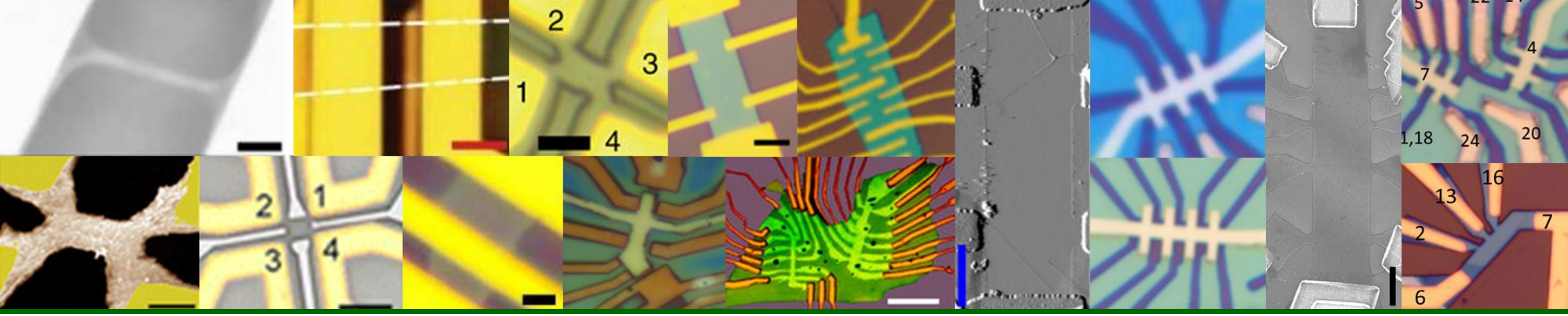


Quantum transport in van der Waals heterostructures

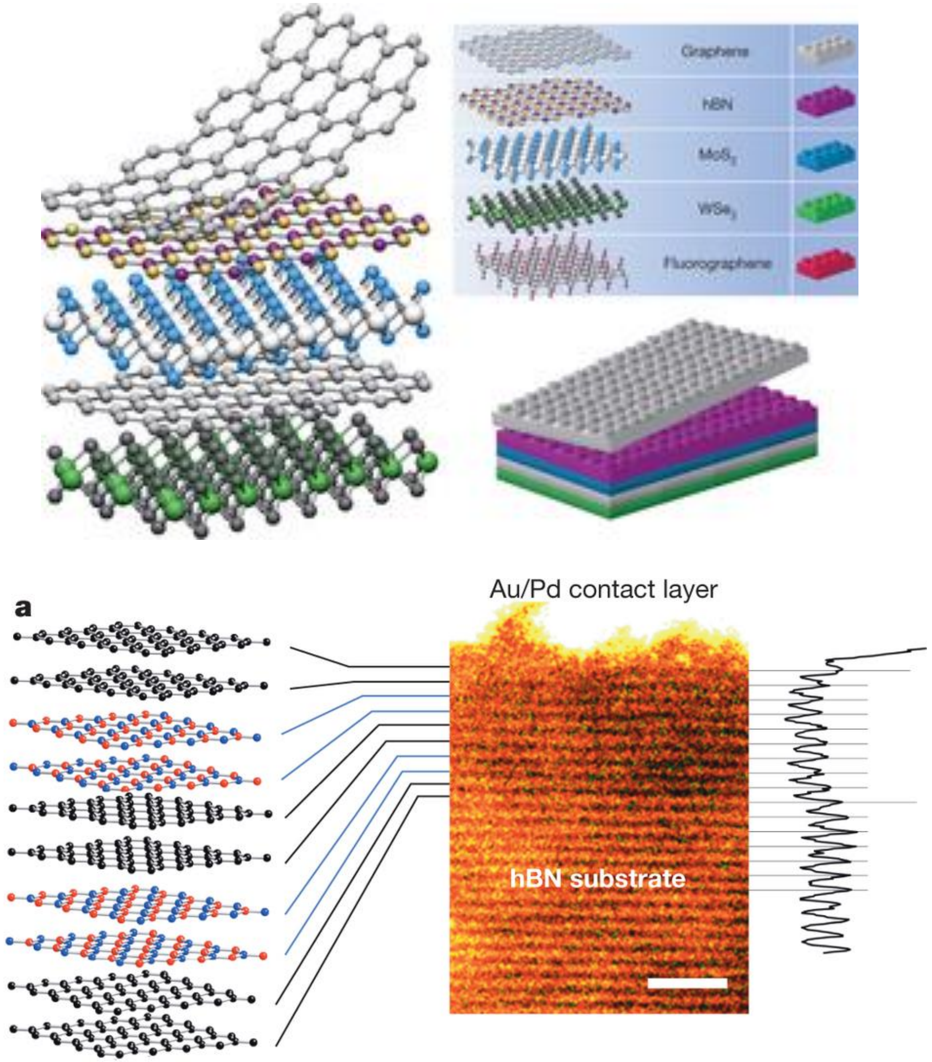
Dong-Keun Ki

May 19, 2023

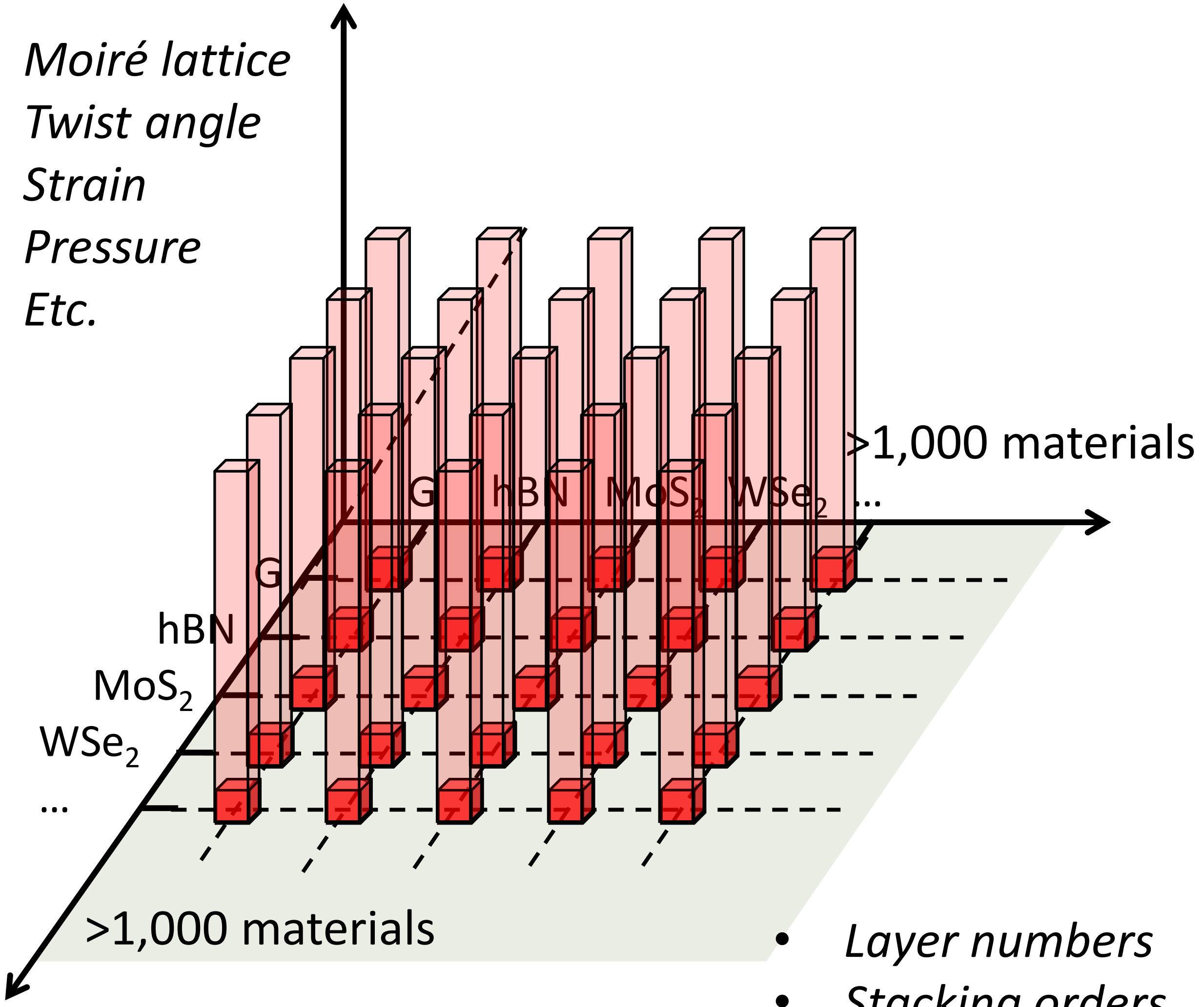
School of Mesoscopic Physics, POSTECH

Department of Physics
The University of Hong Kong

Plenty of important progresses and infinite possibilities in vdW heterostructures



Review by A.K. Geim and I.V. Grigoriea, Nature 2013



Moiré lattice
Twist angle
Strain
Pressure
Etc.

>1,000 materials

G hBN MoS_2 WSe_2 …

G
hBN
MoS_2
WSe_2
…

>1,000 materials

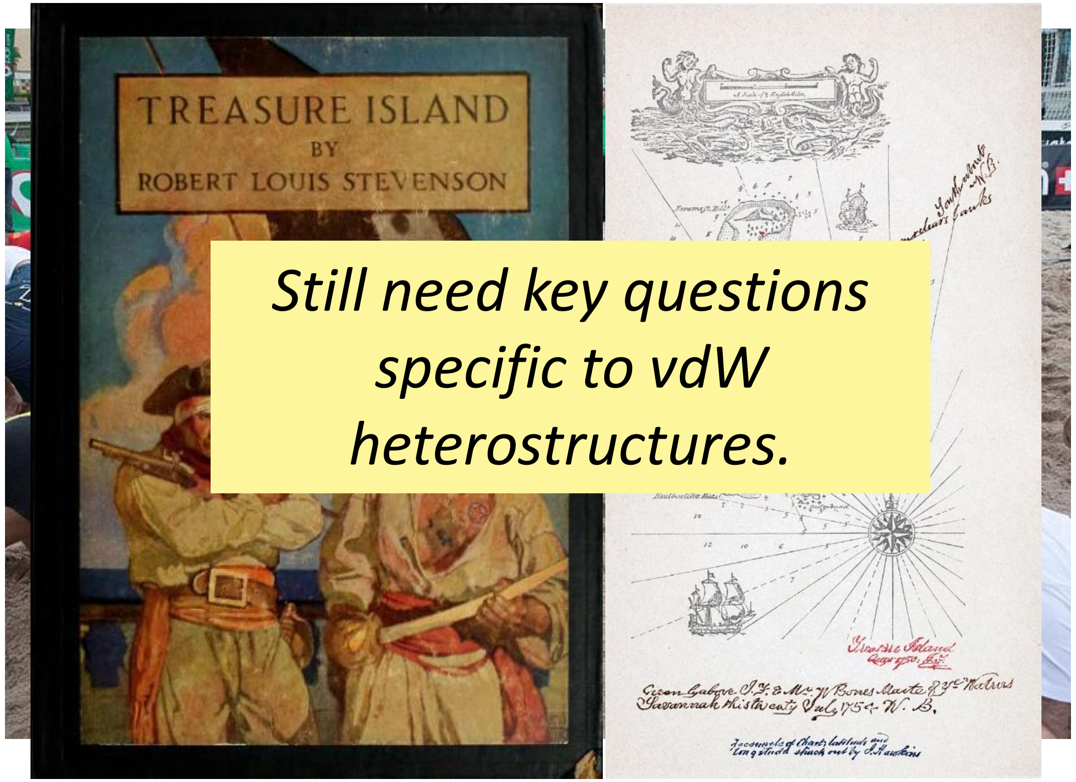
- *Layer numbers*
- *Stacking orders*
- *Dimensions*

Grand challenges or questions?

Grand goals

- *High figure of merits: energy efficiency, etc.*
- *Mass of virtual particles >> Dark energy*
- *……*

Hunting treasures

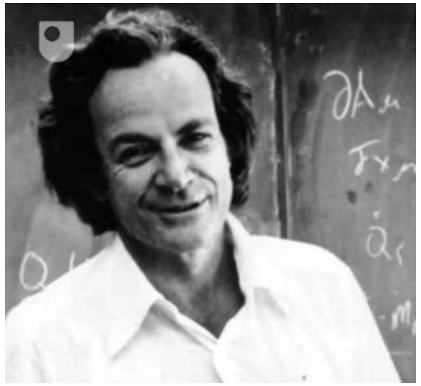


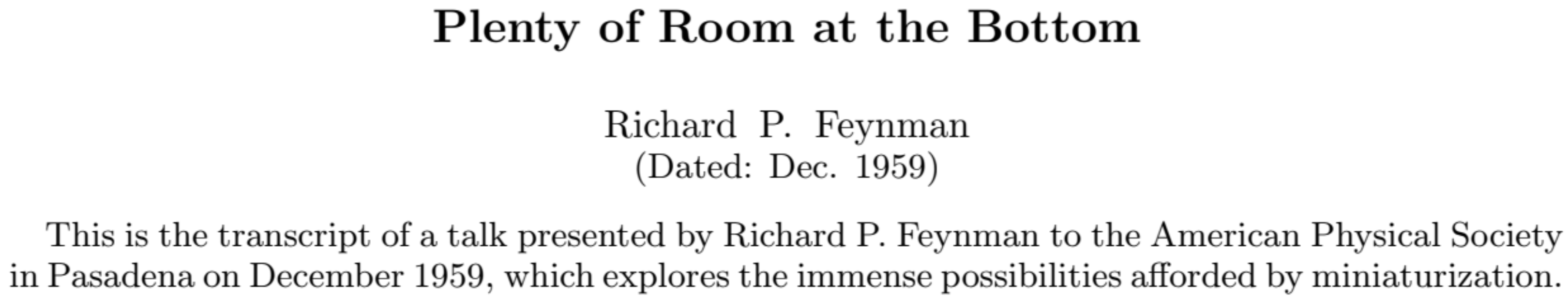
Still need key questions specific to vdW heterostructures.

Obey the laws of physics

Current technologies

Words from giant

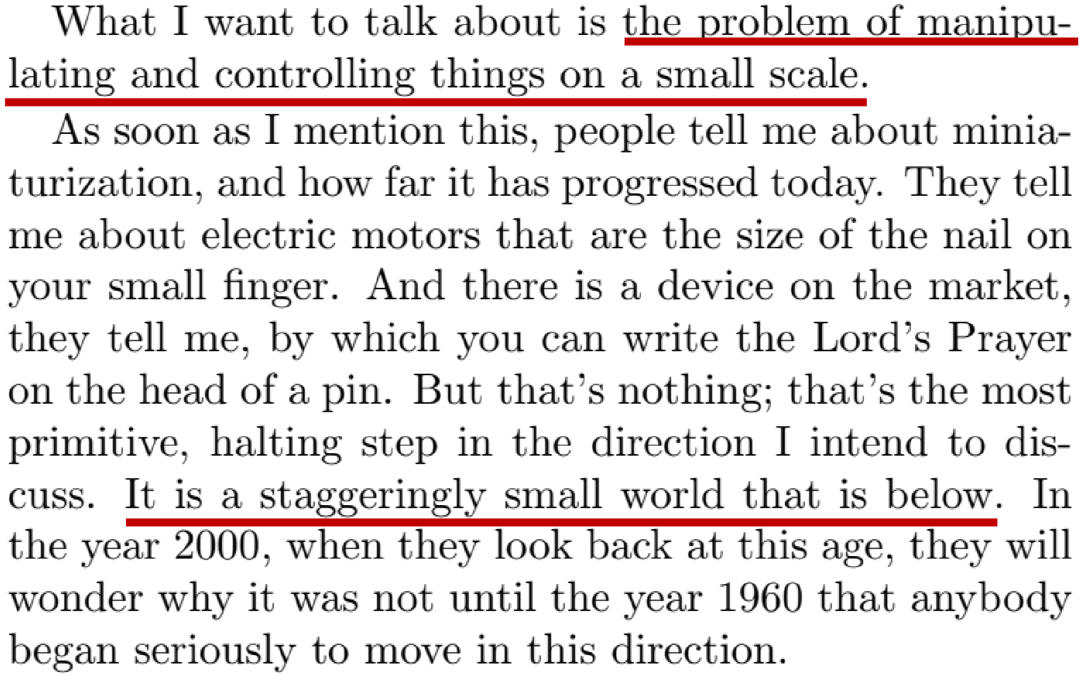




Plenty of Room at the Bottom

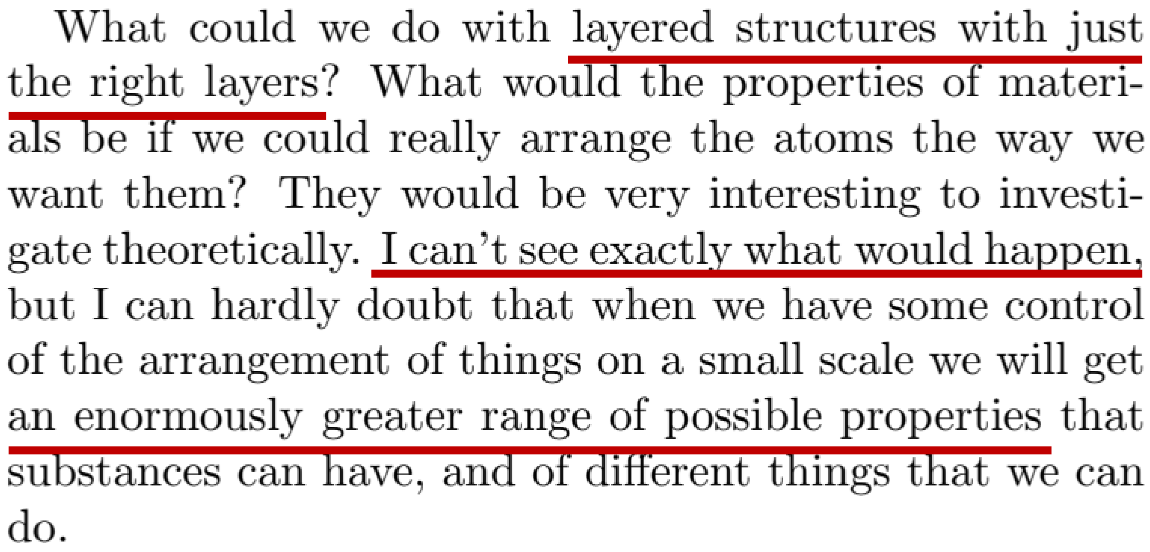
Richard P. Feynman
(Dated: Dec. 1959)

This is the transcript of a talk presented by Richard P. Feynman to the American Physical Society in Pasadena on December 1959, which explores the immense possibilities afforded by miniaturization.



What I want to talk about is the problem of manipulating and controlling things on a small scale.

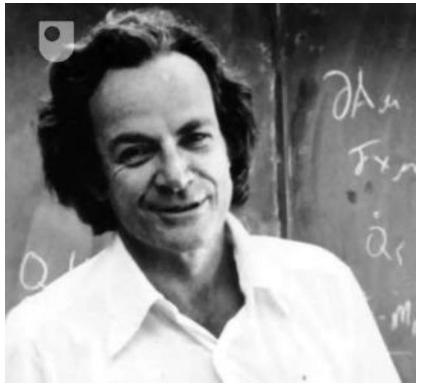
As soon as I mention this, people tell me about miniaturization, and how far it has progressed today. They tell me about electric motors that are the size of the nail on your small finger. And there is a device on the market, they tell me, by which you can write the Lord's Prayer on the head of a pin. But that's nothing; that's the most primitive, halting step in the direction I intend to discuss. It is a staggeringly small world that is below. In the year 2000, when they look back at this age, they will wonder why it was not until the year 1960 that anybody began seriously to move in this direction.

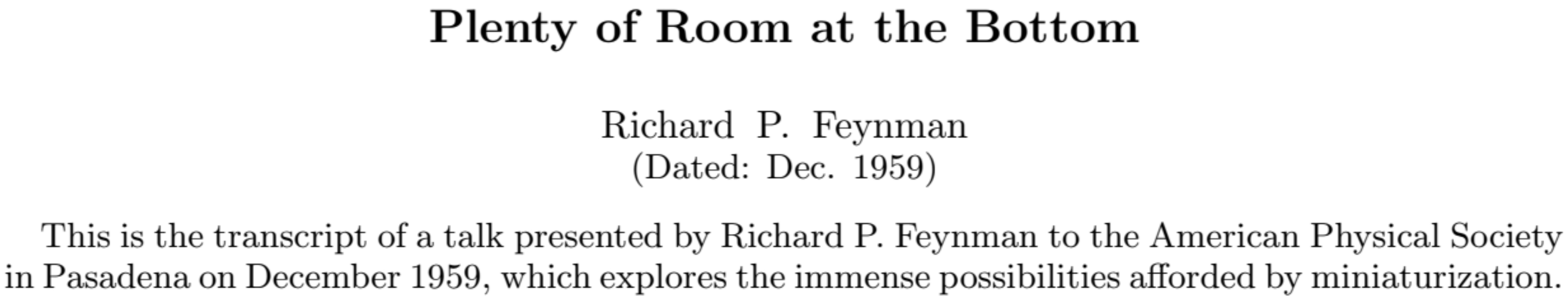


What could we do with layered structures with just the right layers? What would the properties of materials be if we could really arrange the atoms the way we want them? They would be very interesting to investigate theoretically. I can't see exactly what would happen, but I can hardly doubt that when we have some control of the arrangement of things on a small scale we will get an enormously greater range of possible properties that substances can have, and of different things that we can do.

https://web.pa.msu.edu/people/yang/RFeynman_plentySpace.pdf

Key questions to answer





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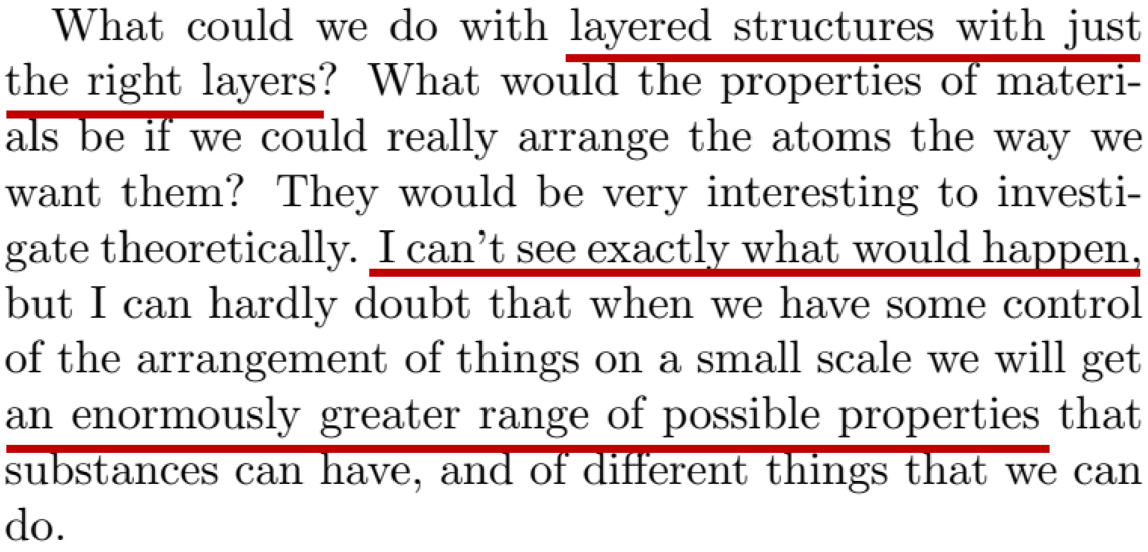

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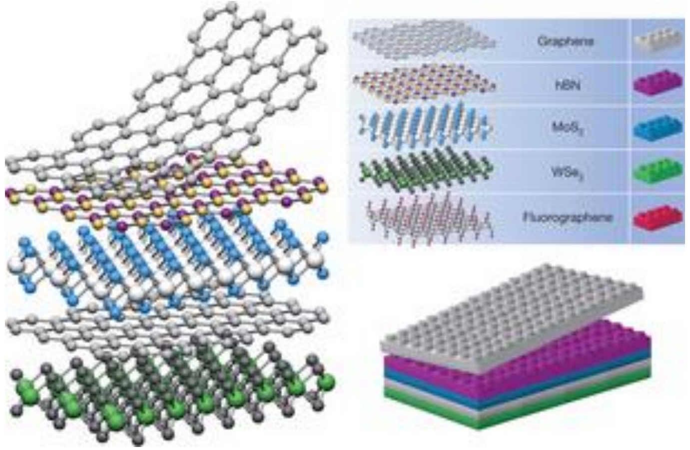
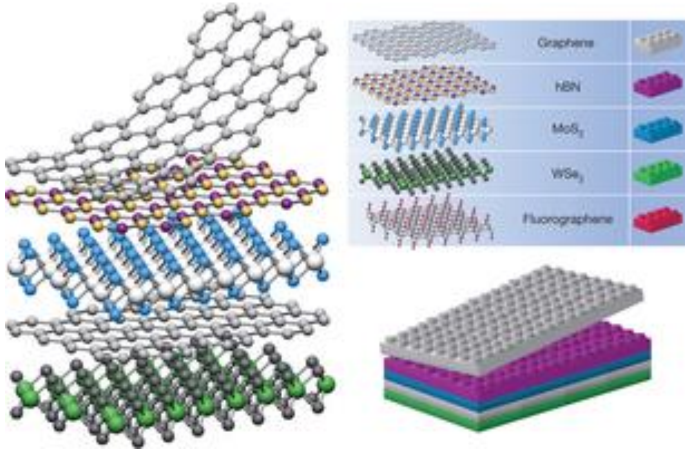


- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

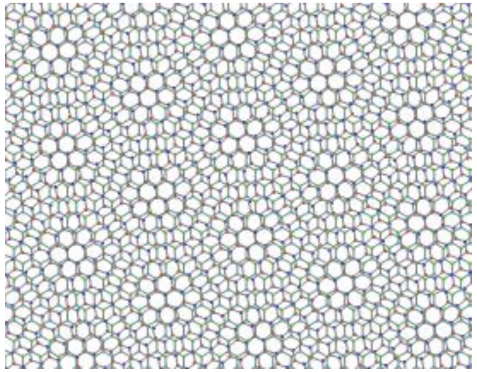
Contents

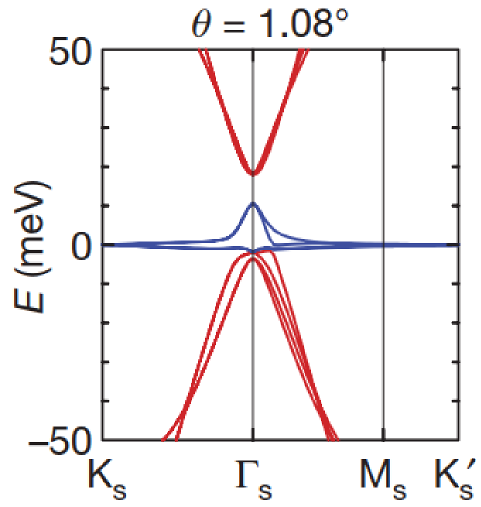
- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

- *Introduction to 2D materials*
- *Their special properties compared with other 2D electron gas systems*
- *Van der Waals assembly techniques*



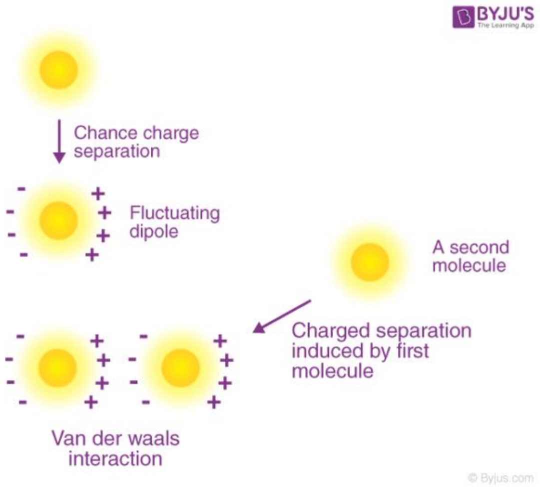
- *Few examples of new properties*
- *Discussions on the mechanisms*
- *Future challenges and directions?*





2D materials

van der Waals coupled layered materials → defect-free, free-standing monolayers

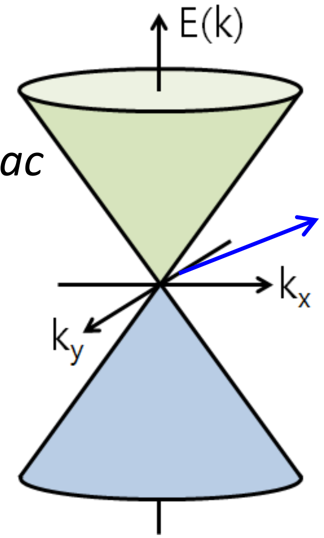


In graphite, each graphene layers are coupled by a van der Waals (vdW) force which is much weaker than chemical bonds.

