Physics of Few-Layer Graphene

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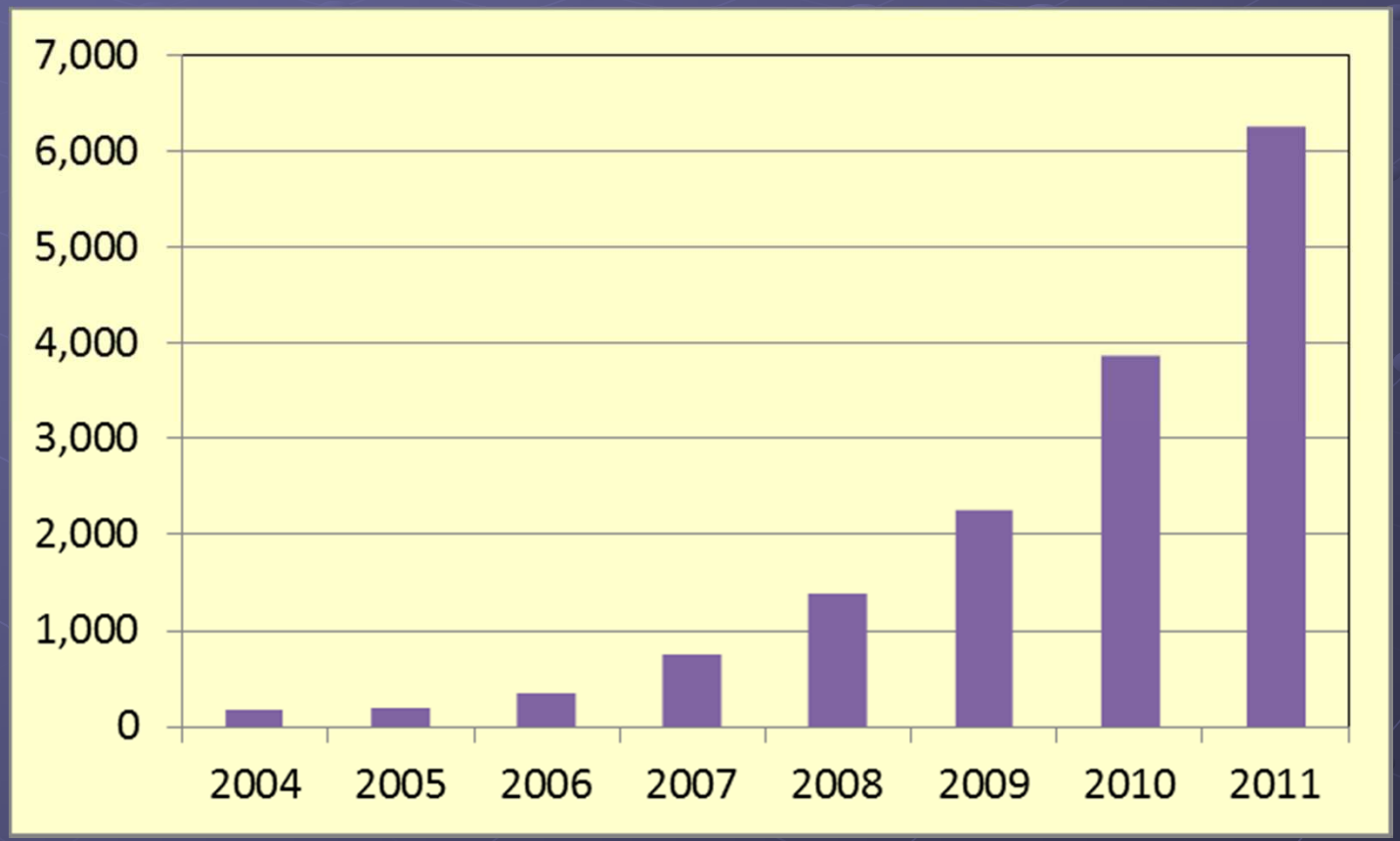
The Graphene Phenomenon (石墨烯現象)

15,000 SCI papers on graphene published since 2004



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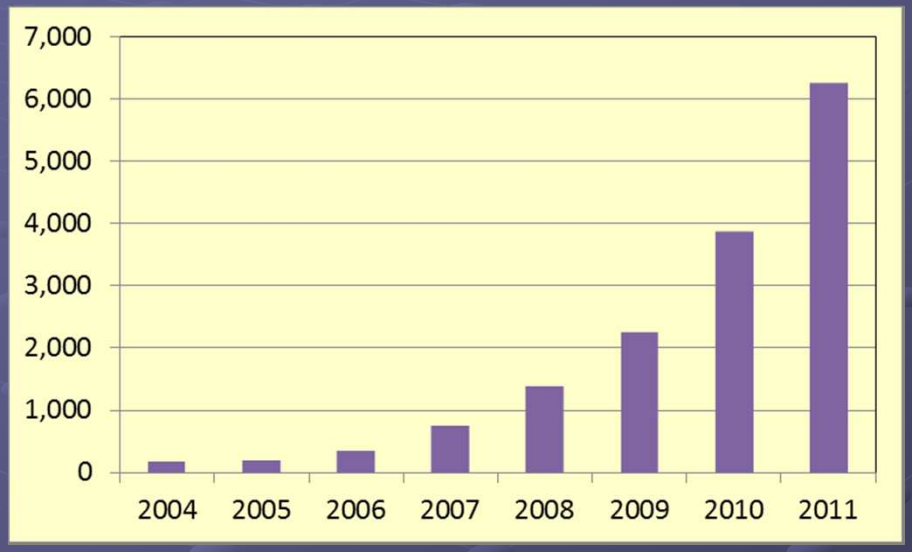
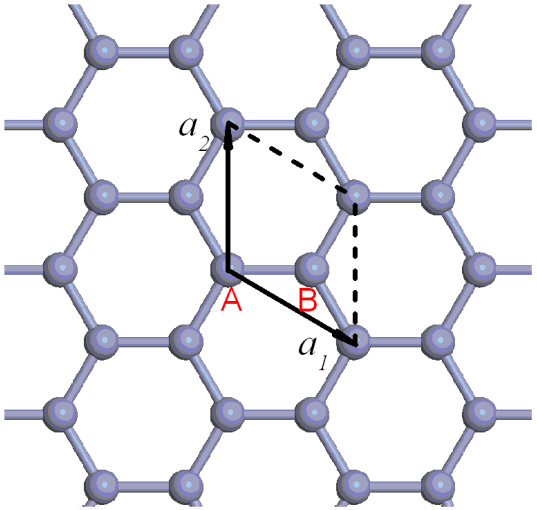


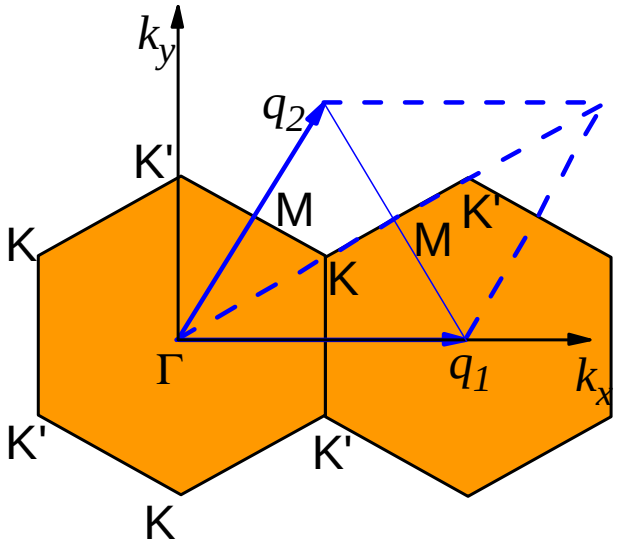
Graphene may turn out to be the Lady Gaga of materials:
it seems to have come from nowhere, everybody's talking about it, and before long it's going to be absolutely everywhere.

Gary Marshall, World of Tech News

Graphene

(Wallace 1947)



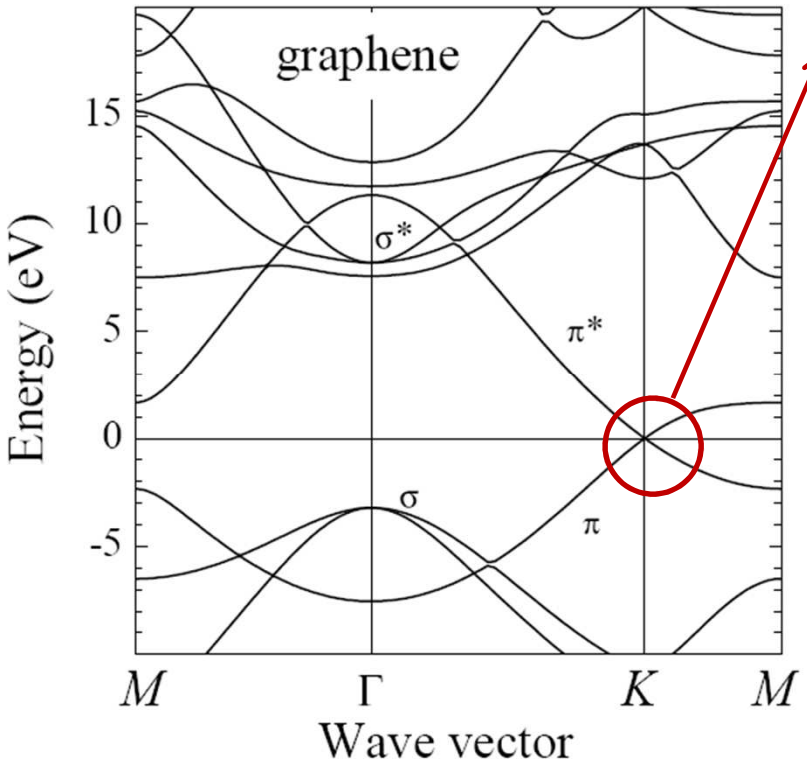


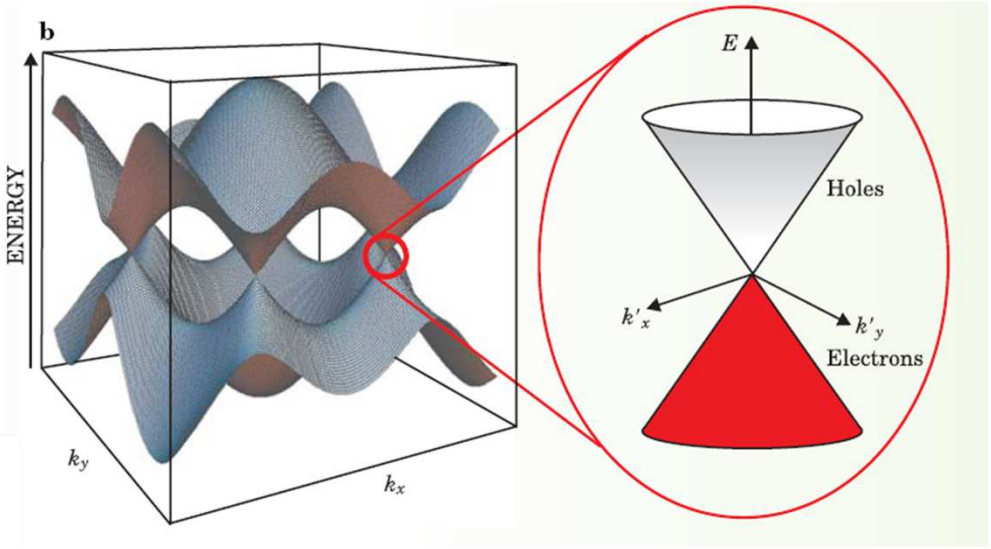
Linear Dispersion

$$E = \hbar v_F k$$

$V_F \approx 10^6$ m/s $\approx c$ /300

$$E = \frac{\hbar^2 k^2}{2m}$$ X





(Mark Wilson, Physics Today Jan 2006, p. 21)

Formal similarity between the excitations in graphene and 2d Dirac-Weyl fermions (Semenoff 1984; DiVincenzo and Mele 1984)

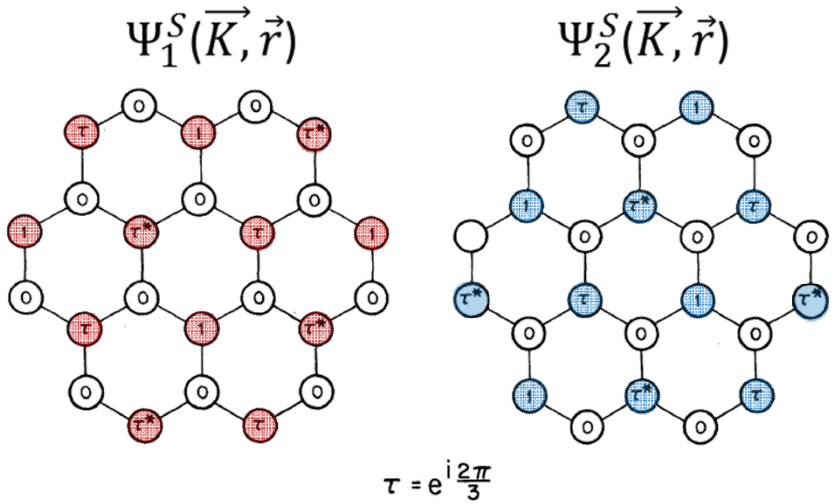


FIG. 3. Two degenerate eigenstates which provide the basis set for the $\vec{\kappa}\cdot\vec{p}$ trial wave function. The coefficients for a one-orbital description of these eigenstates are shown. They are the two Bloch functions at $E=0$ at the K point (see Fig. 2).

$$\psi(\vec{k},\vec{r}) = f_1(\vec{\kappa})e^{i\vec{\kappa}\cdot\vec{r}}\psi_1^S(K,\vec{r}) + f_2(\vec{\kappa})e^{i\vec{\kappa}\cdot\vec{r}}\psi_2^S(K,\vec{r}) \ . \qquad (1)$$

Inserting ψ into the Schrödinger equation, keeping terms of order $\vec{\kappa}$, and taking $E_F=0$ gives the secular equation,

$$\frac{\hbar}{m}\vec{\kappa}\cdot\begin{bmatrix}\vec{p}_{11} & \vec{p}_{12}\\ \vec{p}_{21} & \vec{p}_{22}\end{bmatrix}\begin{bmatrix}f_1(\vec{\kappa})\\ f_2(\vec{\kappa})\end{bmatrix} = E(\vec{\kappa})\begin{bmatrix}f_1(\vec{\kappa})\\ f_2(\vec{\kappa})\end{bmatrix} . \qquad (2)$$

Here $\vec{p}_{ij} \equiv \int \psi_1^{S*}(K,\vec{r})\vec{p}\psi_j^S(K,\vec{r})d\vec{r}$. It can be shown from group-theoretic arguments (and we can verify directly within the one-orbital tight-binding model) that the momentum matrix may be written in the form

$$\bar{p}\begin{bmatrix}0 & \hat{x}-i\hat{y}\\ \hat{x}+i\hat{y} & 0\end{bmatrix} . \qquad (3)$$

$$\sigma_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} \quad \sigma_y = \begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}$$

For $\vec{k} = \vec{K} + \vec{\kappa}$ close to K, expand the wave function using Ψ_1^S and Ψ_2^S.

