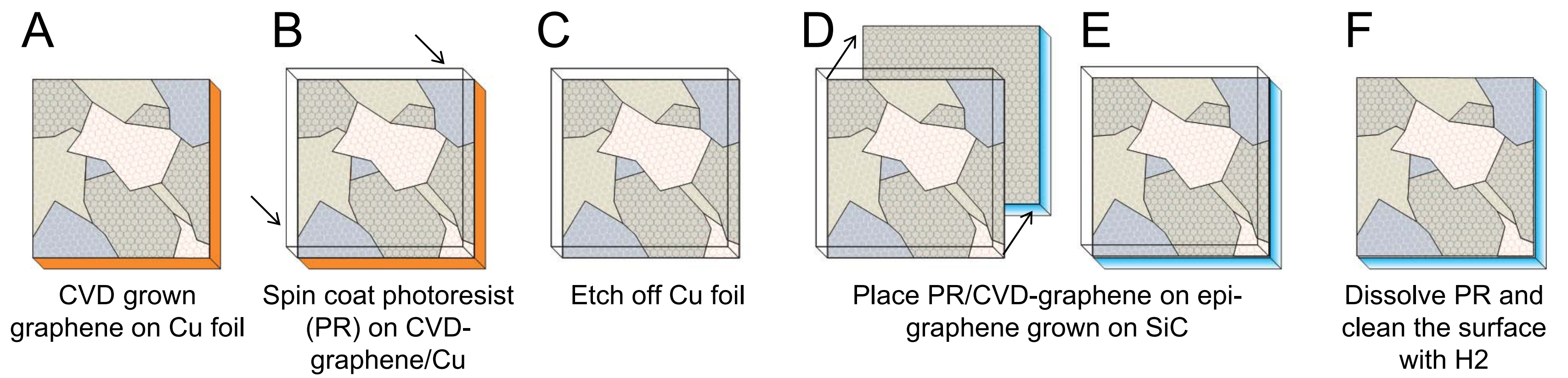


Hicks et al., PRB 83, 205403 (2011)

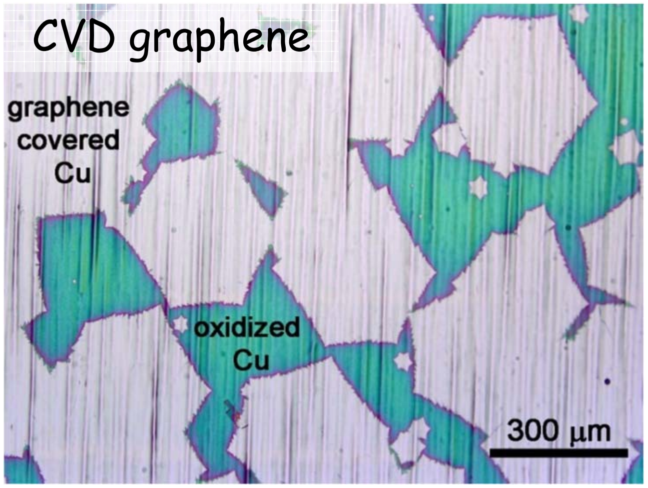
  - ARPES suggest non-interacting states for twisted multilayer graphene on SiC (C-face)

How we make TBG

- Transferring graphene overlayer (grown on Cu foil) onto epi-graphene underlayer (grown on SiC substrate)

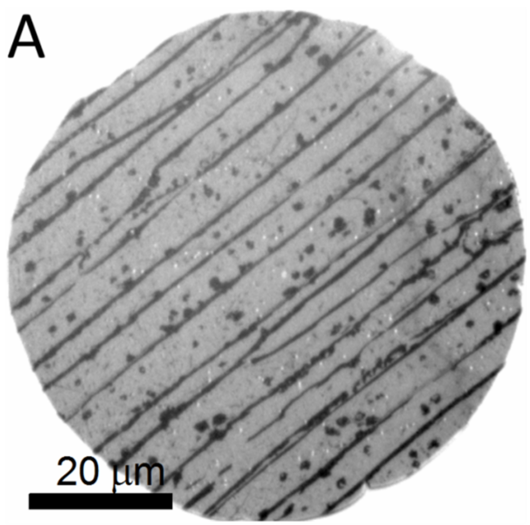


  - Monolithic underlayer and large-area overlayer (>100um-size domain)
  - Large TBG domains with random twist angles