Thus, it can be exfoliated to produce "defect-free" atomically thin layers

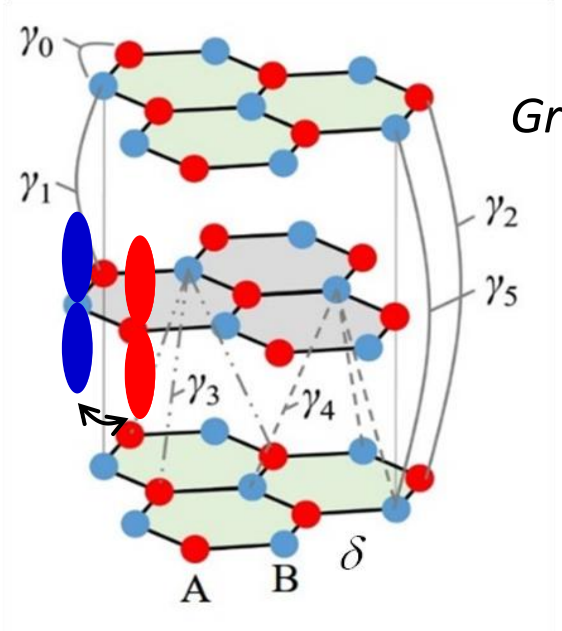
Carriers move between atomic orbitals: ***atomic registry/potential is important***

Massless Dirac electrons

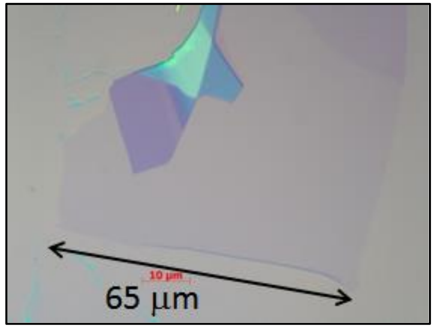


Charge neutrality point (CNP)

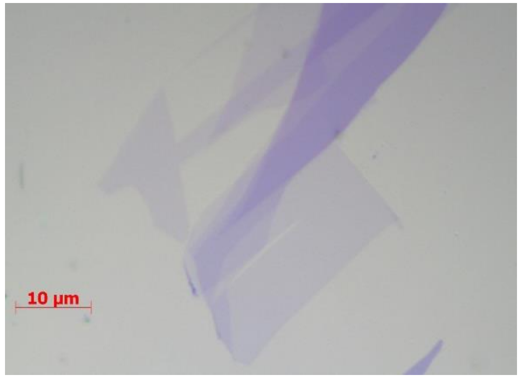
Graphite

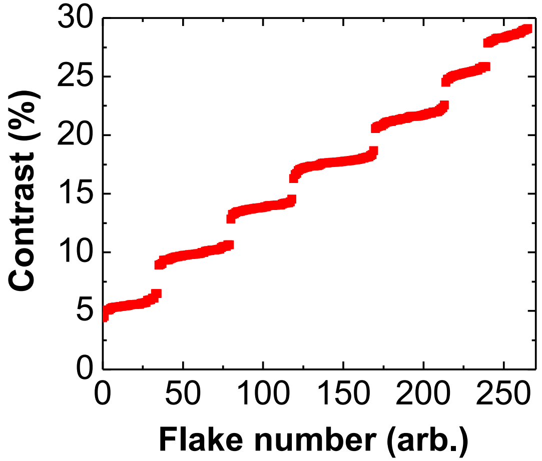


Mechanical, Chemical → *Graphene*



N-layer graphene, a family of closely related electronic systems

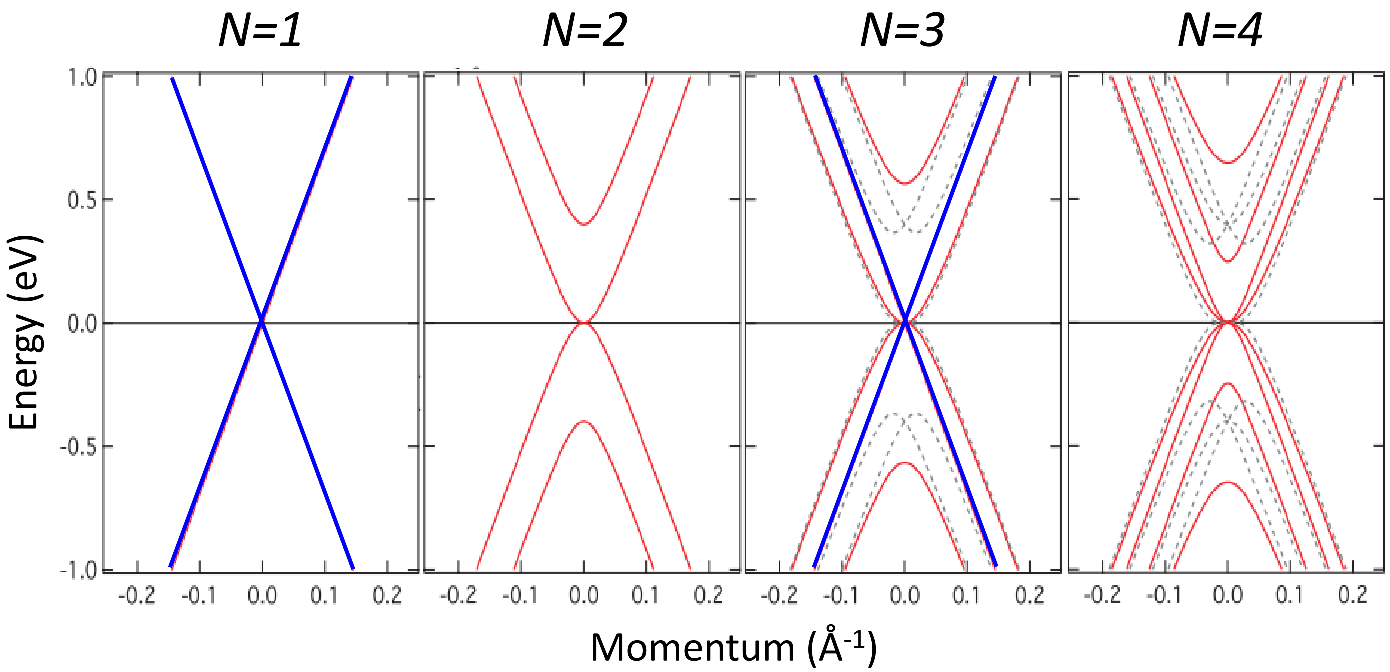




***Graphite**: Wallace (1947), McCluire (1957), Slonczewski & Weiss (1958), M.S. Dresselhaus & G. Dresselhaus (1965).*

***Few-layers**: McCann & Fal'ko PRL (2006), Latil et al. PRL (2006), Guinea et al. PRB (2006), Nilsson et al. PRL (2006), Partoens & Peeters PRB (2007), Koshino & Ando PRB (2007), …*

*Carriers move between atomic orbitals: **atomic registry/potential is important***



A. Bostwick et al., NJP 2007

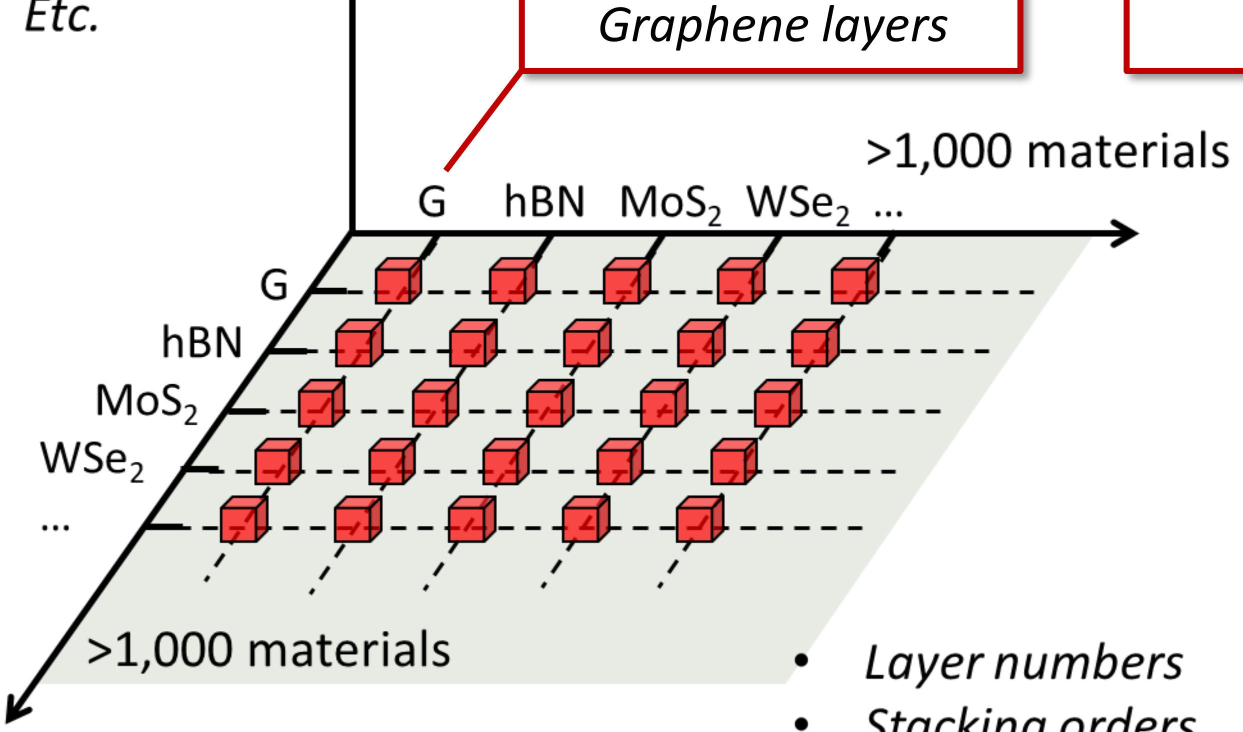
- *N=odd: **1 monolayer-like**, (N-1)/2 bilayer-like bands*
- *N=even: N/2 bilayer-like bands*

- *For ABC stacking,* $E \propto k^N$

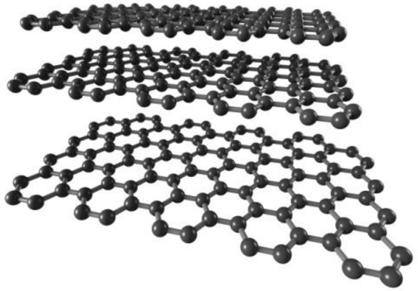
*Carriers move between atomic orbitals: **chemical composition changes the properties***

Vast number of choices

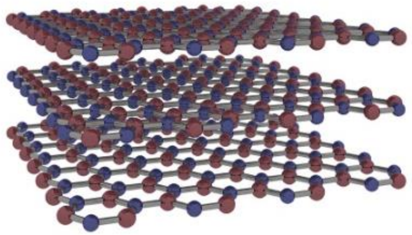
Moiré lattice
Twist angle
Strain
Pressure
Etc.



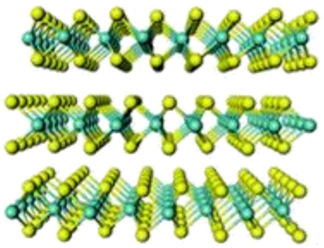
- *Layer numbers*
- *Stacking orders*
- *Dimensions*



Graphene layers

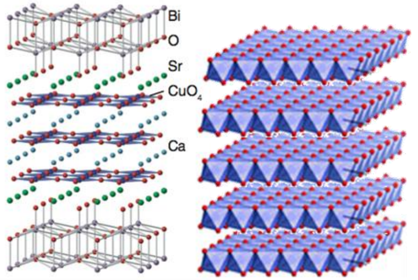


hBN layers



TDMCs (MX_2)

- *Semiconducting*
- *Semimetallic*
- *Superconducting*

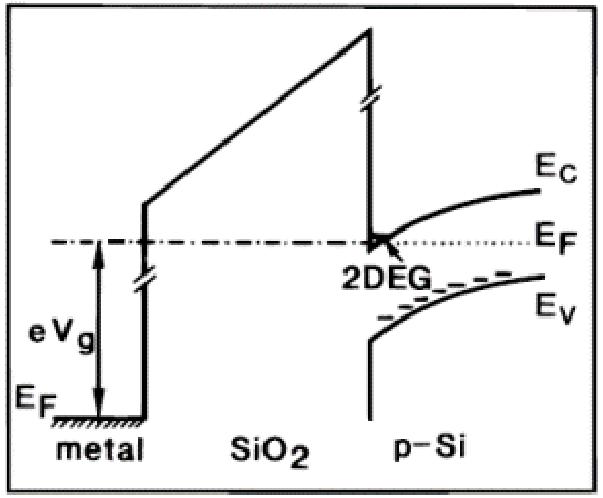


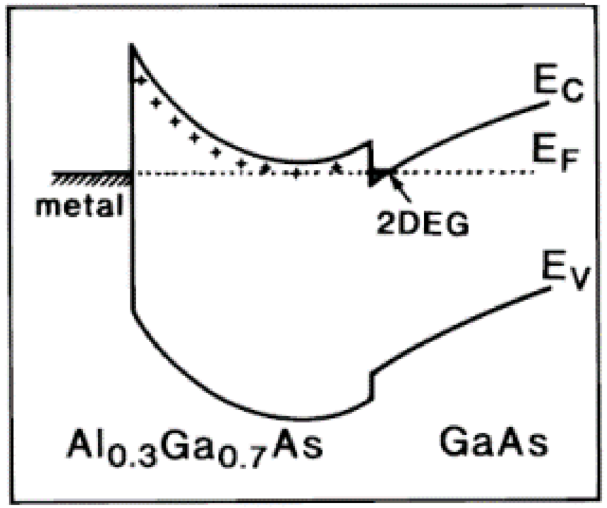
2D oxides

2D magnets (CrI3, FGT, etc.)

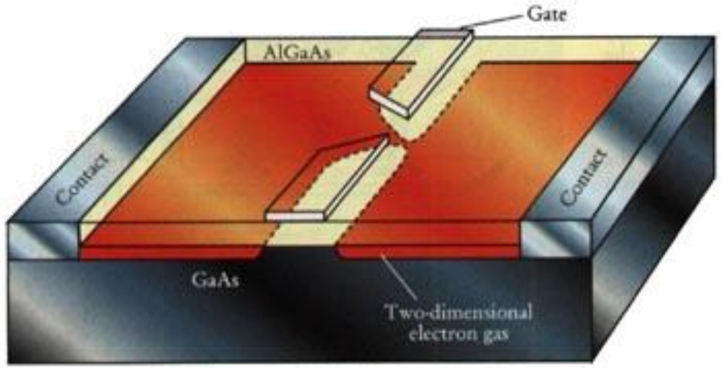
MX: layered semiconductors (GaSe, GaTe, InSe, …)
Bi_2Se_3: topological insulator
and others….

Comparison with conventional 2D electron gas





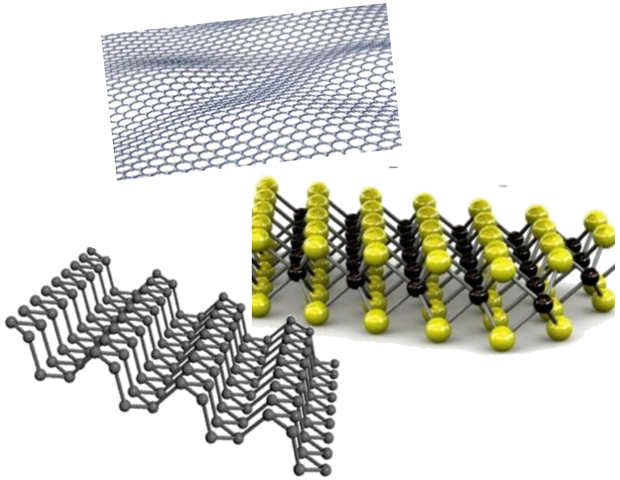
- *Excellent electrostatic control*



- *Very high electron mobility*
- *An excellent (nearly perfect) platform to study quantum behaviour of free electrons in low dimensions*

$$E_0 = \frac{p^2}{2m^*}$$

2D family



- *Diverse band structures and material properties*
- *Charge carriers move at the surface not at the interface*
- *Atomic registry/potential is important*
- *Weak van der Waals interactions*

True 2D nature promotes interactions with environment

Charge transport occurs at the ***atomically flat*** *surface*

Large experimental accessibility

- *Electrostatic gating*
- *Scanning probes*
- *Optical investigations,*

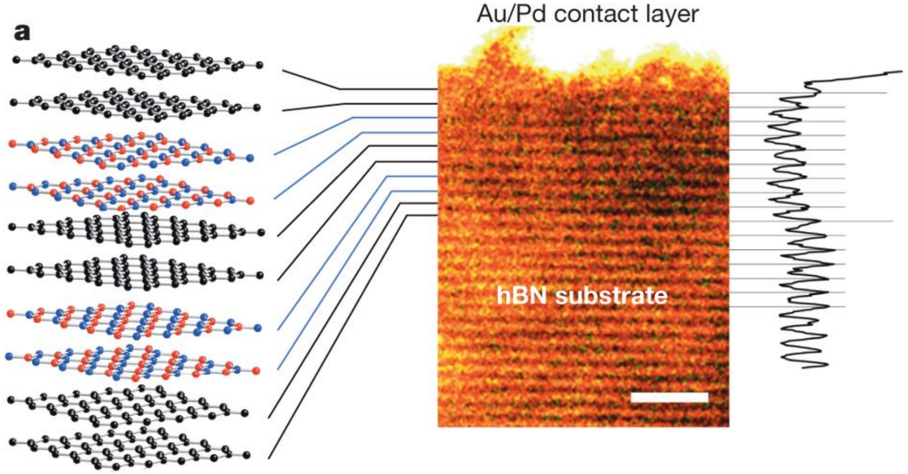
…

Easy contact engineering

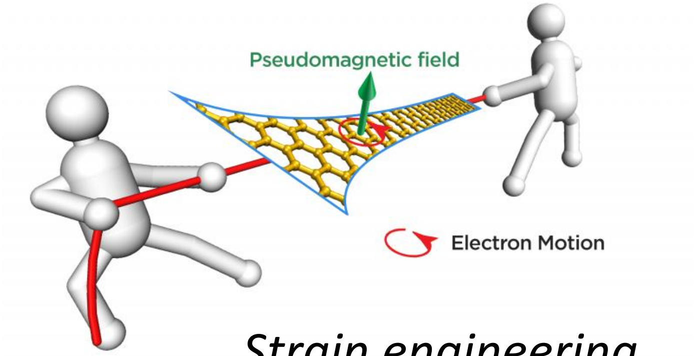
- *Superconducting contacts*
- *Magnetic contacts*
- *Other 2D crystals*

New functional heterostructures

(without suffering from the lattice mismatch, strain, etc)



Atomically thin = Flexible



Strain engineering
Folding—Unfolding

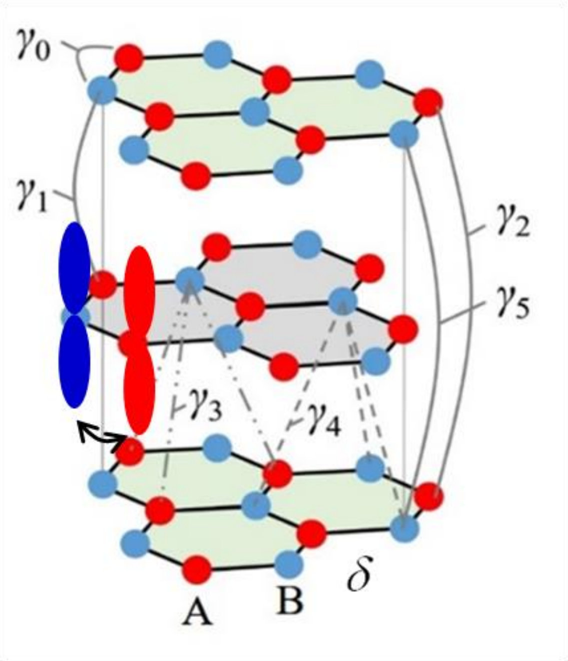
Chemical or surface engineering

***Flatlands 2D crystals**:*
a remarkable 2D platform with large experimental advantages

Contents

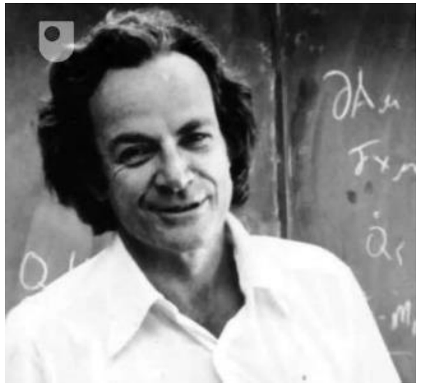
- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

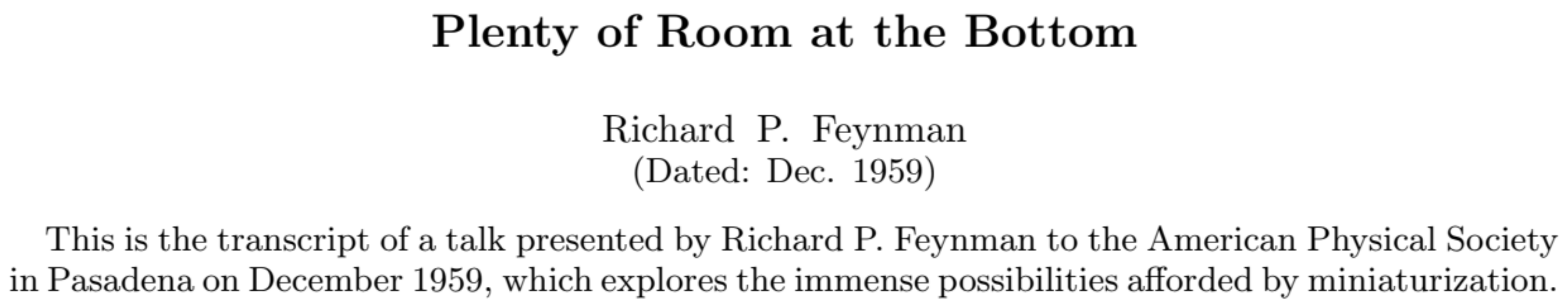
van der Waals coupled layered materials → defect-free monolayers



- *Carriers move between atomic orbitals:* ***atomic registry/potential is important (chemical composition)***
- *Carriers move at the atomically flat surface:* ***highly sensitive to the environment***
- *Surface is defect free in principle:* ***no dangling bonds and strong vdW interactions***

Van der Waals assembly technique





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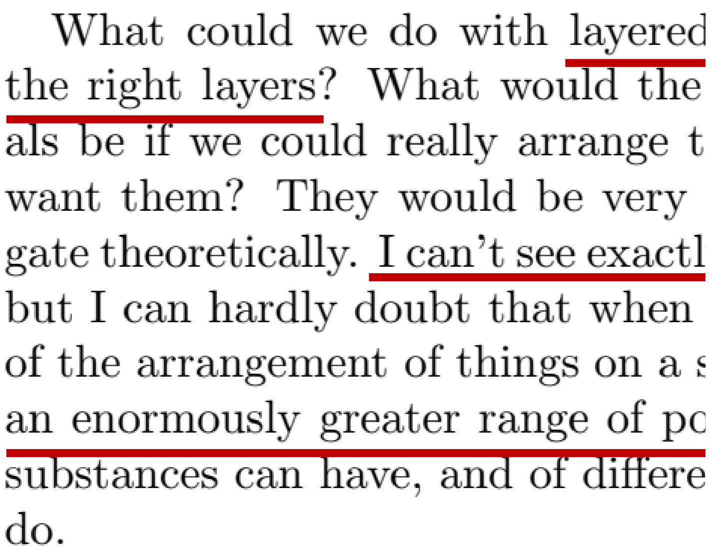

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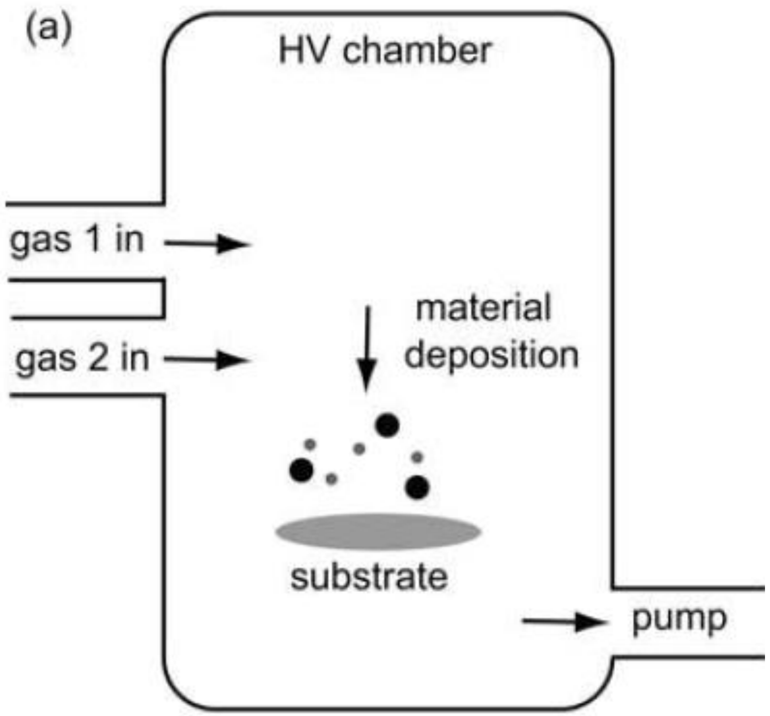
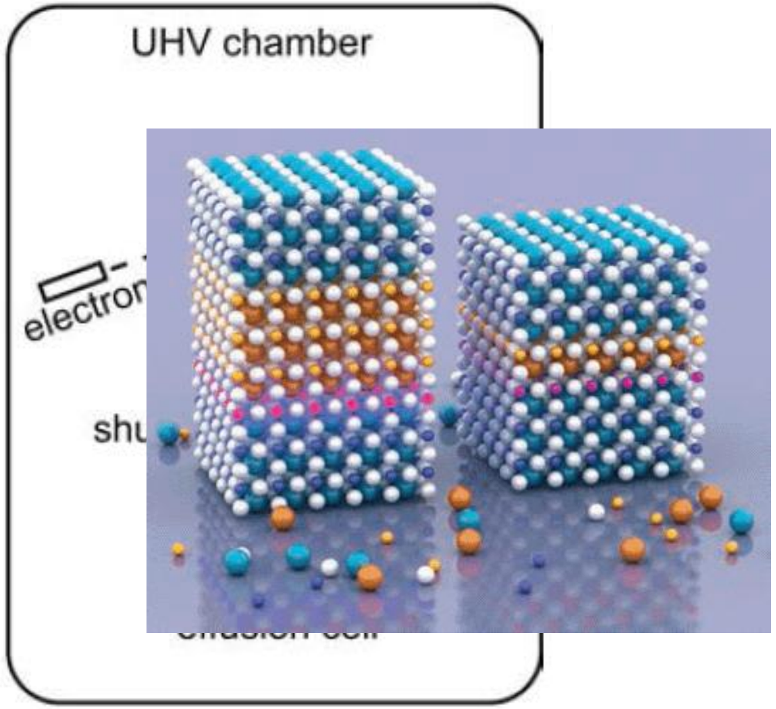
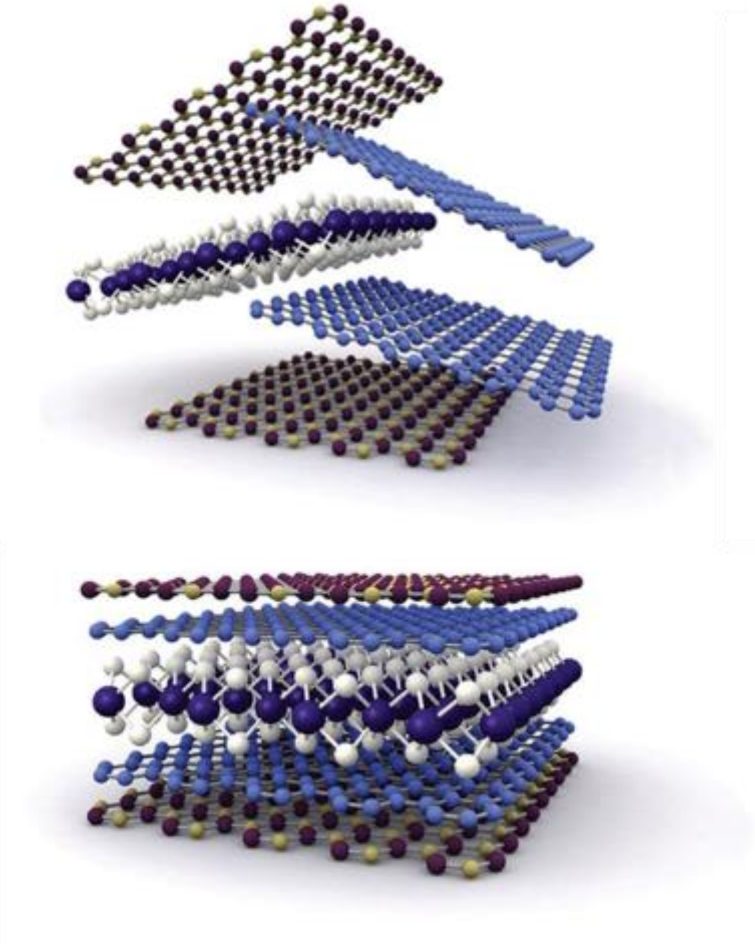
MOCVD

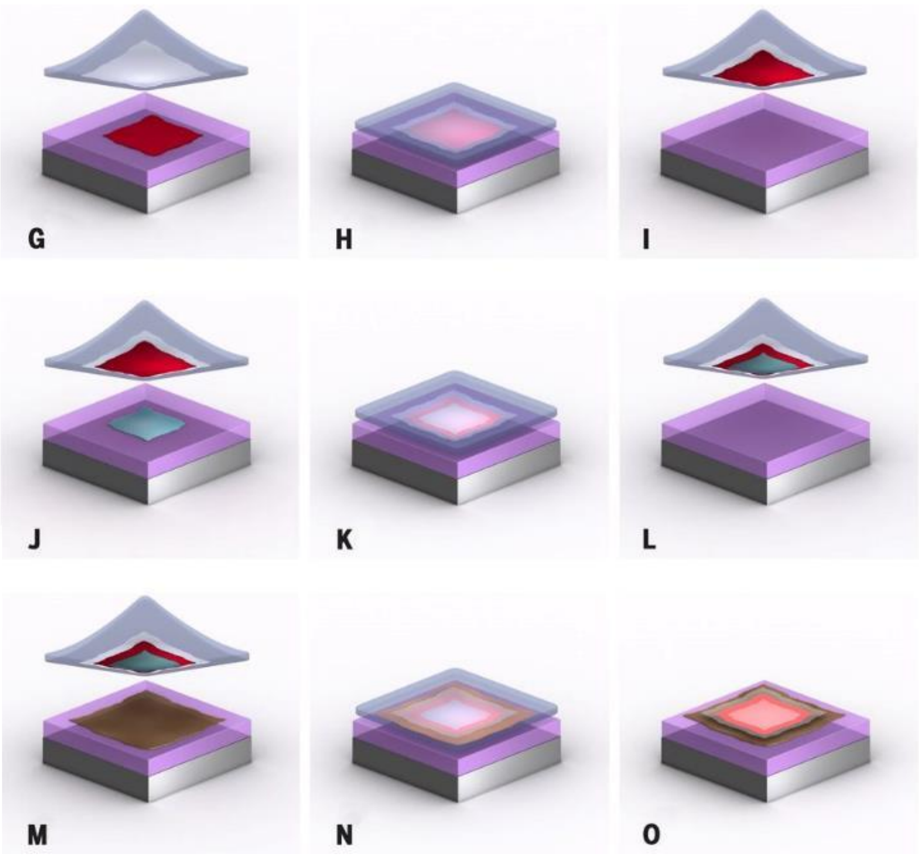


MBE

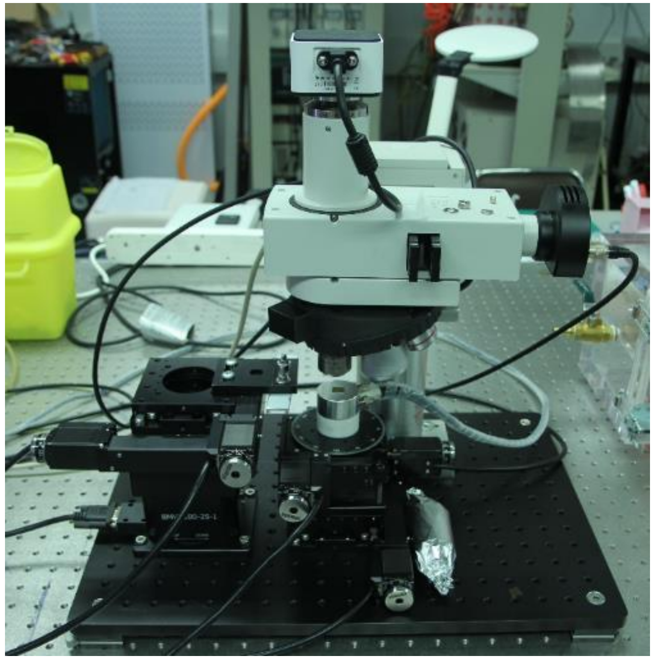


Van der Waals assembly technique



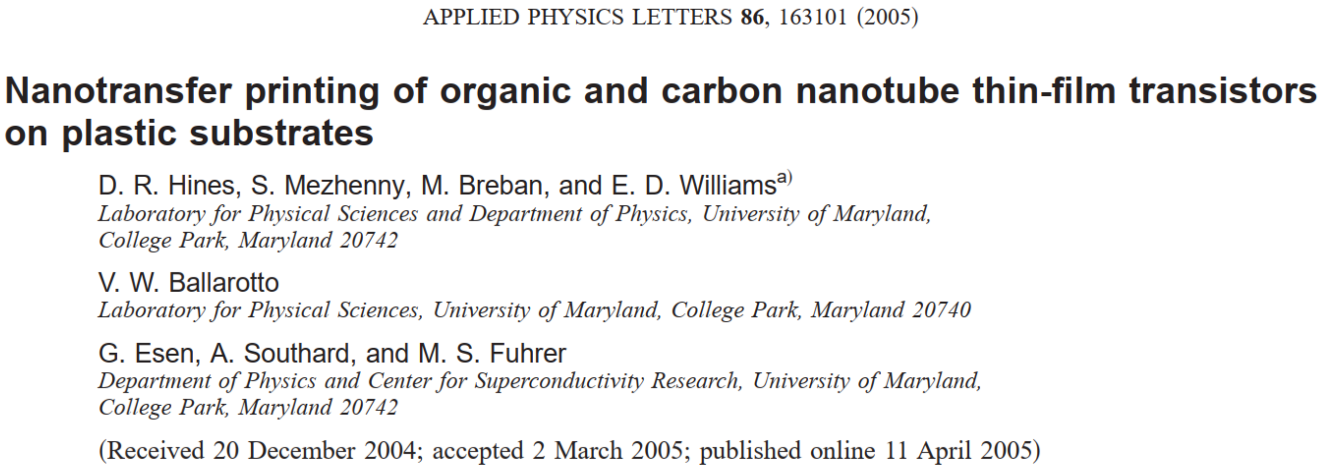


Science **353**, aac9439 (2016)



- Cheap
- Clean interface
- Less concern on lattice mismatch

Van der Waals assembly technique



APPLIED PHYSICS LETTERS **86**, 163101 (2005)

Nanotransfer printing of organic and carbon nanotube thin-film transistors on plastic substrates

D. R. Hines, S. Mezhenny, M. Breban, and E. D. Williams[a)]
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College Park, Maryland 20742

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Laboratory for Physical Sciences, University of Maryland, College Park, Maryland 20740

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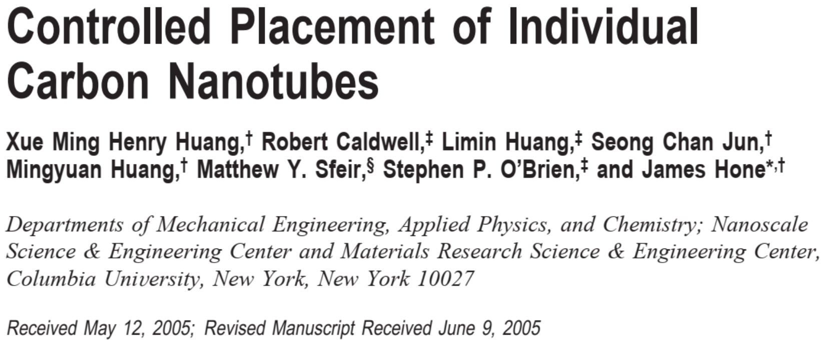
(Received 20 December 2004; accepted 2 March 2005; published online 11 April 2005)



Controlled Placement of Individual Carbon Nanotubes

Xue Ming Henry Huang,[†] Robert Caldwell,[‡] Limin Huang,[‡] Seong Chan Jun,[†] Mingyuan Huang,[†] Matthew Y. Sfeir,[§] Stephen P. O'Brien,[‡] and James Hone[*,†]

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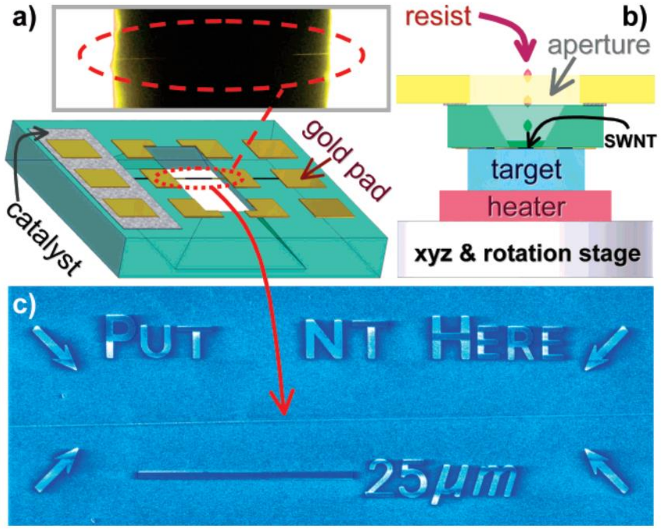
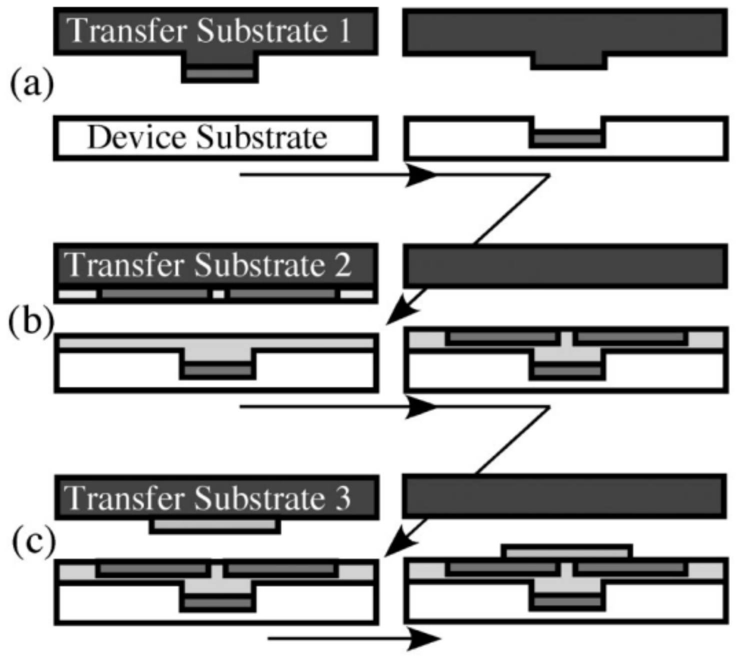
Received May 12, 2005; Revised Manuscript Received June 9, 2005



