Commensuration and Incommensuration in the van der Waals Heterojunctions

Philip Kim
Physics Department
Harvard University
Rise of 2D van der Waals Systems

- **Semiconducting materials**: $WSe_2$, $MoSe_2$, $MoS_2$, …
- **Complex-metallic compounds**: $TaSe_2$, $TaS_2$, …
- **Magnetic materials**: $Fe-TaS_2$, $CrSiTe_3$, …
- **Superconducting**: $NbSe_2$, $Bi_2Sr_2CaCu_2O_{8-x}$, $ZrNCl$, …
Graphen/hBN Moire Pattern

Moire wavelength:

\[ \lambda = \frac{(1 + \delta)a}{\sqrt{2(1 + \delta)(1 - \cos(\phi)) + \delta^2}} \]

Not commensurated except for special angles
**Moire pattern in Graphene on hBN: Scanning Probe Microscopy**

Graphene on BN exhibits clear Moiré pattern


Minigap formation near the Dirac point due to Moire superlattice
Harper’s Equation: Competition of Two Length Scales

The General Motion of Conduction Electrons in a Uniform Magnetic Field, with Application to the Diamagnetism of Metals

BY P. G. HARPER
Department of Mathematical Physics, University of Birmingham

Communicated by R. E. Peierls; MS. received 19th January 1955 and in amended form 27th April 1955

Tight binding on 2D Square lattice with magnetic field

\[ \tilde{H} = \left( \frac{\tilde{p} - eA/c}{2m} \right)^2 + U(r) \]

Harper’s Equation

\[ 2\psi_i \cos(2\pi lb - \kappa) + \psi_{i+1} + \psi_{i-1} = E\psi_i \]

For \( b \ll \mu^*H \), the broadening factor may be written approximately \( \exp[-(bn/\mu^*H)^2] \) and the broadening effect becomes additive to that due to collision as described by Dingle (1952 b).

The level structure in the vicinity of an energy gap near a zone boundary is all-important for the de Haas–van Alphen effect. Unfortunately, this seems very difficult to determine in detail, even for a sinusoidal potential. Apart from the regularity already mentioned there seems little one can say. It is likely, however, that if periodicity exists, the period will give rise to effective mass parameters much smaller than one would otherwise expect. This is because the level structure will consist of irregular groups, regularly repeated. Since the period is large, the oscillatory period will also be large and the effective mass correspondingly smaller.
Commensuration / Incommensuration of Two Length Scales

Spirograph

\[ \frac{a}{l_B} = \frac{p}{q} \]
Hofstadter's Butterfly

Energy levels and wave functions of Bloch electrons in rational and irrational magnetic fields*

Douglas R. Hofstadter
Physics Department, University of Oregon, Eugene, Oregon 97403
(Received 9 February 1976; revised 15 May 1976)

Harper’s Equation

\[ 2\psi_l \cos \left( 2\pi lb - \kappa \right) + \psi_{l+1} + \psi_{l-1} = E\psi_l \]

When \( b = p/q \), where \( p, q \) are coprimes, each LL splits into \( q \) sub-bands that are \( p \)-fold degenerate

Energy bands develop fractal structure when magnetic length is of order the periodic unit cell

Diophantine equation for gaps

\[ \left( \frac{n}{n_0} \right) = t\left( \frac{\phi}{\phi_0} \right) + s \]

\( t, s \in \mathbb{Z} \)

Wannier (1978)

Quantum Hall Conductance

\[ \sigma_{xy}^Q = e^2c \frac{\partial n(E)}{\partial B} \bigg|_{E=E_F} = \frac{e^2}{h} \]

TKNN (1982); Stredar (1982)

Quantum Hall Effect with Two Integer Numbers

\( \frac{n}{n_0} \): density per unit cell; \( \phi \): flux per unit cell

\((t, s)\)

\[ R_{xx} \text{ (k}\Omega\text{)\;0\;15} \]

Fractal Quantum Hall Effect

\[ \text{Quantization of } \sigma_{xx} \text{ and } \sigma_{xy} \]

\( n/n_0 = t(\phi/\phi_0) + s \)

\( t, s \in \mathbb{Z} \)