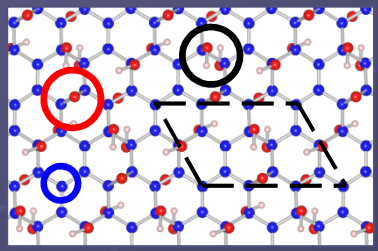
$$H = \hbar v_F \hat{\sigma}\cdot\vec{\kappa}$$
$$E = \pm\hbar v_F\left|\vec{\kappa}\right|$$

Dirac-Weyl equation for massless relativistic fermions (e.g., neutrinos);
Here pseudospin = $\frac{1}{2}$ (two sublattices)

Electronic and Transport Properties of Graphene Systems

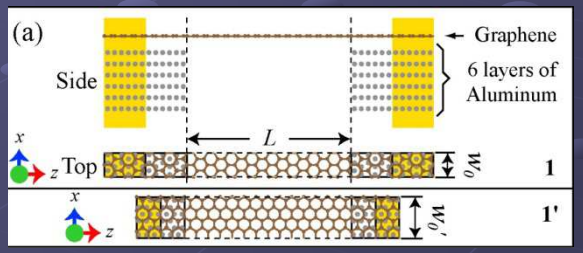
- Structural and electronic properties of oxidized graphene

  PRL 103, 086802 (2009)



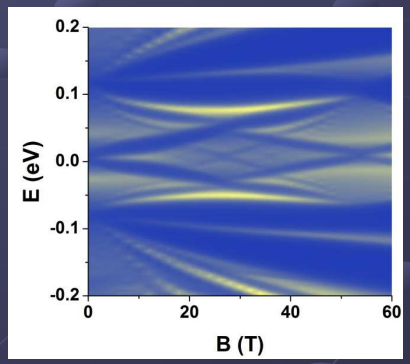
- Effects of metallic contacts on electron transport through graphene

  PRL 104, 076807 (2010)
  Nano Letters (in press)



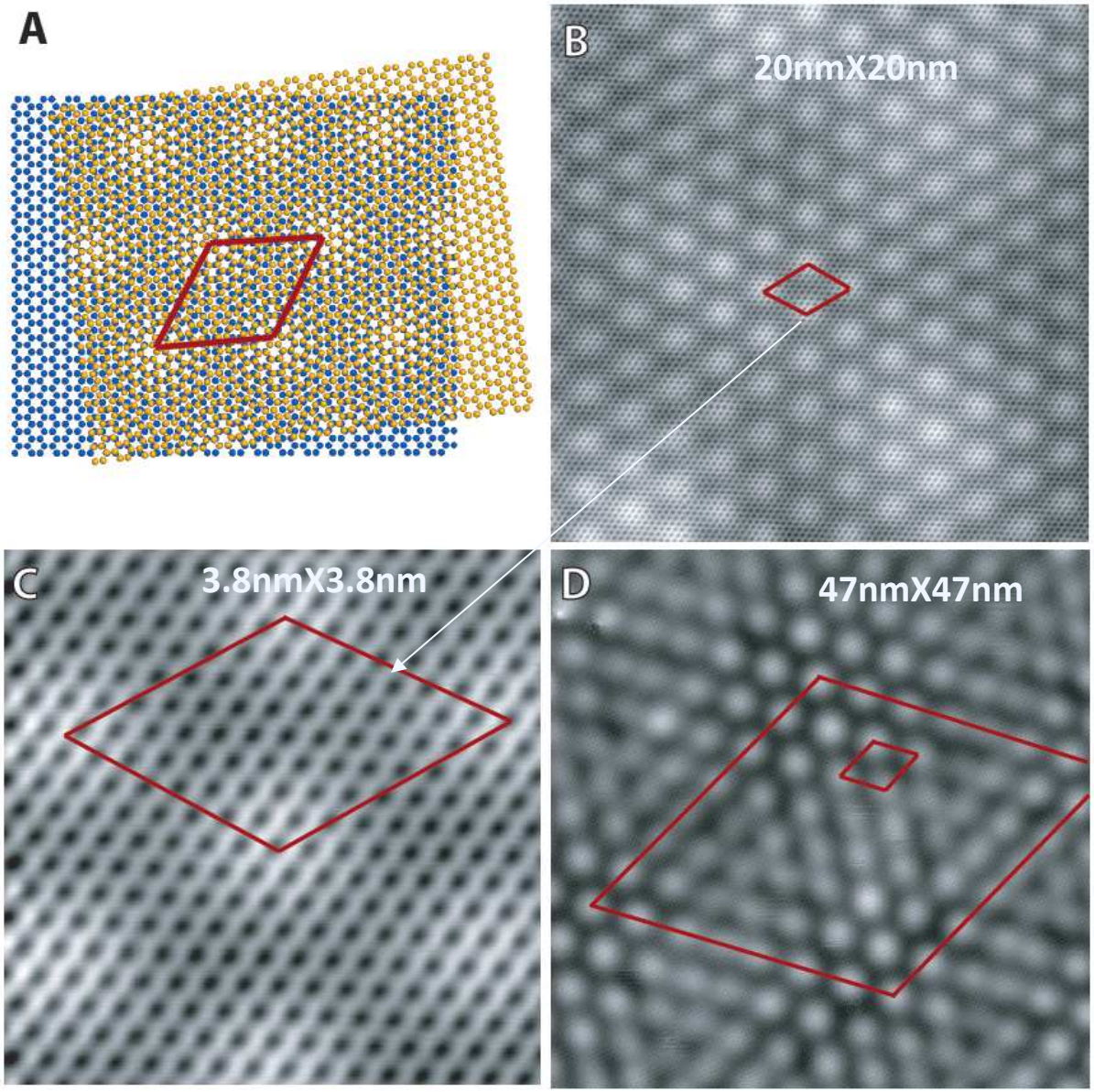
- Landau levels in twisted bilayer graphene

  Nano Letters (in press)



- Wave-packet dynamics in twisted bilayer graphene
- B-N co-doping in graphene

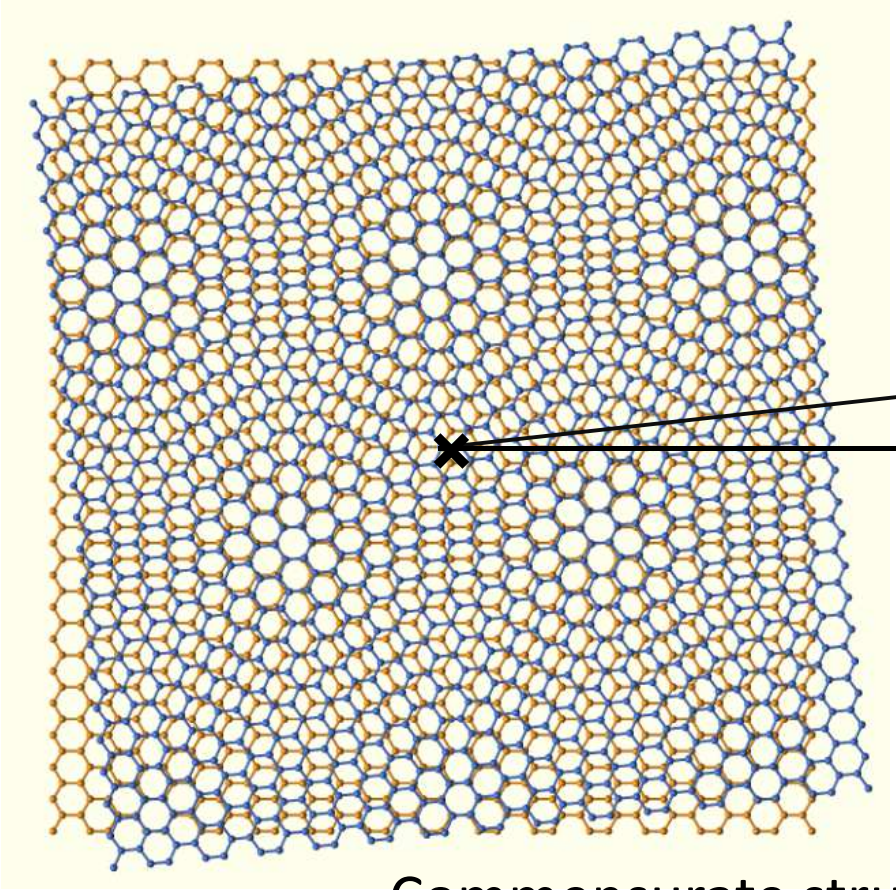
Multilayer Epitaxial Graphene



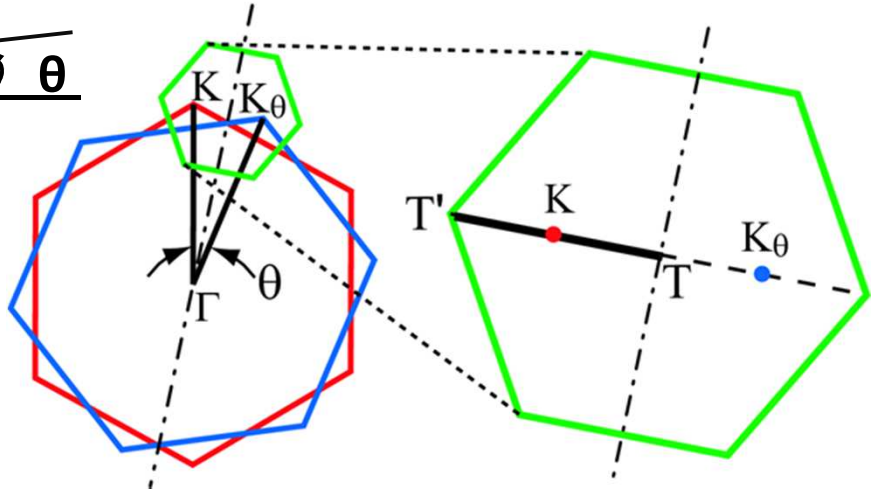
L. Miller et al.
Science 324, 924 (2009)

J. Hass, et al.
Phys. Rev. Lett **100** 125504 (2008)

Twisted Bilayer Graphene



Starting from AB-stacked bilayer graphene, the bottom layer is fixed and the top layer is rotated.

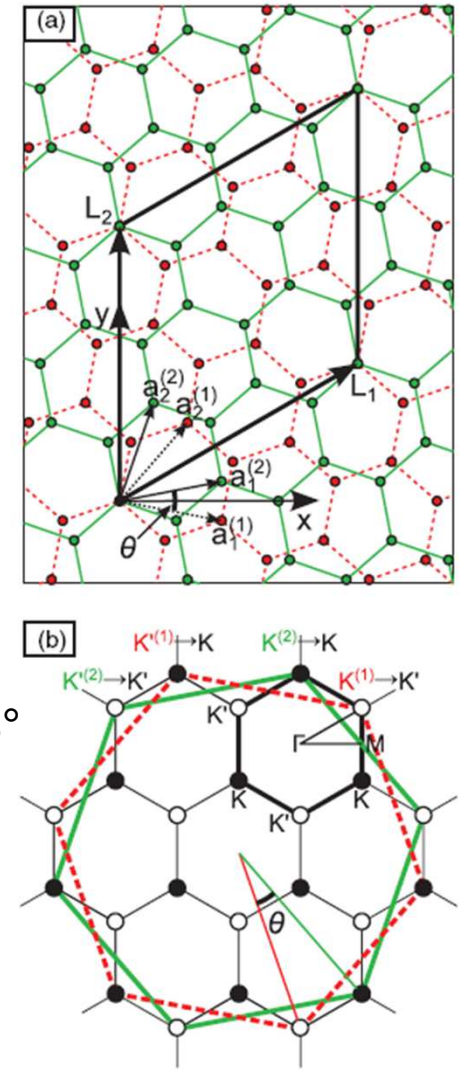


Commensurate structures labeled by (*m,n*):