Rule of thumb
Higher T → Enhance adhesion



- *Transfer medium (polymer)*
- *Manipulators & Heaters*

$$E_{Ad}^{A-C} > E_{Ad}^{A-B}$$

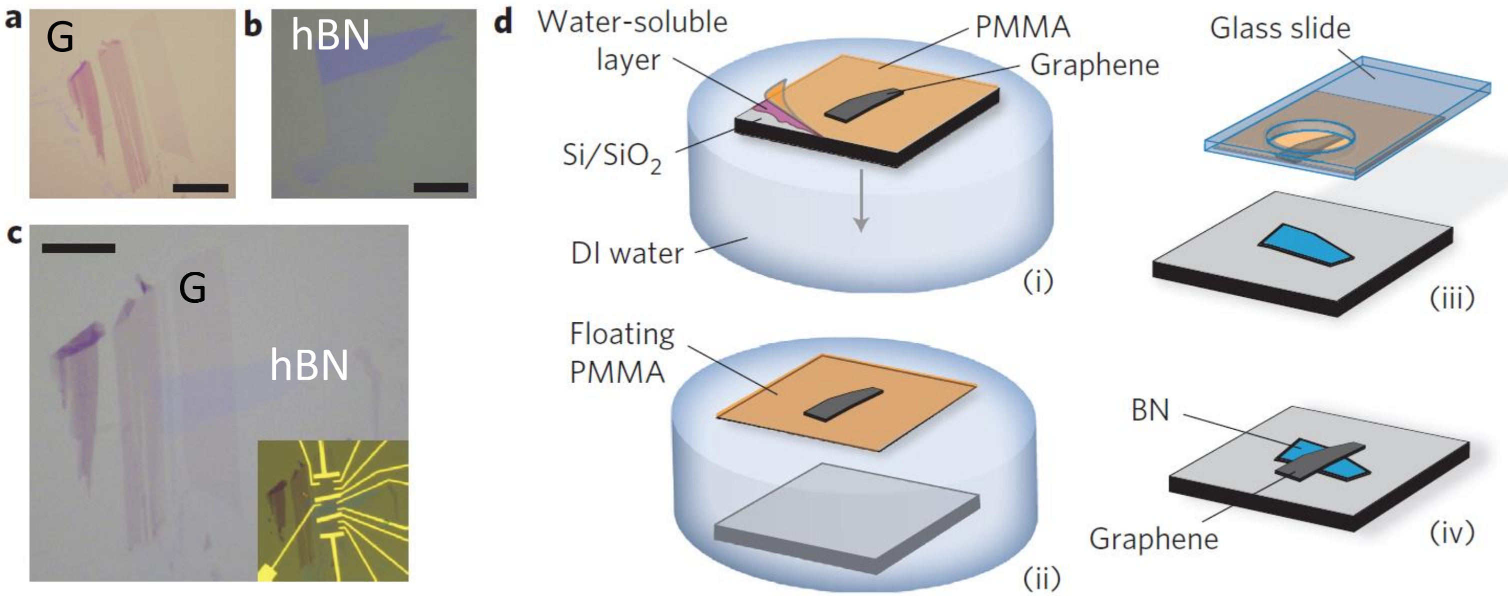
$$E_{Ad}^{A-B} = \gamma_A + \gamma_B - \gamma_{AB}$$

Surface free E *Interface free E*

Van der Waals assembly technique

Transfer technique

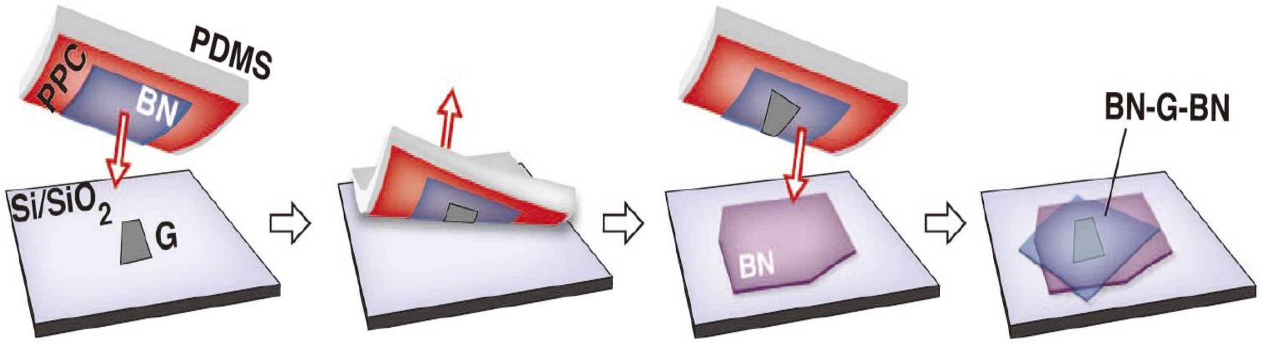
C.R. Dean et al., Nat. Nanotech. 2010

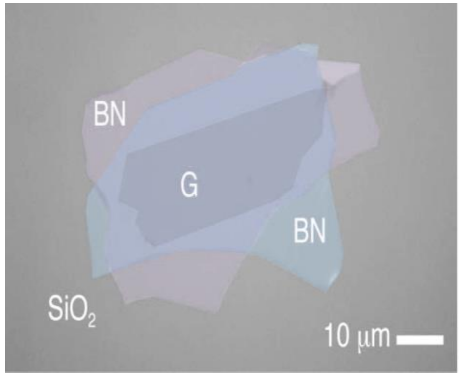


- *Exfoliate materials on transfer medium (PMMA) on top of water soluble polymer*
- *The material touches PMMA*
- *Heater used to control* $E_{Ad}^{G-hBN} > E_{Ad}^{G-PMMA}$

Pick-up technique

L. Wang et al., Science 2013



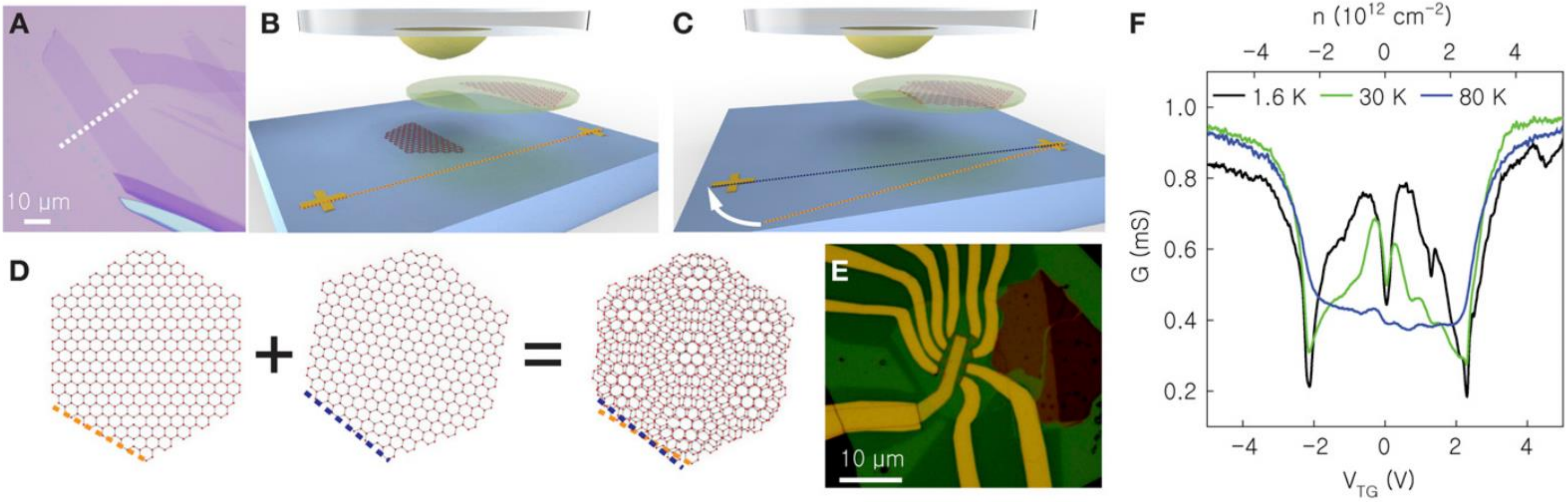


- *Exfoliate materials on substrate: easier to identify*
- *The material never touches chemicals*
- *Heater used to control*

$$E_{Ad}^{G-SiO2} < E_{Ad}^{G-hBN}, E_{Ad}^{hBN-PPC}$$

Tear-and-stack method

Nano Lett. 16, 1989 (2016), PNAS 114, 3364 (2017)

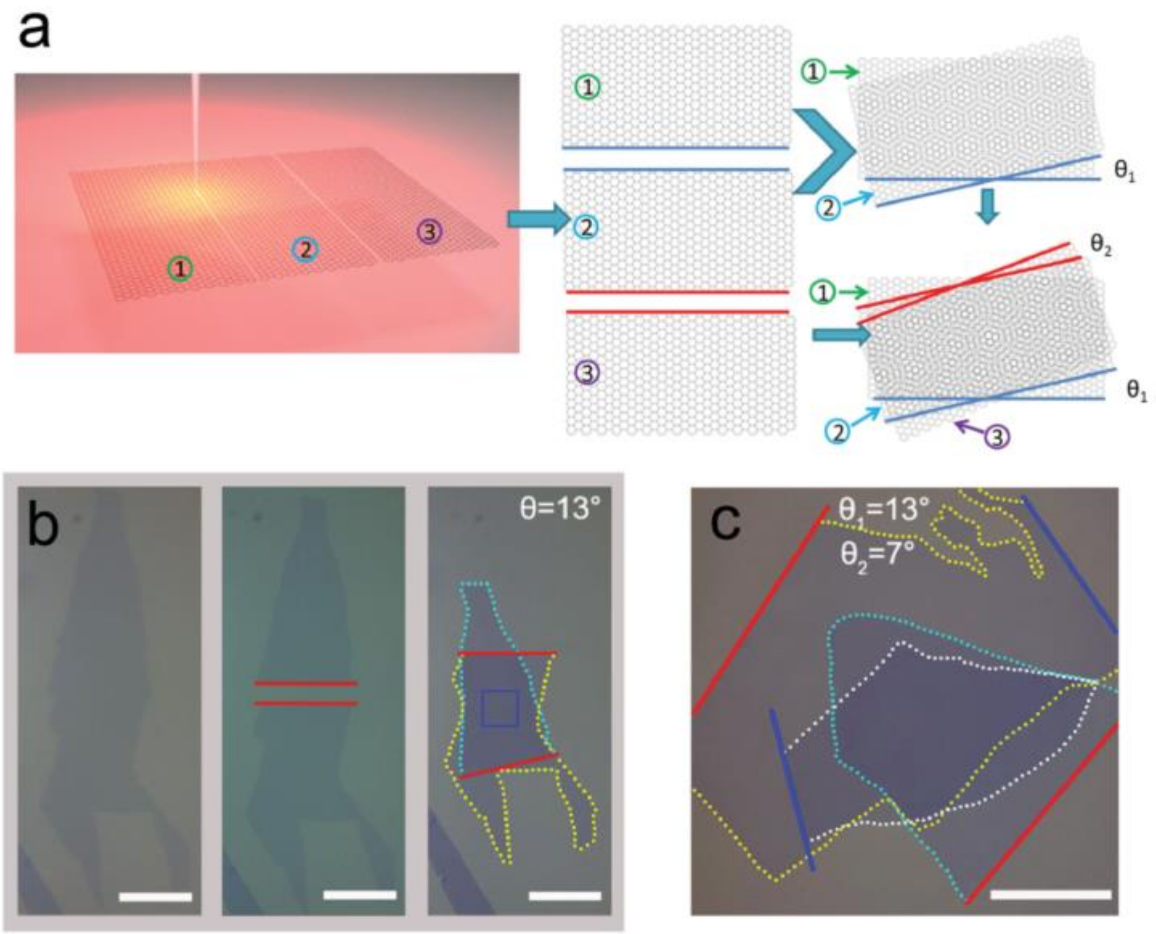


$$E_{Ad}^{G-hBN} > E_{Ad}^{G-PC} \qquad E_{Bonding}^{G-G}?$$

- *Great method to fine-tune the twist angle*
- *May not work for other 2D materials depending on their bonding strength*
- *Tearing doesn't always work*
- *Can be some rotation of the layer on the substrate due to tearing*

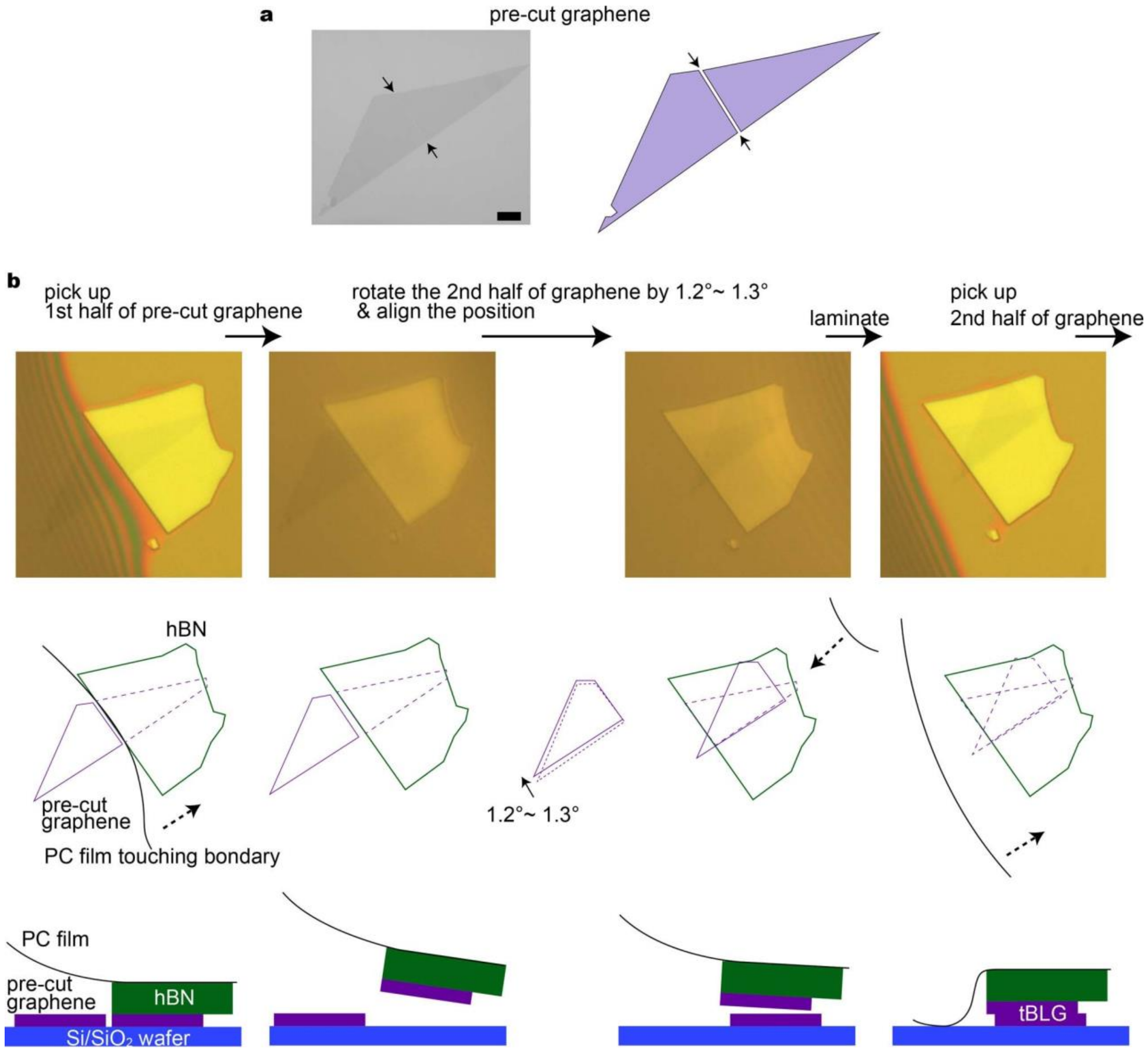
Cut-and-stack method

Laser *Adv. Mater.* ***28****, 2563 (2016)*

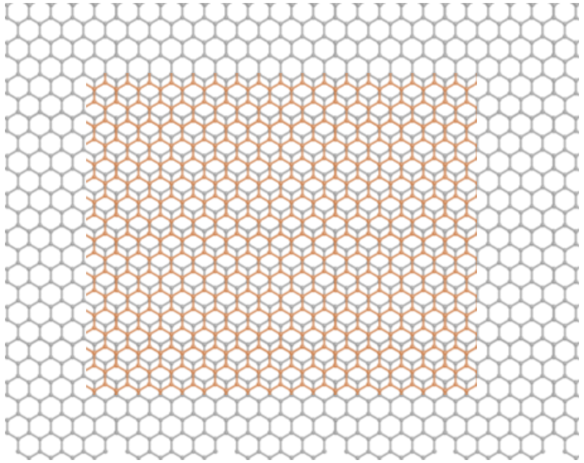


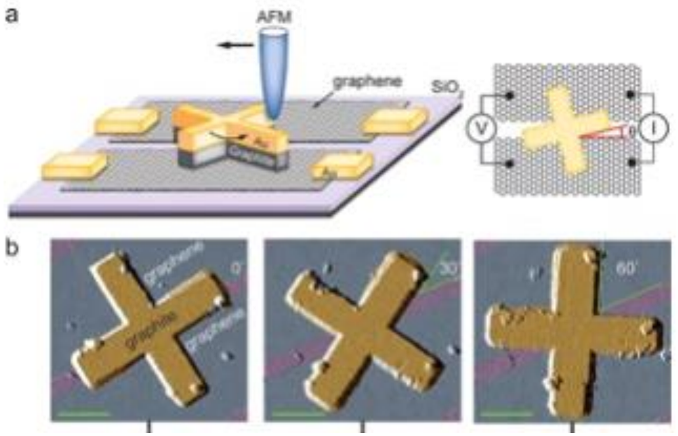
- *Better control but expensive*

AFM tip *Nat. Phys.* ***16****, 926 (2020)*

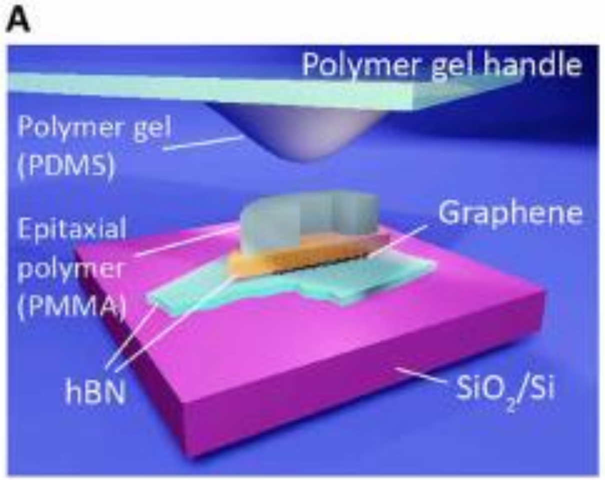


Post processing methods





Nano Lett. **16**, 4477 (2016)



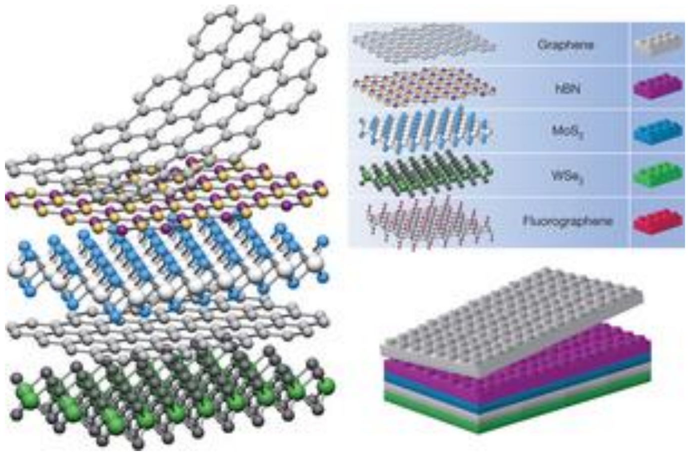
Sci. Adv. **6**, eabd3655 (2020)

- *Transfer techniques for large area 2D materials*
- *Controlled folding*
- *Direct growth of vdW heterostructures*

Contents

- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

- *Introduction to 2D materials*
- *Their special properties compared with other 2D electron gas systems*
- *Van der Waals assembly techniques*



van der Waals coupled layered materials
→ *defect-free monolayers*

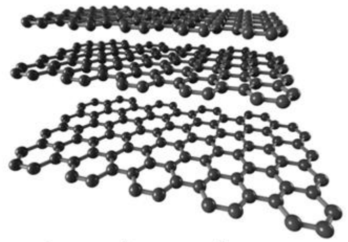
- *Carriers move between atomic orbitals: **atomic registry/potential is important (chemical composition)***
- *Carriers move at the atomically flat surface: **highly sensitive to the environment***
- *Surface is defect free in principle: **no dangling bonds and strong vdW interactions***

Various techniques are developed to have a better control on vdW assembly

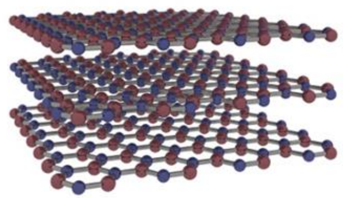
Contents

- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

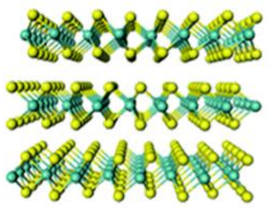
- *Few examples of new properties*
- *Discussions on the mechanisms*
- *Future challenges and directions?*



Graphene layers

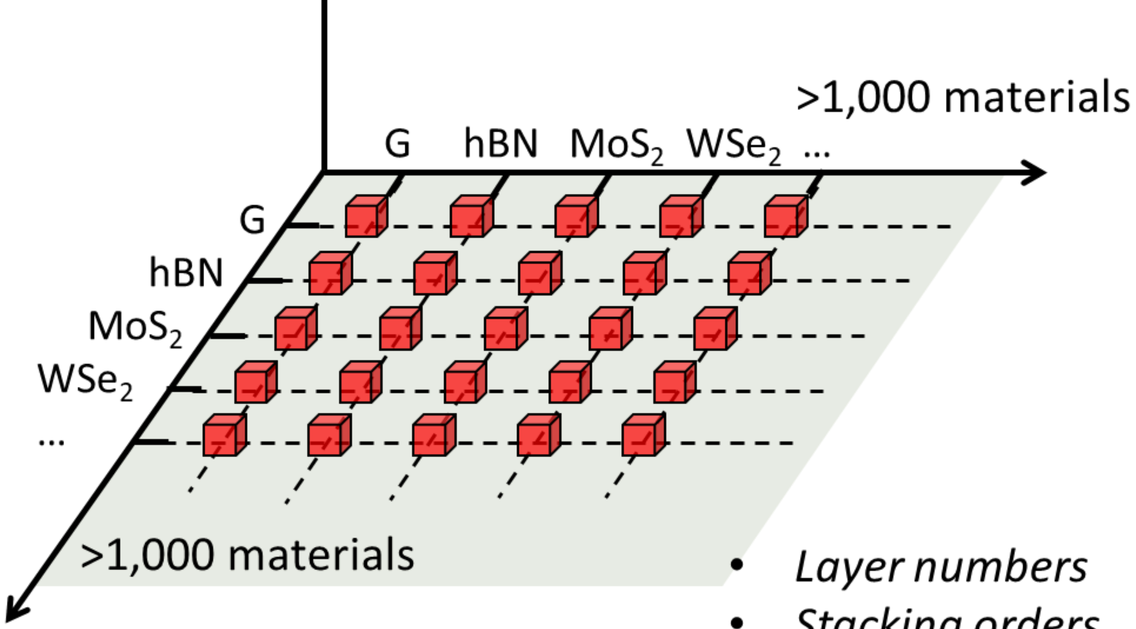


hBN layers

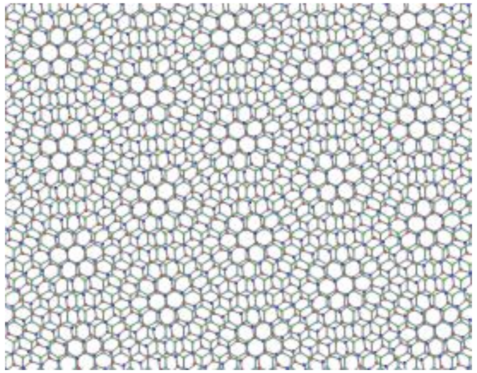


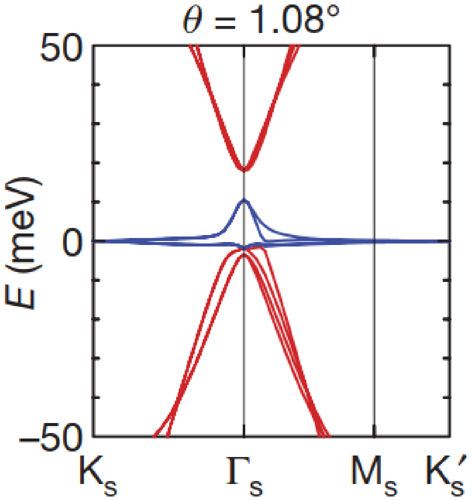
TDMCs (MX$_2$)

Etc.