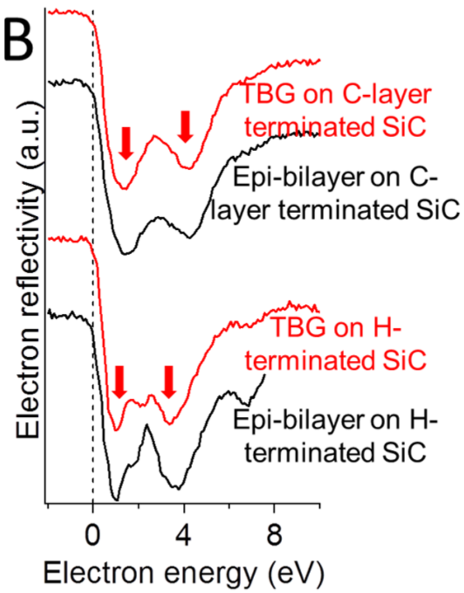


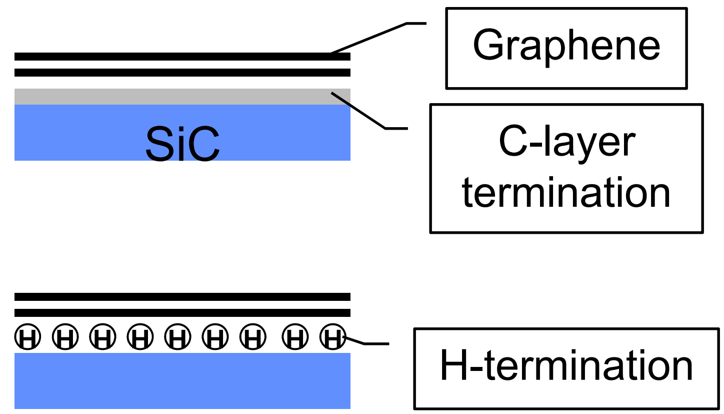
TBG shows characteristic electron reflectivity of bilayer graphene

- Two dips in electron reflectivity spectra are characteristics of bilayer graphene on SiC
  - *Low energy electron microscopy (LEEM) measurement*

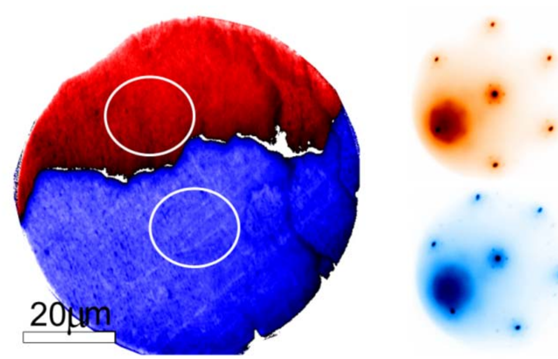


LEEM image of TBG





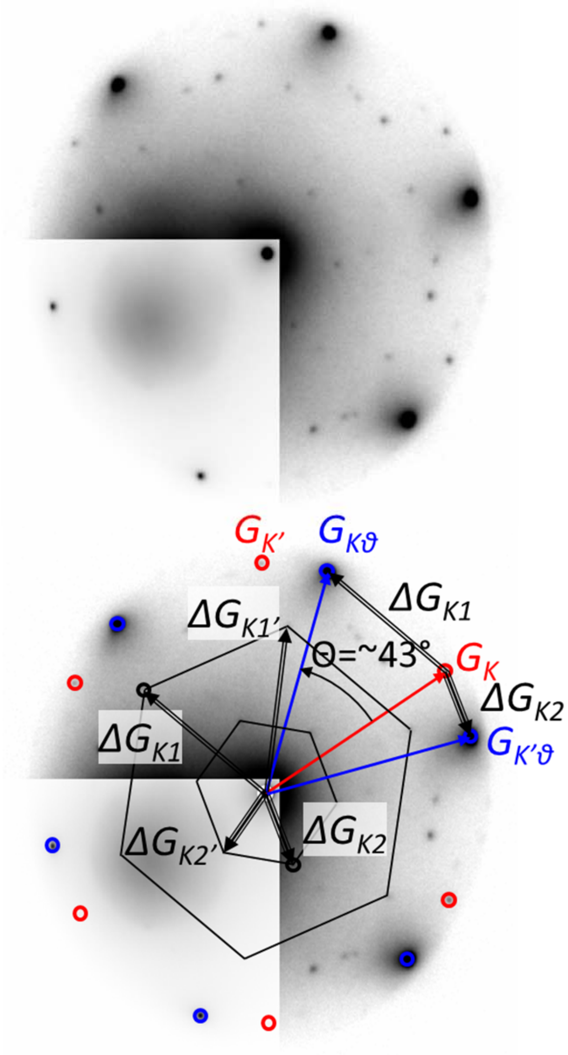
  - Diffraction experiments (area-selected diffraction (5 µm diameter) and dark-field imaging) shows large domains each with an unique twist angle