Wannier (1978)

Dean et al. Nature (2103)
Charge Density Waves in 1T-TaS$_2$

Series of first order phase transition indicated by resistance changes

Ma, arXiv1507.01312
**Charge Density Waves in 1T-TaS$_2$: Icommensurate CDW**

Incommensurate CDW

\[ Q_{\text{Bragg}} \sim \frac{1}{a} \]

\[ Q_{\text{CDW}} \sim \frac{1}{\lambda_{\text{CDW}}} \]

\[ \lambda_{\text{CDW}} \neq ma\hat{x} + na\hat{y} \]

T = 375 K

Tsen et. al., PNAS (2016)
Charge Density Waves in 1T-TaS$_2$: Commensurate CDW

Commensurate CDW

$T = 100$ K

$Q_{\text{Bragg}} \approx \frac{1}{a}$

$Q_{\text{CDW}} \approx \frac{1}{\lambda_{\text{CDW}}}$

$\lambda_{\text{CDW}} = ma\hat{x} + na\hat{y}$
Charge Density Waves in 1T-TaS$_2$: Nearly Commensurate CDW

Nearly Commensurate CDW

$Q_{\text{Bragg}} \sim \frac{1}{a}$

$Q_{\text{CDW}} \sim \frac{1}{\lambda_{\text{CDW}}}$

$T = \sim 300$ K

Wu, Lieber, Science 1989

Discommensuration lines

Tsen et al., PNAS (2016)
C-IC Charge Density Waves Phase Transition in Atomically Thin 1T-TaS$_2$

Tsen et. al., PNAS (2016)
Imaging AB/AC domain in Bernal stacked bilayer graphene

Strain solitons and topological defects in bilayer graphene

Jonathan S. Alden, Adam W. Tsen, Pinshane Y. Huang, Robert Hovden, Lola Brown, Jiwoong Park, David A. Muller, and Paul L. McEuen

School of Applied and Engineering Physics, Department of Chemistry and Chemical Biology, Kavli Institute at Cornell for Nanoscale Science, and Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, NY 14853

Contributed by Paul L. McEuen, May 23, 2013 (sent for review April 28, 2013)

Image from Spiecker (FAU)
Topological valley transport at bilayer graphene domain walls

Long Lu, Zhiwen Shi, Nityan Nair, Yinchuan Lyu, Chenhao Jin, Jairo Velasco Jr, Claudia Ojeda-Aristizabal, Hans A. Bechtel, Michael C. Martin, Alex Zettl, James Analytis & Feng Wang

Propagation 1-D Transport Channel Along the Domain Boundaries
Controlled Stacking of Graphene-Graphene Layers

Controlling Twisting Angle: Tear and Stack

Integrate with SiN membrane for TEM study

Y. Cao et al., (PRL in press)
Dark Field Imaging Graphene Moire

Transmission Electron Microscopy

- Instrument and ray diagram

Dark field imaging

Graphene/graphene/hBN

Incommensurate Moire structures

Graphene/hBN

J. M. Yuk et al., Carbon 80, 755 (2014)

Graphene/graphene

Commensurate Moire structures??

Dark field imaging
Graphene on Graphene With a Small Twisting Angle

- TEM dark field image \( (g = 10\overline{1}0) \) and selected area electron diffraction pattern

Rotational displacement + Uniaxial displacement = Both Rotational and Uniaxial displacement
Twisting Engineering C/IC Domain Boundaries
Twisted Bilayer Graphene: Satellite Diffraction Peaks

TEM diffraction pattern Two graphene sheets with twist Angle: 0.41 deg on hBN substrate
Real Space Versus Fourier Space: Satellite Peaks

Selected Area Diffraction patterns zoomed up (x50) on each graphene peak
Twisting Angle Dependent Satellite Peak Intensities

First Order:

Second Order:

Third Order:

$I_{\text{satellite}} / I_{\text{Bragg}}$

Intensity ratio

Angle (degrees)
Relaxation of moiré patterns for slightly misaligned identical lattices: graphene on graphite

M M van Wijk, A Schuring, M I Katsnelson and A Fasolino
Radboud University Nijmegen, Institute for Molecules and Materials, Heyendaalseweg 135, 6525 AJ Nijmegen, The Netherlands