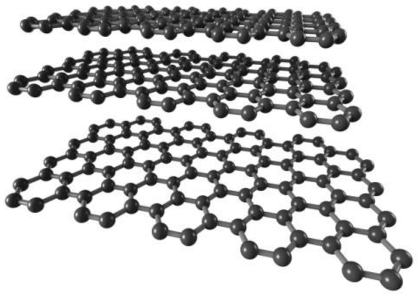


- *Layer numbers*
- *Stacking orders*
- *Dimensions*



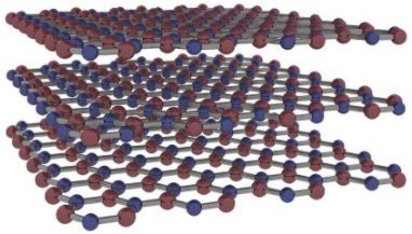


Graphene-hBN heterostructures: high-quality graphene samples



Graphene layers

- Semimetal
- Massless or massive electrons



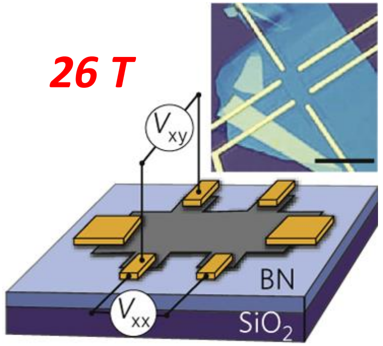
hBN layers

- Insulator ~ 5 eV
- Chemically inert

Quantum correlations

Fractional QHE

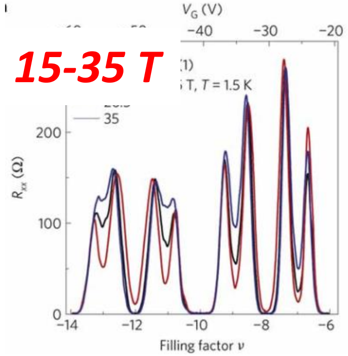
26 T



Kim 2011

Symmetry broken QHE

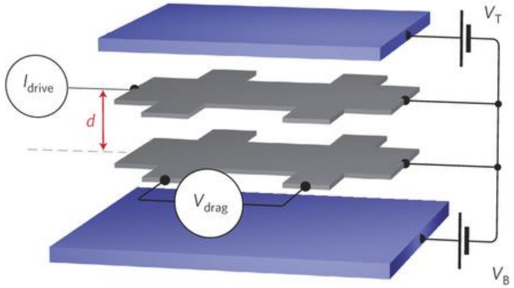
15-35 T



Kim 2012

Classical correlations

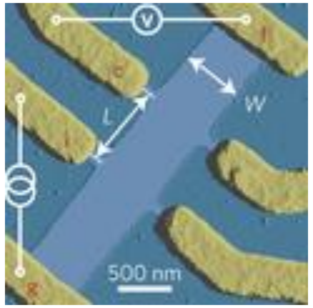
Coulomb drag



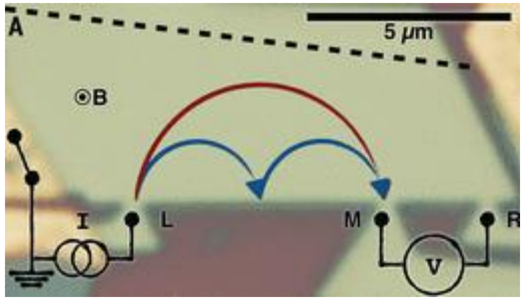
Geim 2012

Ballistic electron optics

Magnetic focusing

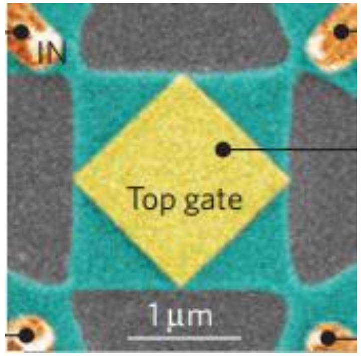


Pablo 2013



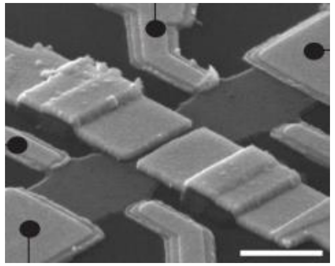
D Goldhaber-Gordon 2016

Negative index



Hu-Jong Lee 2015

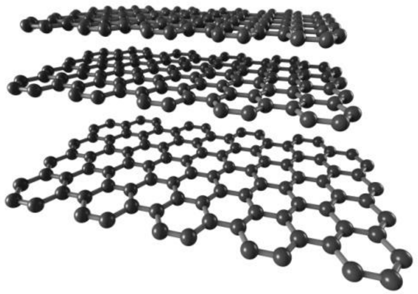
Guiding



Hu-Jong Lee 2016

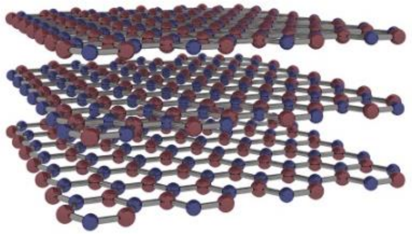
and more …

Graphene-hBN heterostructures: moiré structure



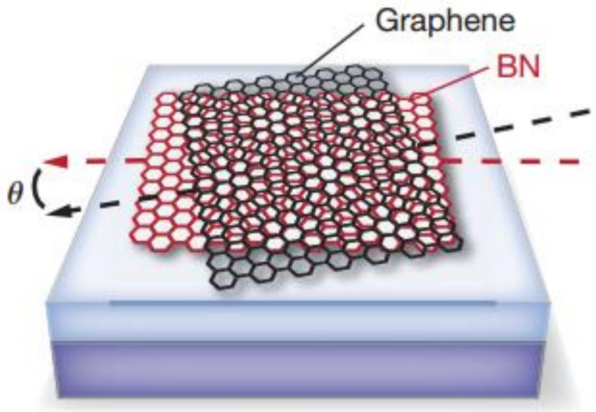
Graphene layers

- Semimetal
- Massless or massive electrons

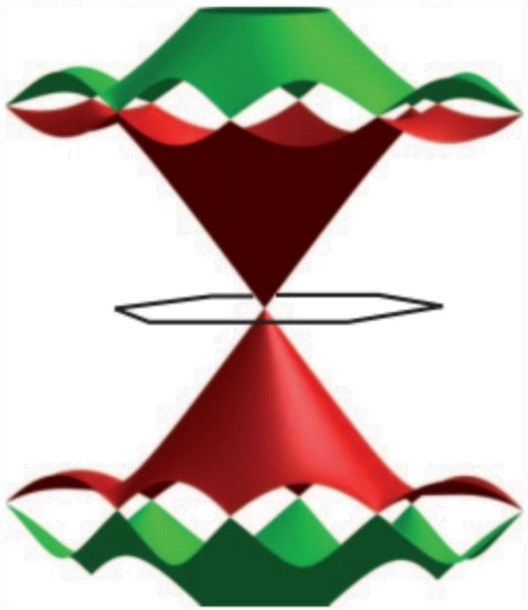


hBN layers

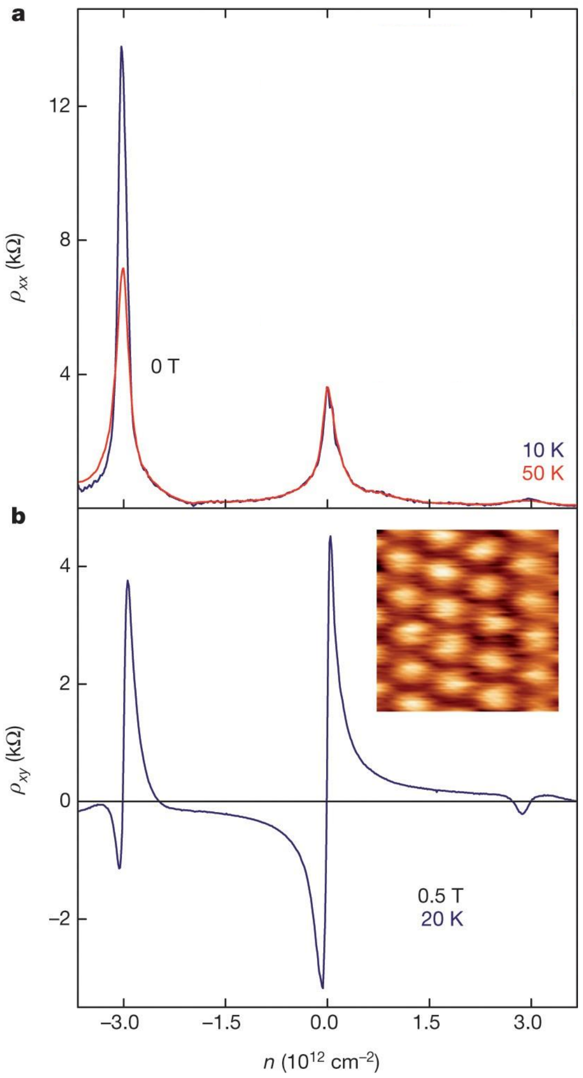
- Insulator ~ 5 eV
- Chemically inert

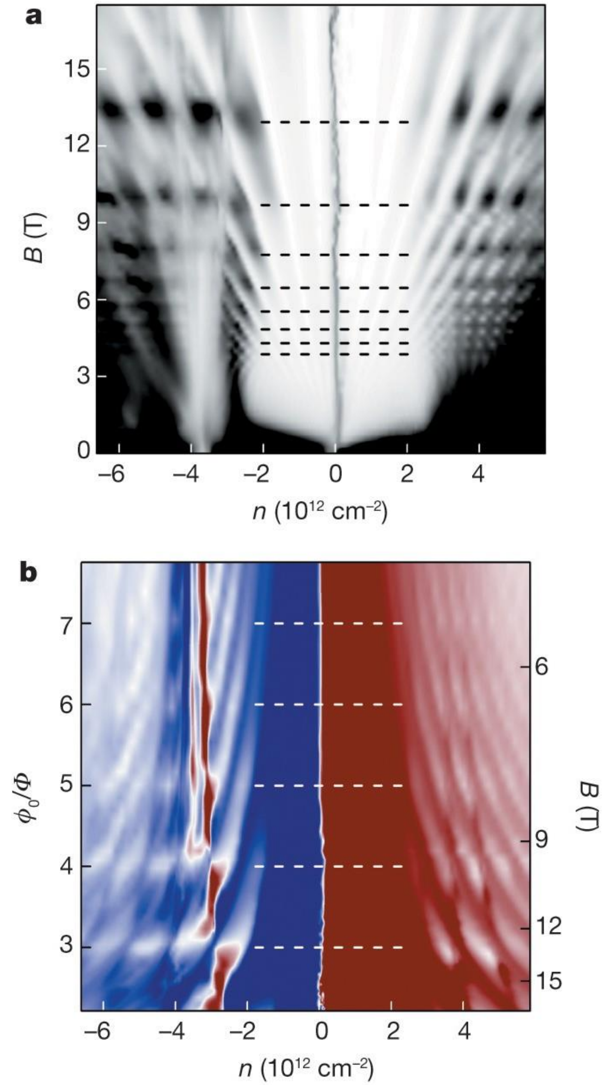


Moiré Miniband

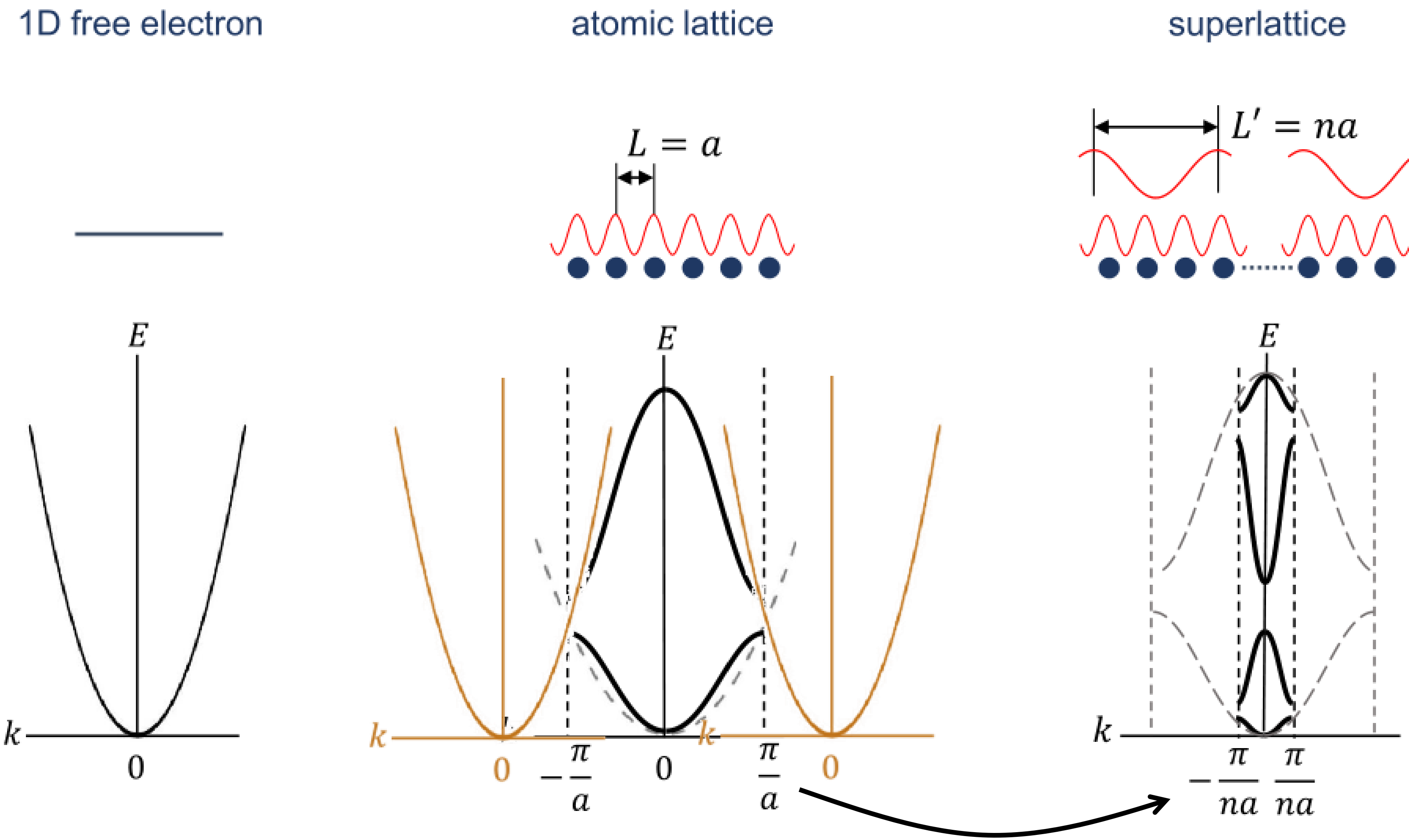


Nat. Phys. 8, 382 (2012); Nature 497, 594; Nature 497, 598; Science 340, 1427 (2013)





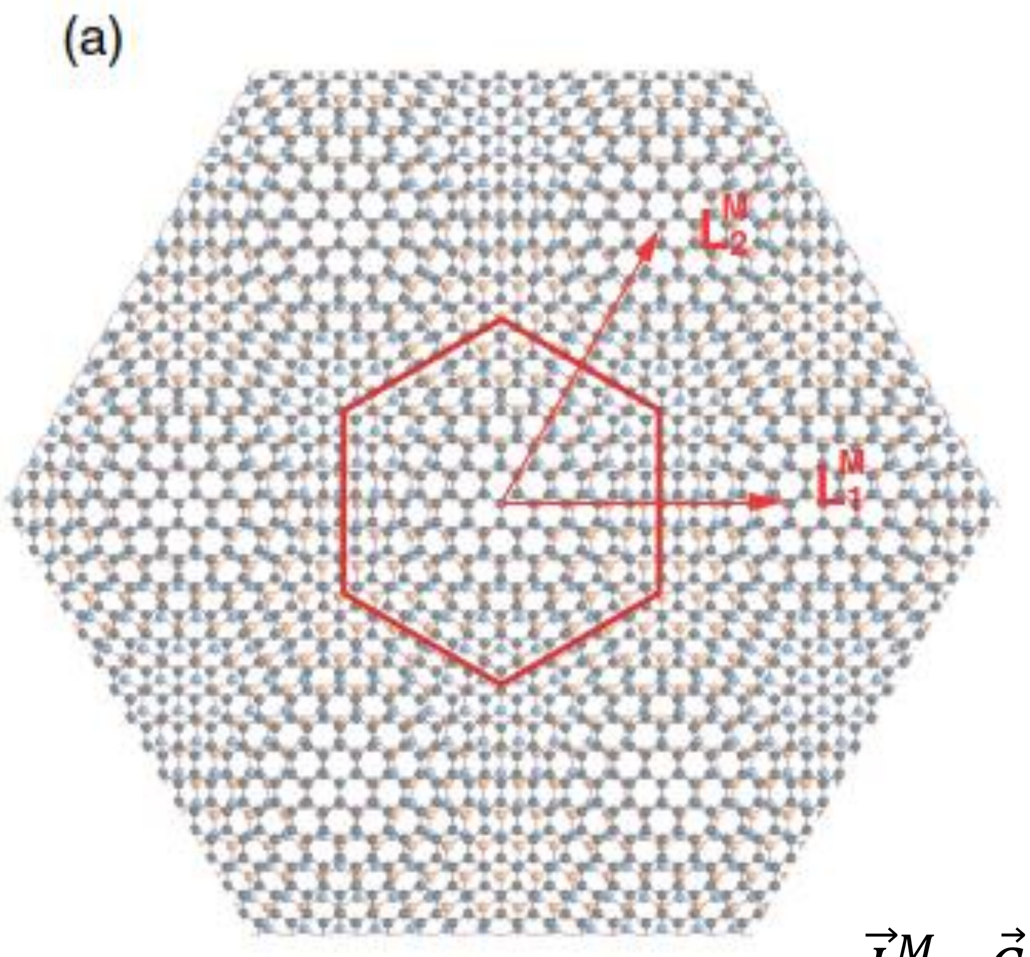
Graphene-hBN heterostructures: moiré structure



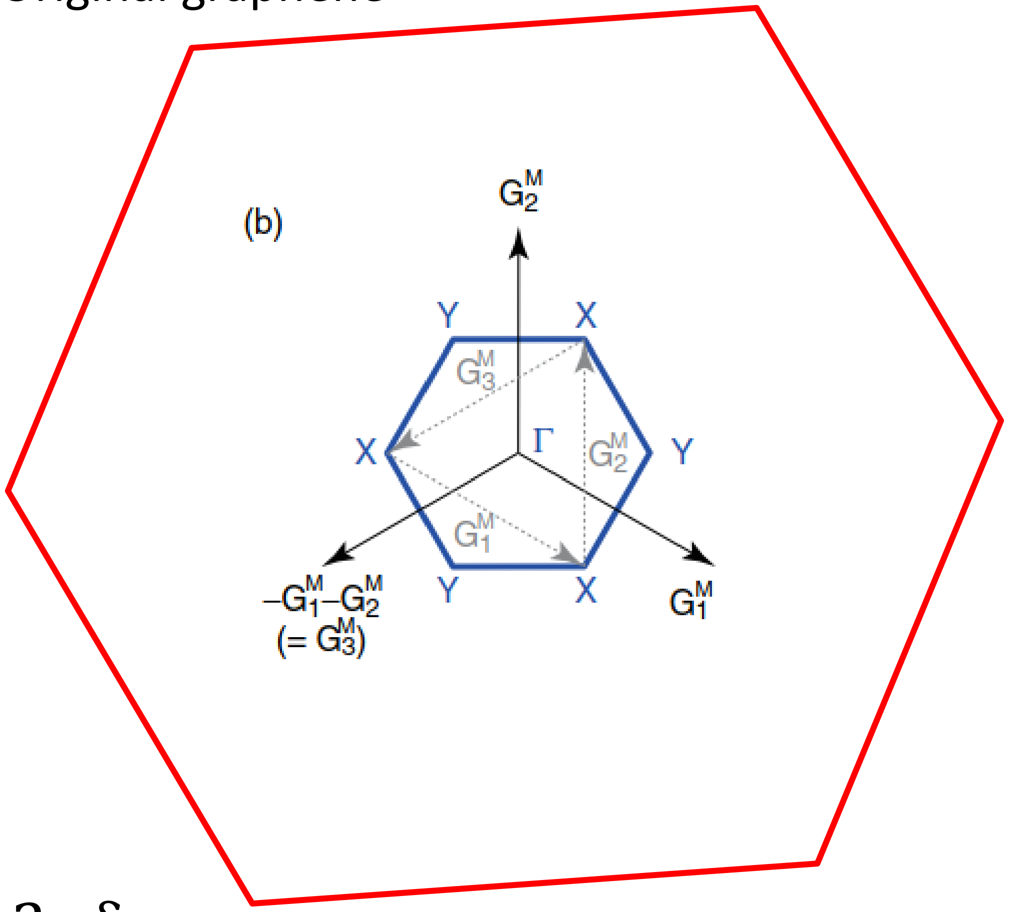
The first Brillouin zone (BZ)

Quantum Frontiers 1, 8 (2022)

Graphene-hBN heterostructures: moiré structure



Original graphene



$$\vec{L}_i^M \cdot \vec{G}_j^M = 2\pi\delta_{ij}$$

PRB 90, 155406 (2014)

Graphene-hBN heterostructures: moiré structure

$$H_{G-hBN} = \begin{pmatrix} H_G & U^\dagger \\ U & H_{hBN} \end{pmatrix} \to H_G + V_{hBN}$$

$$V_{hBN} = V(\vec{r}) + M(\vec{r})\sigma_z + ev\vec{A}(\vec{r})\cdot\vec{\sigma}_\xi$$

$$\xi = \pm 1$$

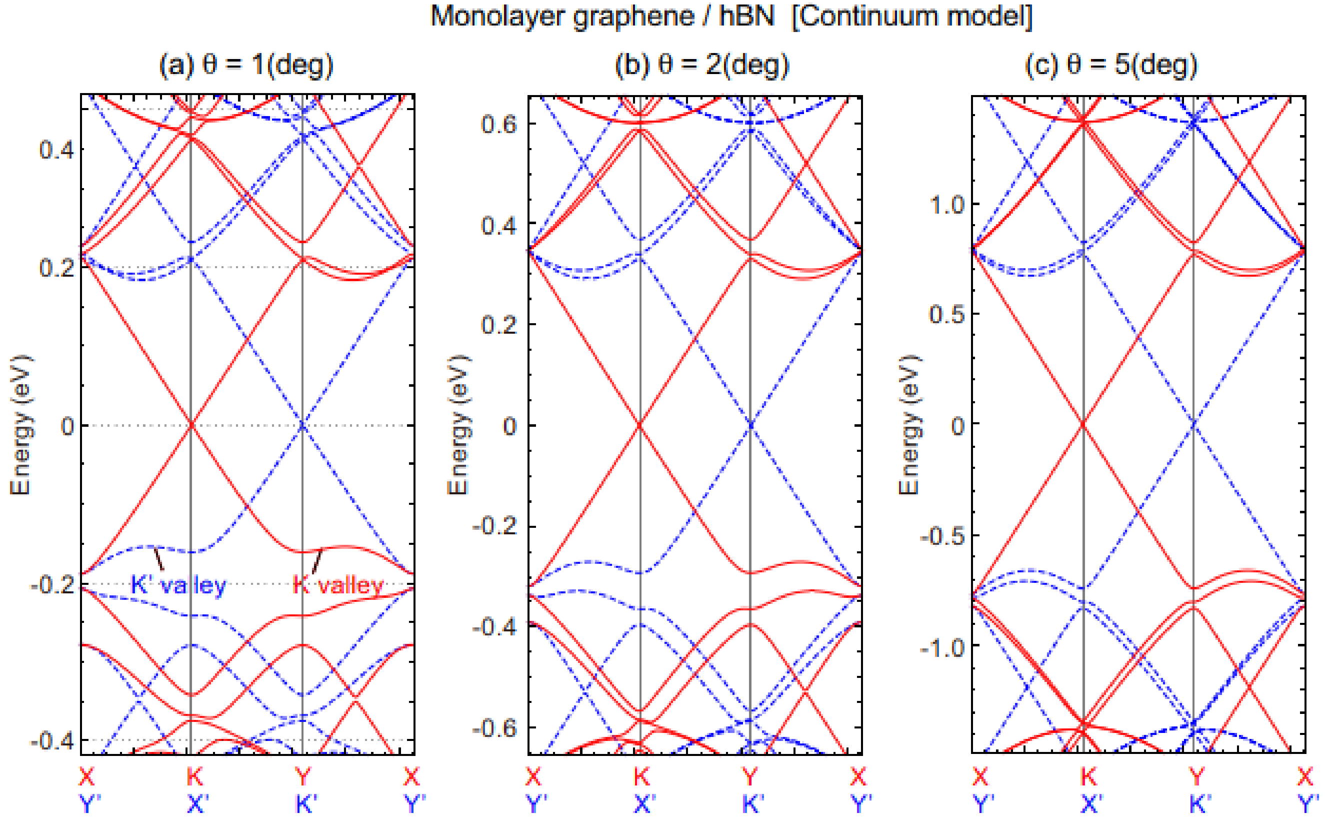
$$V^{\mathrm{eff}}(\mathbf{r}) = V_0 - V_1 \sum_{l=1}^{3} \cos\alpha_l(\mathbf{r})$$

$$M^{\mathrm{eff}}(\mathbf{r}) = \sqrt{3}V_1 \sum_{l=1}^{3} \sin\alpha_l(\mathbf{r}) \qquad (20)$$

$$ev\mathbf{A}^{\mathrm{eff}}(\mathbf{r}) = 2\xi V_1 \sum_{l=1}^{3} \begin{pmatrix} \cos[2\pi(l+1)/3] \\ \sin[2\pi(l+1)/3] \end{pmatrix} \cos\alpha_l(\mathbf{r})$$

$$V_0 = -3u_0^2\left(\frac{1}{V_{\mathrm{N}}} + \frac{1}{V_{\mathrm{B}}}\right), \qquad \alpha_l(\mathbf{r}) = \mathbf{G}_l^{\mathrm{M}}\cdot\mathbf{r} + \psi + \frac{2\pi}{3},$$

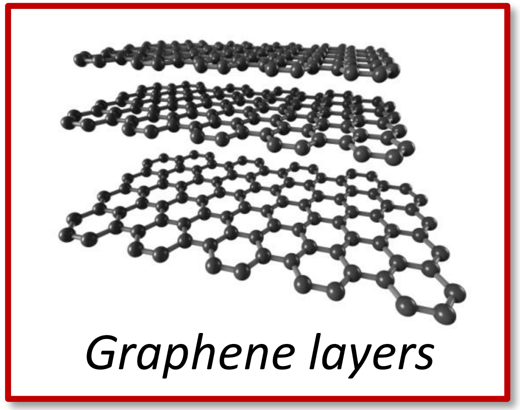
$$V_1 e^{i\psi} = -u_0^2\left(\frac{1}{V_{\mathrm{N}}} + \omega\frac{1}{V_{\mathrm{B}}}\right). \qquad \omega = e^{2\pi i/3}$$



- *Charge neutrality points at higher energies*
- *Electron-hole asymmetry*
- *Inversion symmetry is broken*

PRB 90, 155406 (2014)

Graphene-hBN heterostructures: moiré structure



Graphene layers

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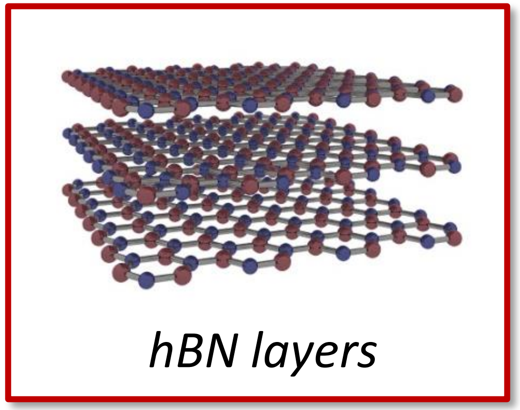
- Massless or massive electrons



hBN layers

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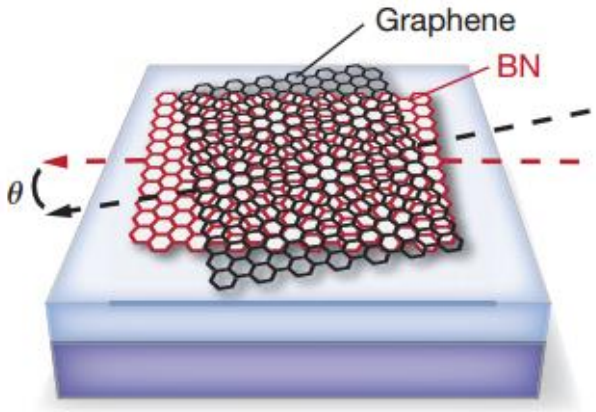

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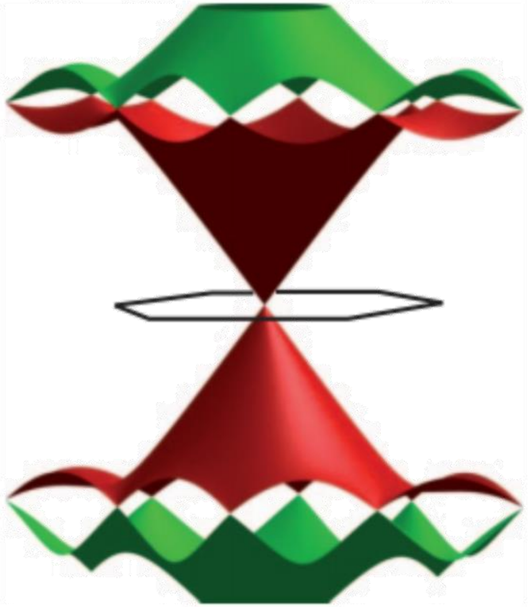

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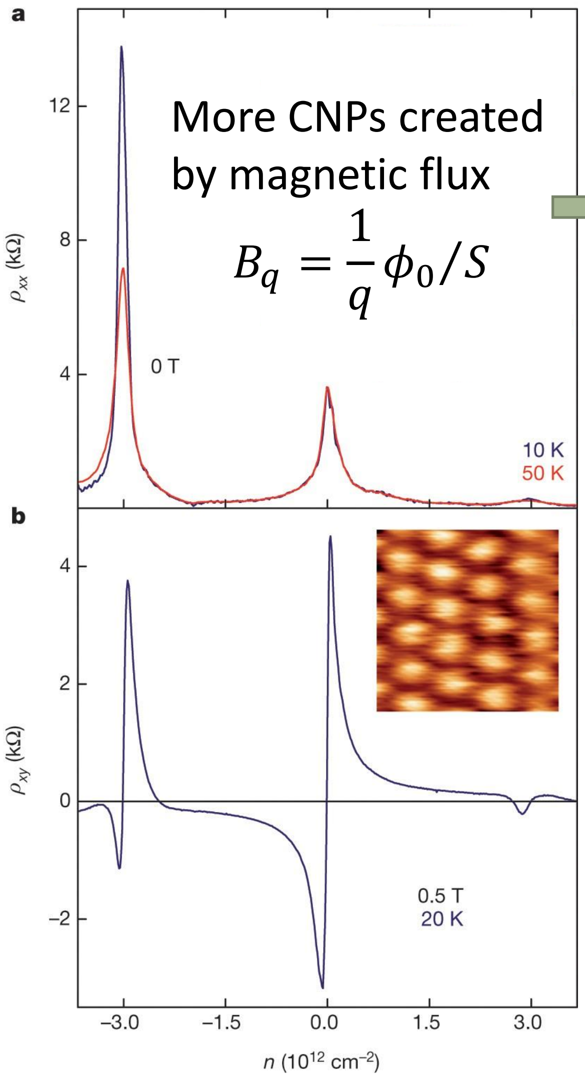
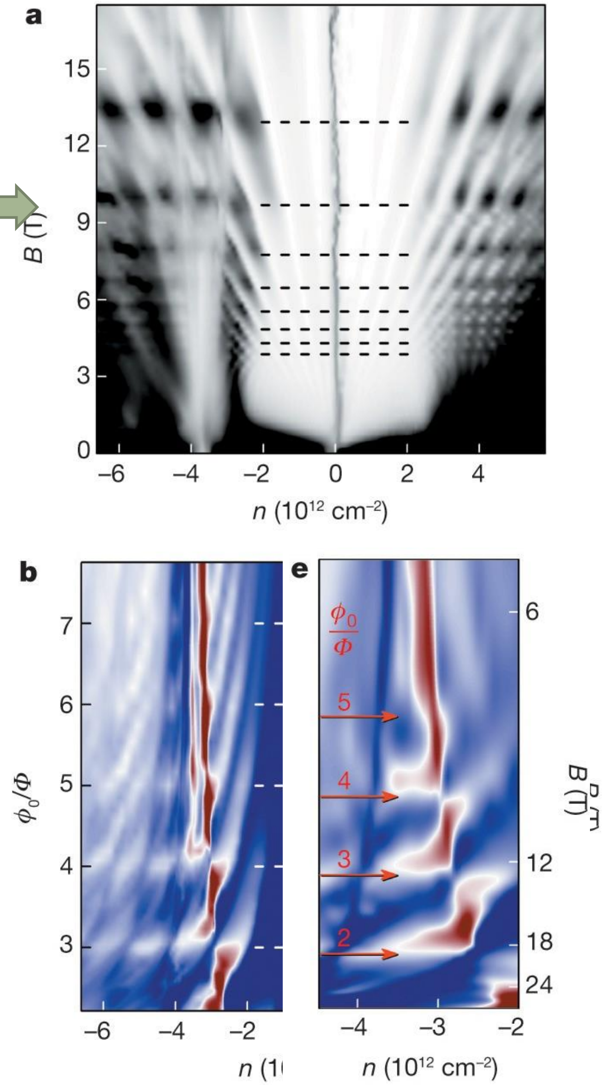
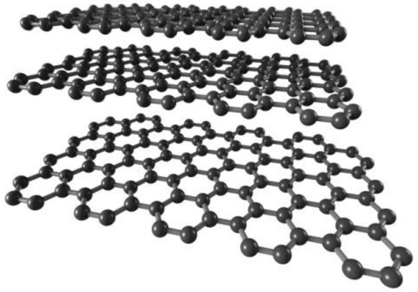


More CNPs created by magnetic flux

$$B_q = \frac{1}{q}\phi_0/S$$



Graphene-Graphene

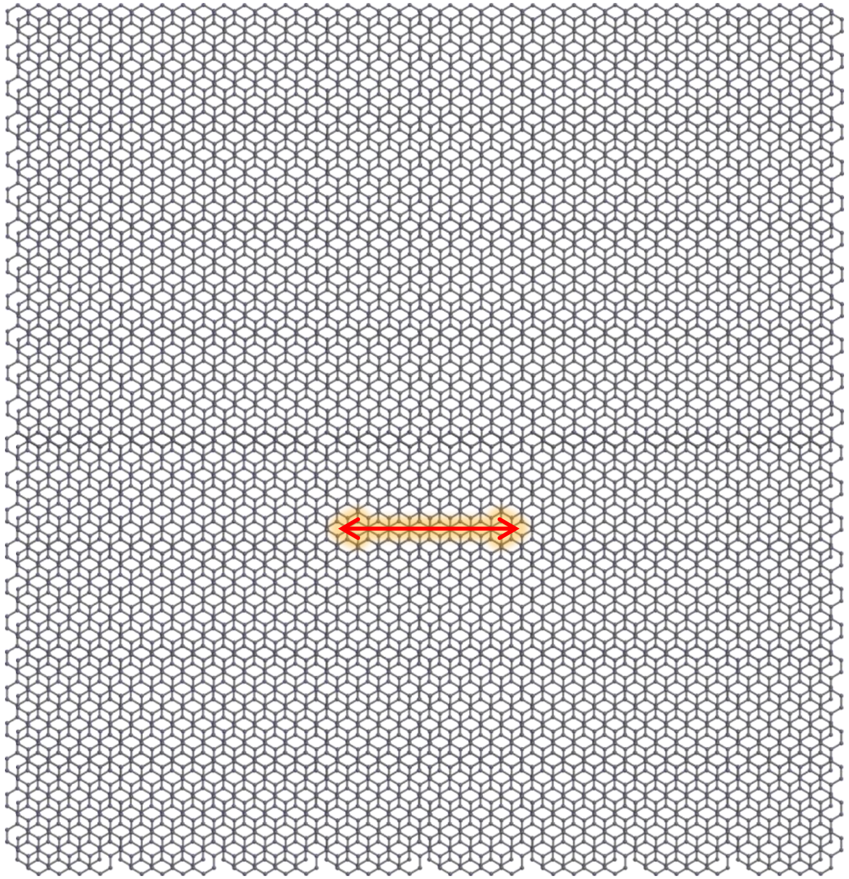


Graphene layers

- *Semimetal*
- *Massless or massive electrons*

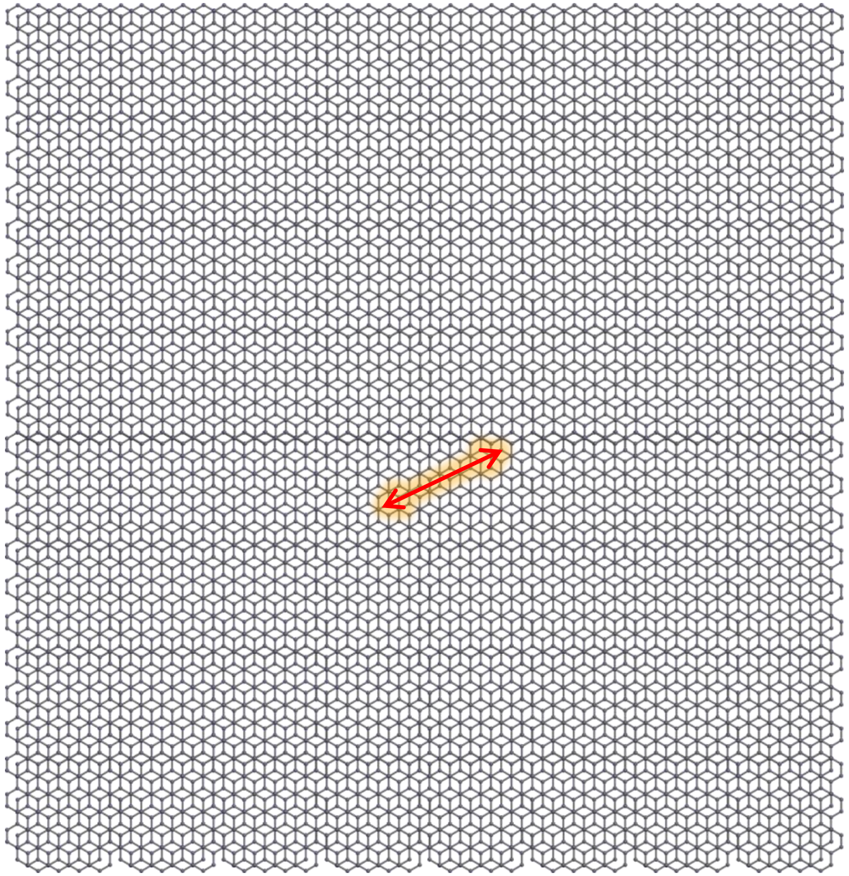
- *Interlayer coupling*
- *Many-body states*
- *Non-Fermi liquid*
- *More…*

Difference in lattice period



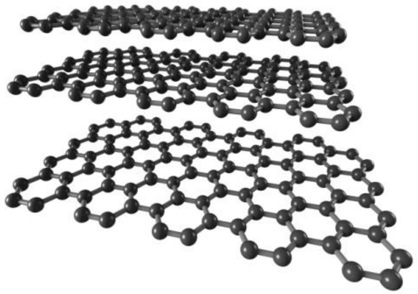
Graphene-hBN heterostructures

Difference in lattice orientation



Twisted bilayers, trilayers, etc.

