Bloch, Landau, and Dirac: Hofstadter’s Butterfly in Graphene

Philip Kim, Department of Physics, Columbia University
Fractional & Integer Quantum Hall effect in graphene: electron correlation

Dean et al, Nature Phys (2011)

Heteroepitaxy of Layered Materials: Andreev reflection between NbSe$_2$ and graphene

Efetov et al (2013)
Graphene Materials and Applications

- Large-Scale CVD Graphene + Graphene Nanoplatelet Composites
- Flexible/Transparent Electrodes/Touch Panels
- Semi-conductors
- Energy Electrodes
- Composites
- Printable Inks
- Gas Barriers
- Heat Dissipation
- Gas barriers for Displays, Solar Cells
- LED Lights, BLU
- ECU, PC ...
- Cars, Aerospace Applications

Images: Royal Swedish Academy

Courtesy: B. H. Hong
Will graphene appear in market soon?
Bloch, Landau, and Dirac: Hofstadter’s Butterfly in Graphene

Prof. Jim Hone
Prof. Ken Shepard

Theory: P. Moon & M. Koshino (Tohoku)

hBN samples: T. Taniguchi & K. Watanabe (NIMS)

Dr. Cory Dean  Lei Wang  Patrick Maher  Fereshte Ghahari  Carlos Forsythe
Bloch Waves: Periodic Structure & Band Filling

Über die Quantenmechanik der Elektronen in Kristallgittern.

Von Felix Bloch in Leipzig.

Mit 2 Abbildungen. (Eingegangen am 10. August 1928.)

Zeitschrift für Physik, 52, 555 (1929)

Periodic Lattice

\[ \tilde{H} = \frac{\hat{p}^2}{2m} + U(x), \quad U(x) = U(x + a) \]

\[ A_0 : \text{unit cell volume} \]

\[ a \]

Block Waves:

\[ \psi_{n,k}(x) = e^{i k x} u_{n,k}(x), \quad u_{n,k}(x + a) = u_{n,k}(x) \]

Band Filling factor

\[ s = -A_0^2 \frac{\partial n(\varepsilon_F)}{\partial A_0} \]

MacDonald (1983)
Free electron under magnetic field

\[ \hat{H} = \frac{(\hat{p} - eA/c)^2}{2m} \]

Energy and orbit are quantized:

\[ \varepsilon_n = \hbar w_c(n + 1/2), \quad w_c = eB/mc \]

Each Landau orbit contains magnetic flux quanta

\[ \phi_0 = \frac{\hbar c}{e} \]

\[ \ell_B = \sqrt{\hbar/eB} \]

Massively degenerated energy level

2-dimensional electron systems

Landau level filling fraction:

\[ \nu = 2\pi \ell_B^2 n(\varepsilon_F) \]
Tight binding on 2D Square lattice with magnetic field

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

Harper’s Equation

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

Two competing length scales:

- \(a\): lattice periodicity
- \(l_B\): magnetic periodicity

For \(b \ll \mu^*H\), the broadening factor may be written approximately as \(\exp[-(b/c\gamma\mu^*H)^2]\) 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)

Harper’s Equation

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

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
Energy Gaps in the Butterfly: Wannier Diagram

Hofstadter’s Energy Spectrum

\[ \frac{n}{n_0} : \text{ # of state per unit cell} \]
\[ \phi / \phi_0 : \text{ magnetic flux in unit cell} \]
\[ n : \text{ electron density} \]


Tracing Gaps in \( \phi \) and \( n \)

Diophantine equation for gaps

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

\( t, s \in \mathbb{Z} \)
What is the physical meaning of the integers $s$ and $t$?

Quantised Hall effect in a two-dimensional periodic potential

P Středa
Institute of Physics, Czechoslovak Academy of Sciences, 180 40 Praha 6, N, Czechoslovakia

Received 6 October 1982

Quantum Hall Conductance

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

Band Filling factor

$$s = -A_0^2 \frac{\partial n(\varepsilon_F)}{\partial A_0}$$

Physical Review Letters

**Quantized Hall Conductance in a Two-Dimensional Periodic Potential**

D. J. Thouless, M. Kohmoto, M. P. Nightingale, and M. den Nijs

Department of Physics, University of Washington, Seattle, Washington 98195

(Received 90 April 1982)

$$\sigma_H = \frac{i e^2}{4\pi h} \sum k \int d^2 k \int d^2 r \left( \frac{\partial u^*}{\partial k_1} \frac{\partial u}{\partial k_2} - \frac{\partial u^*}{\partial k_2} \frac{\partial u}{\partial k_1} \right)$$

$$= \frac{i e^2}{4\pi h} \sum k \int d^2 r \left( u^* \frac{\partial u}{\partial k_j} - \frac{\partial u^*}{\partial k_j} u \right),$$