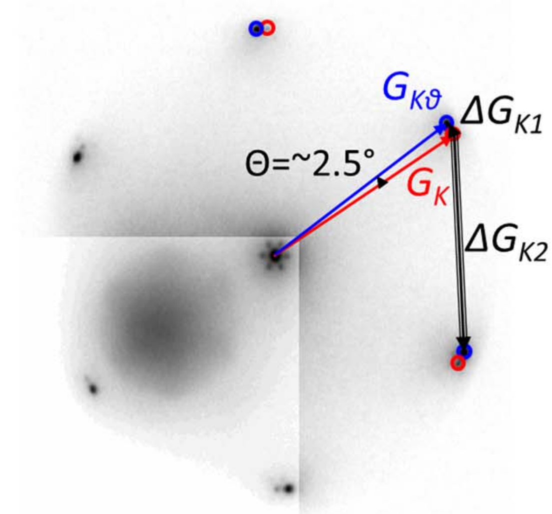


Ohta et al., PRB, 85, 075415 (2012)

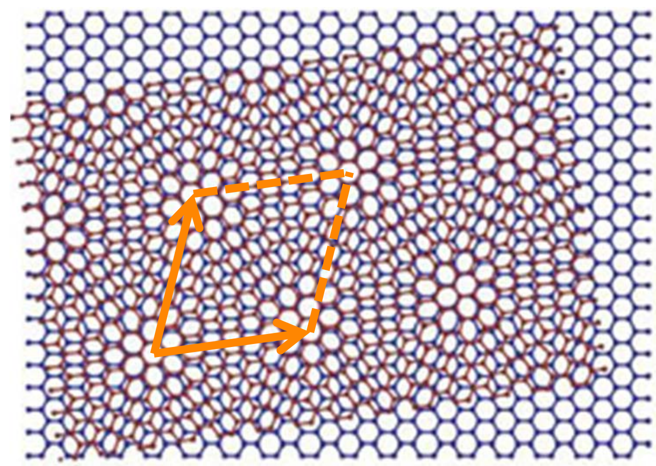
TBG has long-range atomic ordering

- Diffraction patterns from TBG with a large and small twist angles
  - Clear diffraction spots due to moiré





- Specular beam
- Underlayer diffraction spots
- Overlayer diffraction spots
- 6√3 x 6√3 cos30° BL diffraction spots
- Moiré diffraction spots

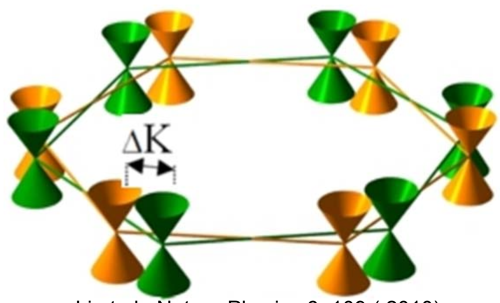


Real-space moiré vectors

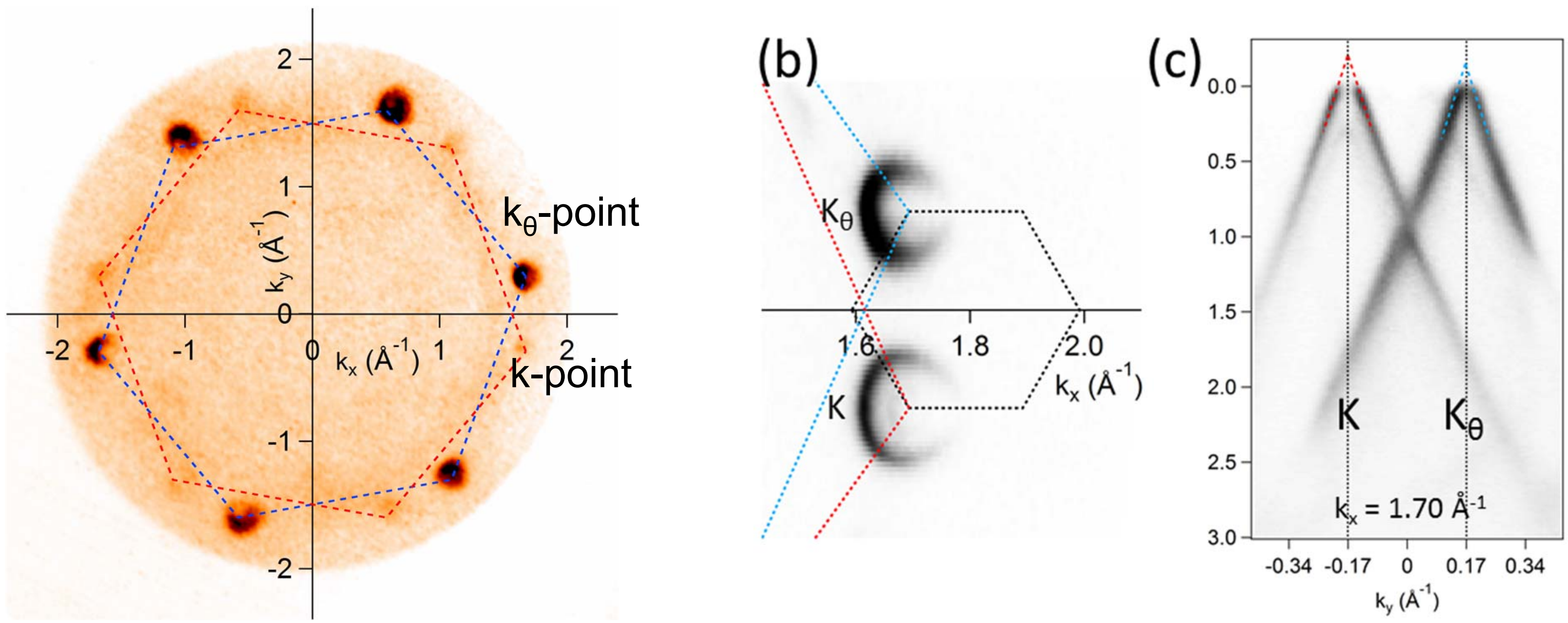
- Graphene's high material quality is also shown using Raman spectroscopy
  - Please see PRB, 85, 075415 (2012) for detail