Graphene-Graphene at large angle



Graphene layers

- *Semimetal*
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- *Interlayer coupling*
- *Many-body states*
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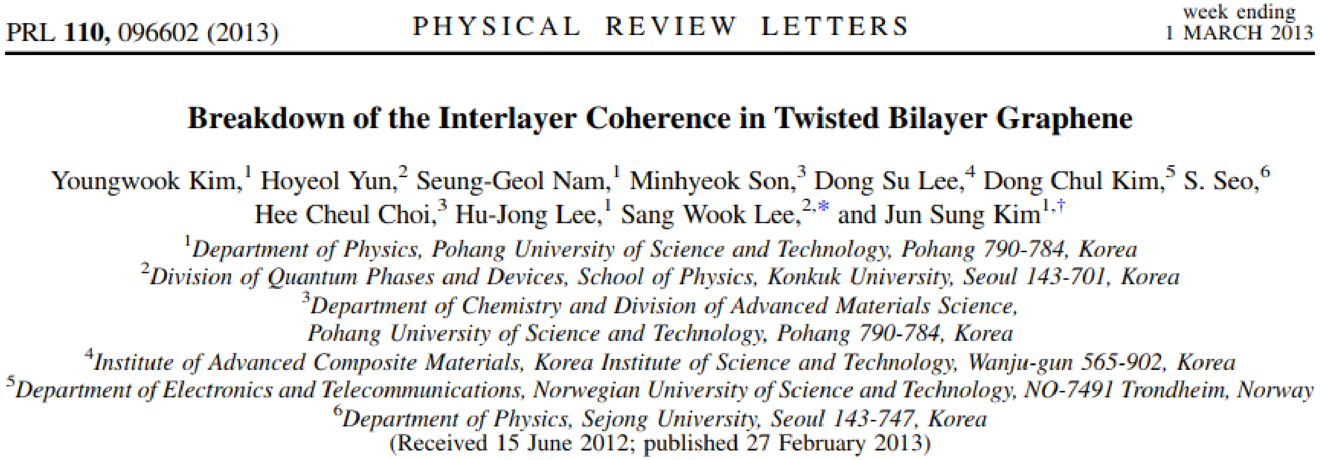
- *More…*

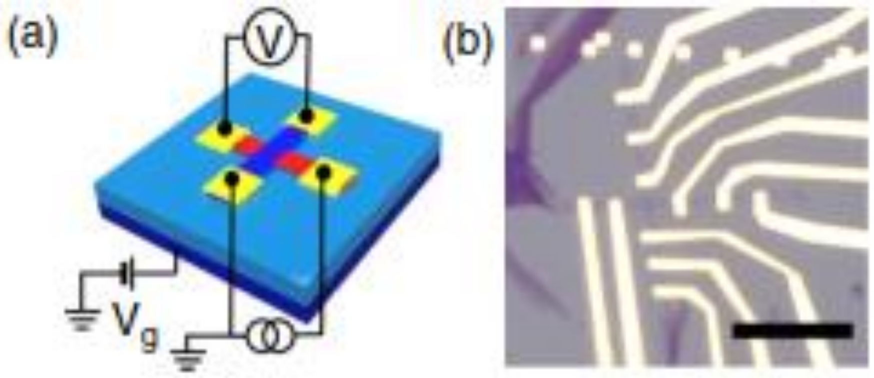


PRL **110**, 096602 (2013) PHYSICAL REVIEW LETTERS week ending 1 MARCH 2013

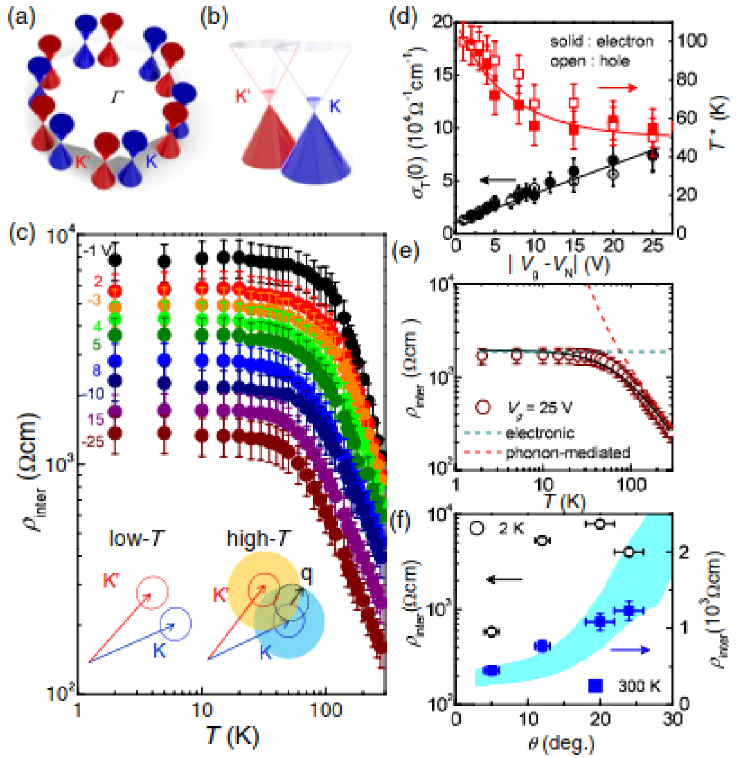
Breakdown of the Interlayer Coherence in Twisted Bilayer Graphene

Youngwook Kim,[1] Hoyeol Yun,[2] Seung-Geol Nam,[1] Minhyeok Son,[3] Dong Su Lee,[4] Dong Chul Kim,[5] S. Seo,[6] Hee Cheul Choi,[3] Hu-Jong Lee,[1] Sang Wook Lee,[2,*] and Jun Sung Kim[1,†]

[1]*Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea*
[2]*Division of Quantum Phases and Devices, School of Physics, Konkuk University, Seoul 143-701, Korea*
[3]*Department of Chemistry and Division of Advanced Materials Science, Pohang University of Science and Technology, Pohang 790-784, Korea*
[4]*Institute of Advanced Composite Materials, Korea Institute of Science and Technology, Wanju-gun 565-902, Korea*
[5]*Department of Electronics and Telecommunications, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway*
[6]*Department of Physics, Sejong University, Seoul 143-747, Korea*
(Received 15 June 2012; published 27 February 2013)

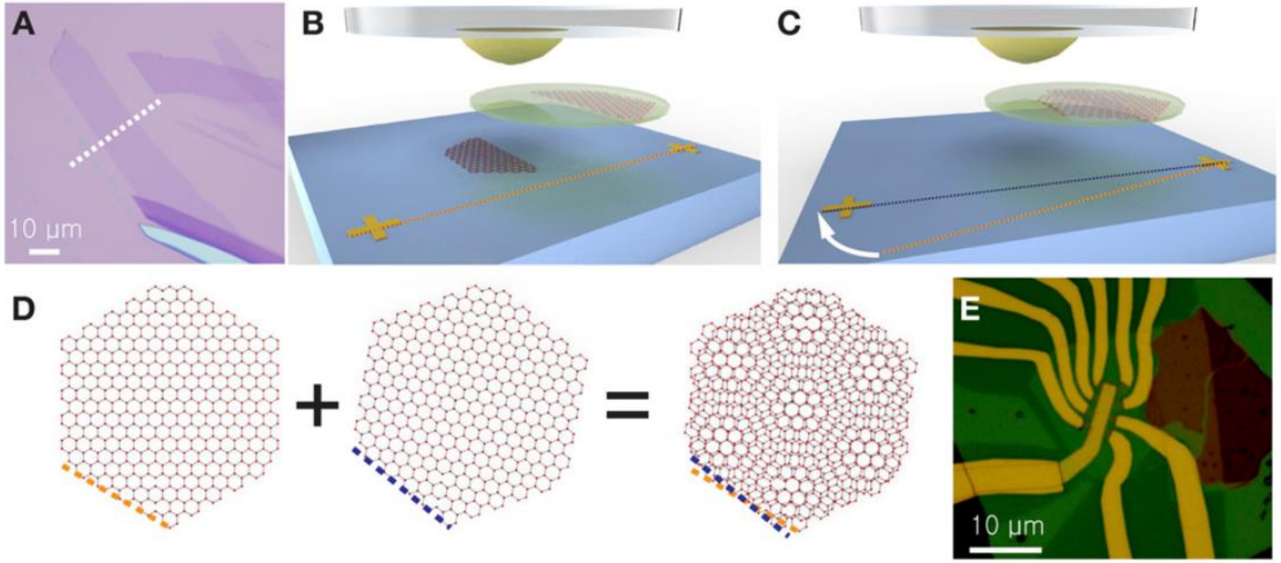


- *Large interlayer R with strong T dependence*
- *At large angle, they are fairly well decoupled*



Graphene-Graphene at small angle

Nano Lett. 16, 1989 (2016), PNAS 114, 3364 (2017)



Moiré bands in twisted double-layer graphene

Rafi Bistritzer and Allan H. MacDonald[1]

Department of Physics, University of Texas at Austin, Austin, TX 78712

Contributed by Allan H. MacDonald, June 7, 2011 (sent for review December 8, 2010)

Layer 1

$$\mathcal{H}_{\mathbf{k}} = \begin{bmatrix} \boxed{h_{\mathbf{k}}(\theta/2)} & T_b & T_{tr} & T_{tl} \\ T_b^{\dagger} & h_{\mathbf{k}_b}(-\theta/2) & 0 & 0 \\ T_{tr}^{\dagger} & 0 & h_{\mathbf{k}_{tr}}(-\theta/2) & 0 \\ T_{tl}^{\dagger} & 0 & 0 & h_{\mathbf{k}_{tl}}(-\theta/2) \end{bmatrix},$$

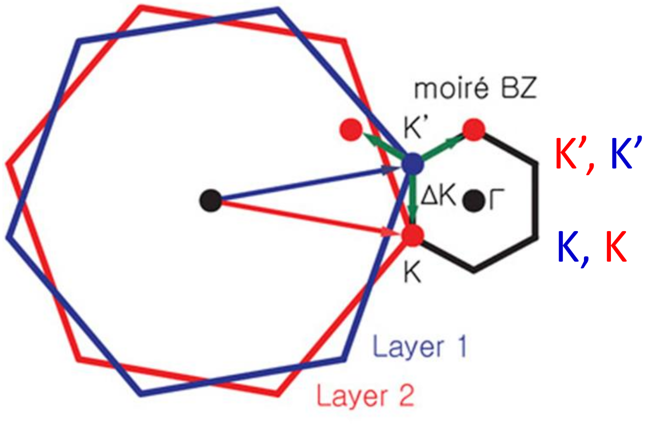
Layer 2

where $\mathbf{k}$ is in the moiré Brillouin-zone and $\mathbf{k_j} = \mathbf{k} + \mathbf{q}_j$.

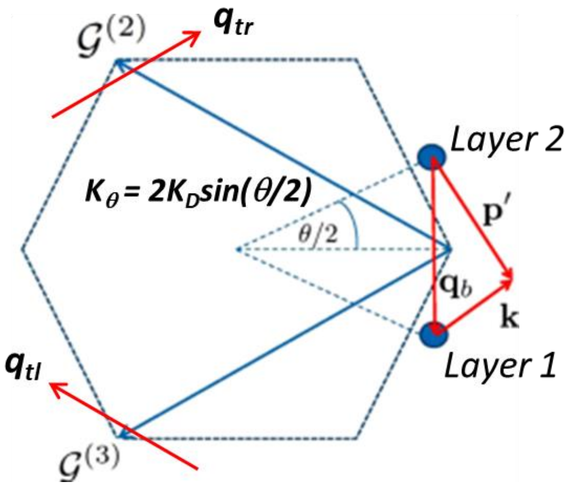
$$h_{\mathbf{k}}(\theta) = -vk \begin{bmatrix} 0 & e^{i(\theta_{\mathbf{k}}-\theta)} \\ e^{-i(\theta_{\mathbf{k}}-\theta)} & 0 \end{bmatrix},$$

hopping

$$\frac{v^{\star}}{v} = \frac{1-3\alpha^2}{1+6\alpha^2}, \qquad \alpha = w/vk_{\theta}$$

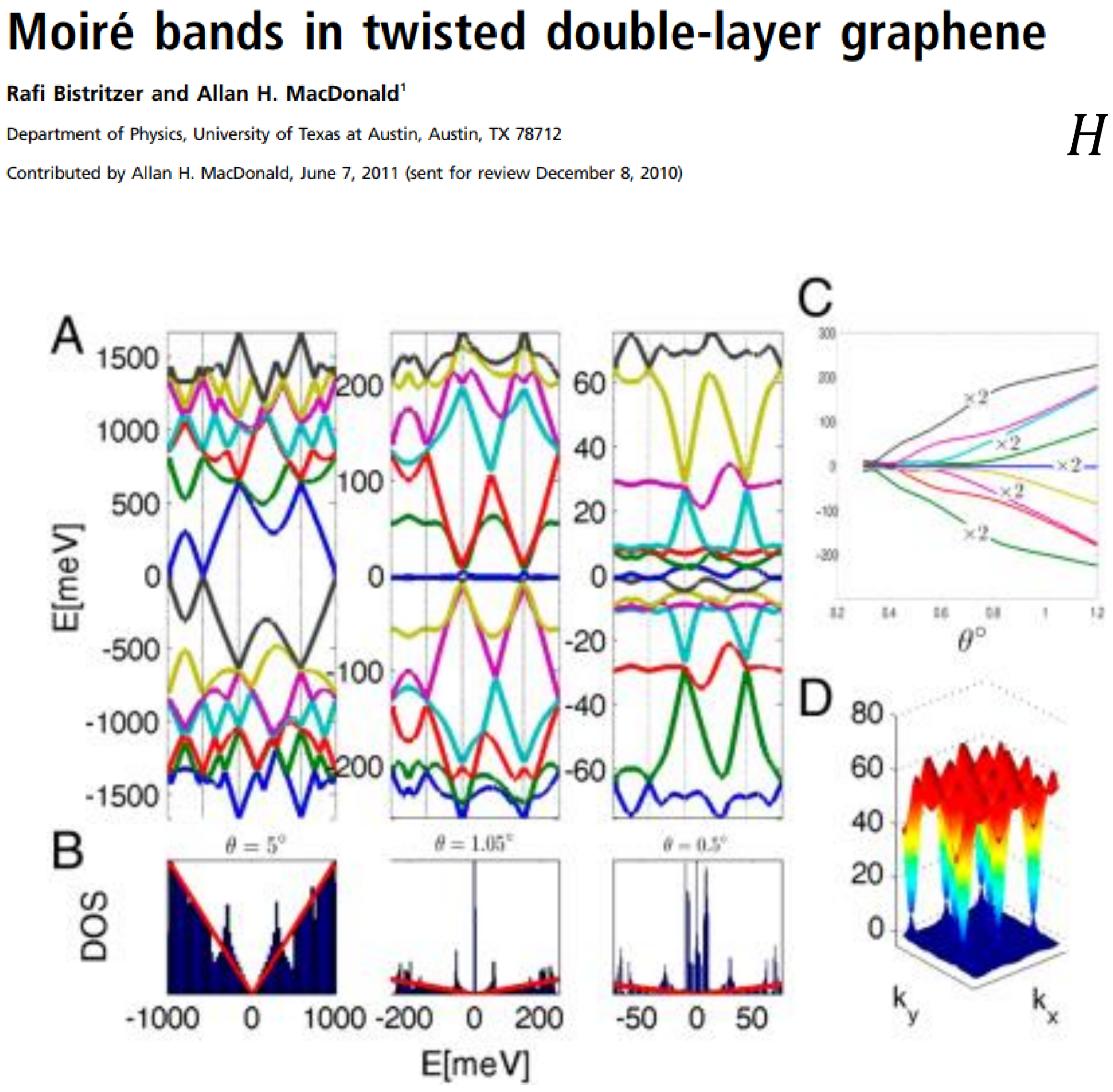


PNAS 114, 3364 (2017)



Other theories on twisted bilayer:
PRL 99, 256802 (2007) by Lopes dos Santos, Peres, and Castro Neto
PRB 81, 161405 (2010) by E.J.Mele and more…..

Graphene-Graphene at small angle



Moiré bands in twisted double-layer graphene

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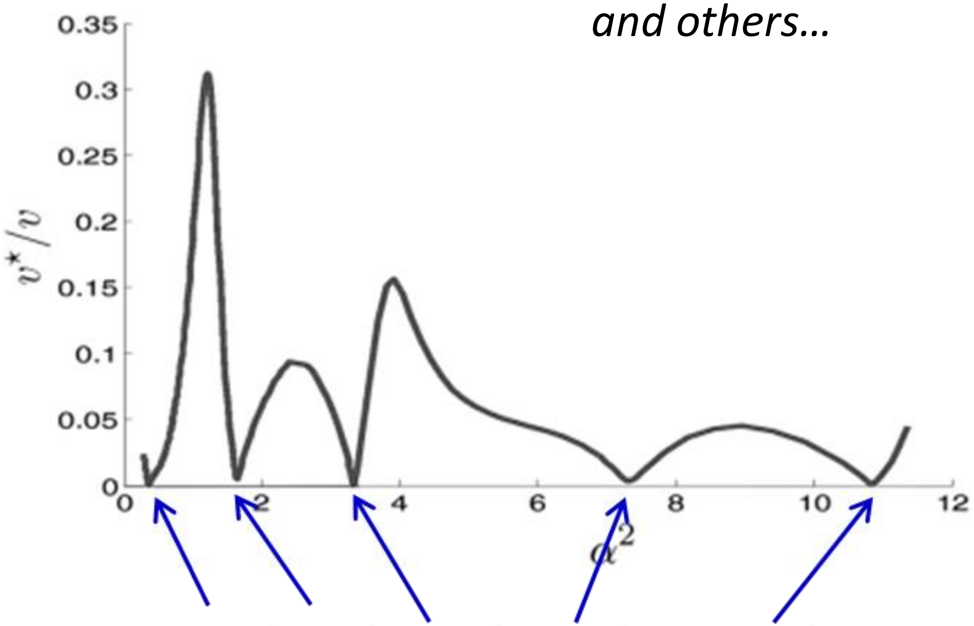
Contributed by Allan H. MacDonald, June 7, 2011 (sent for review December 8, 2010)

$$H = \hbar v_F\left(k_x\sigma_x + k_y\sigma_y\right)$$ *+ Interactions*

~~Kinetic energy~~

Flat bands

Coulomb interaction
Spin-orbit coupling
Electron-phonon (BCS)
Spin-spin interaction (Ising)
and others…



θ ≈ **1.05** °, 0.5 °, 0.35 °, 0.24 °, and 0.2 °

Strong interaction in flat bands

$$H = \hbar v_F\left(k_x\sigma_x + k_y\sigma_y\right)$$ + *Interactions*

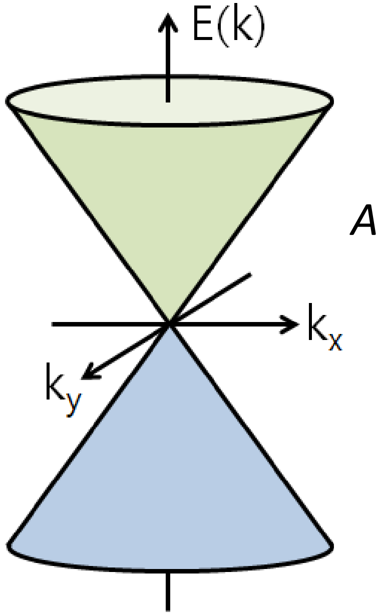
~~*Kinetic energy*~~

Flat bands

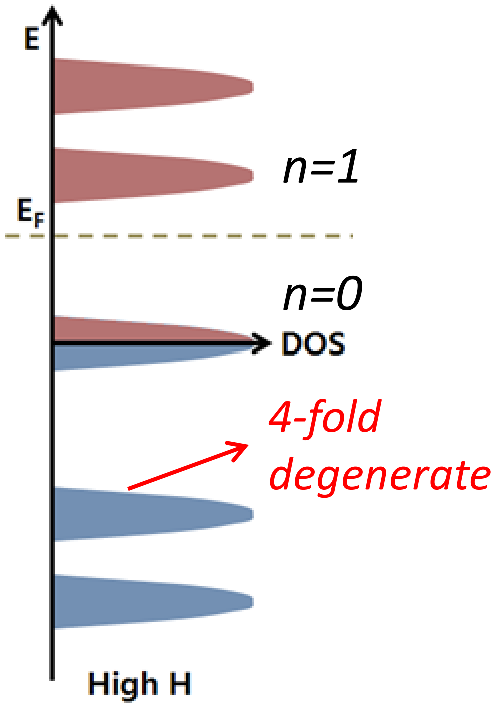
Coulomb interaction
Spin-orbit coupling
Electron-phonon (BCS)
Spin-spin interaction (Ising)
and others…

Approaching low-energy limit

Geometry	Δn (cm^{-2})	ΔE (1LG)	ΔE (2LG)
On SiO$_2$	10^{12}	~120 meV	~30 meV
On hBN	10^{10}	~12 meV	~0.3 meV
Suspended	**$10^{8\text{-}9}$**	**1~4 meV**	**3~30 μeV**



Apply B field

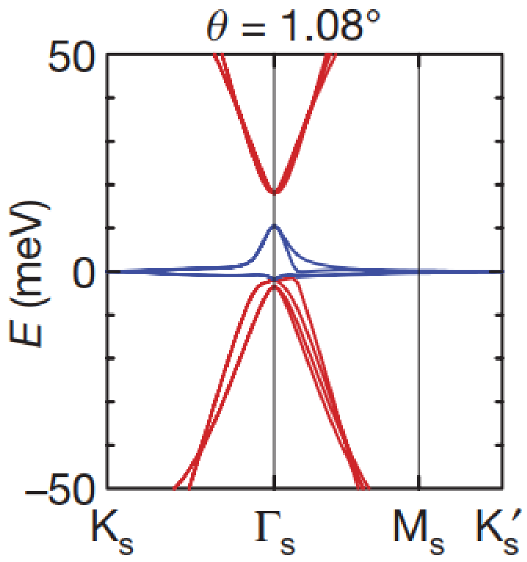


$$E_n = \pm v_F\sqrt{2e\hbar Bn}$$

$$\sigma_{xy} = 4(n + 1/2) \times e^2/h$$

Kinetic energy is quenched into a flat Landau level.

Fractional quantum Hall effect

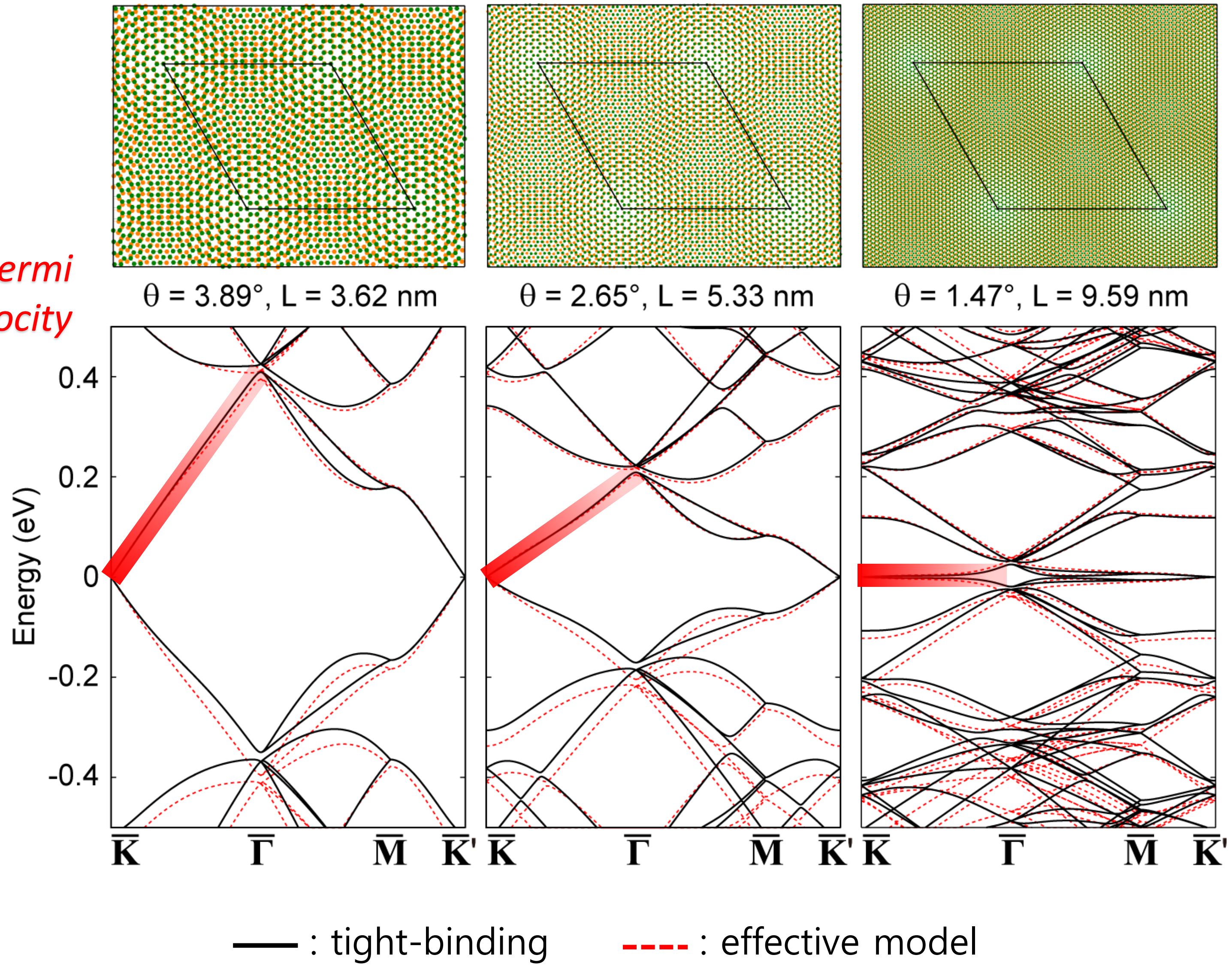


Graphene-Graphene at small angle

Moon and Koshino, Phys. Rev. B 85, 195458 (2012), Phys. Rev. B 87, 205404 (2013).

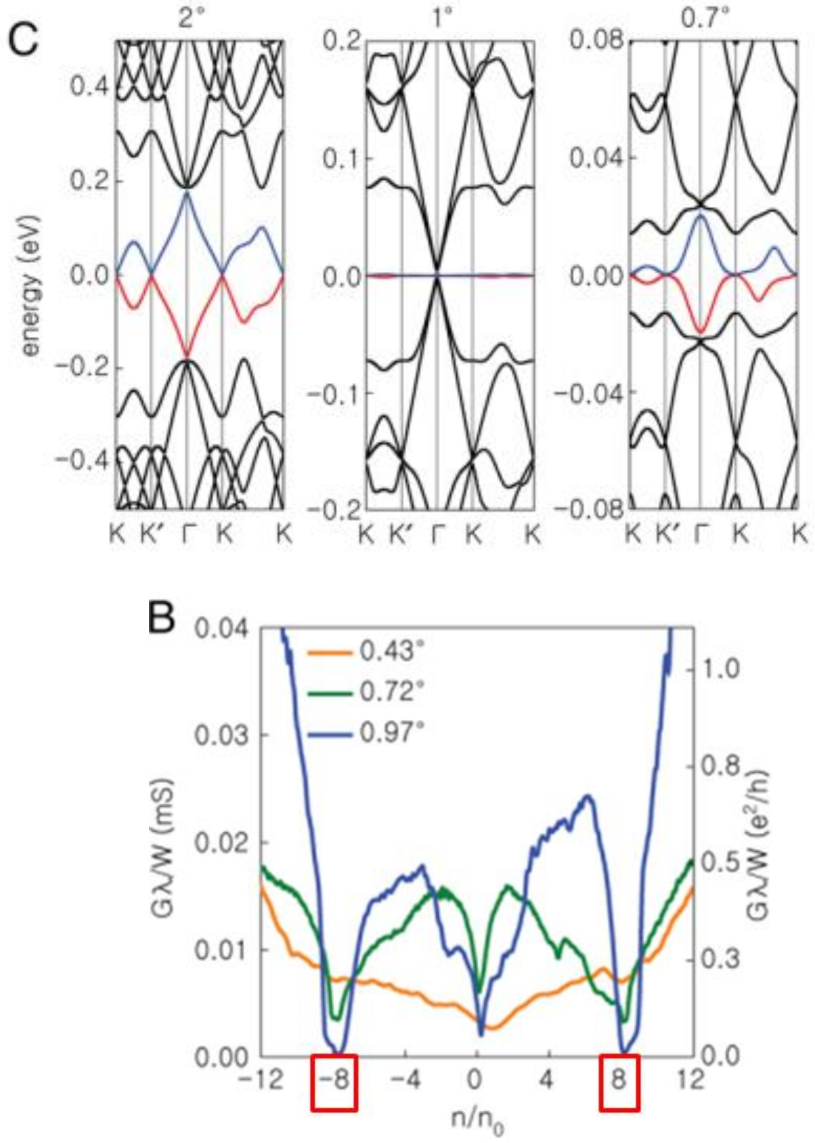
PNAS 114, 3364 (2017)

Fermi velocity


θ = 3.89°, L = 3.62 nm

θ = 2.65°, L = 5.33 nm

θ = 1.47°, L = 9.59 nm



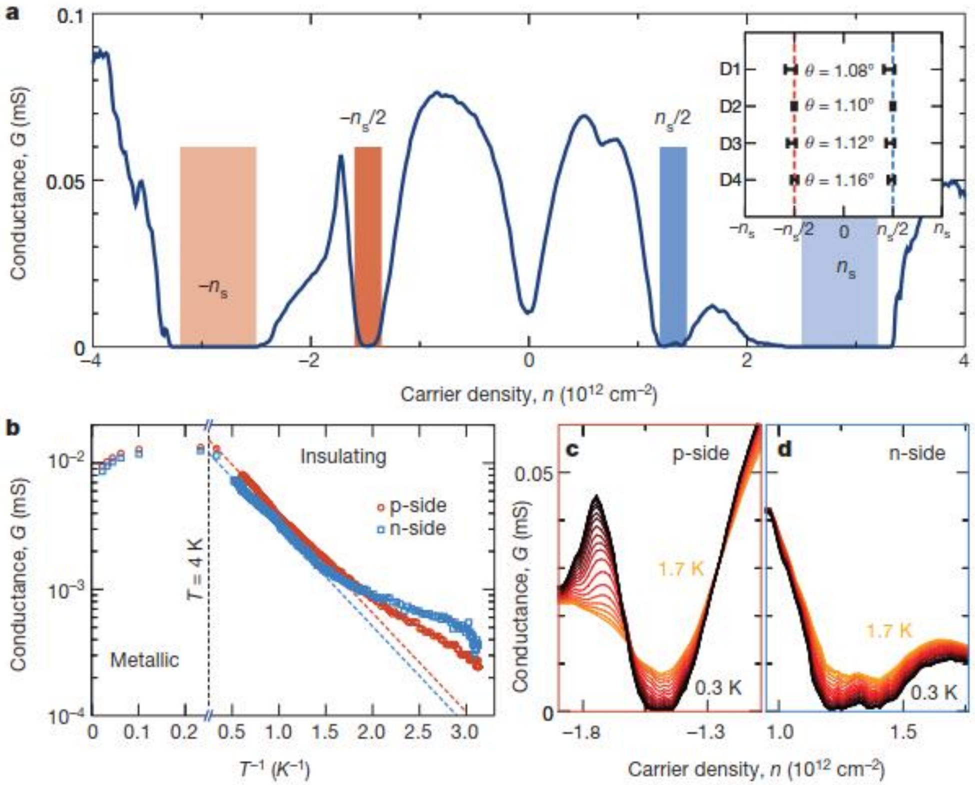
Graphene-Graphene at magic angle

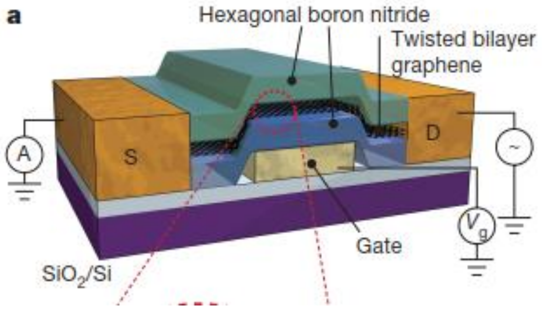
LETTER

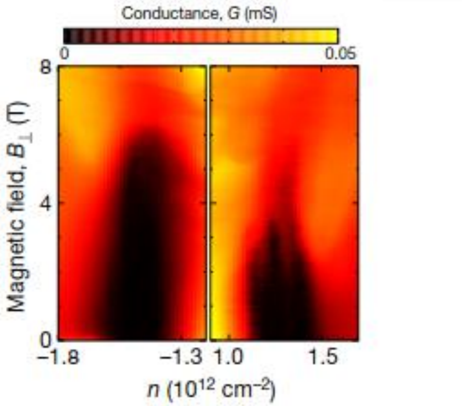
doi:10.1038/nature26154

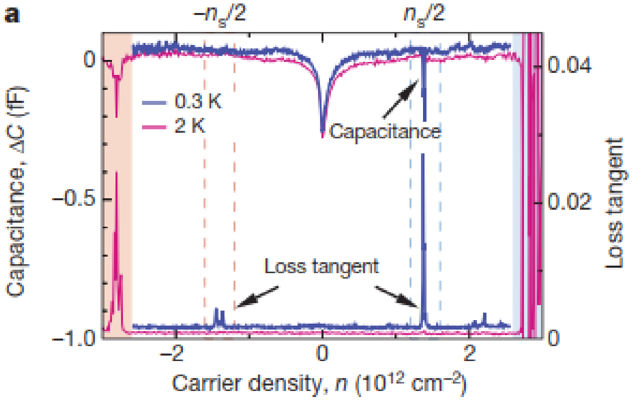
Correlated insulator behaviour at half-filling in magic-angle graphene superlattices

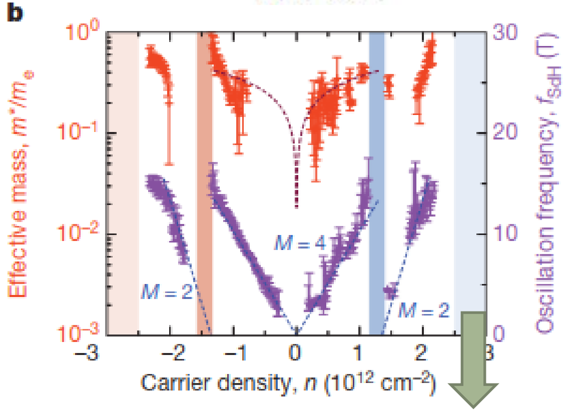
Yuan Cao[1], Valla Fatemi[1], Ahmet Demir[1], Shiang Fang[2], Spencer L. Tomarken[1], Jason Y. Luo[1], Javier D. Sanchez-Yamagishi[2], Kenji Watanabe[3], Takashi Taniguchi[3], Efthimios Kaxiras[2,4], Ray C. Ashoori[1] & Pablo Jarillo-Herrero[1]

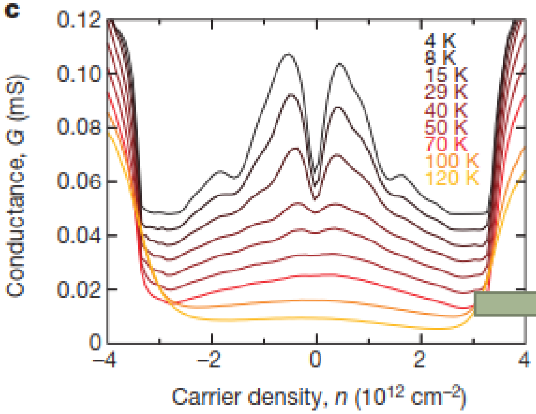












- *Degeneracy broken as expected for the Mott insulator*
- *Valley degeneracy is broken not spin*
- *Flattened due to thermal excitation*