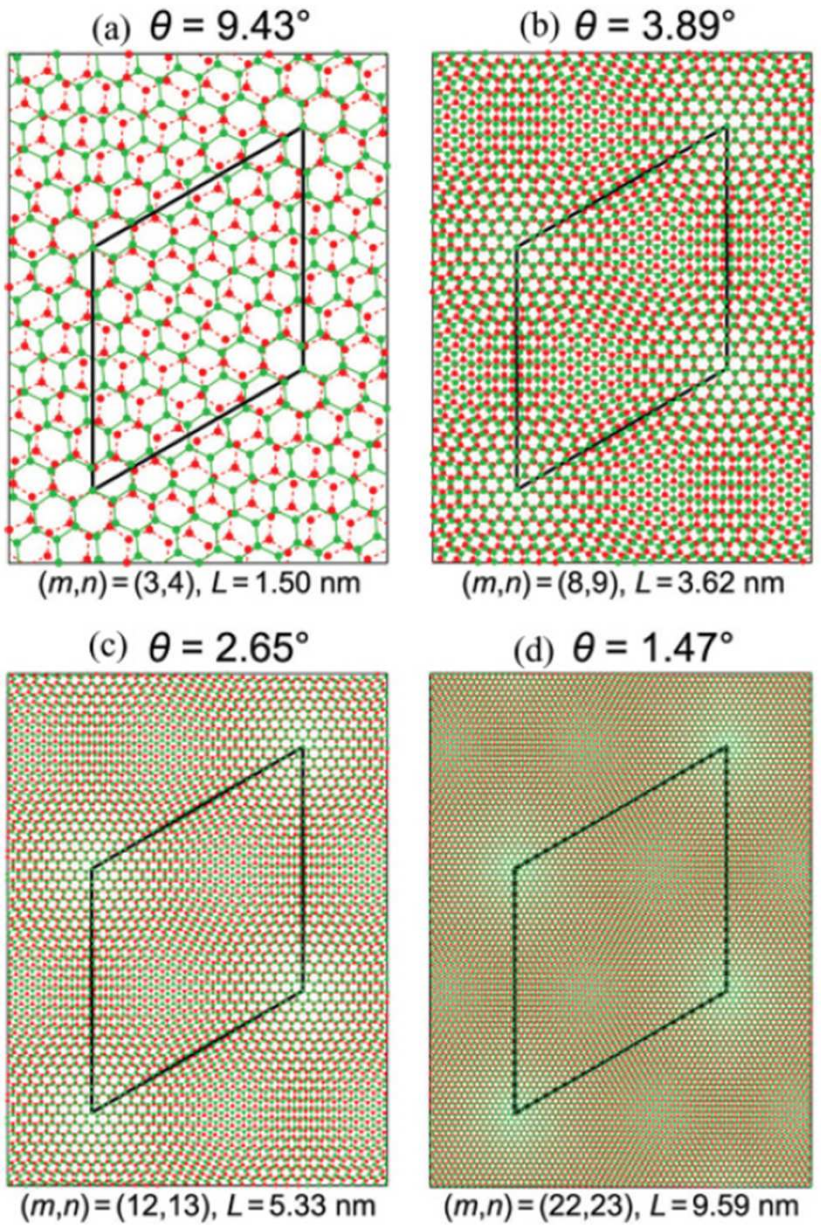
$$\mathbf{t_1} = n\mathbf{a_1} + m\mathbf{a_2}\ ;\ \ t_2 = -\,m\mathbf{a_1} + (n+m)\,\mathbf{a_2}$$

$$\cos(\theta) = \frac{n^2+4nm+m^2}{2(n^2+nm+m^2)}$$

Twisted Bilayer Graphene



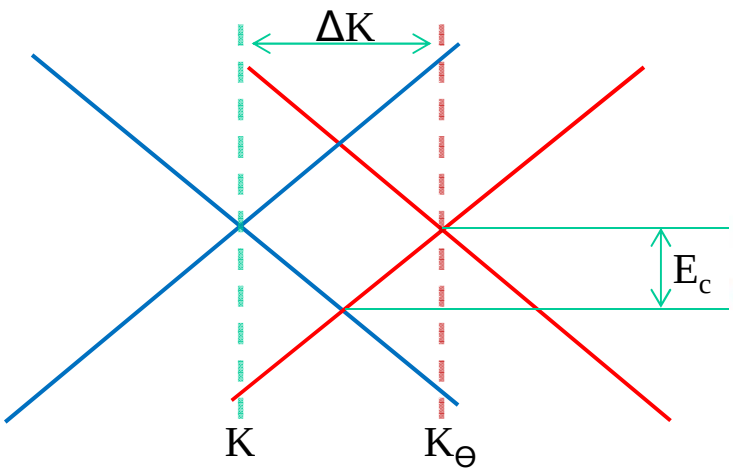
θ = 21.8°



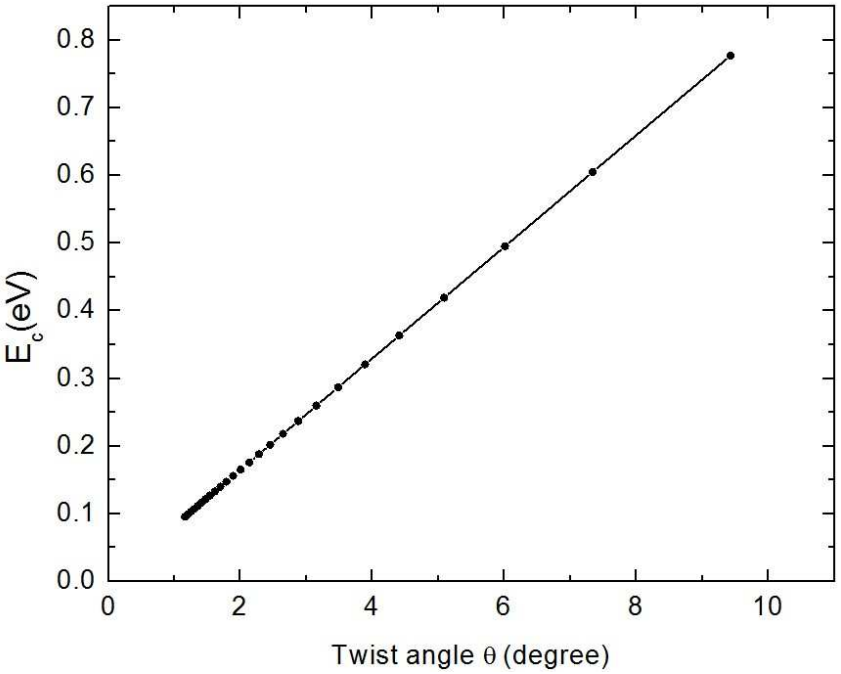
Moon et al. PRB 85, 195458 (2012)

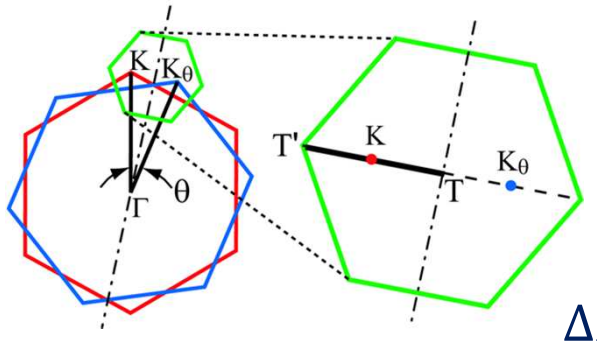
Electronic Structure of Twisted Bilayer Graphene

$$E_c = \hbar v_f \times \Delta K/2$$



WITHOUT interlayer coupling

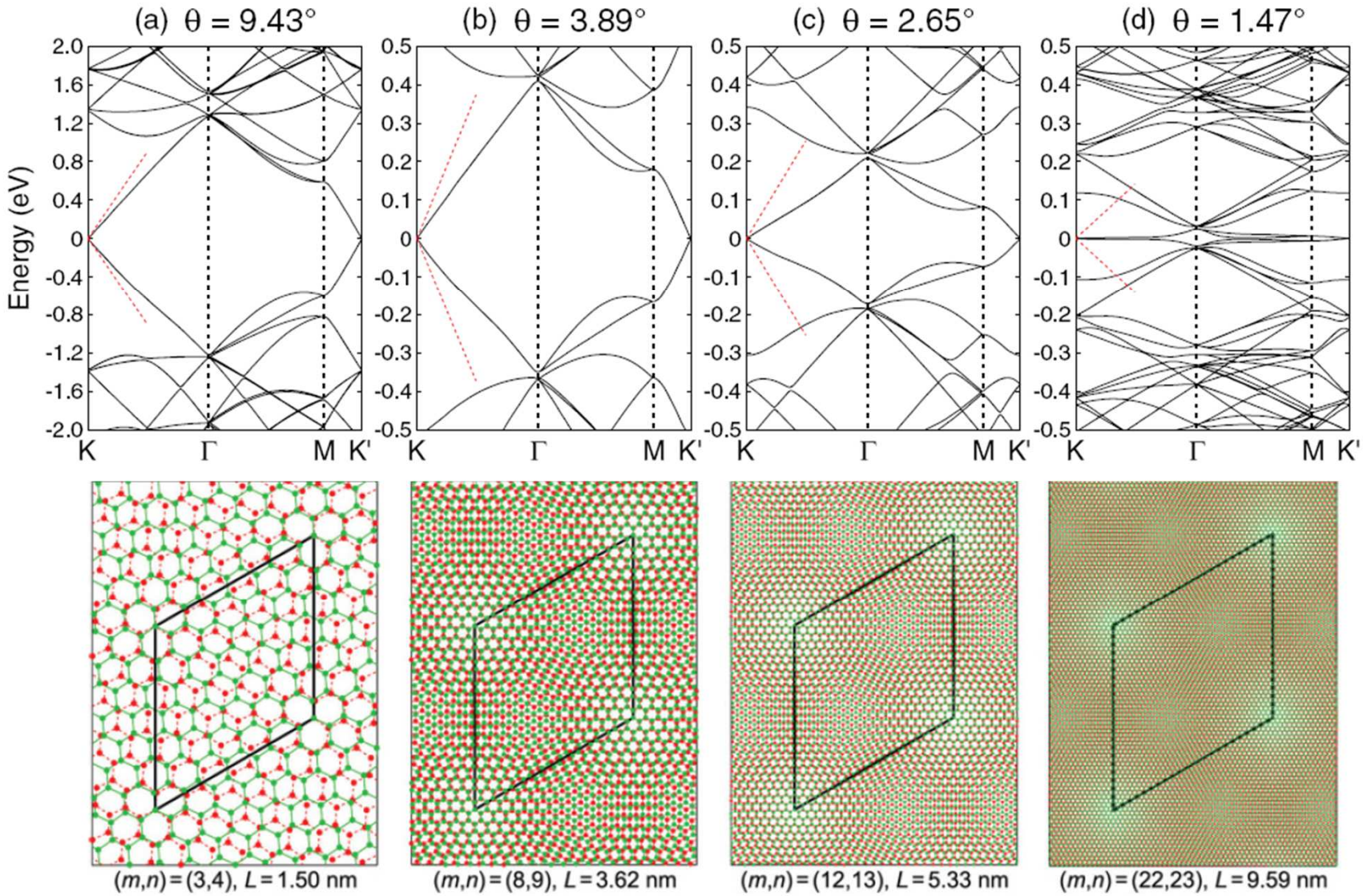




Estimated anisotropic energy region for different twist angle

$$\Delta K = 2\,|K|\,\sin(\theta/2)$$

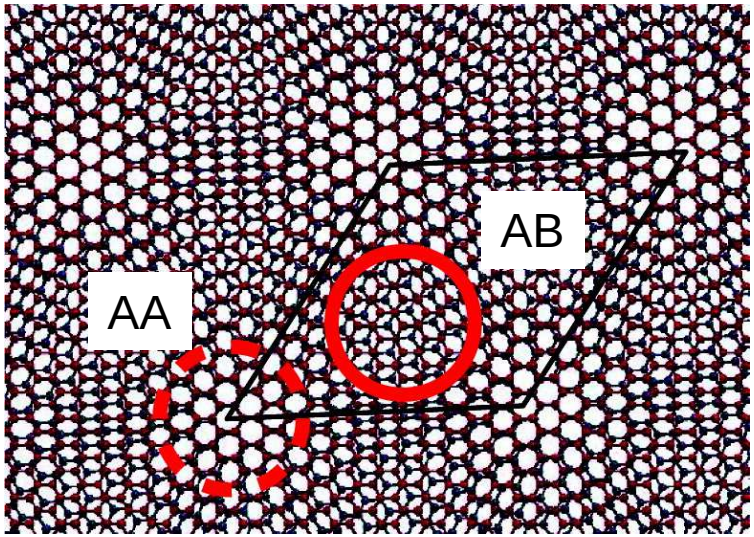
Electronic Structure of Twisted Bilayer Graphene



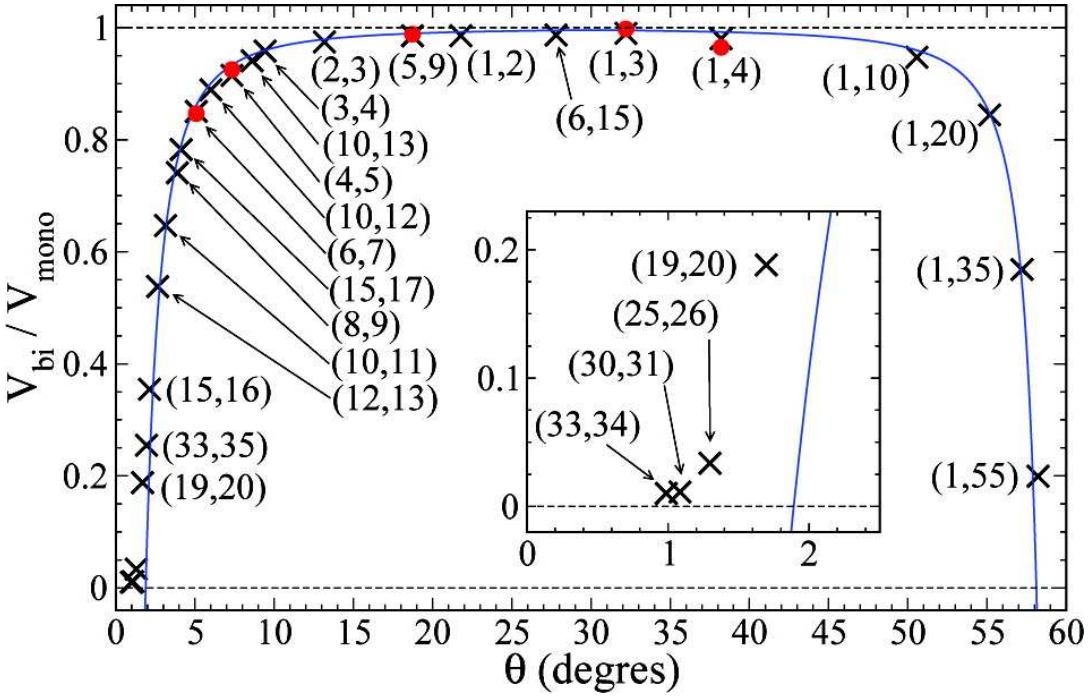
Moon et al. PRB 85, 195458 (2012)

Localization of Dirac Electrons in Rotated Graphene Bilayers

Trambly de Laissardiere et al. Nano Lett. 2010

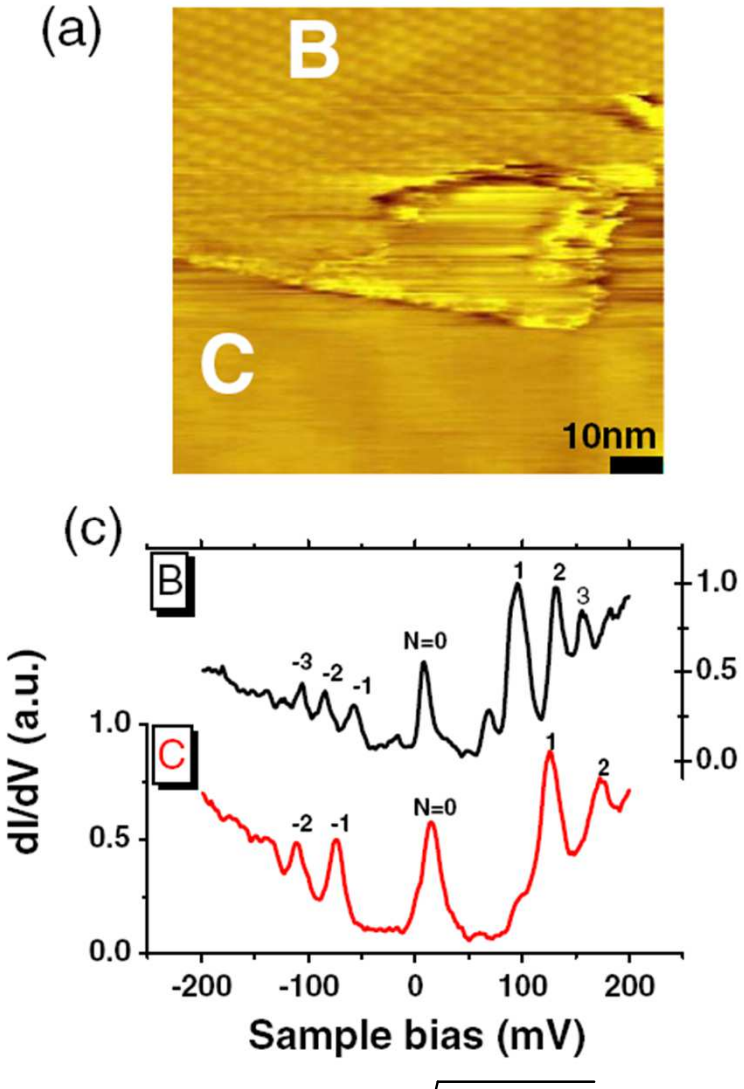


Commensurate bilayer cell (n, m) = (6, 7) for a rotation of θ = 5.08°. Full (dashed) line circle AB (AA) region.