Figure 3. Bondlengths of relaxed configurations for samples with \( z = 3 \) Å. The supercell is shown in black. The bottom panels show the bond length along the dashed diagonal line. (a) \( \theta = 2.1^\circ \), \((n,m)=(47,1)\), \(a_m = 66.4\) Å. (b) \( \theta = 1.2^\circ \), \((n,m)=(82,1)\), \(a_m = 115.3\) Å. (c) \( \theta = 0.46^\circ \), \((n,m)=(216,1)\), \(a_m = 302.6\) Å.

Figure 6. \( \vec{u} \) as in equation 8 for (a) \( \theta = 5.7^\circ \) and (b) \( \theta = 0.46^\circ \).

- Atomic scale relaxation
- Buckling out of plane
- Electronic properties
Atomic Scale Relaxation Modeling: Commensurate Lattice

vdW interactions modeled using discrete–continuum effective potentials for Lennard–Jones (LJ) and Kolmogorov–Crespi (KC) for full atomistic resolution.

Simulated Diffractions
Qualitative agreement with experiments!

Kuan Zhang and Ellad B. Tadmor
Mathematical Modeling: Inversion Problem

- How we can model general incommensurate twisted lattices?
- Can we recover the microscopic relaxation pattern from satellite diffraction?

- **Atoms of type A of layer 2** are modulated following the rule
  \[ \mathbf{r}_{2,A} = \mathbf{R}_{2,A} + \phi_{2,A}(\mathbf{R}_{2,A}) \]

  - \( \mathbf{R}_{2,A} \) unmodulated positions
    (empty red circles on the figure)
  - \( \mathbf{r}_{2,A} \) modulated positions
    (full red circles on the figure)
  - \( \phi_{2,A} \) modulation field with the periodicity of layer 1

[Experimental diffraction spots]

- **AB/BA** Symmetric
- **AB/BA** Asymmetric
Transport Along Domain Boundaries: Topological Mode

L. Ju et al., Nature 520, 650 (2015)

Vertical electric field

Bulk energy band

Network resistivity
\sim \frac{h}{4e^2} = 6.4 \, k\Omega
Transport at the Neutrality of Twisted Bilayers

**Bernal stacking**
- $V_{\text{top}} = +5 \text{ V}$
- $V_{\text{top}} = -5 \text{ V}$

**Decoupled stacking**
- $V_{\text{top}} = +10 \text{ V}$
- $V_{\text{top}} = -10 \text{ V}$

**IC/C domains**
- $V_{\text{top}} = +5 \text{ V}$
- $V_{\text{top}} = -5 \text{ V}$
Twisting Engineering C/IC Domain Boundaries in MoS$_2$


✓ 5 different possible configurations of bilayer stacking of MoS$_2$.

Bulk Stacking Method

Controlling Twisting Angle: Tear and Stack

Y. Cao et al., (PRL 2016)
MoS$_2$ on MoS$_2$ With a Small Twisting Angle

AB stack with partial dislocations
MoS$_2$ on MoS$_2$ With 180° Twisting Angle
Summary and Outlook

• Commensuration and incommensuration between electric potential and magnetic length creates fractalized quantum Hall effect

• C/IC phase transition in CDW lattice in TaS2 can be tuned by dimensionality changes in the samples.

• In graphene bilayer, tunable domain structures of C/IC boundary can be controlled with twist angle control.

• Observation of different types of domain structures in bilayer MoS2 formed as a result of commensurate transition.

Going Forward:
Symmetry elements in the materials can be engineered by twist angle control.

Can we generate topologically non-trivial electronic structures from twisted vdW stacks?
Acknowledgements

Hofstadter Butterfly


Charge Density Waves in TaS$_2$


Twisted vdW Layers

H. Yoo, R. Engelke, M. Kim, G. –C, Yi K. Zhang, E. Tadmor P. Cazeaux, M. Luskin R. Hovden

Funding

Harvard University Center for Nanoscale Systems

NATIONAL HIGH MAGNETIC FIELD LABORATORY

NSF

Gordon and Betty Moore Foundation