Graphene-Graphene at magic angle

LETTER

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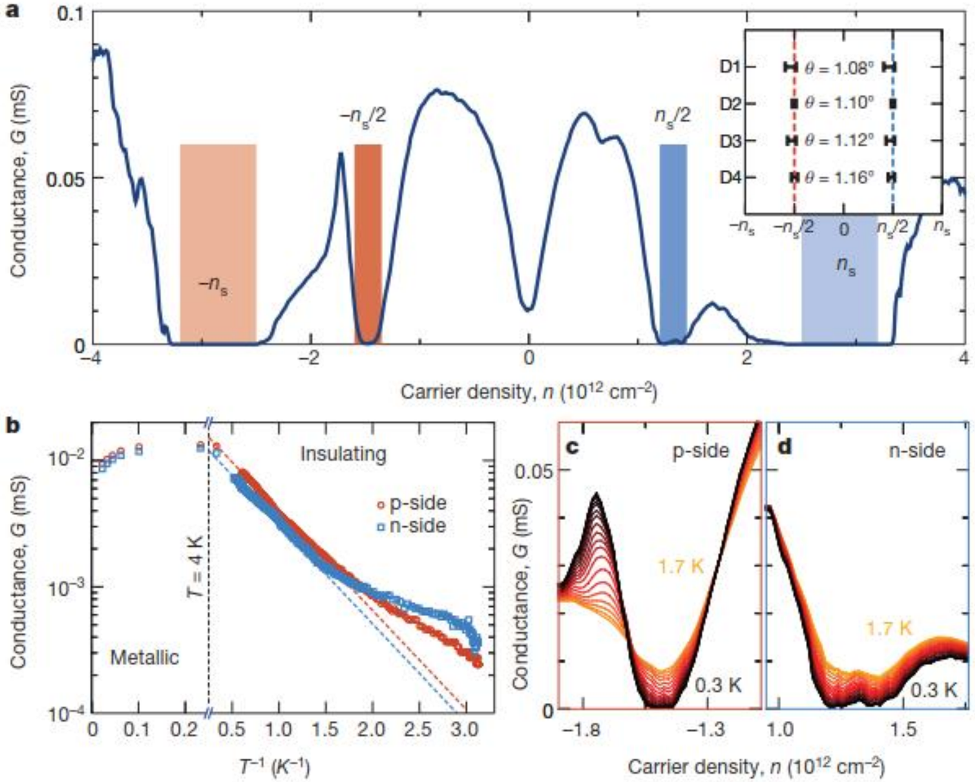
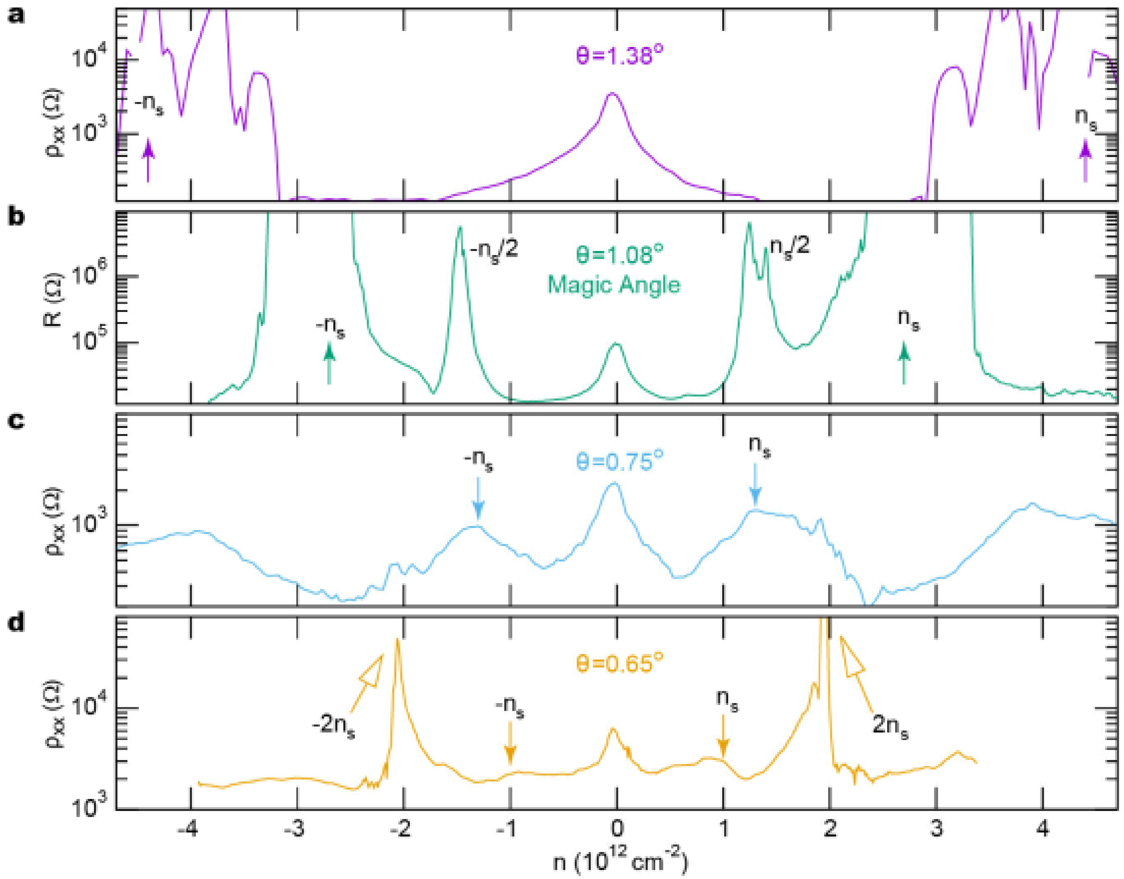
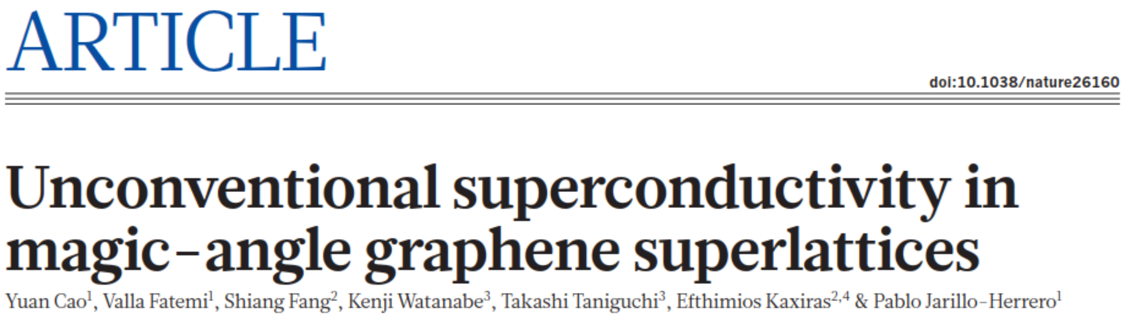




Graphene-Graphene at magic angle

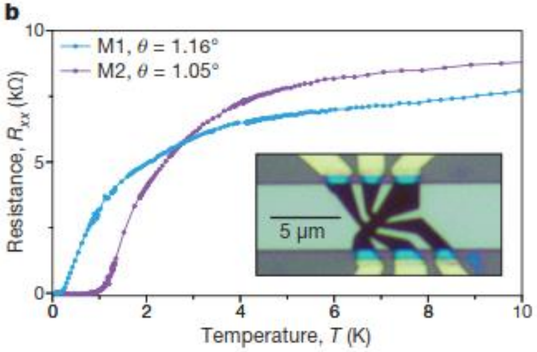


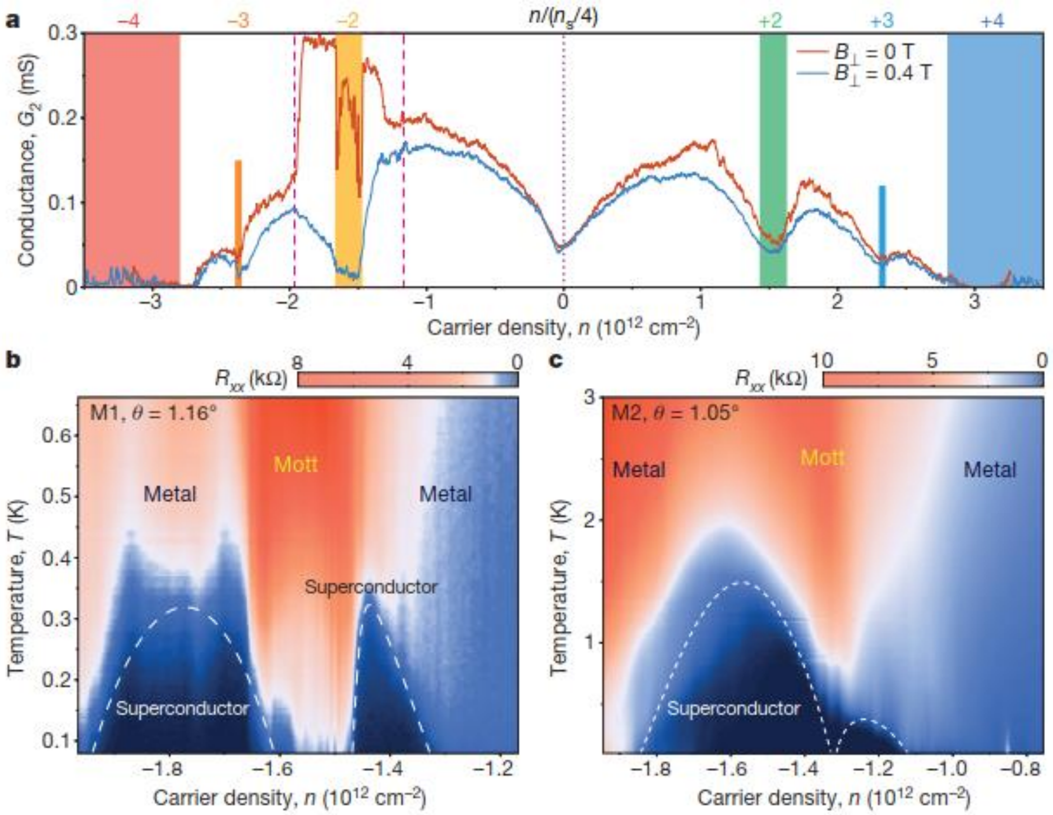
ARTICLE

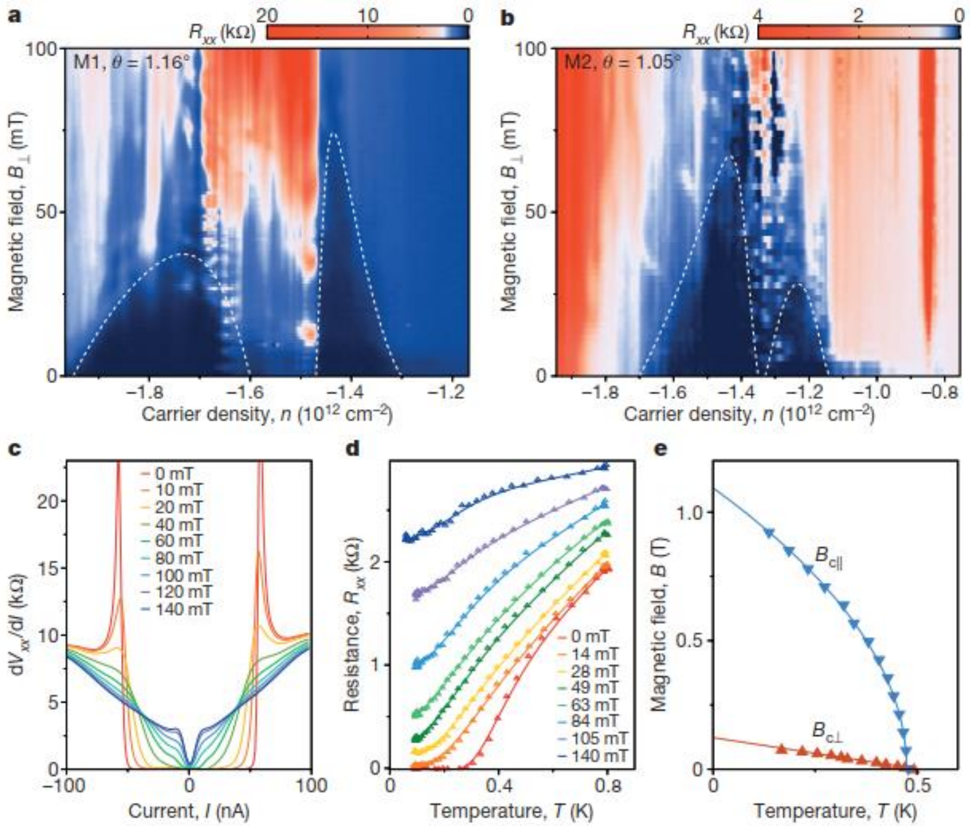
doi:10.1038/nature26160

Unconventional superconductivity in magic-angle graphene superlattices

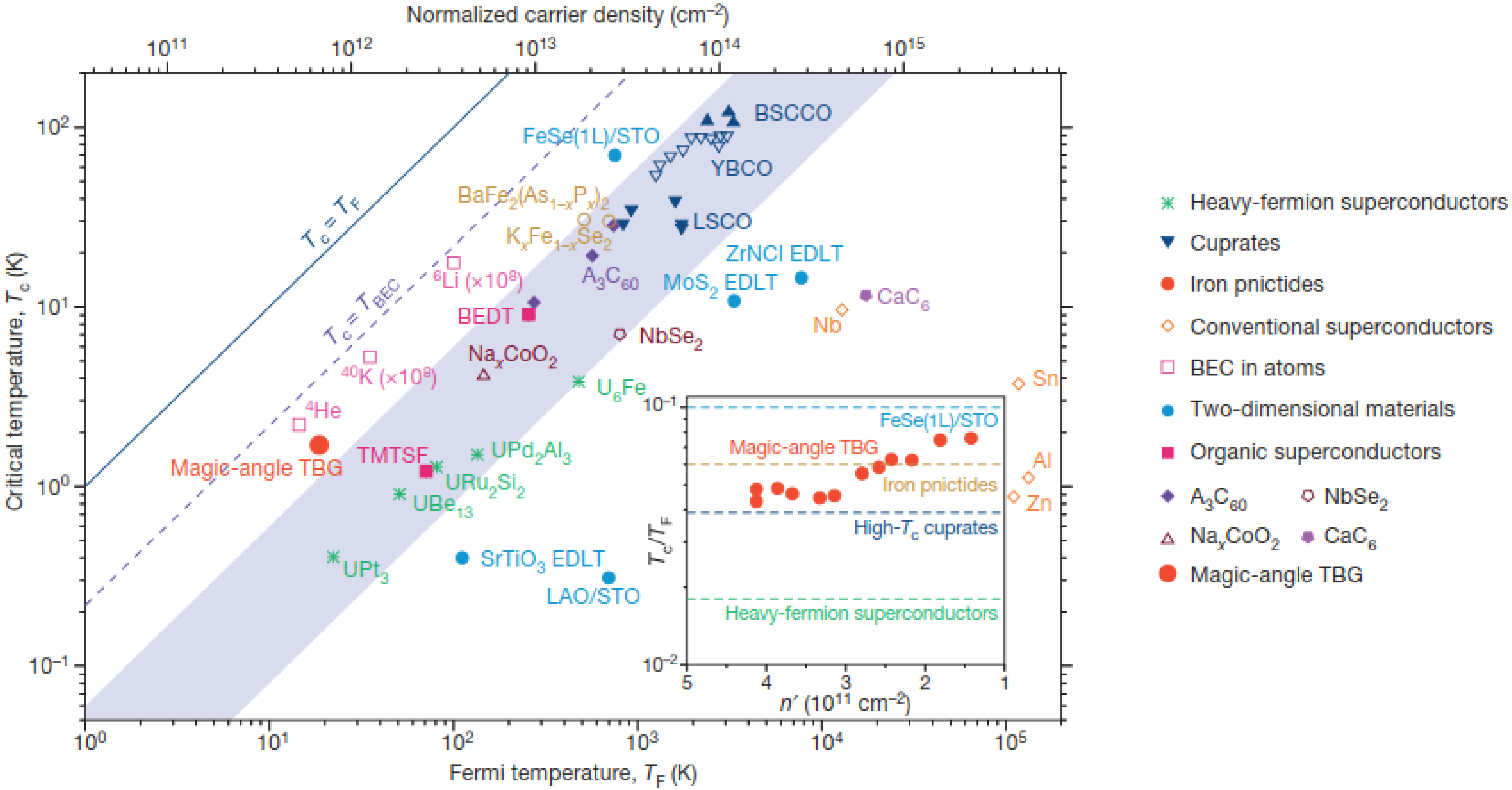
Yuan Cao[1], Valla Fatemi[1], Shiang Fang[2], Kenji Watanabe[3], Takashi Taniguchi[3], Efthimios Kaxiras[2,4] & Pablo Jarillo-Herrero[1]



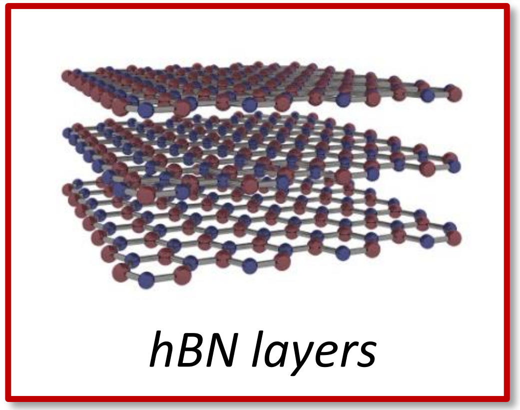




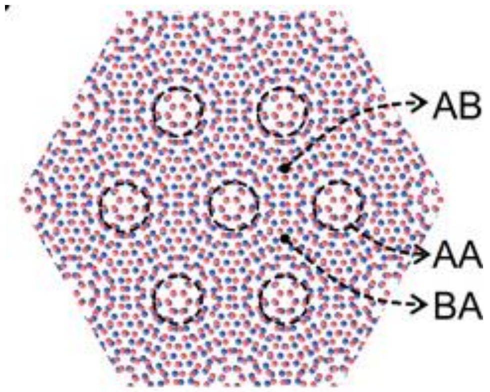
Graphene-Graphene at magic angle



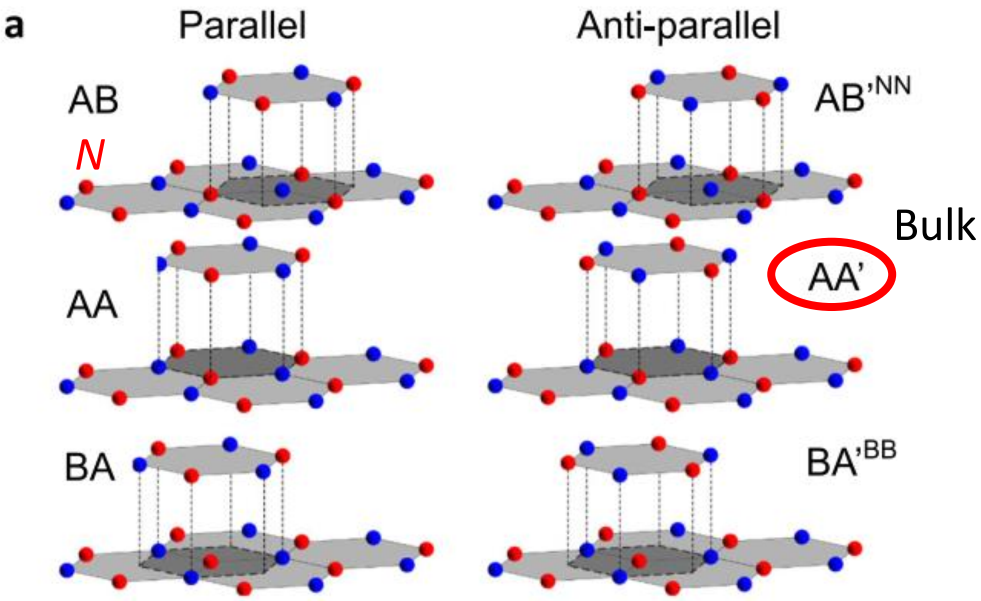
hBN-hBN heterostructures: sliding ferroelectricity



hBN layers



- *Insulator ~ 5 eV*
- *Chemically inert*



ARTICLE

https://doi.org/10.1038/s41467-020-20667-2 OPEN

Check for

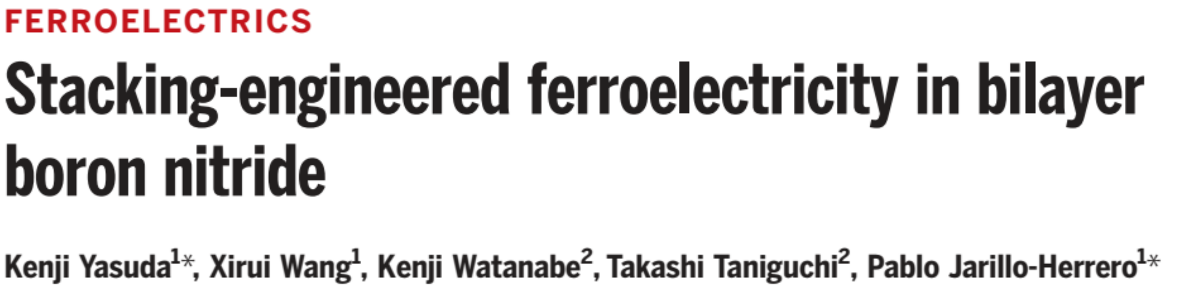
Charge-polarized interfacial superlattices in marginally twisted hexagonal boron nitride

C. R. Woods[1,2]✉, P. Ares[1,2], H. Nevison-Andrews[1,2], M. J. Holwill[1,2], R. Fabregas[1], F. Guinea[3,4], A. K. Geim[1,2], K. S. Novoselov[1,2,5,6], N. R. Walet[1] & L. Fumagalli[1,2]✉

FERROELECTRICS

Interfacial ferroelectricity by van der Waals sliding

M. Vizner Stern[1], Y. Waschitz[1], W. Cao[2], I. Nevo[1], K. Watanabe[3], T. Taniguchi[3], E. Sela[1], M. Urbakh[2], O. Hod[2], M. Ben Shalom[1]*



FERROELECTRICS

Stacking-engineered ferroelectricity in bilayer boron nitride

Kenji Yasuda[1]*, Xirui Wang[1], Kenji Watanabe[2], Takashi Taniguchi[2], Pablo Jarillo-Herrero[1]*

hBN-hBN heterostructures: sliding ferroelectricity

ARTICLE

Check for

https://doi.org/10.1038/s41467-020-20667-2 OPEN

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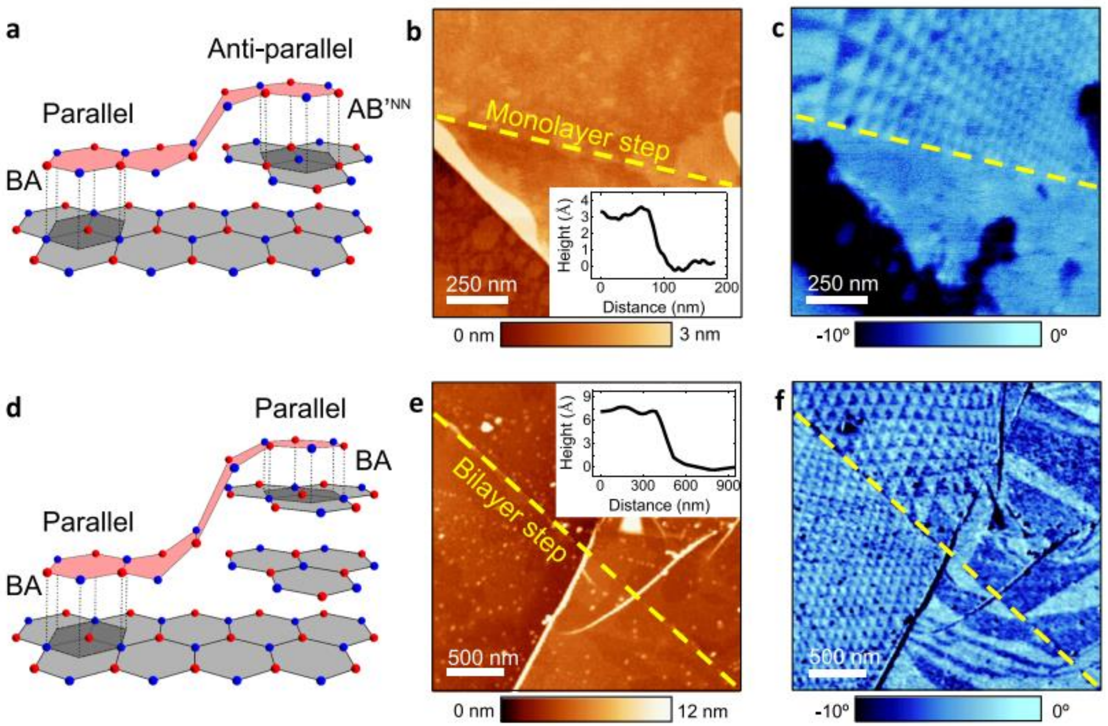

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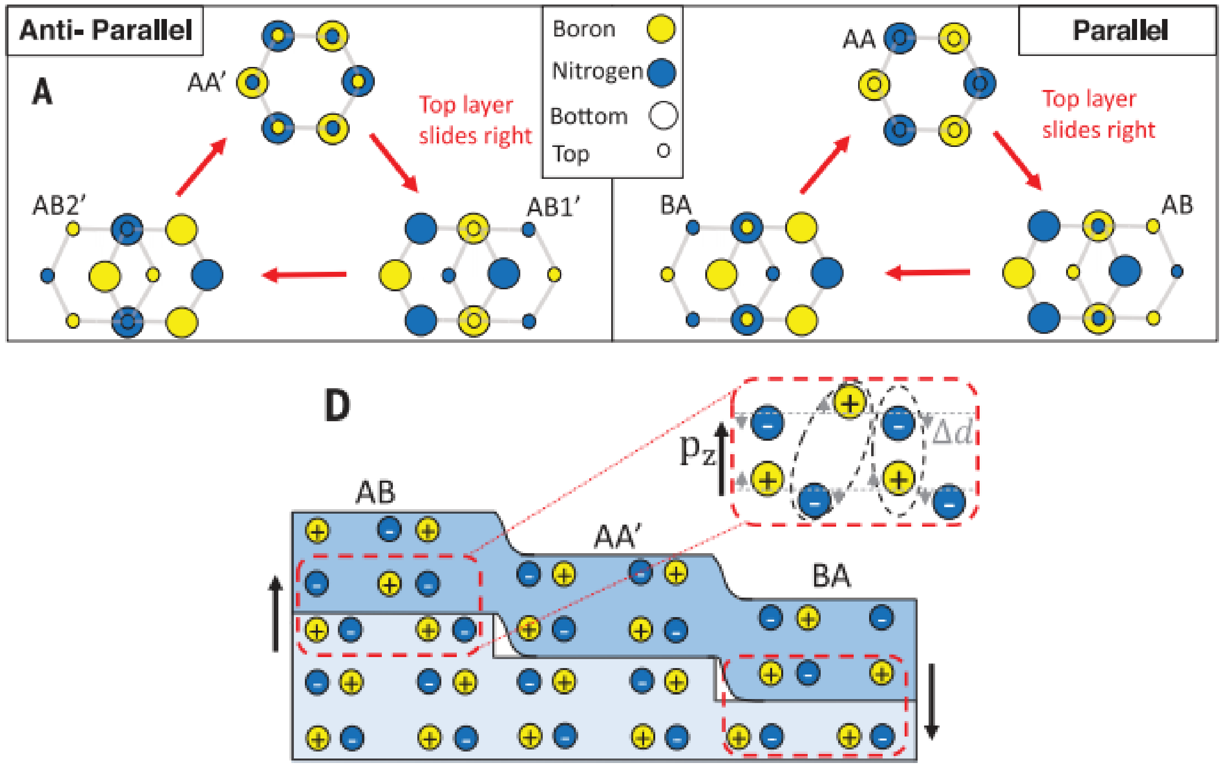

hBN-hBN heterostructures: sliding ferroelectricity

FERROELECTRICS

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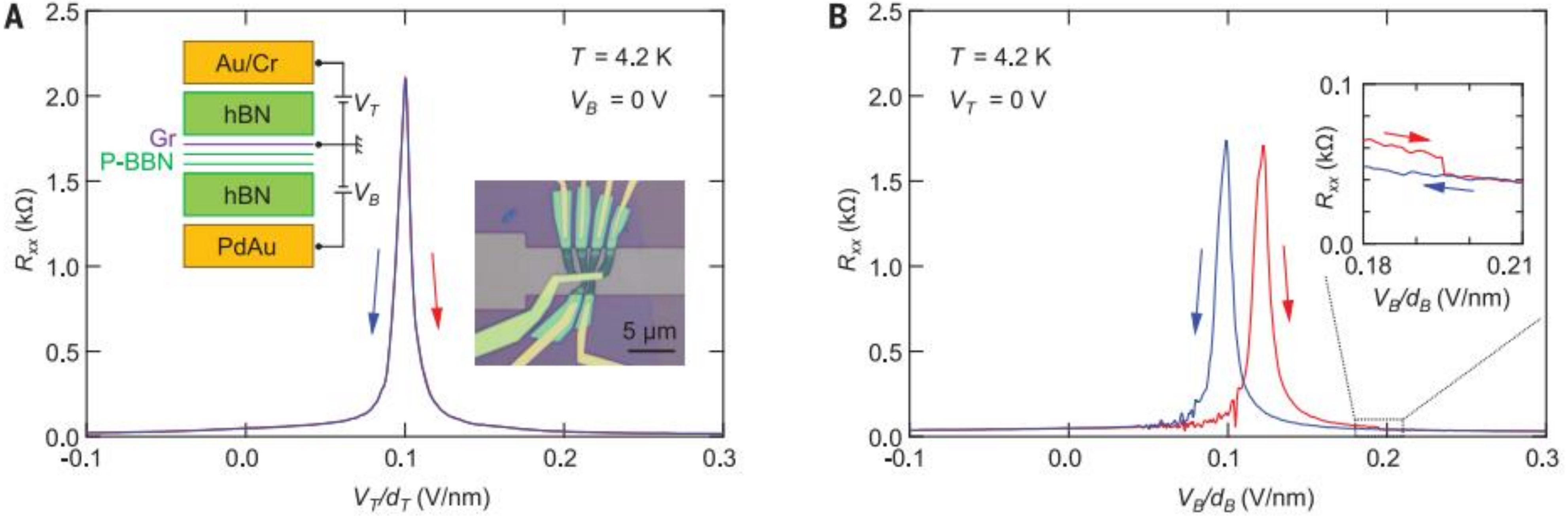

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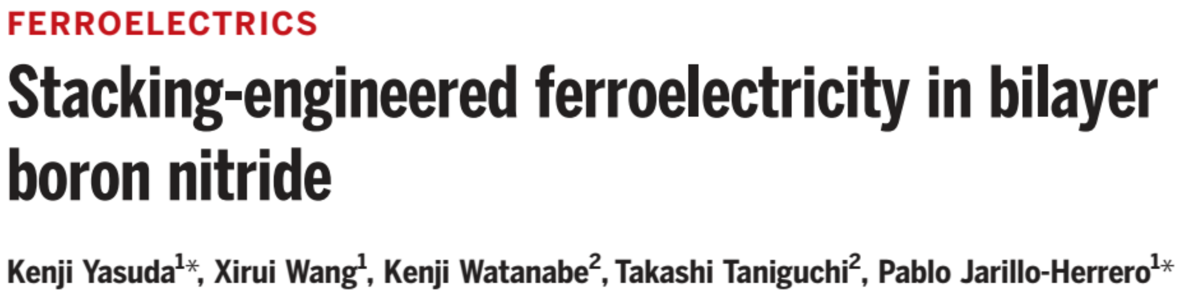