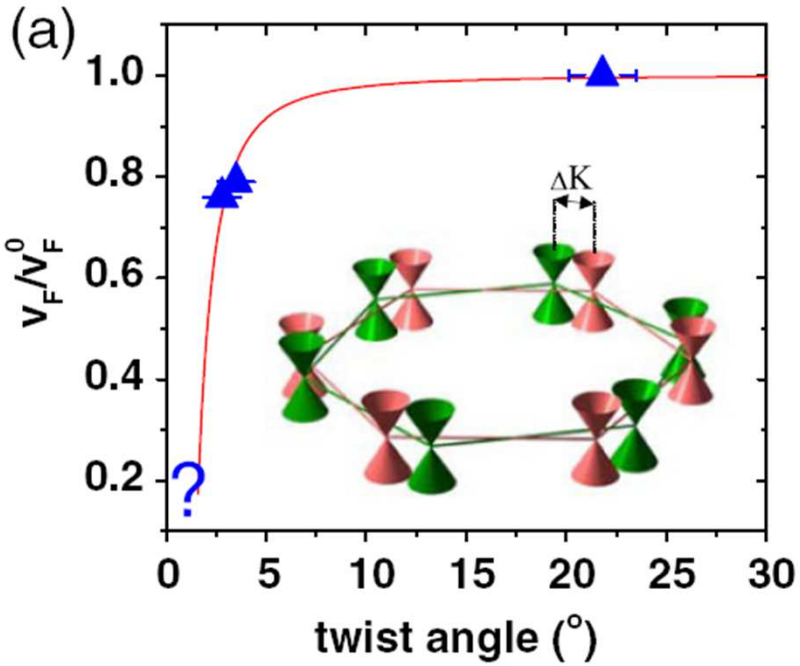


Velocity ratio Vbi /Vmono for a commensurate (n, m) bilayer cell versus rotation angle θ: circle, VASP; cross, TB calculations. The line is the model of Lopez dos Santos et al.:(19) $V\text{bi}/V\text{mono} = 1 - 9\,[\tilde{t}/(V\text{mono}K \sin(\theta/2))]2$, with $\tilde{t}$ = 0.11 eV and $V\text{mono}K = 2\gamma 0\pi(31/2)$ = 9.8 eV.

Reduction of Fermi Velocity in Rotated Graphene Bilayers



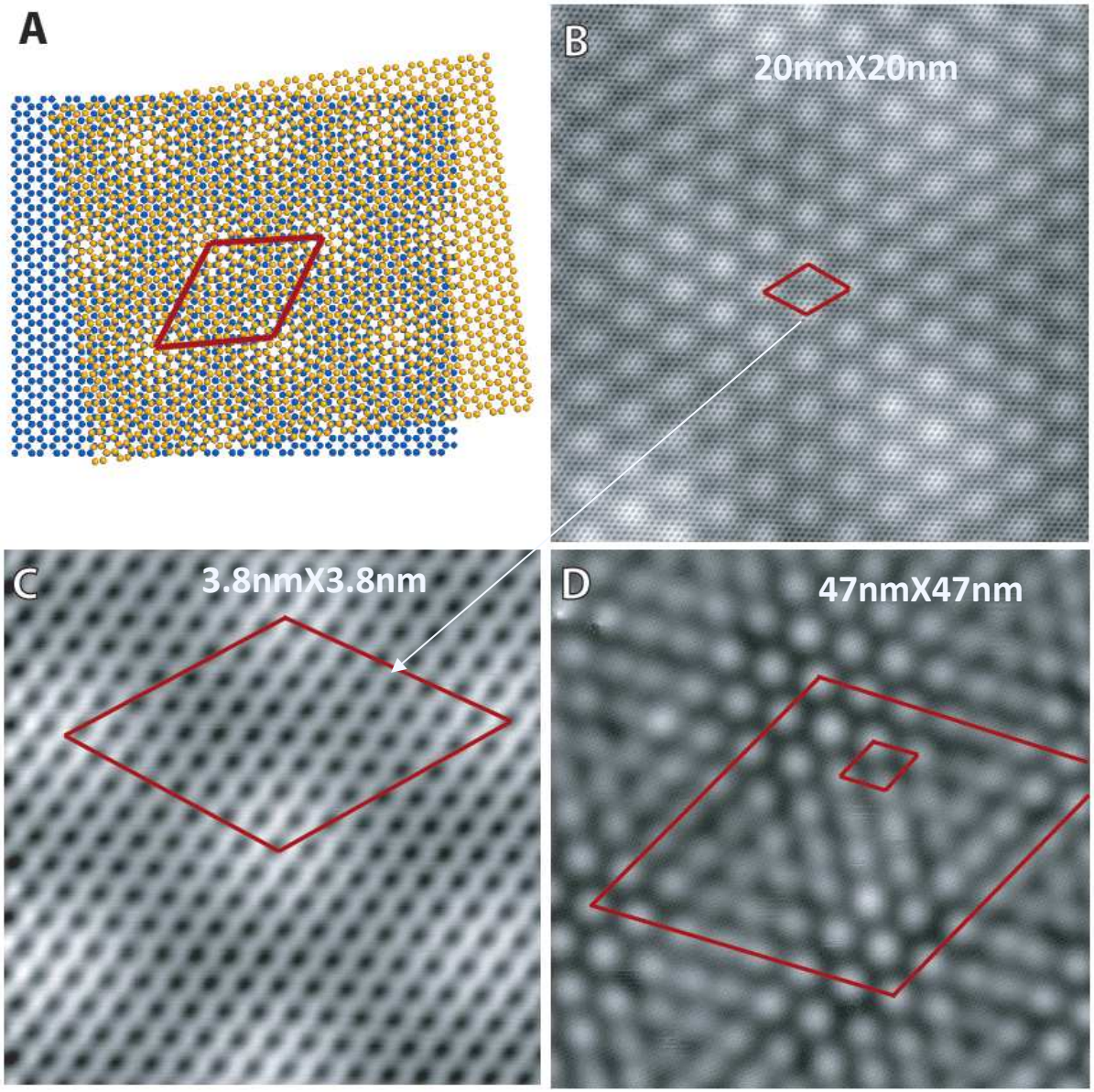
$$E_n = c * \sqrt{2n\hbar eB}$$



STM/STS measurements for few-layer graphene grown via ambient pressure CVD on polycrystalline Ni films

Luican et al. PRL 106, 126802 (2011)

Multilayer Epitaxial Graphene

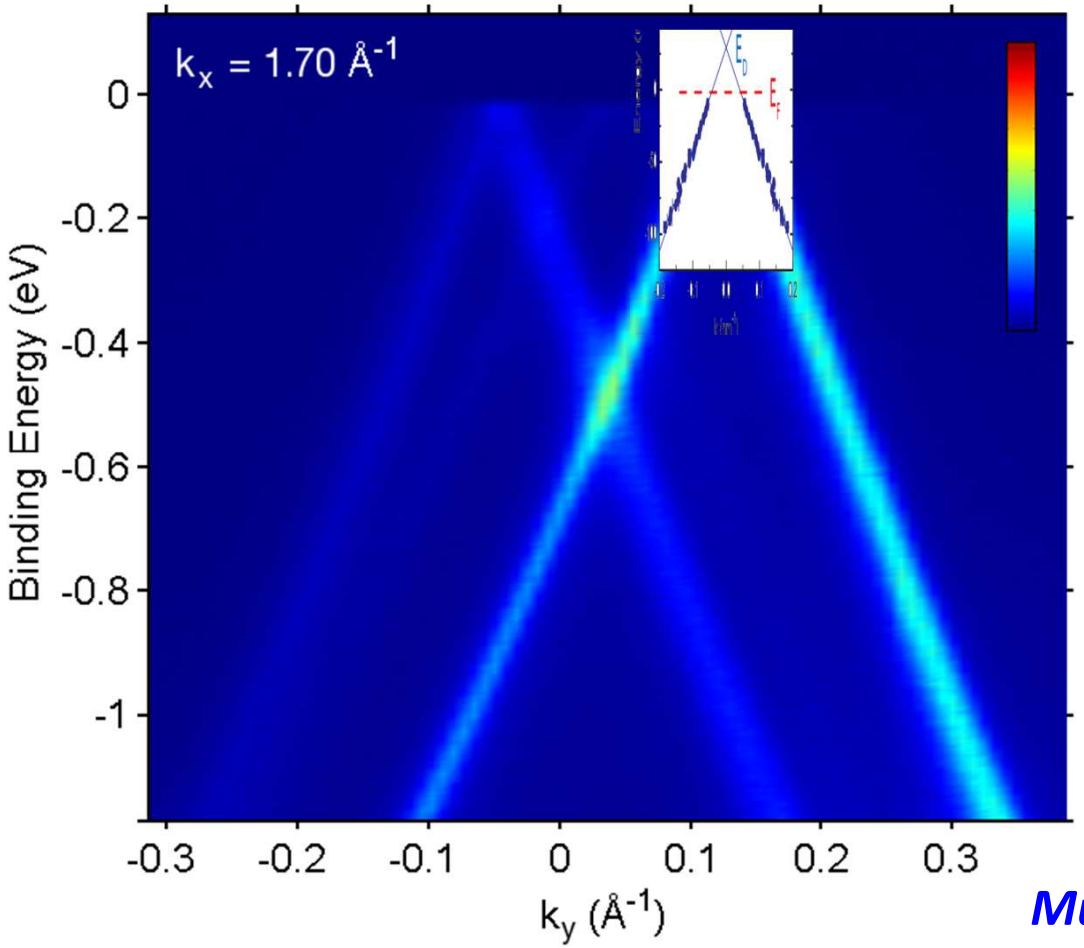


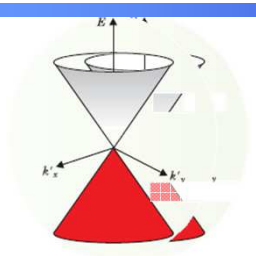
L. Miller et al.
Science 324, 924 (2009)

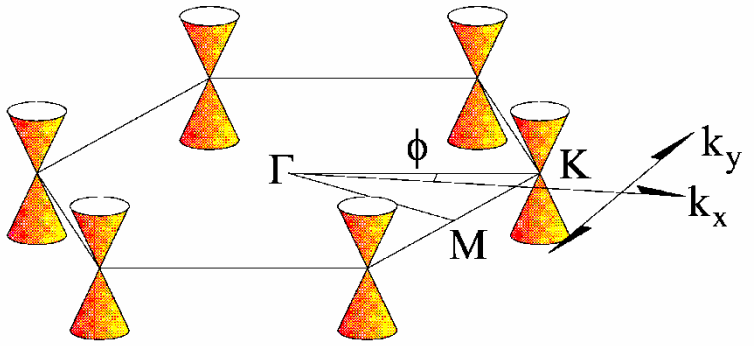
J. Hass, et al.
Phys. Rev. Lett **100** 125504 (2008)

ARPES of C-Face

T=6K; near K-point







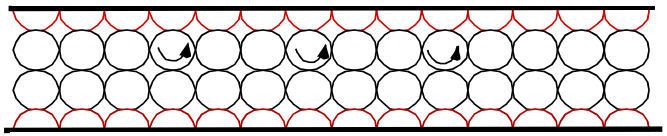
"Slicing" the Dirac cone

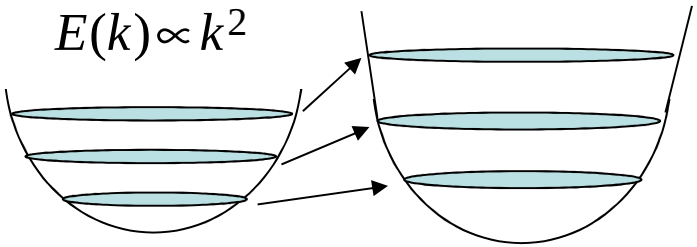
Multi-layer Epitaxial Graphene (MEG)

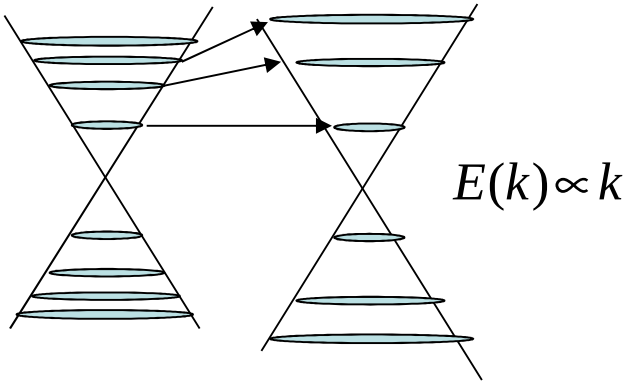
M. Sprinkle *et al., PRL,* **103,** 226803 (2009)