TBG has two sets of Dirac cones

- Electronic dispersion measured using photoemission electron microscopy and angle-resolved photoemission spectroscopy (ARPES)
  - Two sets of Dirac cones come from upper (k_θ-point) and lower (k-point) graphene sheets



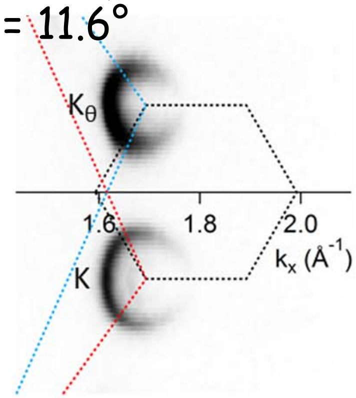
Li et al., Nature Physics 6, 109 (2010)

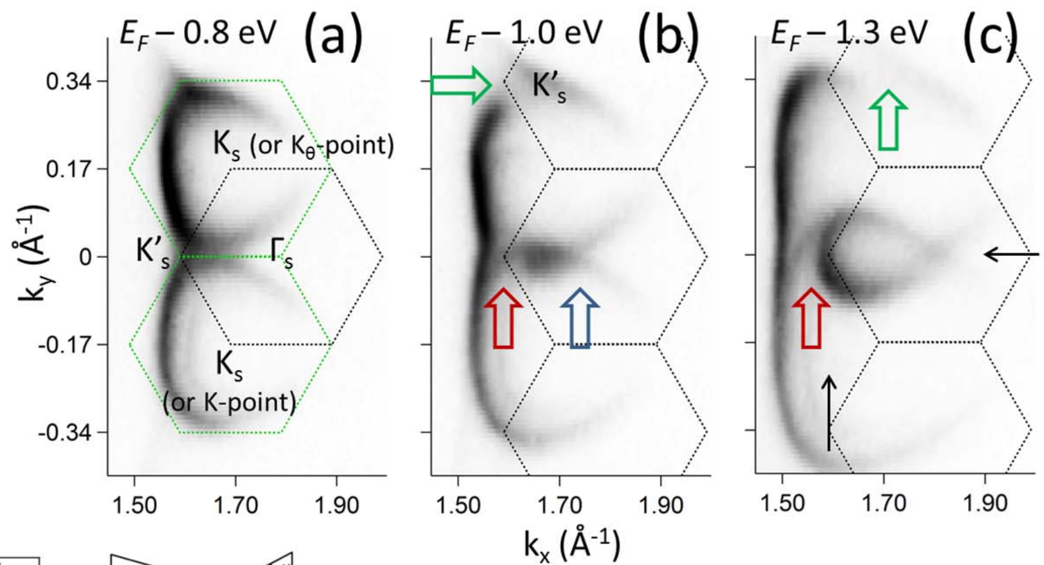


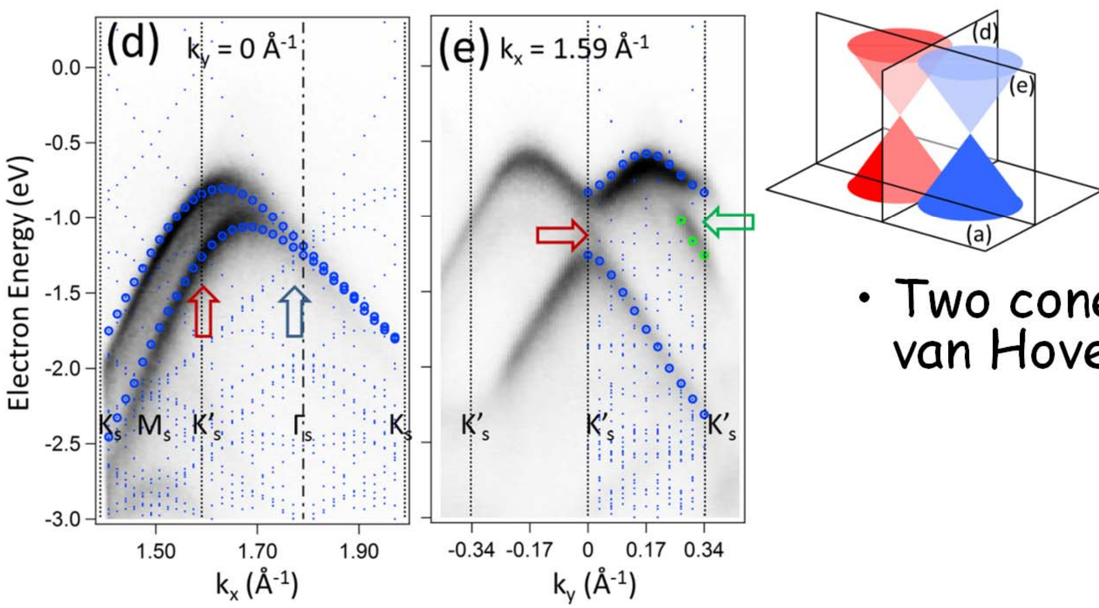
Ohta et al., PRL, accepted

Two Dirac cones display anti-crossing

- Photoemission intensity contours show the departure from the simple Dirac cone picture
  - Twist angle θ = 11.6°