FERROELECTRICS

Stacking-engineered ferroelectricity in bilayer boron nitride

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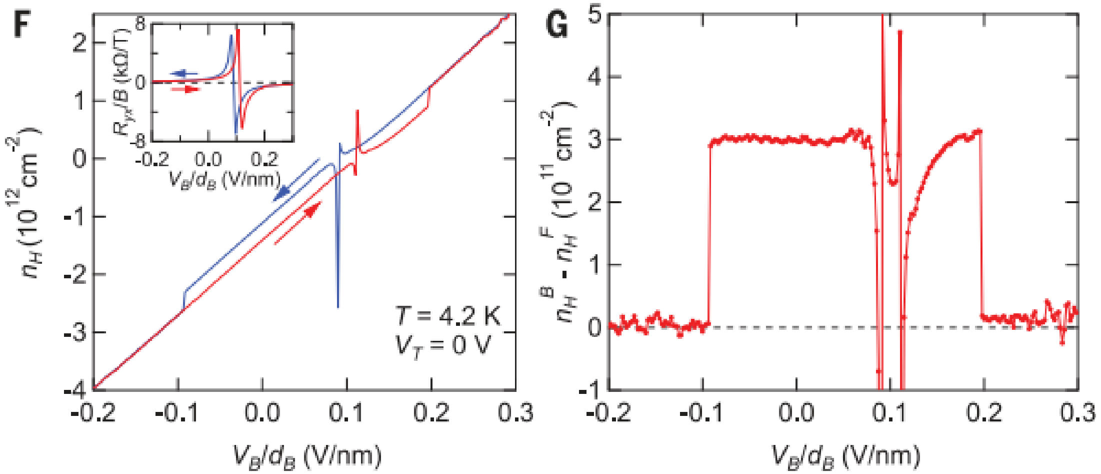
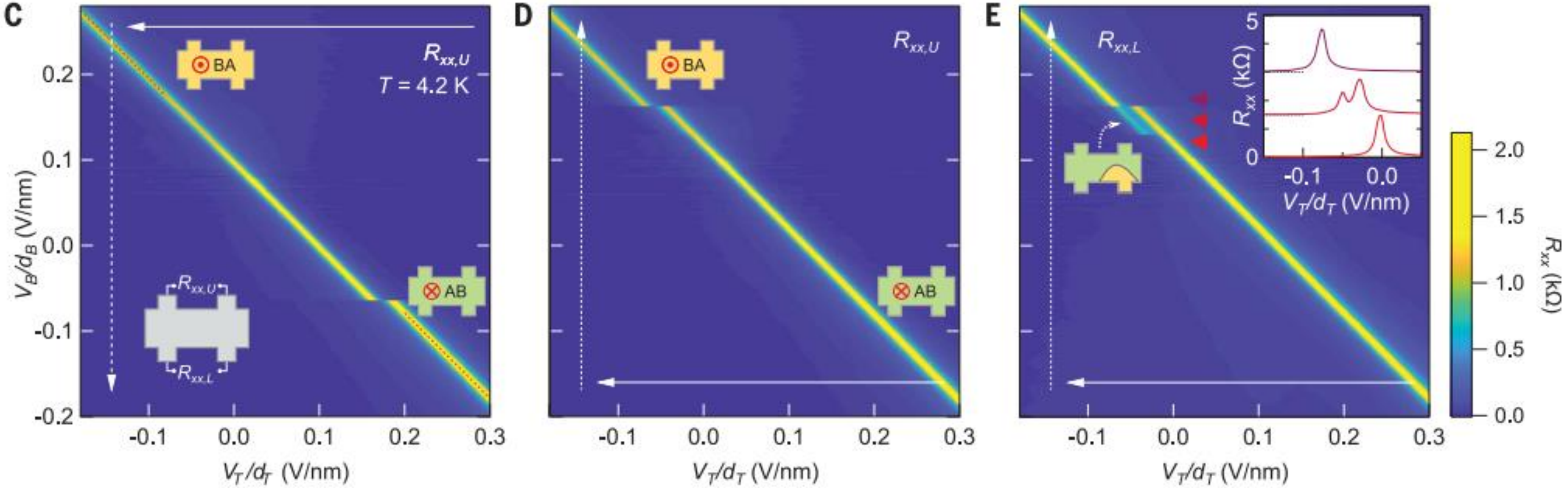
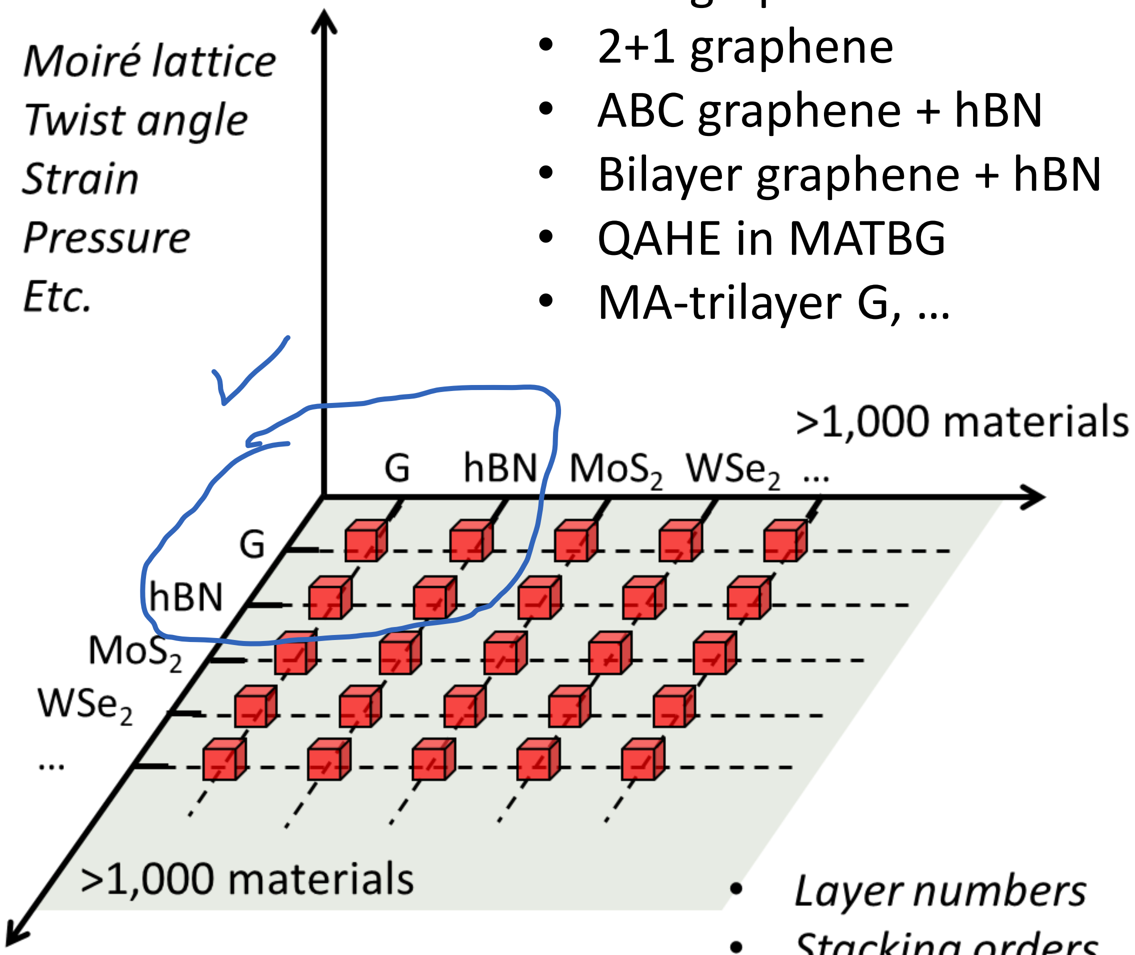




What we have learnt so far.



- 2+2 graphene
- 2+1 graphene
- ABC graphene + hBN
- Bilayer graphene + hBN
- QAHE in MATBG
- MA-trilayer G, …

- *Layer numbers*
- *Stacking orders*
- *Dimensions*

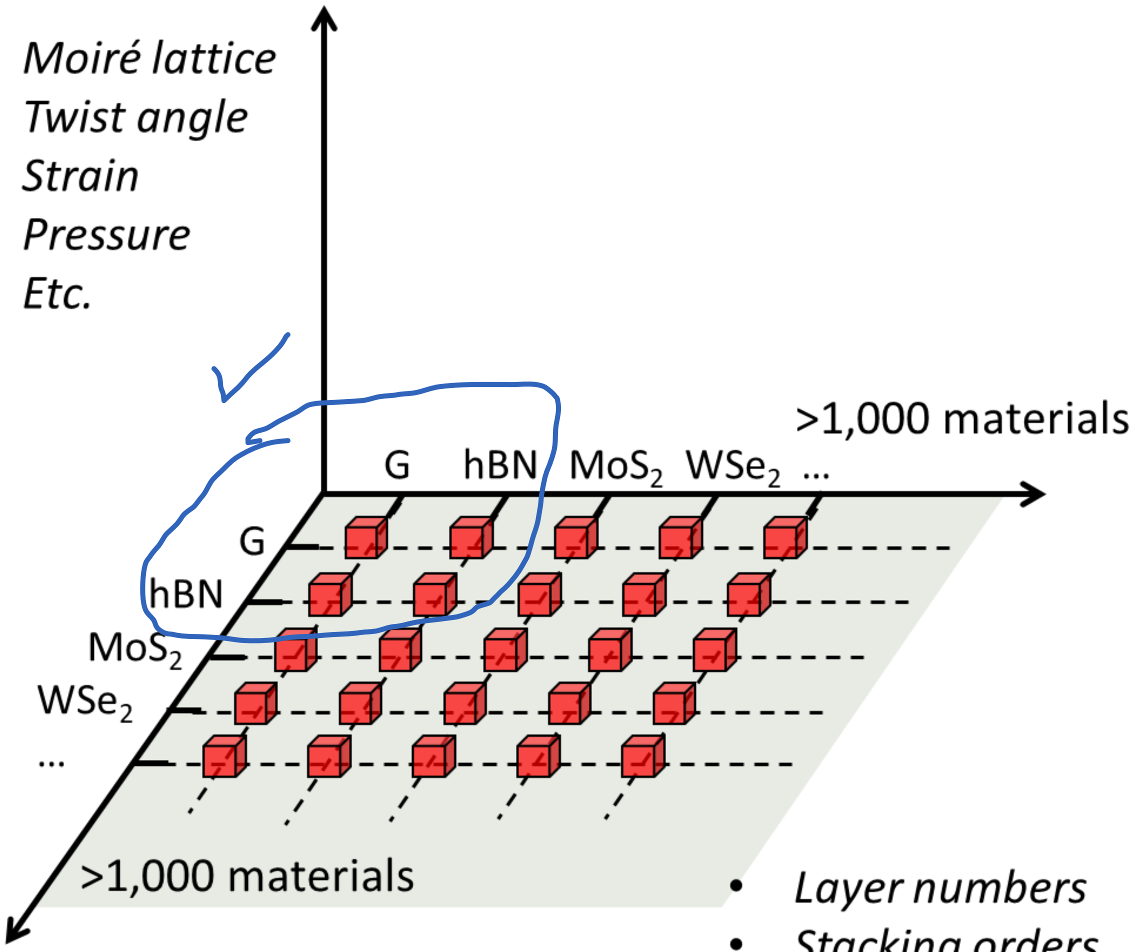
- Graphene-hBN
  - hBN as a substrate for high-quality graphene
  - Moiré structure: folding bands
  - Satellite Dirac point
  - Hofstadter’s butterfly effect

- Graphene-Graphene
  - At large angle: fairly well decoupled
  - At small angle: band structure changes significantly, satellite Dirac point
  - At magic angle: flat bands occur, various many body states appear

- hBN-hBN
  - Sliding ferroelectricity

- *Carriers move between atomic orbitals:* ***atomic registry/potential is important***

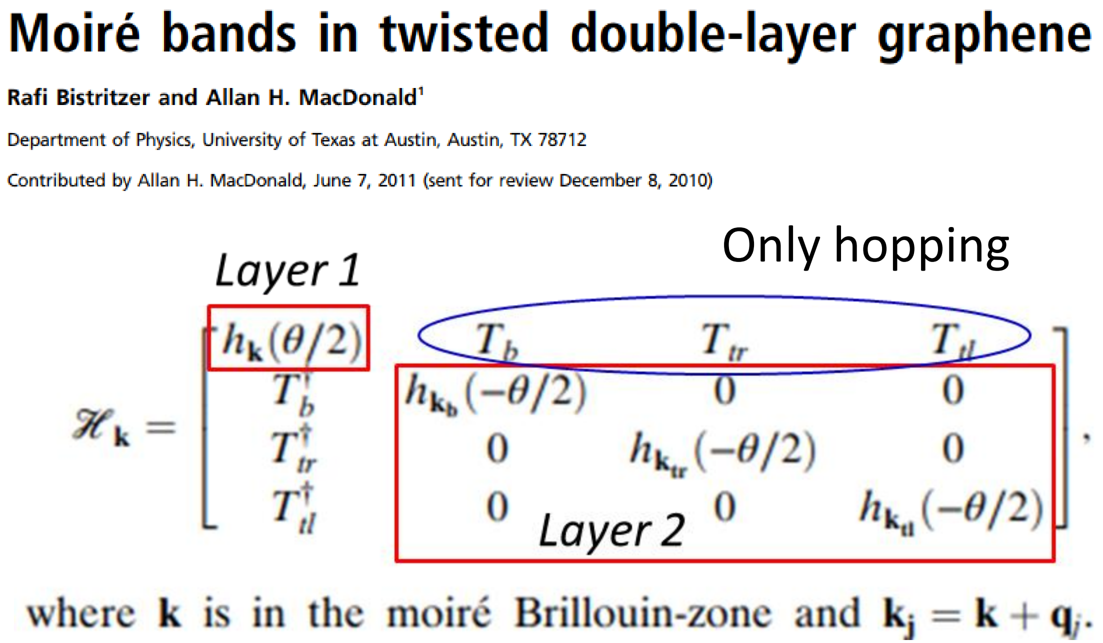
To have moiré



- *Layer numbers*
- *Stacking orders*
- *Dimensions*

- *Similar or identical lattice constant*
- *Small angle*
- *Atomically flat*
- *Weak vdW interaction*

To preserve electronic properties of the individual layers



Moiré bands in twisted double-layer graphene

Rafi Bistritzer and Allan H. MacDonald[1]

Department of Physics, University of Texas at Austin, Austin, TX 78712

Contributed by Allan H. MacDonald, June 7, 2011 (sent for review December 8, 2010)

Only hopping

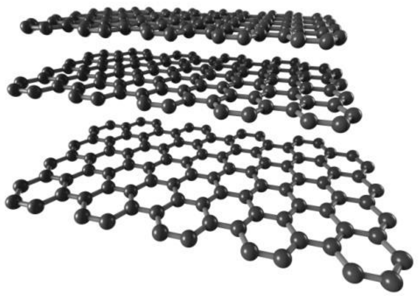
Layer 1

$$\mathscr{H}_{\mathbf{k}} = \begin{bmatrix} h_{\mathbf{k}}(\theta/2) & T_b & T_{tr} & T_{tl} \\ T_b^{\dagger} & h_{\mathbf{k}_b}(-\theta/2) & 0 & 0 \\ T_{tr}^{\dagger} & 0 & h_{\mathbf{k}_{tr}}(-\theta/2) & 0 \\ T_{tl}^{\dagger} & 0 & 0 & h_{\mathbf{k}_{tl}}(-\theta/2) \end{bmatrix},$$

Layer 2

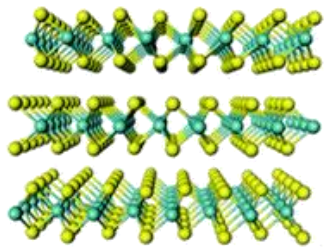
where $\mathbf{k}$ is in the moiré Brillouin-zone and $\mathbf{k_j} = \mathbf{k} + \mathbf{q}_j$.

Graphene-TMDCs



Graphene layers

- *Semimetal*
- *Massless or massive electrons*



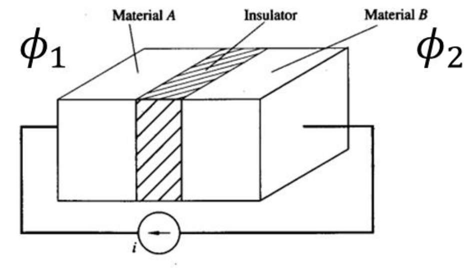
TDMCs (MX_2)

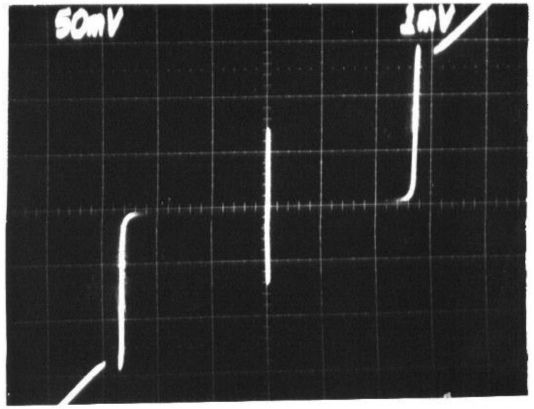
- *Semiconductor ~ 1 eV*
- *Spin-orbit coupling*

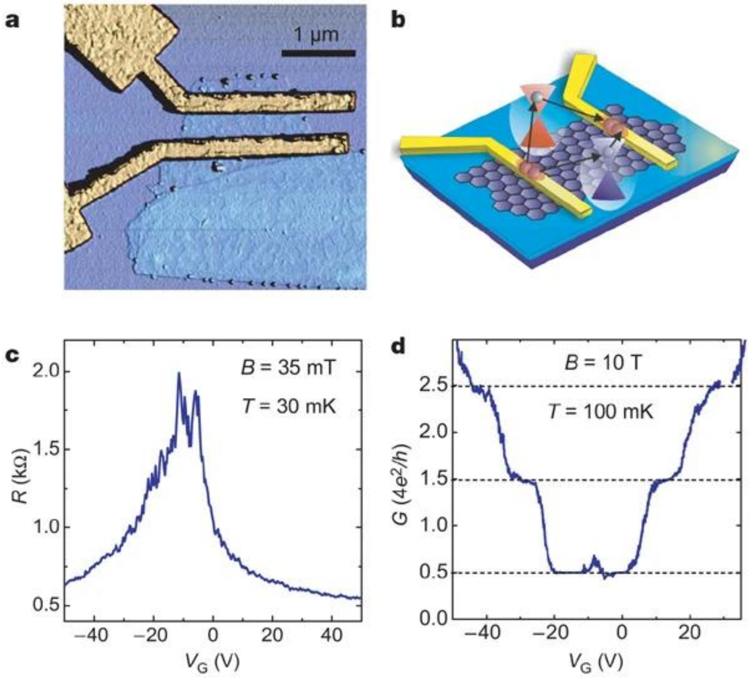
- *Similar or identical lattice constant*
- *Small angle*
- *Atomically flat*
- *Weak vdW interaction*

→ *Strong proximity effect while keeping most of the properties intact*

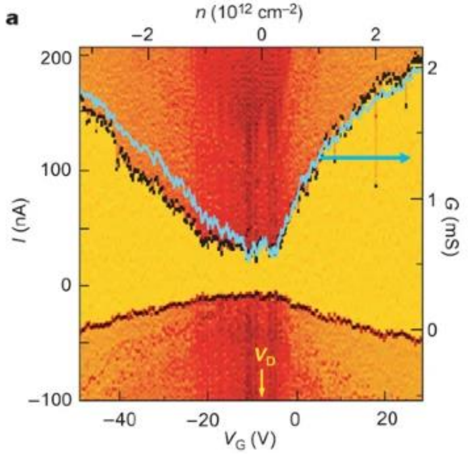
To preserve electronic properties of the individual layers



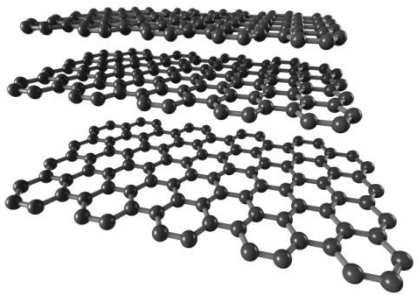




Nature 446, 56 (2007)

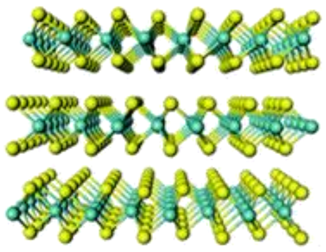


Graphene-TMDCs: induced SOC by proximity



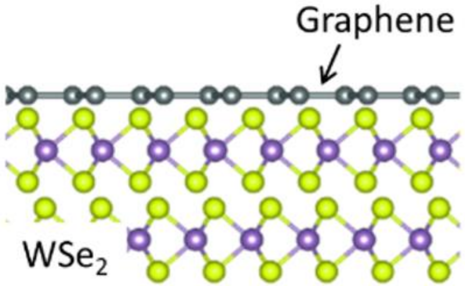
Graphene layers

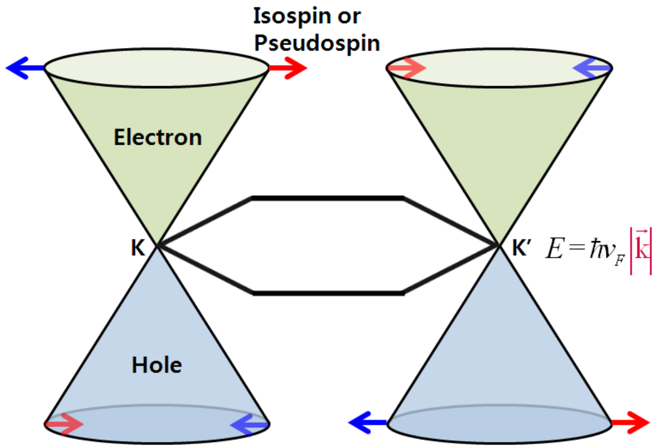
- *Semimetal*
- *Massless or massive electrons*



TDMCs (MX$_2$)

- *Semiconductor ~ 1 eV*
- *Spin-orbit coupling*



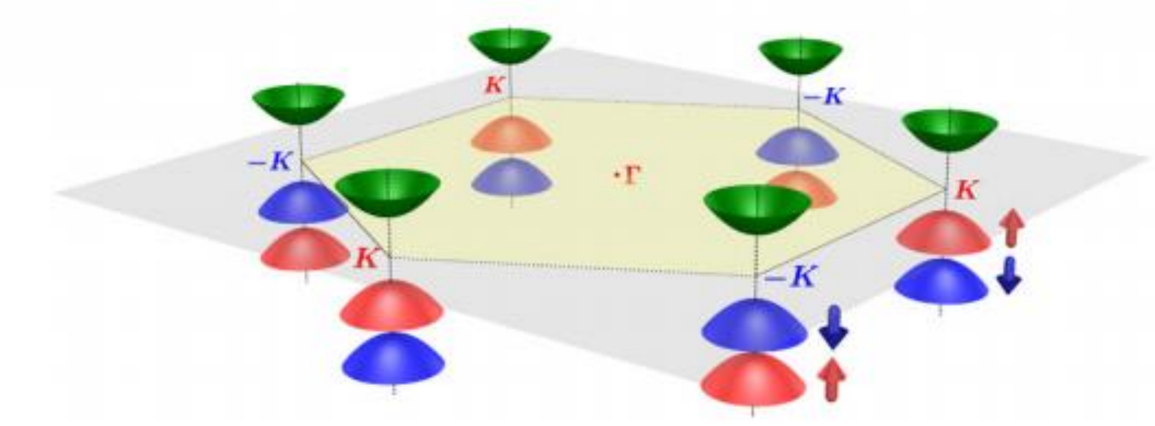


$$H = H_0 + \Delta\sigma_z + \lambda\tau_z s_z + \lambda_R(\tau_z\sigma_x s_y - \sigma_y s_x)$$

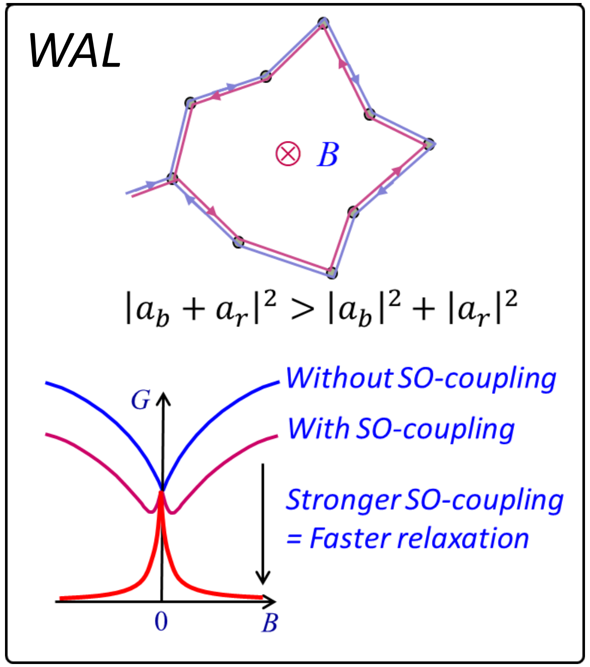
*Nat. Commun. **6**, 8339 (2015)*

*Phys. Rev. X **6**, 041020 (2016)*

$$\hat{H} = at(\tau k_x\hat{\sigma}_x + k_y\hat{\sigma}_y) + \frac{\Delta}{2}\hat{\sigma}_z - \lambda\tau\frac{\hat{\sigma}_z - 1}{2}\hat{s}_z,$$

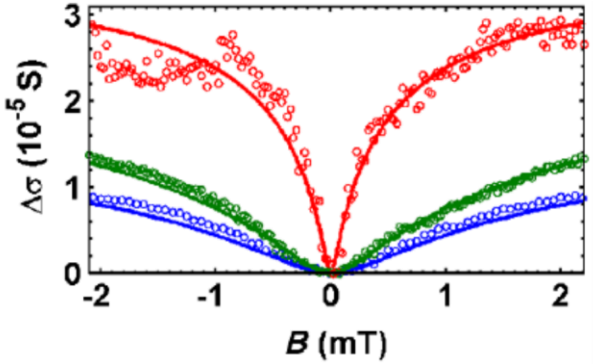


Graphene-TMDCs: evidences



$|a_b + a_r|^2 > |a_b|^2 + |a_r|^2$

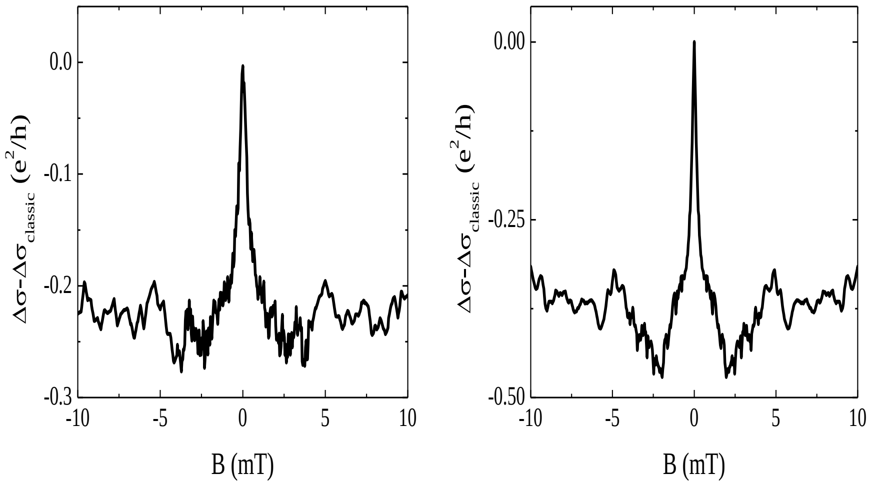
Monolayer on hBN



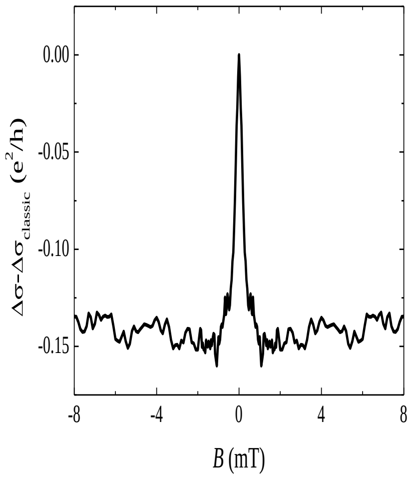
Monolayer on WSe_2

hole side

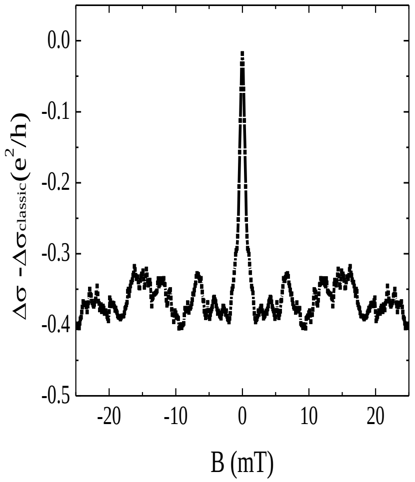
electron side



Monolayer on WS_2



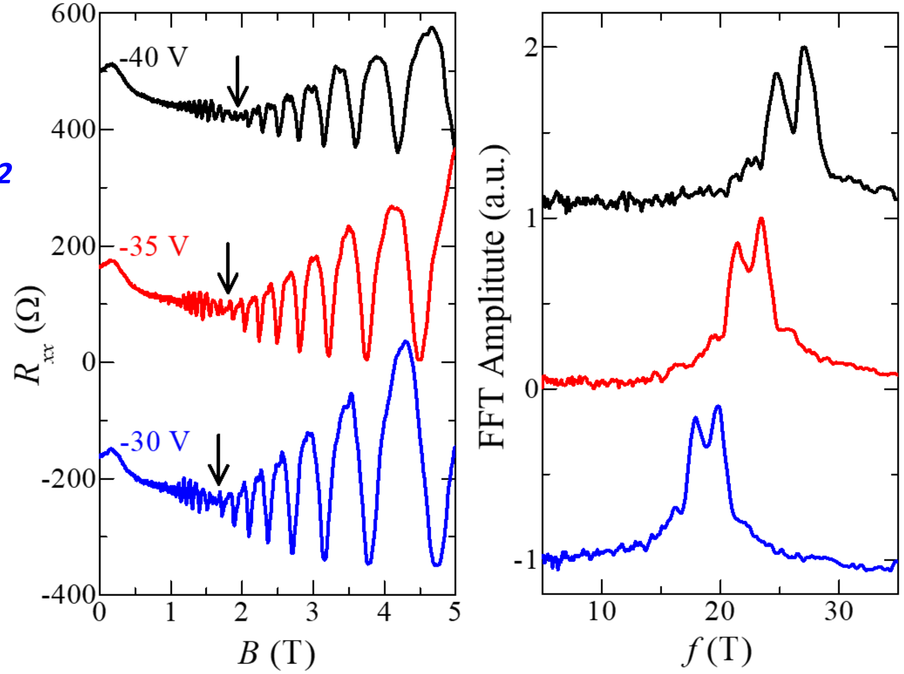
Monolayer on MoS_2



SdH

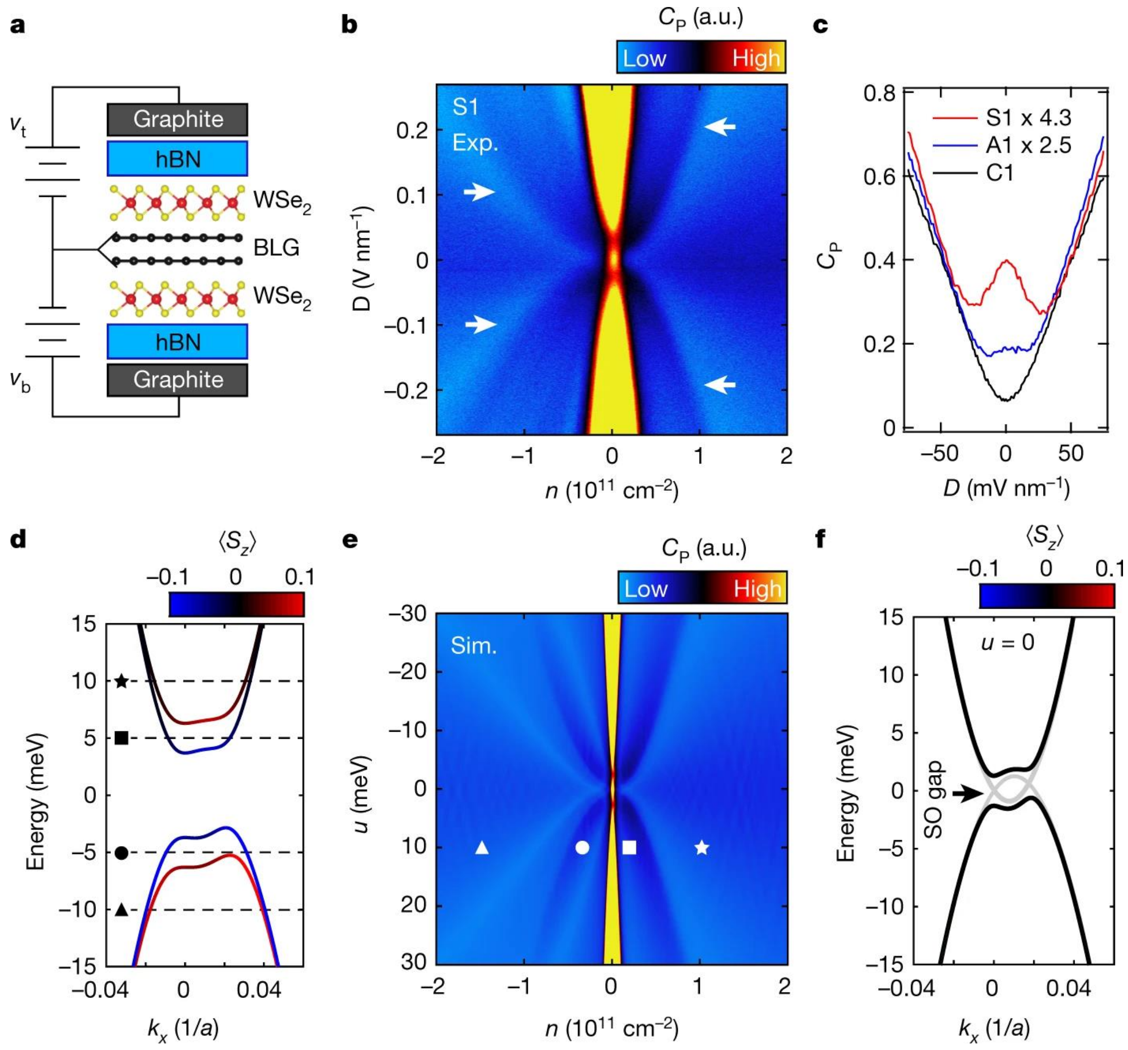
Spin-split bands due to SOC

$$\Delta\left(\frac{1}{B}\right) = \frac{2\pi e}{\hbar S}$$



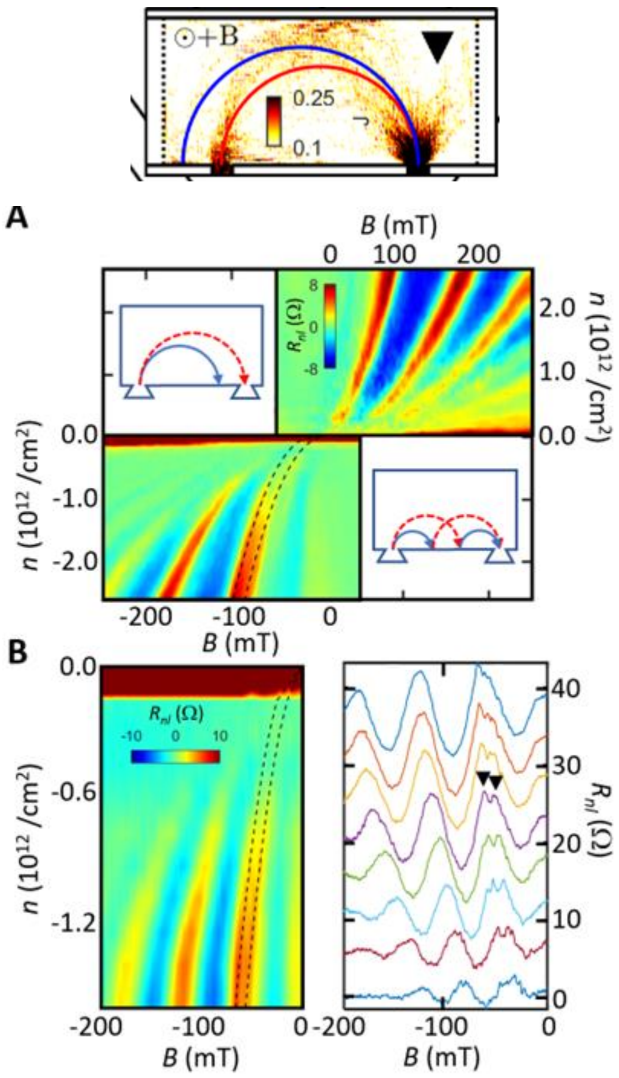
Graphene-TMDCs: evidences

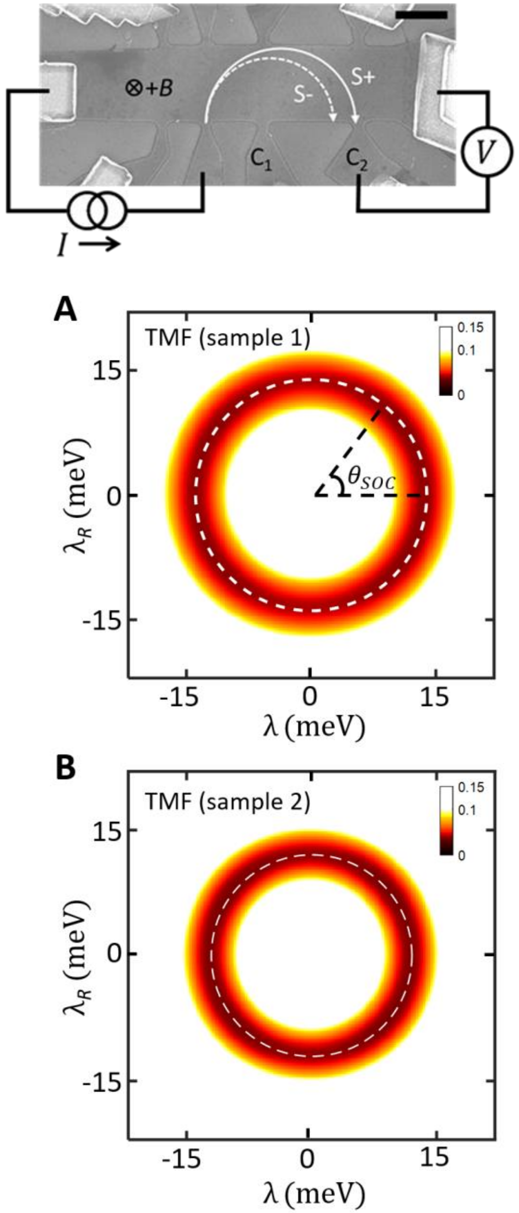
Quantum capacitance



Nature **571**, 85 (2019)

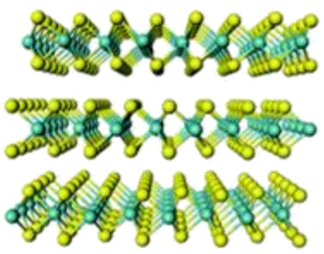
Ballistic TMF





arXiv:2303.01018 (2023).