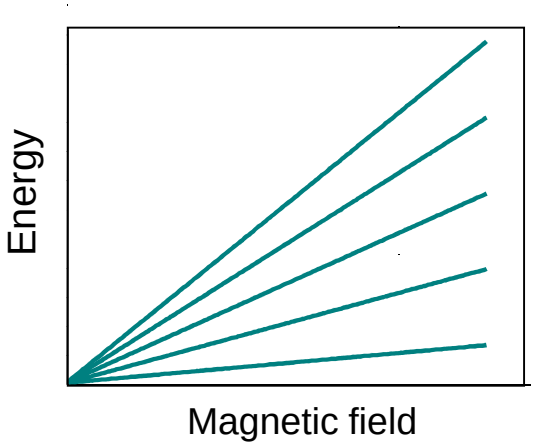
Free Electrons in a magnetic field (Landau levels)

quantization of cyclotron orbits

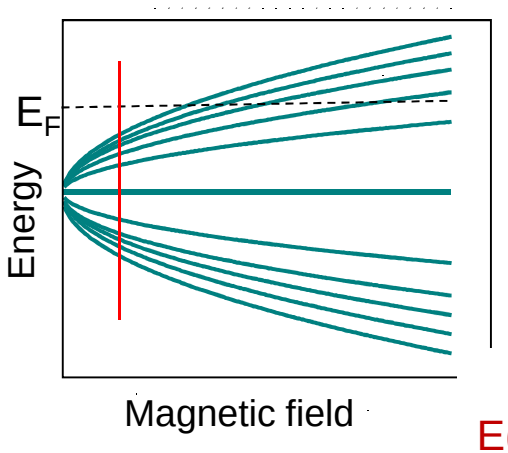








(0.1 meV per Tesla)



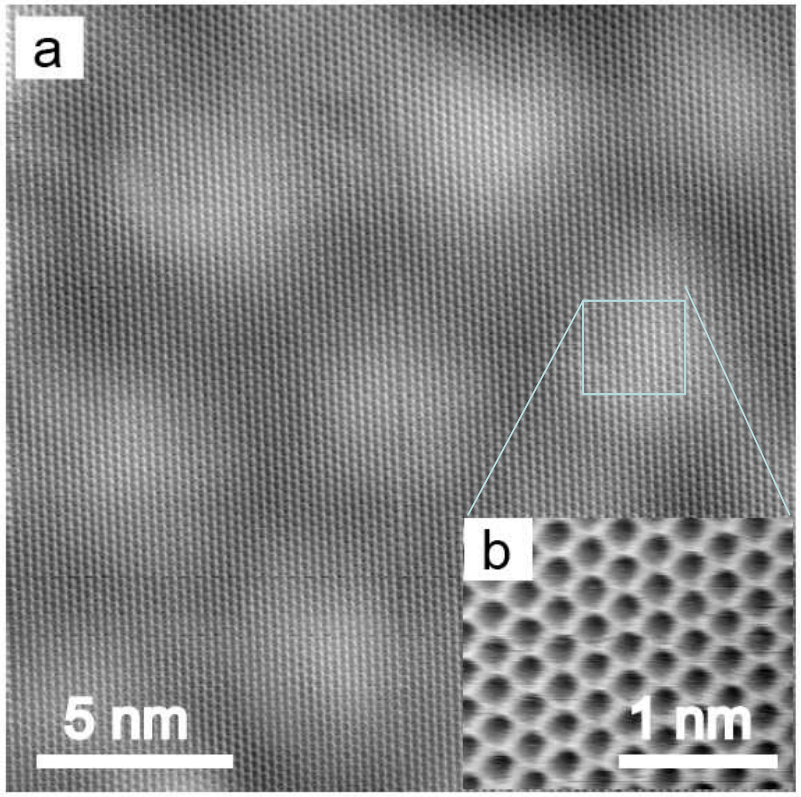
B=1 T
E(n=1) = 40 meV

$$E_n = (n + \frac{1}{2})\,\hbar\,(\frac{eB}{m*})$$

$$E_n = \pm\hbar v\sqrt{eB/\hbar}\,\sqrt{2n}$$

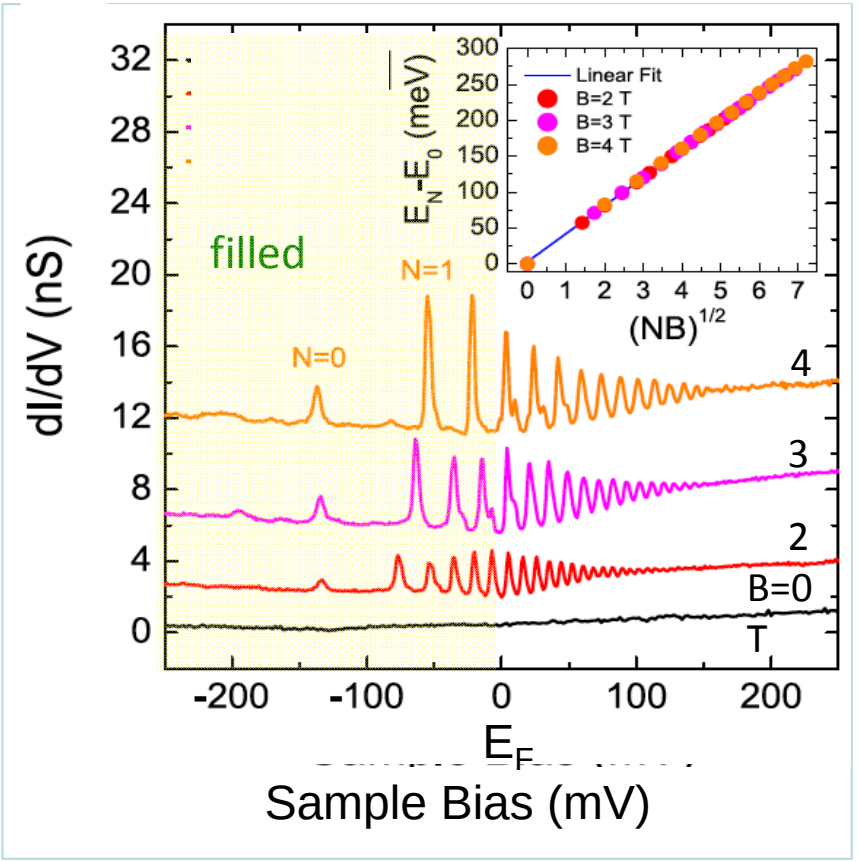
Scanning Tunneling Microscopy and Spectroscopy In a magnetic field (Landau levels)

STM (T=13 mK)



Sample Bias=-250 mV

STS (B=0-4 T)



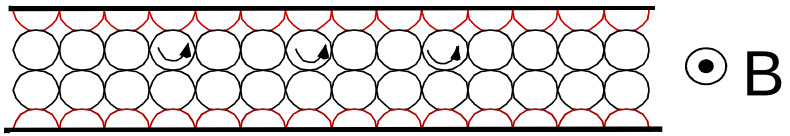
$$E_n = c^* \sqrt{2n\hbar eB}$$

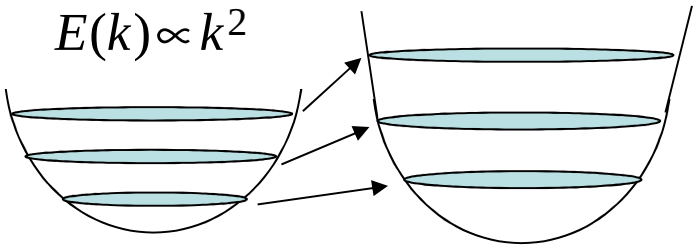
c^*= (1.08 ± 0.03) x 10^6 m/s;

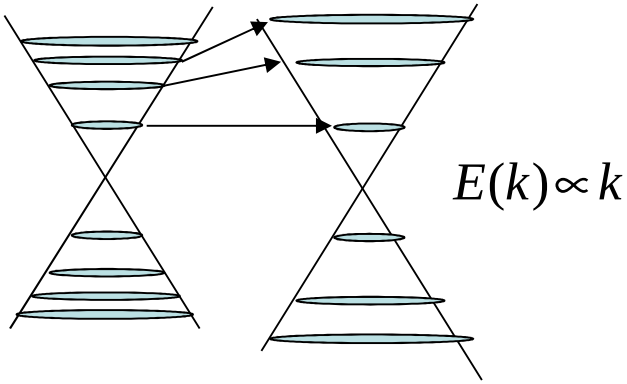
Free Electrons in a magnetic field (Landau levels)

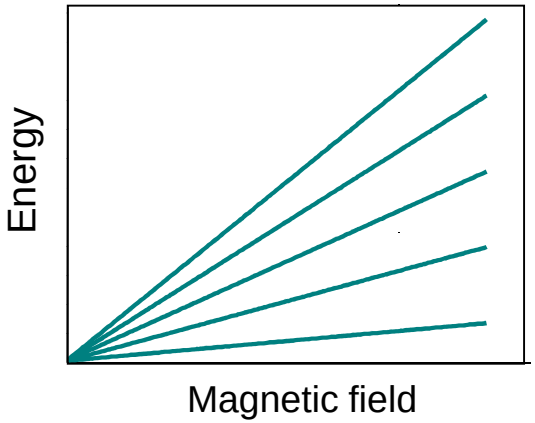
No Lattice!!

quantization of cyclotron orbits

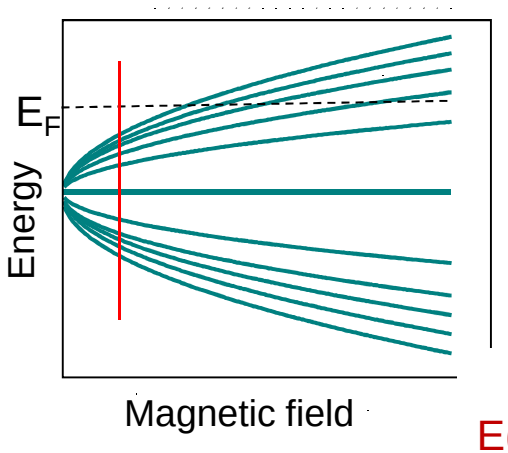








(0.1 meV per Tesla)



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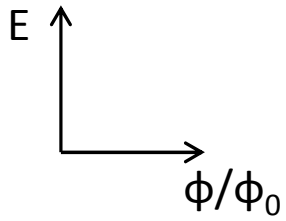
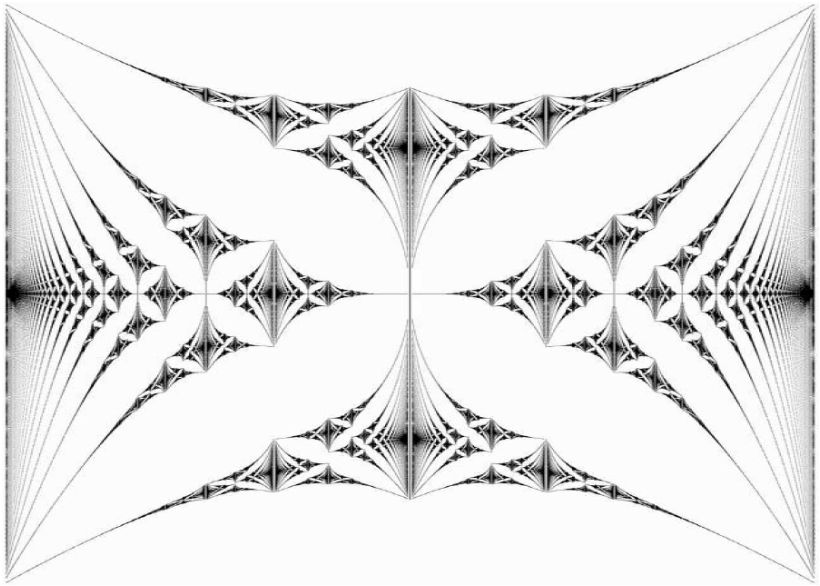
Hofstadter Butterfly (Hofstadter, PRB 14, 2239, 1976)

- A rare occurrence of a nice fractal-like picture in quantum mechanics
- 2D electrons with a periodic potential in the presence of a strong magnetic field

ϕ = magnetic flux
through one unit cell





The energy bands are clustered into subgroups and subcells; spectra of almost Mathieu operator; self-similarity maps; gaps are labeled using a Diophantine equation with parameters related to Chern numbers.

A possibility to measure this spectrum on ultra cold neutral atoms
D. Jaksch and Peter Zoller, New Journal of Physics 2003

Assume a 2D square lattice (representing the periodic potential) with lattice constant *a* and a perpendicular homogeneous magnetic field H.