X. POSSIBLE EXPERIMENTAL TEST

Finally, I would like to comment on the possibility of looking for the features predicted by this model experimentally. At first glance, the idea seems totally out of the range of possibility, since a value of $\alpha = 1$ in a crystal with the rather generous lattice spacing of $a = 2 \ \text{Å}$ demands a magnetic field of roughly $10^6 \text{ G}$. It has been suggested, however (by Lowndes among others), that one could manufacture a synthetic two-dimensional lattice of considerably greater spacing than that which characterizes real crystals. The technique involves applying an electric field across a field-effect transistor (without leads). The effect of

Hofstadter (1976)


- Unit cell limited to ~100 nm
- Limited field and density range accessible, weak perturbation
- Do not observe ‘fully quantized’ mingaps in fractal spectrum
Electrons in Graphene: Effective Dirac Fermions

Graphene, ultimate 2-d conducting system

Effective Dirac Equations

\[ H_{\text{eff}} = \pm \hbar v_F \begin{pmatrix} 0 & k_x - ik_y \\ k_x + ik_y & 0 \end{pmatrix} = \pm \hbar v_F \vec{\sigma} \cdot \vec{k}_\perp \]

DiVincenzo and Mele, PRB (1984); Semenov, PRL (1984)
Graphene: Under Magnetic Fields

Quantum Hall Effect

DOS

Energy

$E_N = \pm \sqrt{2e\hbar v_F^2 |N| B}$

Quantization Condition

$R_{xy}^{-1} = \frac{4e^2}{\hbar} (N + \frac{1}{2})$

$\nu = \pm 2, \pm 6, \pm 10, \ldots$

Novoselov et al (2005)
Zhang et al (2005)
Bilayer Graphene

Bernal stacked bilayer graphene


Low energy approximation in 1st BZ

Energy (eV)

\( E_n^\pm = \pm \hbar \omega_c \sqrt{|n|(|n|-1)} \)

\[ R_{xy}^{-1} = \frac{4Ne^2}{h} \]

\( \nu = \pm 4, \pm 8, \pm 12, \ldots \)

Novoselov et al., Nature Physics (2006)
Hofstadter’s Butterfly in Twisted Graphene

-Moon and Koshino, PRB (2012); See also Bistrizer and MacDonald (2011)

Moiré wavelength is determined by angle between the graphene layers

\[
\theta = 9.43°, \quad \theta = 3.89°, \quad \theta = 2.65°, \quad \theta = 1.47°
\]

\[
\lambda = \frac{(1 + \delta)a}{\sqrt{2(1 + \delta)(1 - \cos\phi)}}
\]
Moire Pattern in Twisted Graphene Layers

Observation of Van Hove singularities in twisted graphene layers


STM observation of Moire pattern

Twisting angle
~1.16,
\( a = 7.7 \, \text{nm} \)
Hexa Boron Nitride: Polymorphic Graphene

Comparison of h-BN and SiO\textsubscript{2}

<table>
<thead>
<tr>
<th></th>
<th>Band Gap</th>
<th>Dielectric Constant</th>
<th>Optical Phonon Energy</th>
<th>Structure</th>
</tr>
</thead>
<tbody>
<tr>
<td>BN</td>
<td>5.5 eV</td>
<td>~4</td>
<td>&gt;150 meV</td>
<td>Layered crystal</td>
</tr>
<tr>
<td>SiO\textsubscript{2}</td>
<td>8.9 eV</td>
<td>3.9</td>
<td>59 meV</td>
<td>Amorphous</td>
</tr>
</tbody>
</table>

\(a_0 = 0.246 \text{ nm}\)

\(a_0 = 0.250 \text{ nm}\)
Stacking graphene on hBN


- Co-lamination techniques
- Submicron size precision
- Atomically smooth interface

Polymer coating/cleaving/peeling

Micro-manipulated Deposition

Remove polymer Anealing

Mobility $> 100,000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$
Moire pattern in Graphene on hBN: a new route to Hofstadter’s butterfly?