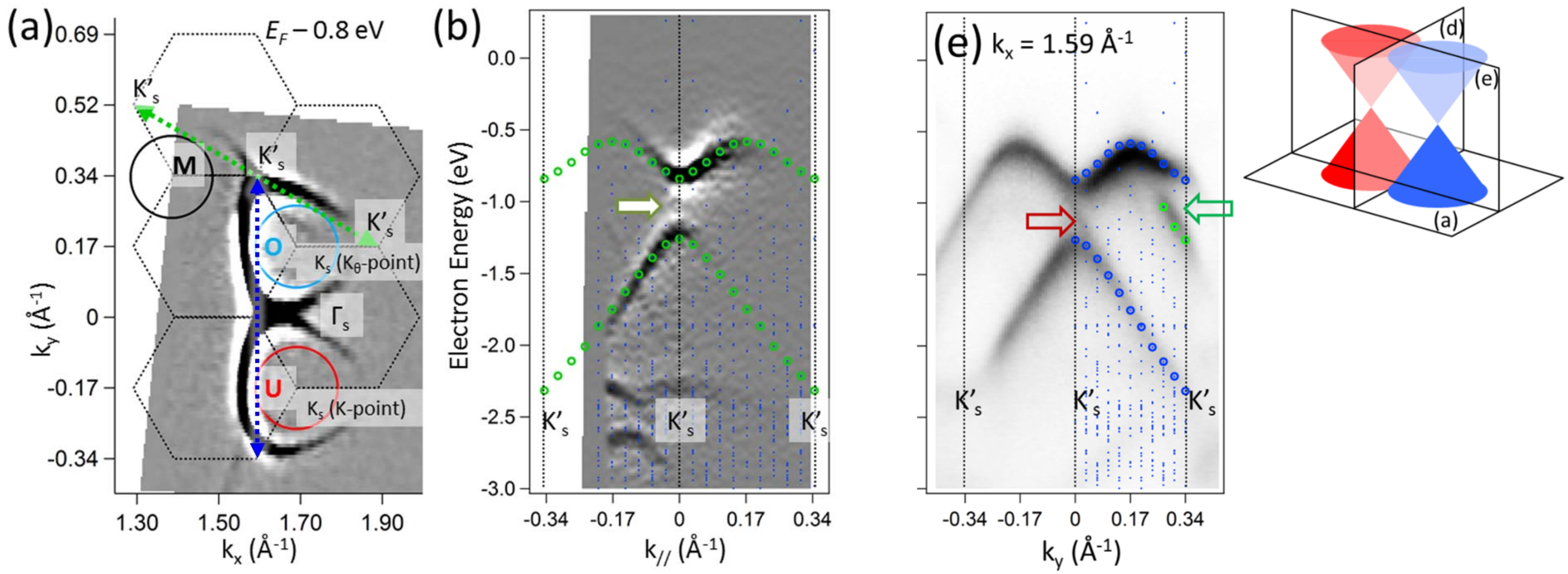




- Two cones' anti-crossing leads to van Hove singularities

Periodic potential renormalizes Dirac cone dispersions

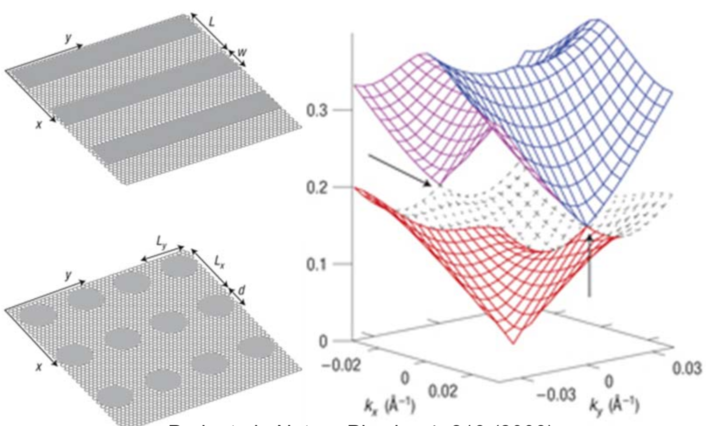
- Superlattice Dirac cone emerges due to the moiré-induced periodic potential
- Additional anti-crossing appears between the original Dirac cone and the superlattice Dirac cone



- Dirac cone dispersion is affected by the periodic potential of moiré superlattice

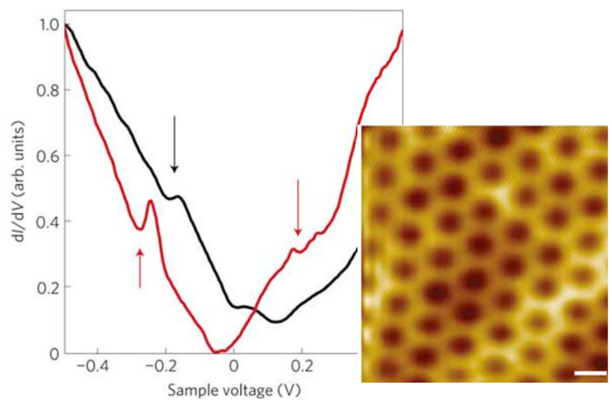
Periodic potential affects electronic dispersion in graphene

- Periodic potentials can be provided by the substrates or neighboring materials
  - Electrons in twisted bilayer graphene feel the periodic potentials induced by the interaction between two graphene sheets



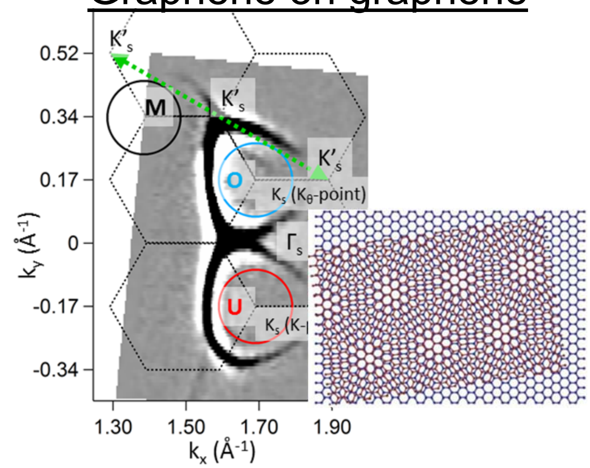
Park et al., Nature Physics 4, 213 (2008)

Graphene on boron-nitride



Yankowitz et al., Nature Physics 8, 382 (2012)