SE describing the system is (LCAO for 1s orbitals and nearest neighbour interaction) is given by

$$\psi(x+a,y)+\psi(x-a,y)+e^{-\frac{ieHax}{\hbar c}}\,\psi(x,y+a)+e^{+\frac{ieHax}{\hbar c}}\,\psi(x,y-a)=\epsilon\,\psi(x,y).$$

Discretize this equation and assume plane wave behaviour along y:

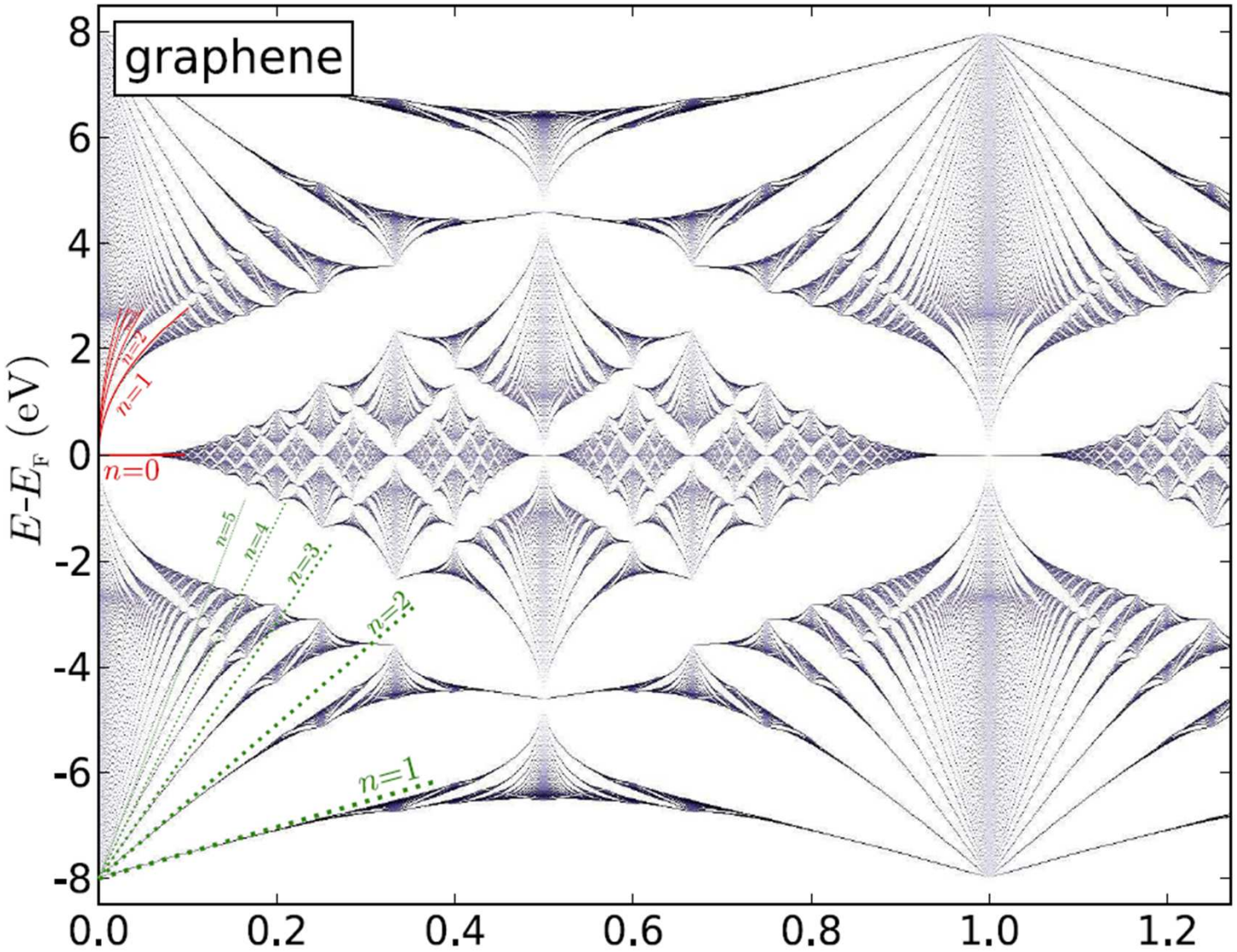
$$x=m\,a,\quad y=n\,a,\quad \psi(ma,na)=e^{i\nu n}\,g(m)$$

$$g(m+1)+g(m-1)+2\cos(2\pi m\alpha-\nu)\,g(m)=\epsilon\,g(m)$$ Harper's equation

Define parameter $\alpha=\frac{a^2H}{hc/e}=\frac{\text{flux through lattice cell}}{\text{flux quantum }\phi_0}$

interesting regime: $\alpha\approx 1$, but for a normal crystal $a\approx 1$ Å that would require a huge magnetic field

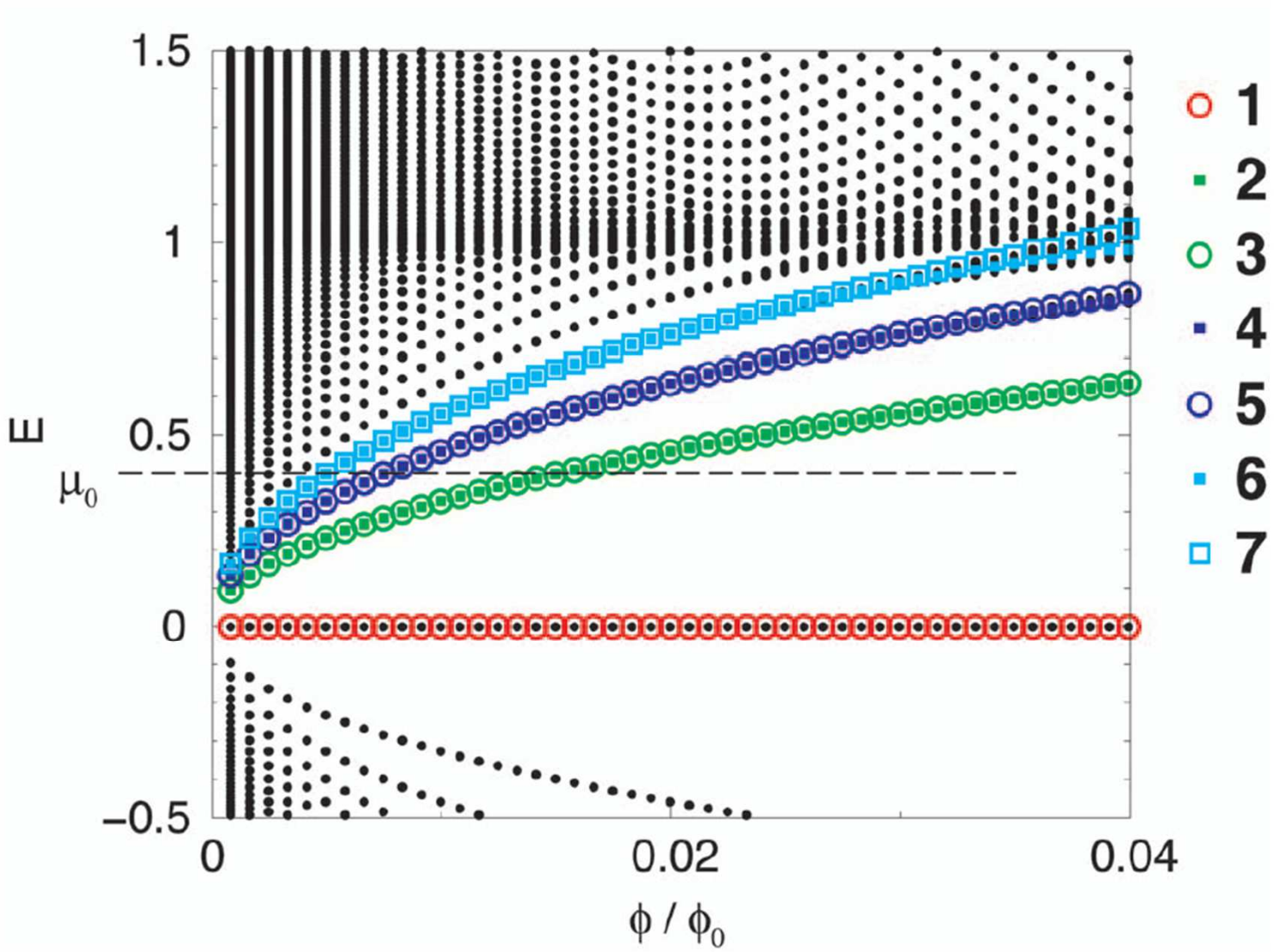
Nemec and Cuniberti, PRB 74, 165411 (2006)



B = 40 T → $\phi/\phi_0 = 0.001$

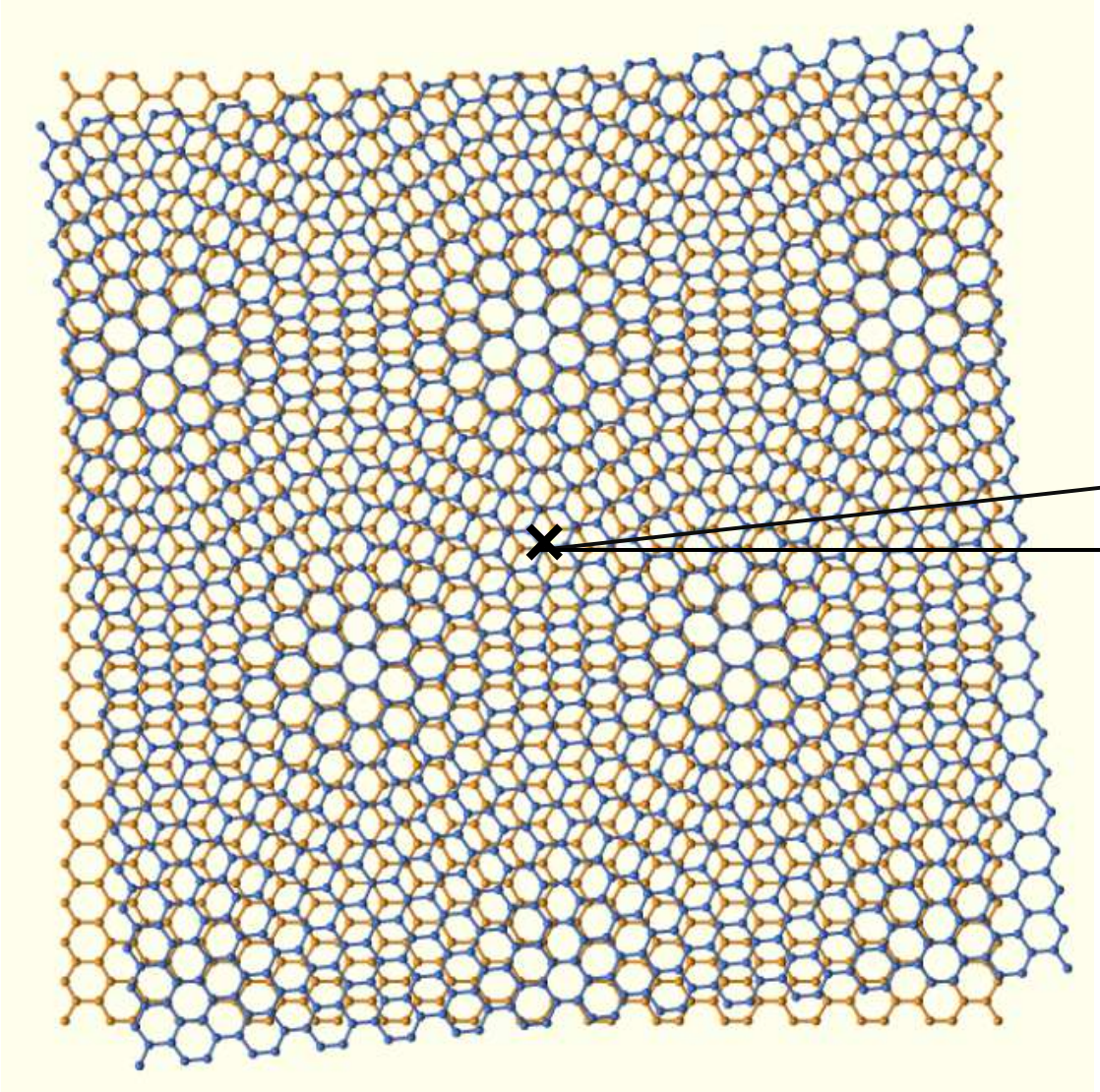
ϕ/ϕ_0

Hasegawa and Kohmoto, PRB 74, 155415 (2006)



B = 40 T → $\phi/\phi_0 = 0.001$

Twisted Bilayer Graphene



Starting from AB-stacked bilayer graphene, the bottom layer is fixed and the top layer is rotated.

Commensurate angle

$$\theta = \cos^{-1}\left(\frac{3q^2 - p^2}{3q^2 + p^2}\right)$$

Shallcross *et al.*
Phys. Rev. B 81, 1 (2010)

Other θ values are incommensurate angles.

Tight Binding Hamiltonian

$$H = \sum_{\mu,\nu=1,2} \sum_{l,j} t_{\mu l,\nu j} exp[\frac{ie}{\hbar} \int_{\vec{r}_{\mu l}}^{\vec{r}_{\nu j}} \vec{A} \cdot d\vec{l}] |\mu l\rangle\langle \nu j|$$

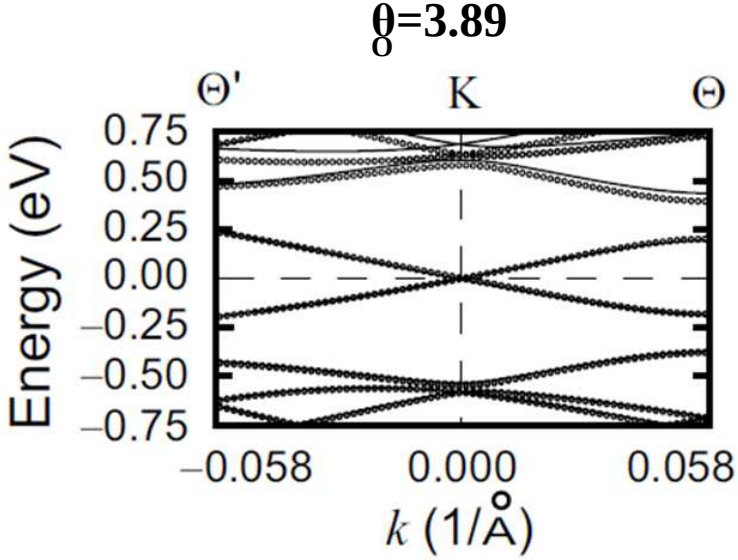
$$\vec{A} = (0, Bx)$$

TB parameters are obtained by fitting the TB bands to reproduce the band structure obtained from first-principles calculations

$$t_{\mu l,\nu j} = \begin{cases} \gamma_1 exp[\lambda_1(1 - |\vec{r}_{\mu l} - \vec{r}_{\nu j}|/a)] & (\mu = \nu) \\ \gamma_2 exp[\lambda_2(1 - |\vec{r}_{\mu l} - \vec{r}_{\nu j}|/c)] & (\mu \neq \nu) \end{cases}$$

$a = 1.42\mathring{A}$ $\quad \gamma_1 = -2.7eV$ $\quad \lambda_1 = 3.15$

$c = 3.35\mathring{A}$ $\quad \gamma_2 = 0.48eV$ $\quad \lambda_2 = 7.42$



Lanczos Recursive Method

Real space Hamiltonian (Hermitian matrix)

$$H = \sum_{i,j} t_{ij} \exp(\frac{ie}{\hbar}\int_i^j \bar{A}\cdot d\bar{l})a_i^+ a_j$$

Construct a new orthogonal basis

$$\left|\tilde{\Phi}_{N+1}\right\rangle = b_{N+1}\left|\Phi_{N+1}\right\rangle = H\left|\Phi_N\right\rangle - a_N\left|\Phi_N\right\rangle - b_N\left|\Phi_{N-1}\right\rangle$$

$$a_N = \left\langle\Phi_N\left|H\right|\Phi_N\right\rangle \qquad b_N = \sqrt{\left\langle\tilde{\Phi}_N\left|\tilde{\Phi}_N\right.\right\rangle}$$

Hamiltonian in the new basis

$$H = \begin{pmatrix} a_0 & b_1 & 0 & \cdots \\ b_1 & a_1 & b_2 & \cdots \\ 0 & b_2 & a_2 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix}$$

Real space Green's function of the first element (continued fraction expansion)