Graphene on BN exhibits clear Moiré pattern

Xue et al, Nature Mater (2011);
Decker et al Nano Lett (2011)

Minigap formation near the Dirac point due to Moire superlattice
Bilayer graphene on BN substrates shows strong signature of satellite peaks...*some times*... (~ 30%)
Abnormal Landau Fan Diagram in Bilayer on hBN

Special Samples with Large Moire Unit Cell

\[ R_{xx} = \frac{V_{xx}}{I} \]

\[ R_{xy} = \frac{V_{xy}}{I} \]

Low Magnetic field regime

\[ |R_{xy}| \ (k\Omega) \]

1.7 K

\[ B \ (\text{Tesla}) \]

\[ V_G \ (\text{Volts}) \]

\[ I \]

\[ V_{ss} \]

1.7 K
How to “Read” Normal Landau Fan Diagram?

Landau Fan Diagram for “typical” graphene

\[ \nu = 2\pi \ell_B^2 n \Rightarrow B = \frac{\phi_0}{\nu} n \]

\[ (R_{xy}^Q)^{-1} = \sigma_{xy}^Q = \frac{e^2}{h} \nu \]
Abnormal Quantum Hall Effect

Quantum Hall-like Transport

\[ R_{xx} = \frac{V_{xx}}{I} \]

Landau level filling factor

\[ \nu = \frac{\phi_0}{B n} \]

Quantum Hall conductance

\[ R_{xy} = \frac{e^2}{h} \]


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

V_G (Volts)

B (Tesla)

\[ R_{xx} (k\Omega) \]

\[ B=18T \]

\[ \nu = t \in \mathbb{Z} \]
Size of the Moire Supper Lattice in Graphene

Yankowitz et al., (2012)

\[ A_0^{-1} = n_0 = C_g V_g^{sb} / 4 \]
\[ A_0 = \frac{\sqrt{3}a^2}{2} \]
\[ \phi = B A_0 \]
\[ a = 14.3 \text{ nm} \]

Zero Field Transport

- \( T = 300 \text{ mK} \)
- \( B = 0 \)

\( V_g^{sb} \approx 30 \text{ V} \)

\( C_g = 118 \text{ aF/\mu m}^2 \)

Confirmed by UHV AFM

Ishigami group (UCF)

\( a = 15 \pm 1 \text{ nm} \)
Normalized Fan Diagrams

\[ n/n_0 = 4V_g/V_g^{sb} \]
\[ \phi/\phi_0 = B/B_0, \quad B_0 = \phi_0/A_0 \]
Normalized Fan Diagrams

\[
n/n_0 = 4V_g/V_g^{sb} \quad \phi/\phi_0 = B/B_0, \quad B_0 = \phi_0/A_0
\]

Wannier diagram:
Tracing gaps in Hofstadter’s Butterfly
Landau Fan in Low Magnetic Field Regime

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

\[ R_{xy}^{-1} = \frac{e^2}{h} t \]
Landau Fan in Low Magnetic Field Regime

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

\[ R_{xy}^{-1} = \frac{e^2}{h} t \]
Landau Fan in Low Magnetic Field Regime

\[ \left( \frac{1}{6} \right) \frac{h}{e^2} \]

\[ \left( \frac{1}{8} \right) \frac{h}{e^2} \]

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

\[ R_{xy}^{-1} = \frac{e^2}{\hbar} t \]
Hall Conductance in Fractal Band

Strong suppression of $R_{xy}$ while $R_{xx}$ is finite!
Recursive QHE near the Fractal Bands

Higher quality sample with lower disorder

At the Fractal Bands
Sign reversal of $\sigma_{xy}$
Large enhancement of $\sigma_{xx}$

Hall conductivity across Fractal Band

Recursive QHE!
Summary

- Graphene on hBN with high quality interface created Moire pattern with supper lattice modulation
- Quantum Hall conductance are determined by two TKNN integers.
- Anomalous Hall conductance at the fractal bands

Open Questions:
- Elementary excitation of the fractal gaps?
- Role of interactions, Hofstadter Butterfly in FQHE?
Fractal Gaps: Energy Scales

Fractal Quantum Hall Effect

\[(t, s)\]

\[R_{xx} (\Omega)\]

Temperature Dependence

Fractal Gap Size

83 K
Similar physics is observed in single layer graphene on hBN.
Acknowledgement

Funding: NSF

hBN samples: T. Taniguchi & K. Watanabe (NIMS)

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