Graphene on graphene

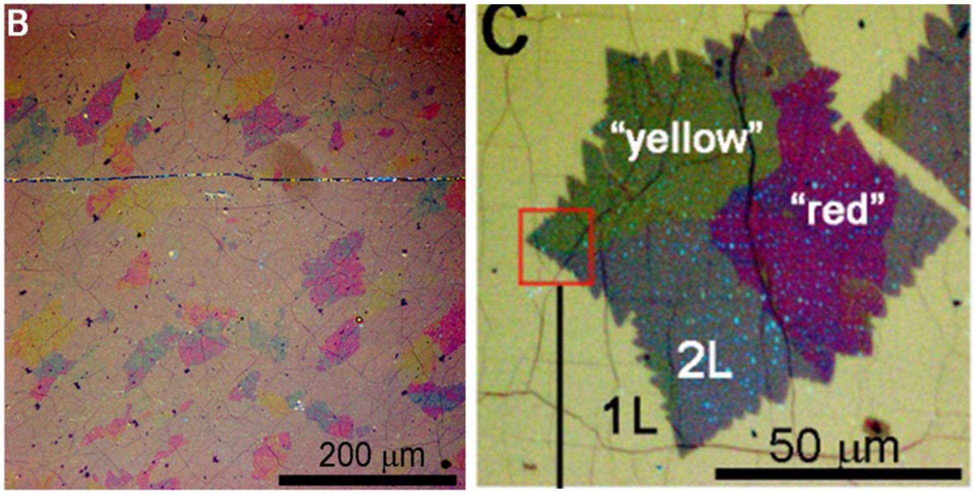


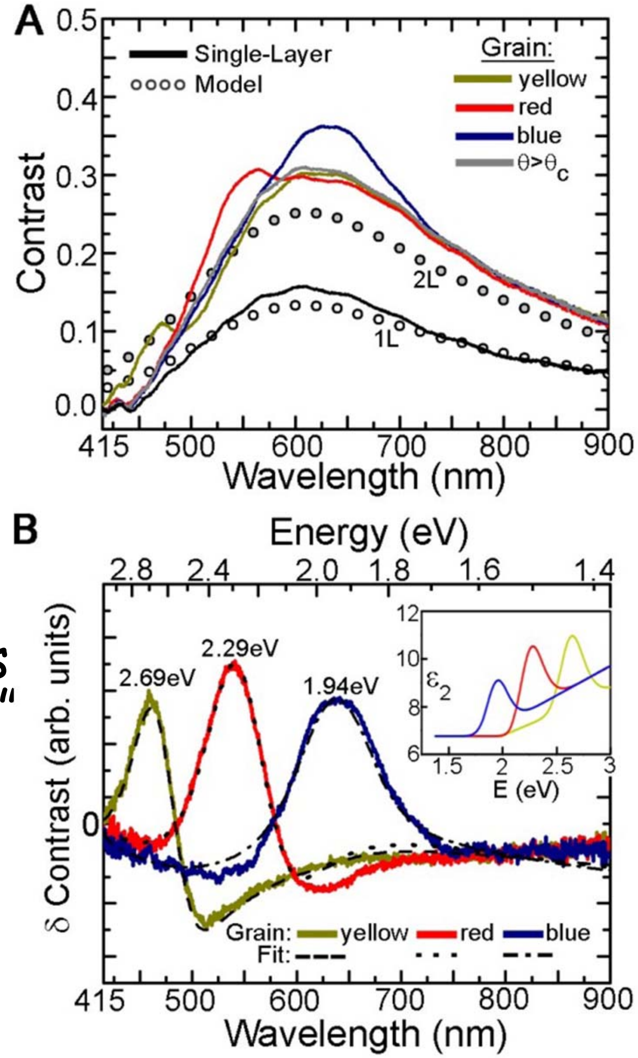
Moiré is ubiquitous!

- Should appear in any hybrid 2D-solids consisting of 2D-crystals with turbostatic disorder
  - New handle to tailor material properties

How does electronic dispersion affect the properties of TBG?

- Patches of "colored grain" observed in optical microscope
  - <u>TBG on SiO_2/Si substrate</u>