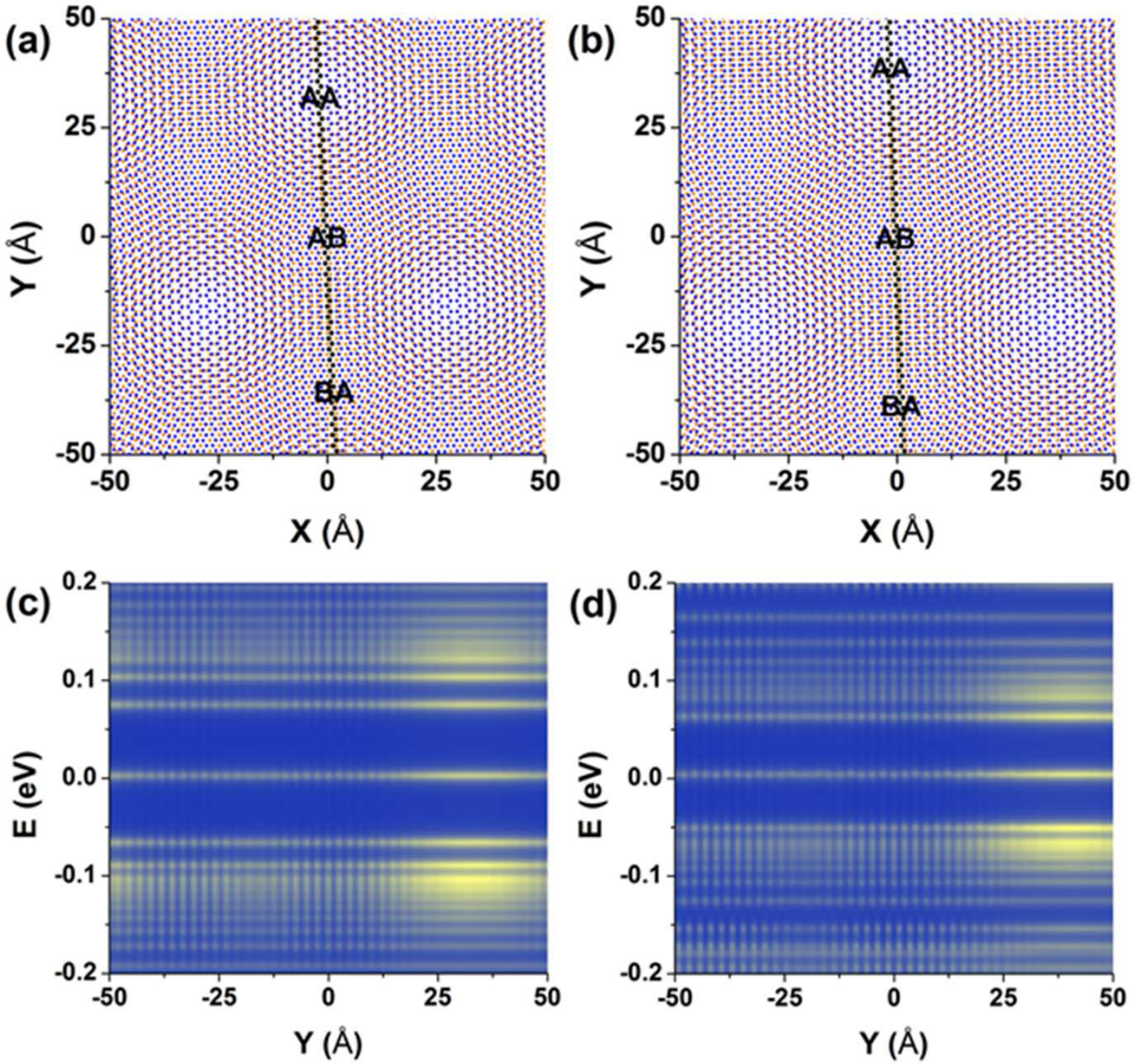
$$\left\langle\Phi_0\left|G^r(E)\right|\Phi_0\right\rangle = \cfrac{1}{E+i\eta-a_0-\cfrac{b_1^2}{E+i\eta-a_1-\cfrac{b_2^2}{E+i\eta-a_2-\cdots}}}$$

$$\cdots$$

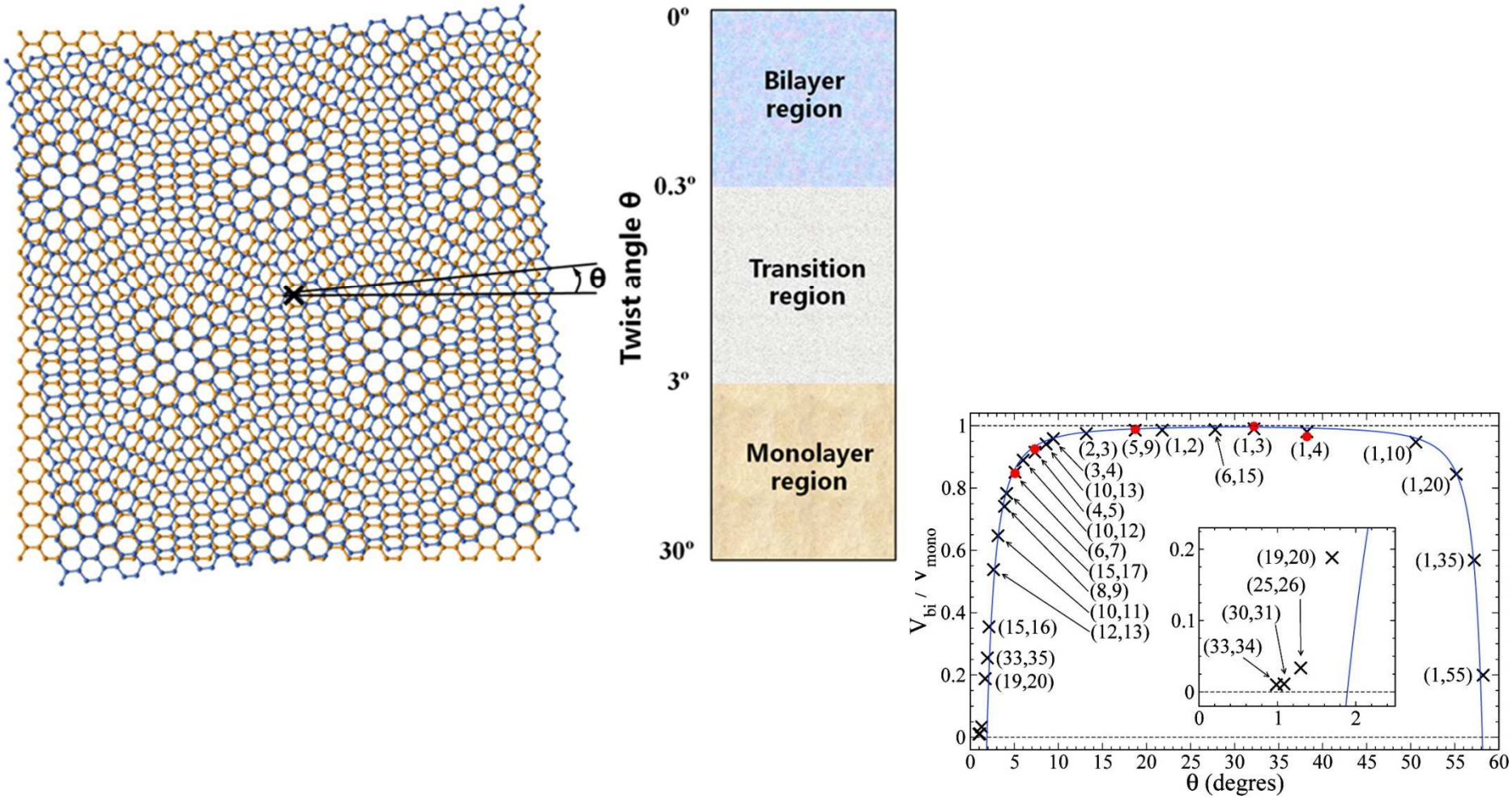
140nm×140nm, over 1.5million atoms

Laudau Levels for Twisted Bilayer Graphene

(a) commensurate angle $\theta = 2.472^{O}$ (b) incommensurate angle $\theta = 2^{O}$



Laudau Levels for Twisted Bilayer Graphene



Trambly de Laissardiere et al. Nano Lett. 2010

Laudau Levels for Twisted Bilayer Graphene

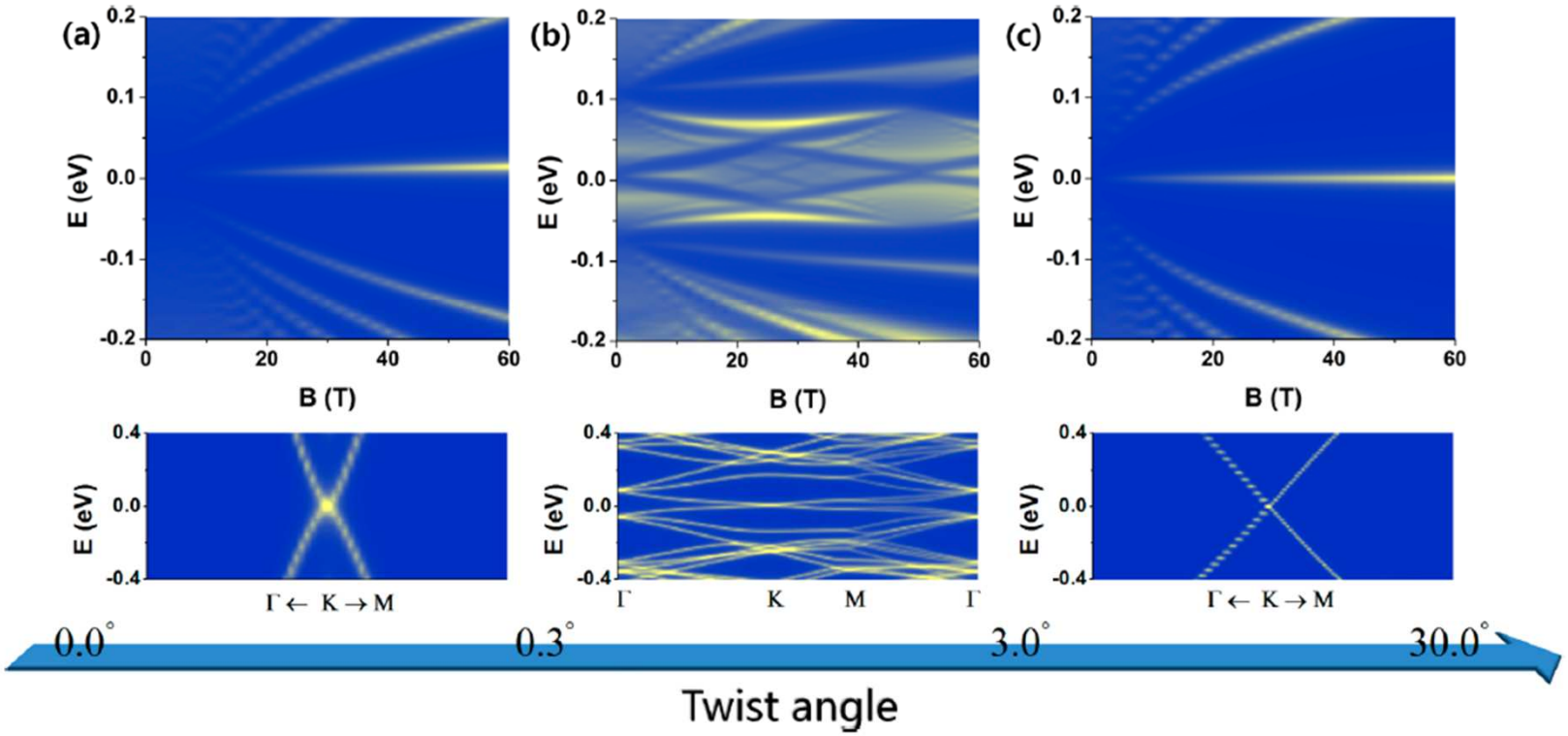
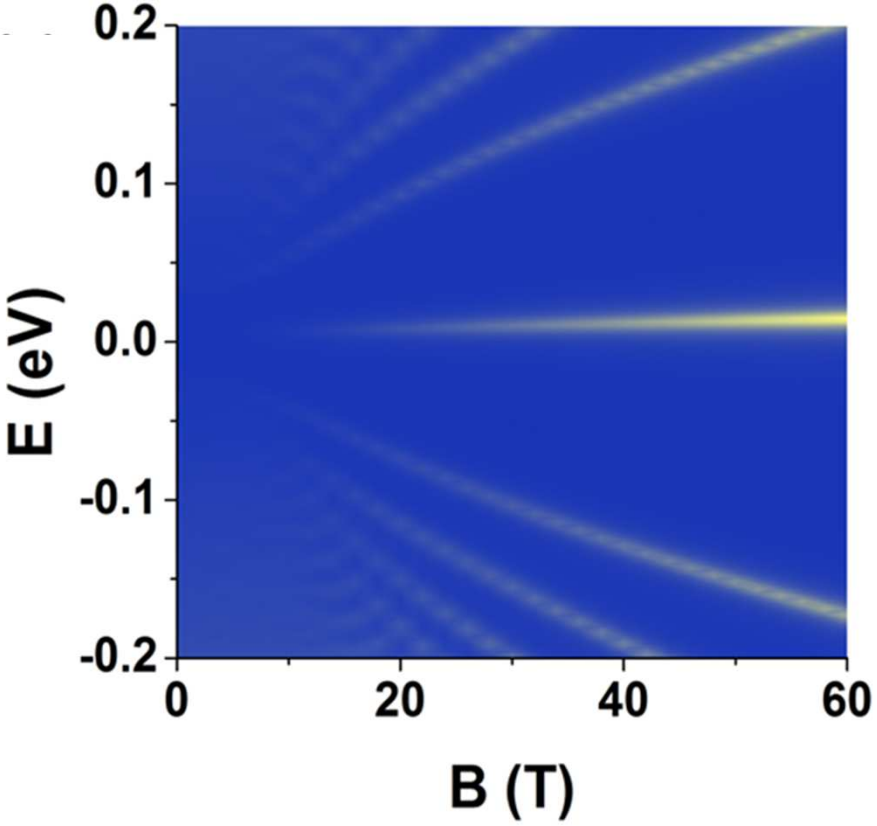
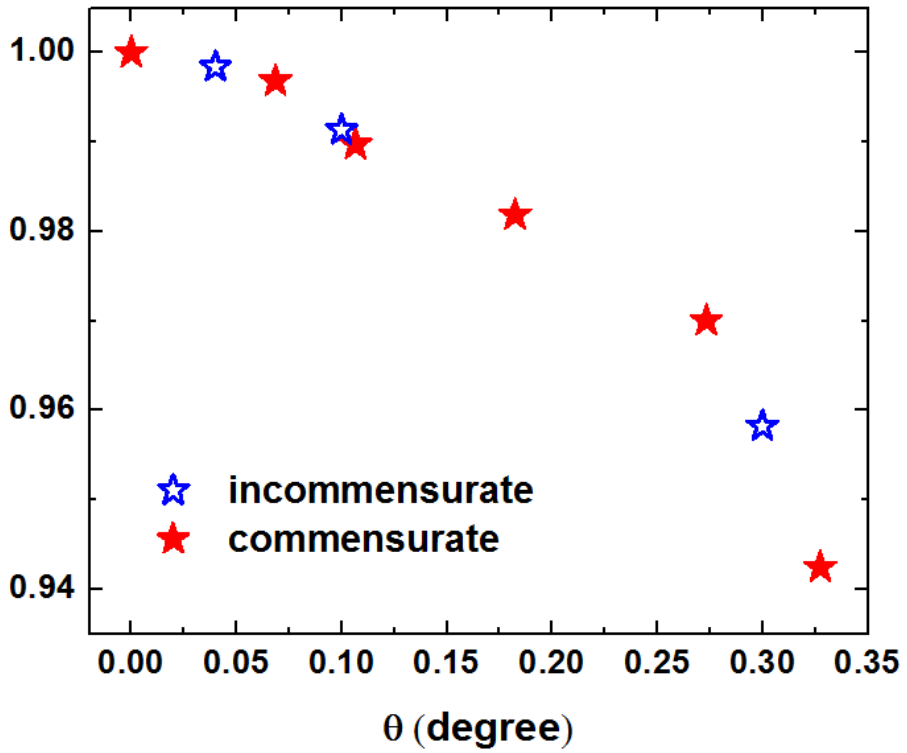