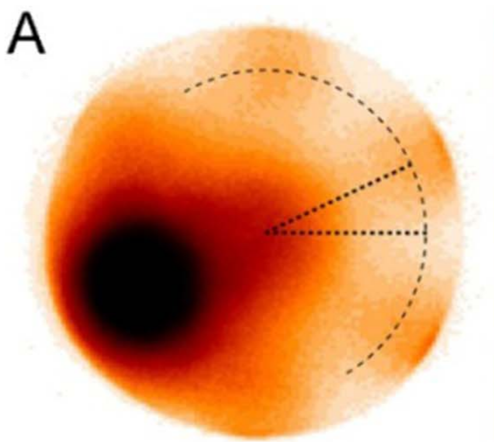


- Change in optical absorption according to the twist angle is responsible for "colored grain"

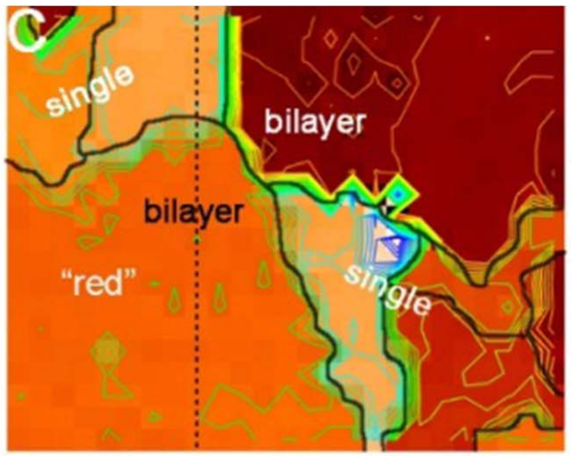
Robinson et al., submitted

We confirmed the twist angle using LEED

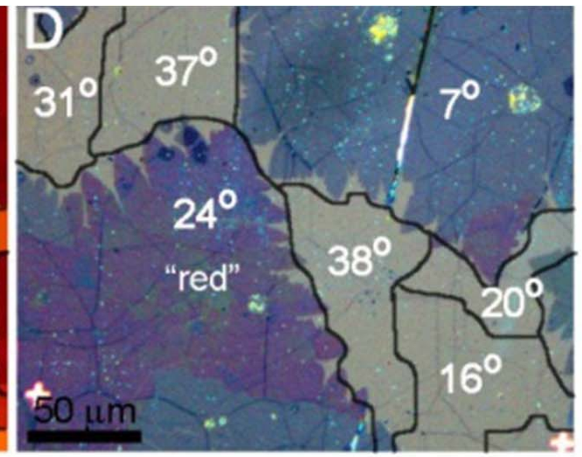
- Twist angle was determined by comparing LEEM pattern orientation map and the information of thickness using optical image
  - LEED is sensitive to only the top layer



Typical LEED pattern of TBG

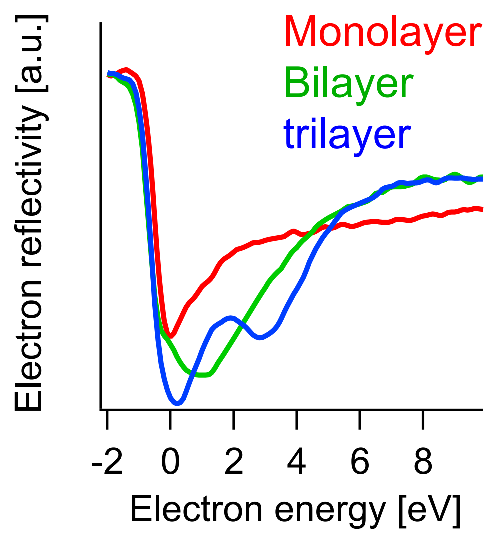


Map of LEED pattern orientations across the sample surface



Optical micrograph of the same area

  - Graphene thickness confirmed using LEEM-IV



Summary

- Long-range ordering and interacting Dirac cones are studied for twisted bilayer graphene (TBG) produced by transfer process
  - TBG displays long-range atomic ordering
  - We observed van Hove singularities
  - Periodic potential influences the band renormalization in TBG
    - For more details, please see:
      - T. Ohta, T. E. Beechem, J. Robinson, G. L. Kellogg, *Long-range atomic ordering and variable interlayer interactions in two overlapping graphene lattices with stacking misorientations*, Phys. Rev. B, 85, 075415, 2012.
      - T. Ohta, J. T. Robinson, P. J. Feibelman, A. Bostwick, E. Rotenberg, T. E. Beechem, *Evidence for interlayer coupling and moiré periodic potentials in twisted bilayer graphene*, accepted to Phys. Rev. Lett.
      - J. T. Robinson, S. W. Schmucker, C. B. Diaconescu, J. P. Long, J. C. Culbertson, T. Ohta, A. L. Friedman, T. Beechem, *Electronic Hybridization of Large-Area Stacked Graphene Films*, submitted.

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  - R. G. Copeland, A. McDonald for experiment support
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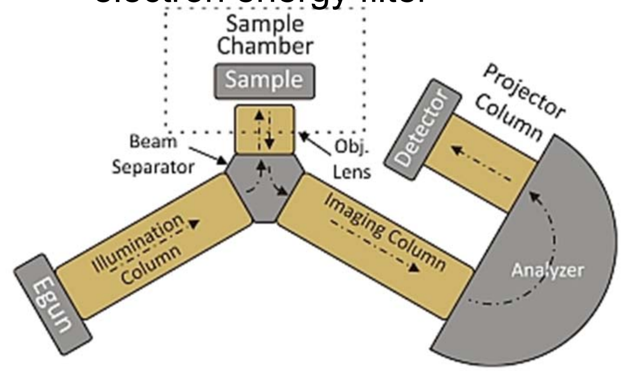
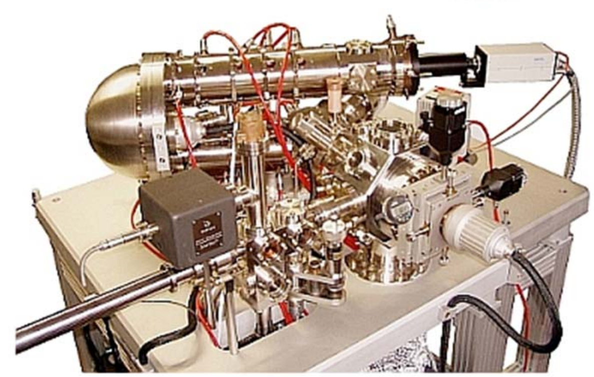
Postdoc opportunities at Sandia National Laboratories

- LEEM-PEEM research programs
  - Surface transport properties of solid ion-conductor oxides for thermo-chemical energy conversion
  - Electronic properties of bilayer graphene
  - Understanding defects in nitride-based semiconductors for lighting applications

- New research capabilities
  - Real-time morphology imaging and diffraction of surface processes
  - Electronic properties of surface using Energy-filtered LEEM: EELS and ARPES (UV-light source)

LEEM-PEEM with electron energy filter





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Thank you for your attention!