Figure 1. Landau-level spectra for twisted bilayer graphene at three commensurate angles representing three ranges of the twist angle θ. (a) θ = 0.069° for the Bernal-bilayer regime 0.0–0.3°, (b) θ = 1.444° for the regime 0.3–3.0°, and (c) θ = 7.565° for the effectively "single layer" regime (3.0–30.0°). The corresponding electronic bands in these ranges are shown below each spectrum with θ = 0.0, 1.538, and 7.341°, respectively. The band structures for all the commensurate angles are obtained by diagonalization of the unit-cell Hamiltonian in reciprocal space with periodic boundary condition.

Small Twist Angles: AB-Bilayer Graphene Region



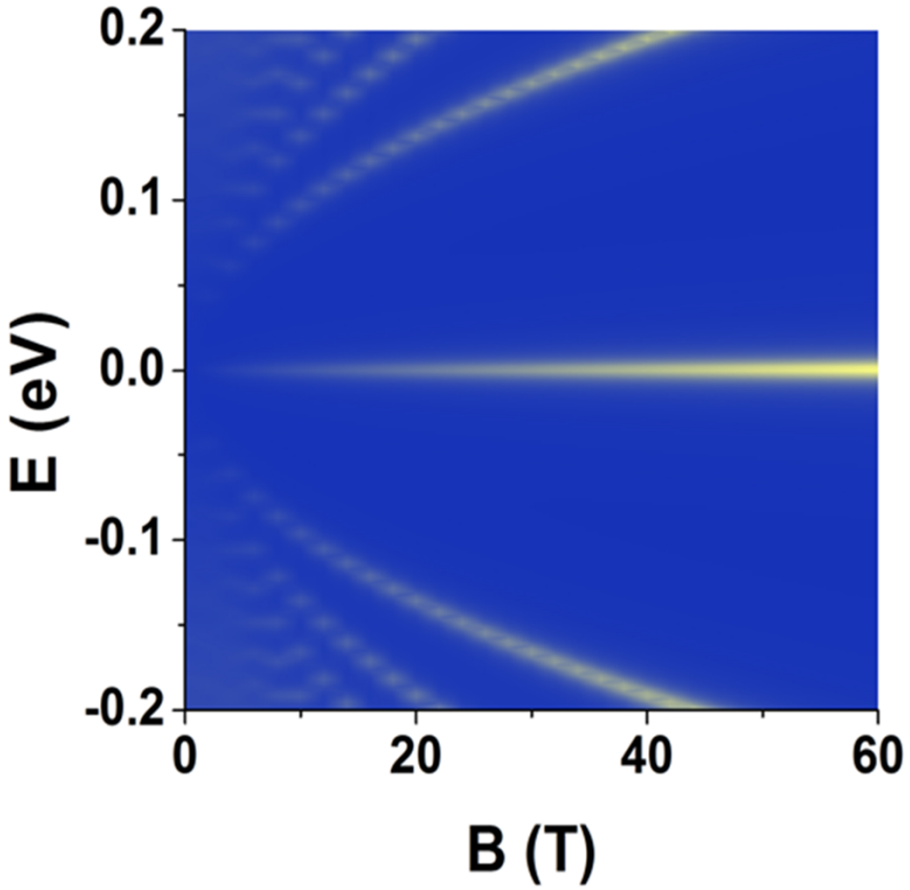


Commensurate
θ=0.06853°

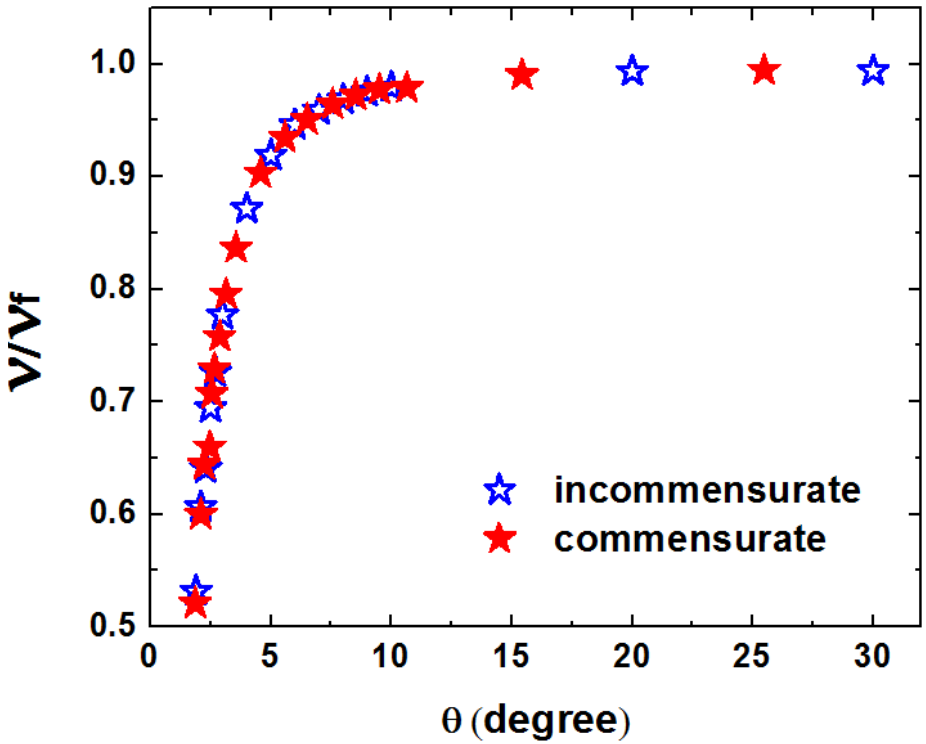
$$E_n = \frac{\mathrm{sgn}(n)}{\sqrt{2}}\left[(2|n|+1)\Delta^2 + \gamma_1^2 - \sqrt{\gamma_1^4 + 2(2|n|+1)\Delta^2\gamma_1^2 + \Delta^4}\right]^{1/2}$$

$$\Delta = \sqrt{2eBv_F^2\hbar}$$

Large Twist Angles: Monolayer Graphene Region



Commensurate
θ=7.56507^{o}



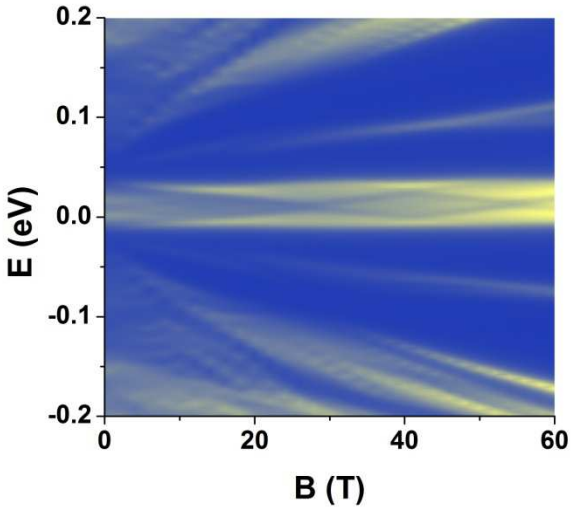
$$E_n = \mathrm{sgn}(n)\Delta\sqrt{|n|}$$

$$\Delta = \sqrt{2eBv_F^2\hbar}$$

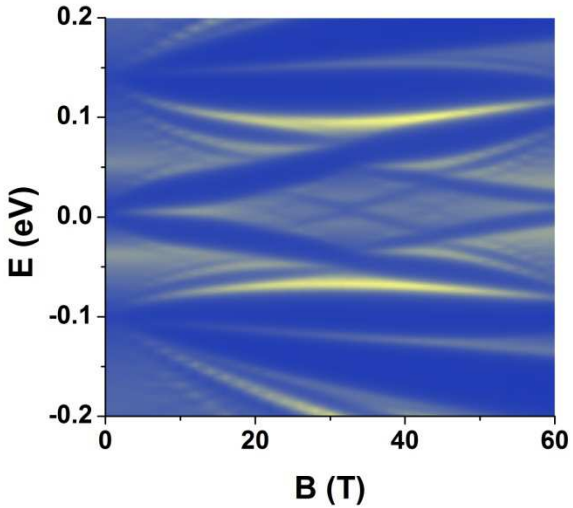
Landau Levels at Small Twist Angles

Commensurate:

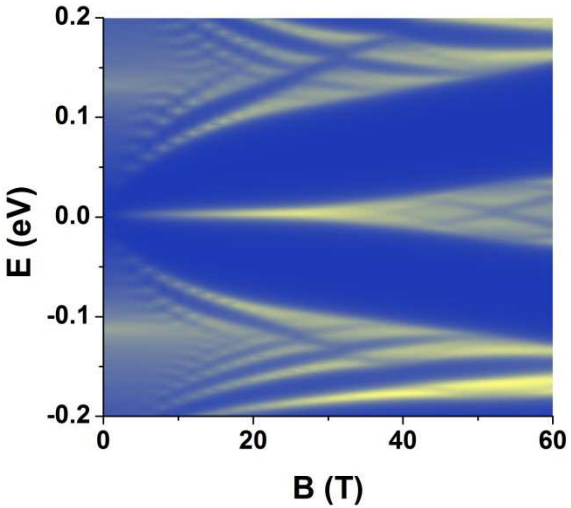
θ=1.06689^{o}



θ=1.64996^{o}

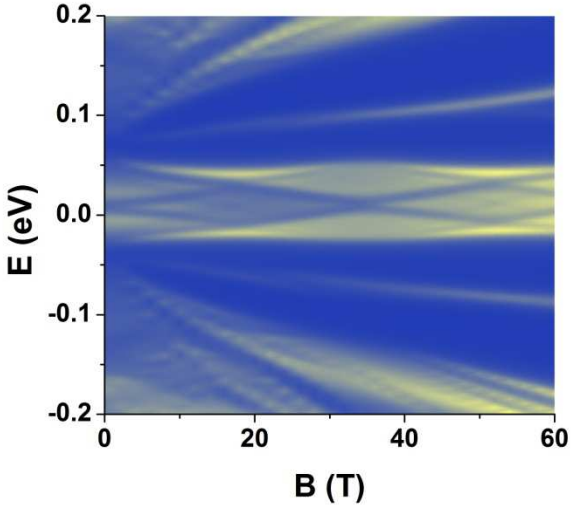


θ=2.56292^{o}

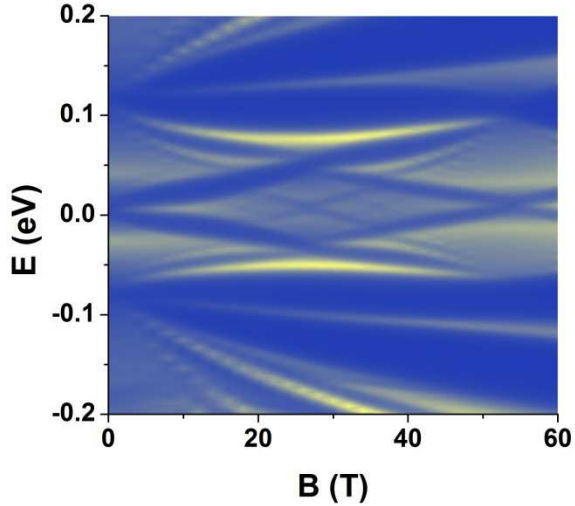


Incommensurate:

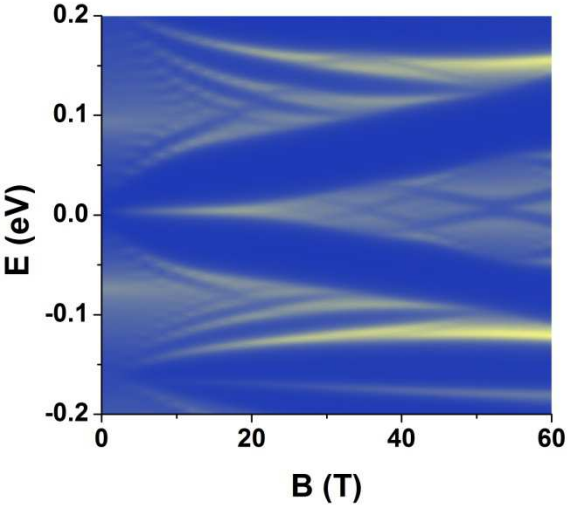
θ=1.2^{o}



θ=1.5^{o}



θ=2.1^{o}



Summary

- Twisted multilayer graphene exhibits intriguing electronic properties.
- Quasi-ordered stacking may preserve single-layer linear energy dispersion.
- A complex Hofstadter butterfly spectra could be observed in twisted graphene bilayer within a certain angular range at laboratory accessible magnitudes of magnetic field.

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