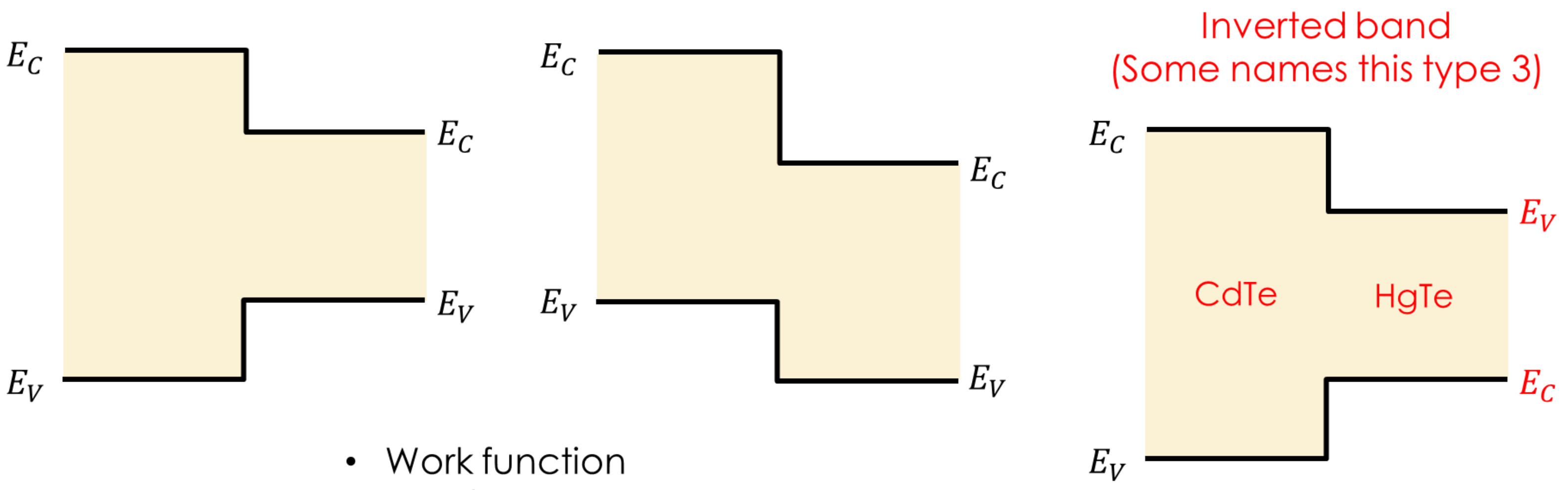
TMDC-TMDC



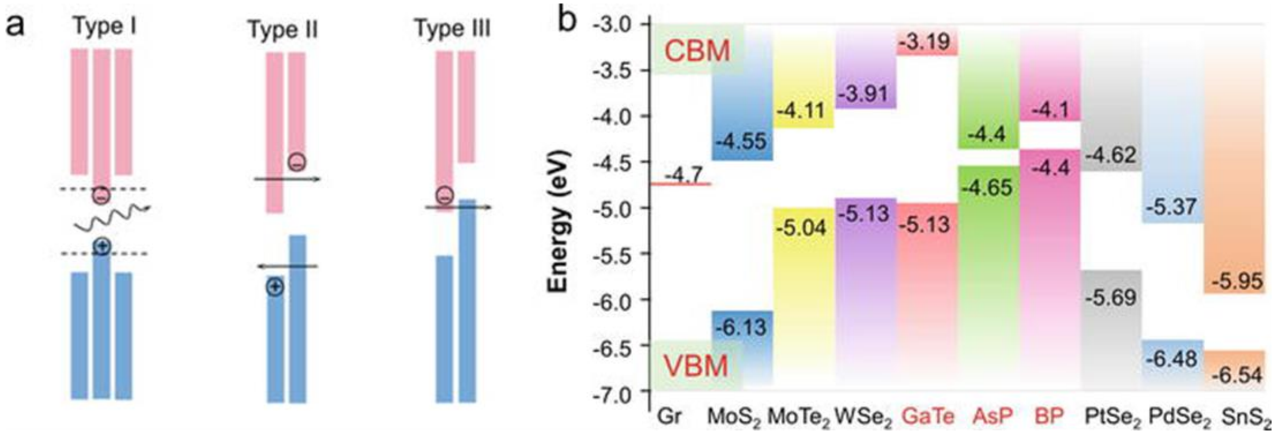
TDMCs (MX_2)

- *Semiconductor ~ 1 eV*
- *Spin-orbit coupling*

Type 1 (Symmetric) Type 2 (staggered) Type 3 (or Type 2 misaligned)



- Work function
- Band gap



10.5772/intechopen.88433

Signatures of moiré-trapped valley excitons in $MoSe_2/WSe_2$ heterobilayers

Kyle L. Seyler[1,7], Pasqual Rivera[1,7], Hongyi Yu[2], Nathan P. Wilson[1], Essance L. Ray[1], David G. Mandrus[3,4,5], Jiaqiang Yan[3,4], Wang Yao[2]* & Xiaodong Xu[1,6]*

Evidence for moiré excitons in van der Waals heterostructures

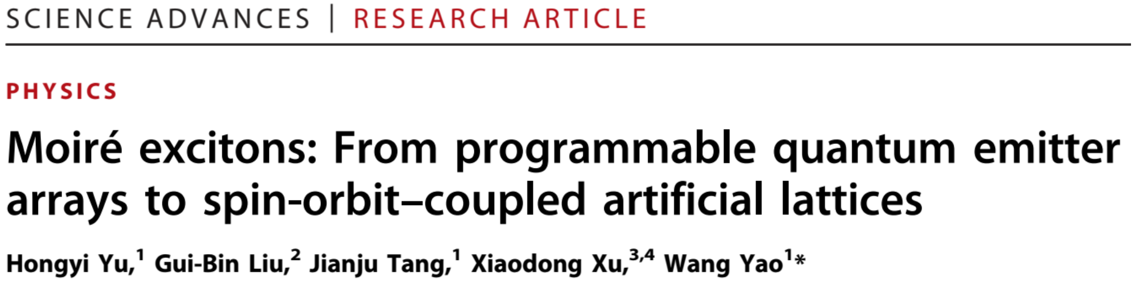
$MoSe_2/WSe_2$

Kha Tran[1,11], Galan Moody[2,11], Fengcheng Wu[3]*, Xiaobo Lu[4], Junho Choi[1], Kyounghwan Kim[5], Amritesh Rai[5], Daniel A. Sanchez[6], Jiamin Quan[1], Akshay Singh[1,10], Jacob Embley[1], André Zepeda[1], Marshall Campbell[1], Travis Autry[2], Takashi Taniguchi[7], Kenji Watanabe[7], Nanshu Lu[6,8], Sanjay K. Banerjee[5], Kevin L. Silverman[2], Suenne Kim[9], Emanuel Tutuc[5], Li Yang[4], Allan H. MacDonald[1] & Xiaoqin Li[1,6]*

Observation of moiré excitons in WSe_2/WS_2 heterostructure superlattices

Chenhao Jin[1,9], Emma C. Regan[1,2,9], Aiming Yan[1,3], M. Iqbal Bakti Utama[1,4], Danqing Wang[1,2], Sihan Zhao[1], Ying Qin[5], Sijie Yang[5], Zhiren Zheng[1], Shenyang Shi[1,6], Kenji Watanabe[7], Takashi Taniguchi[7], Sefaattin Tongay[5], Alex Zettl[1,3,8] & Feng Wang[1,3,8]*

TMDC-TMDC: moiré excitons



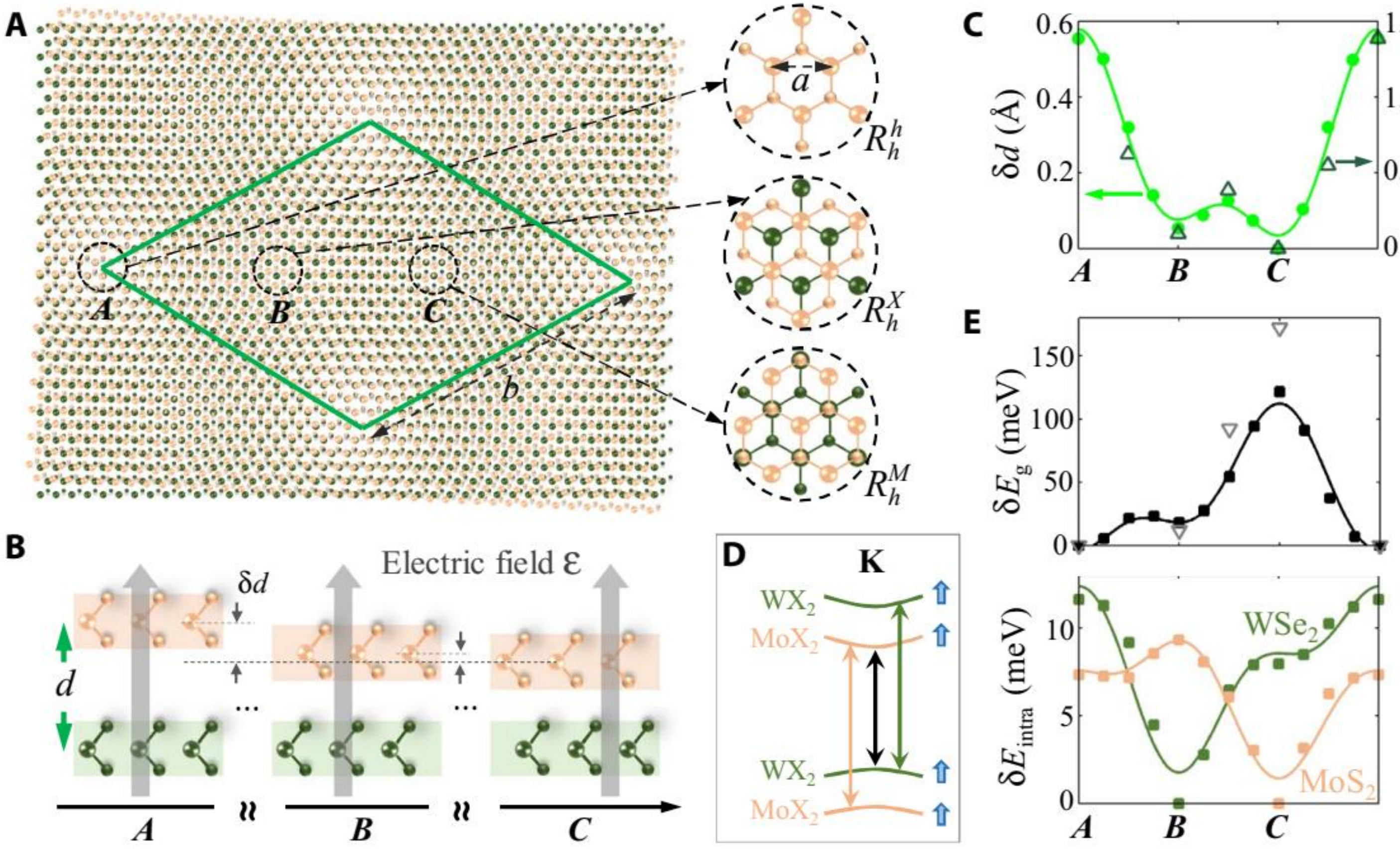
SCIENCE ADVANCES | RESEARCH ARTICLE

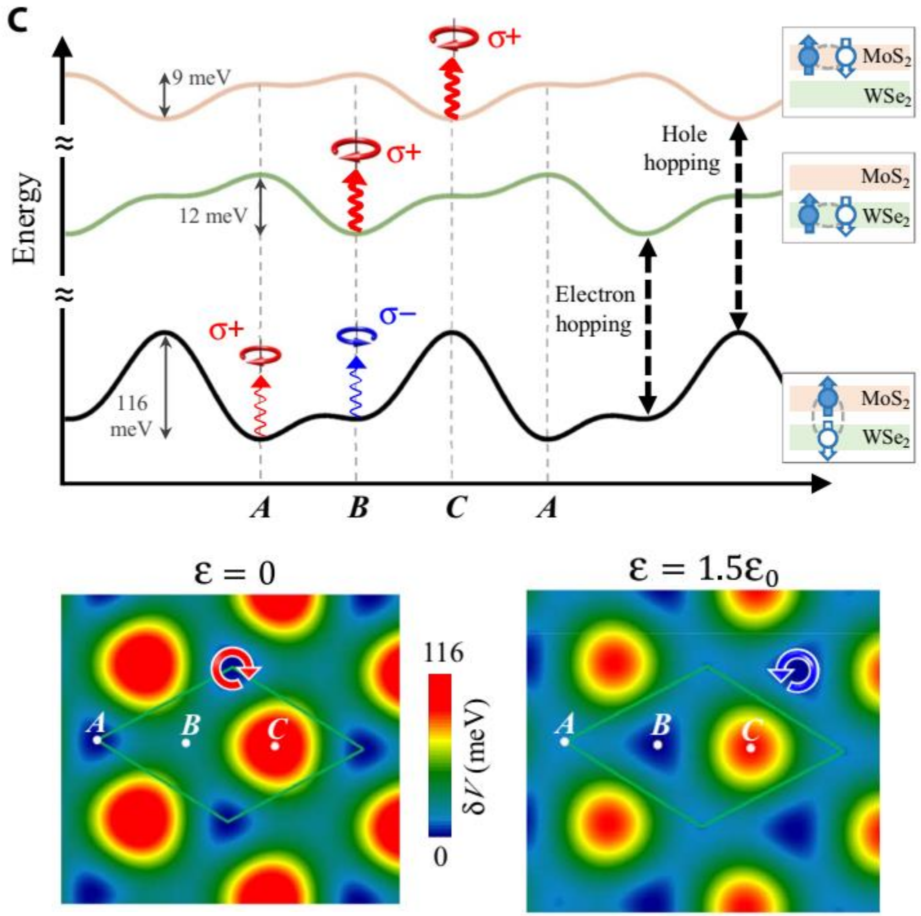
PHYSICS

Moiré excitons: From programmable quantum emitter arrays to spin-orbit–coupled artificial lattices

Hongyi Yu,[1] Gui-Bin Liu,[2] Jianju Tang,[1] Xiaodong Xu,[3,4] Wang Yao[1]*

- Type II configuration: favorable interlayer exciton
- Electric field control possible
- Quantum emitter





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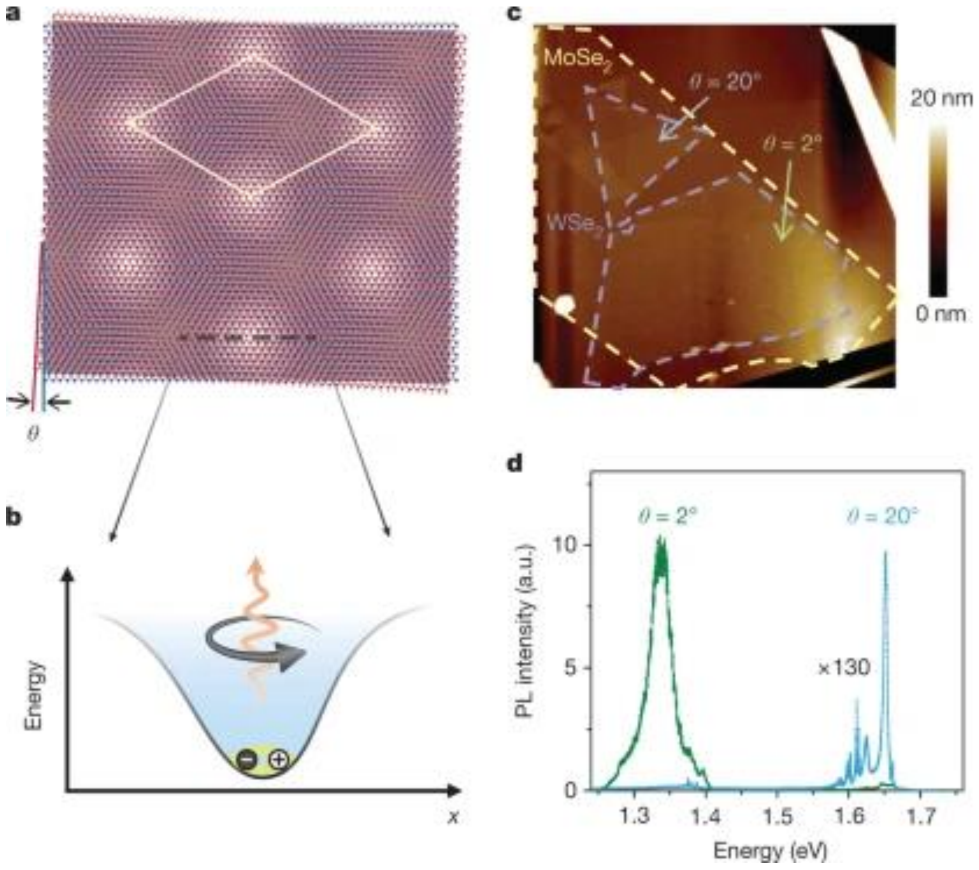
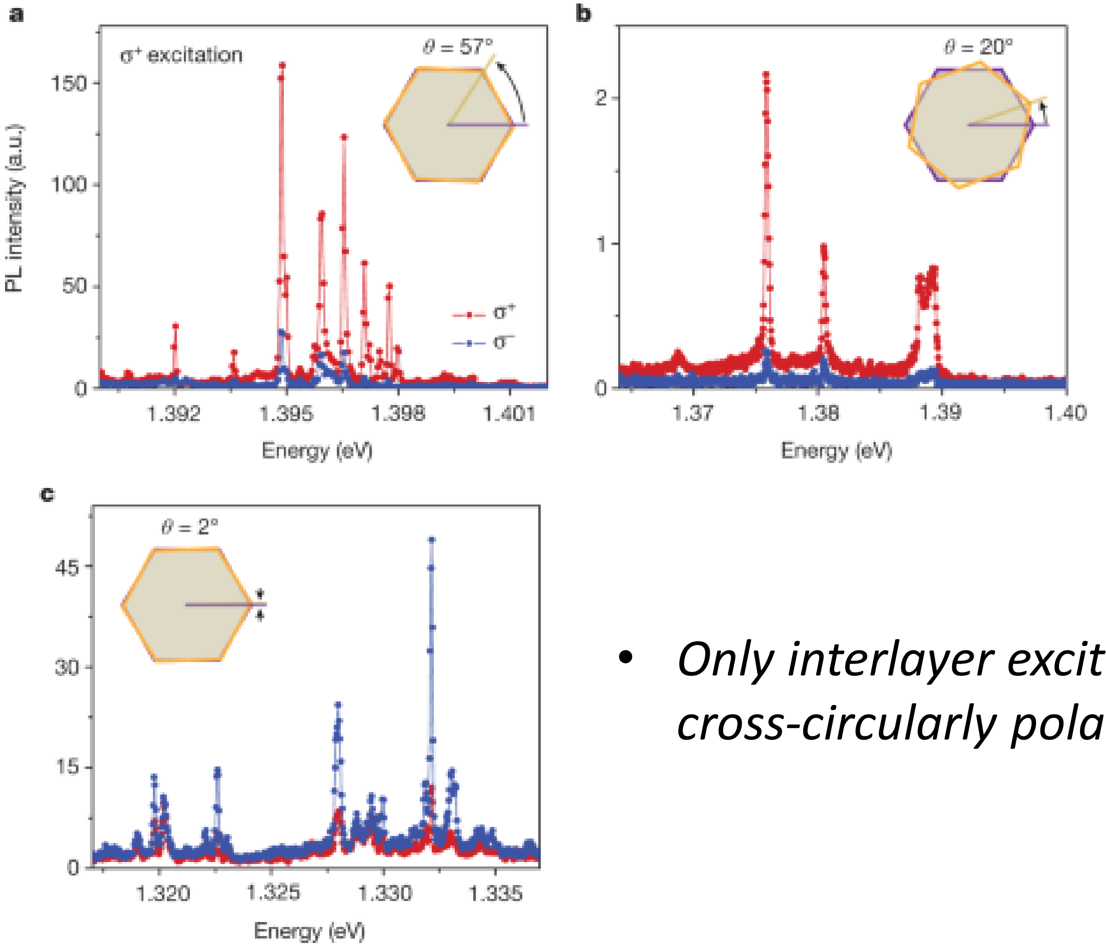
- *When moiré lattice is larger than ~ 1 nm (the size of an exciton), the exciton will experience moiré superlattice potential.*





- *Only interlayer excitons show cross-circularly polarized PL*

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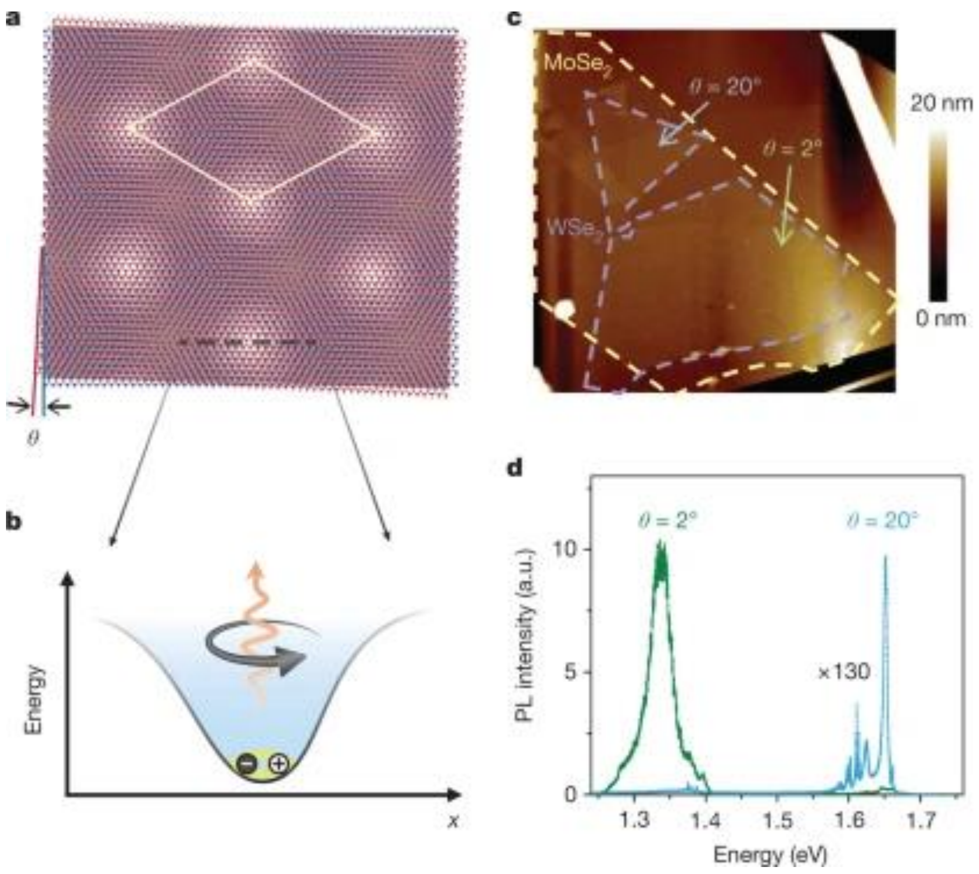
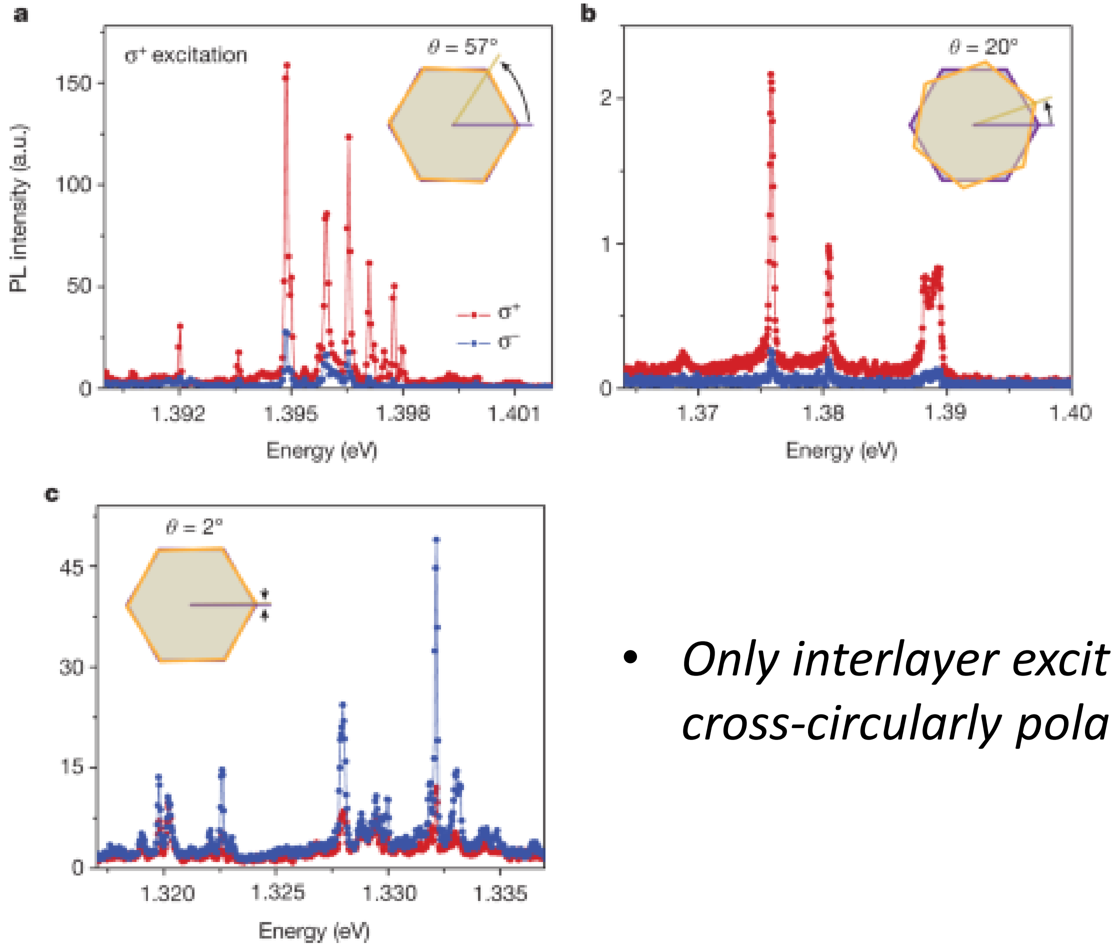
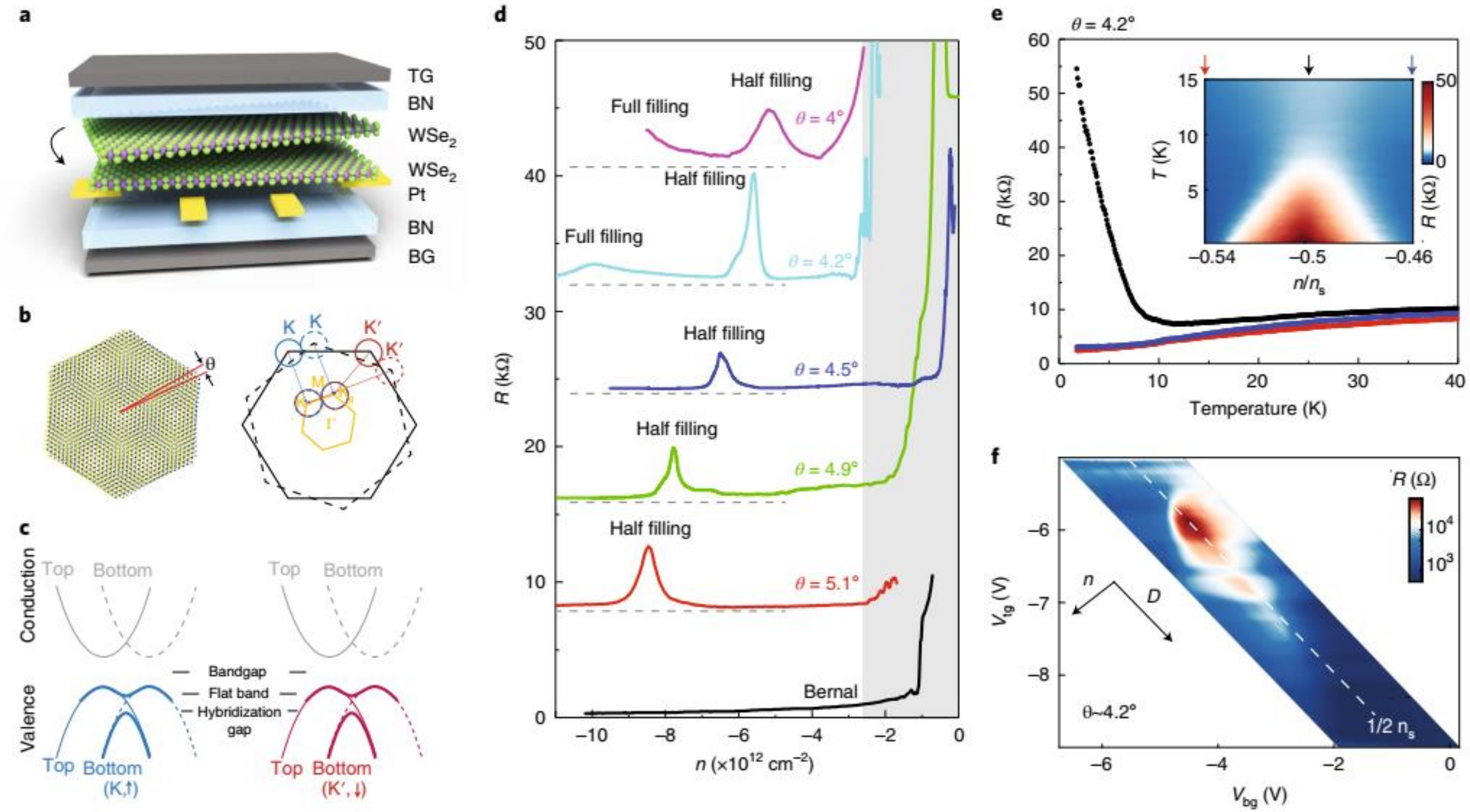


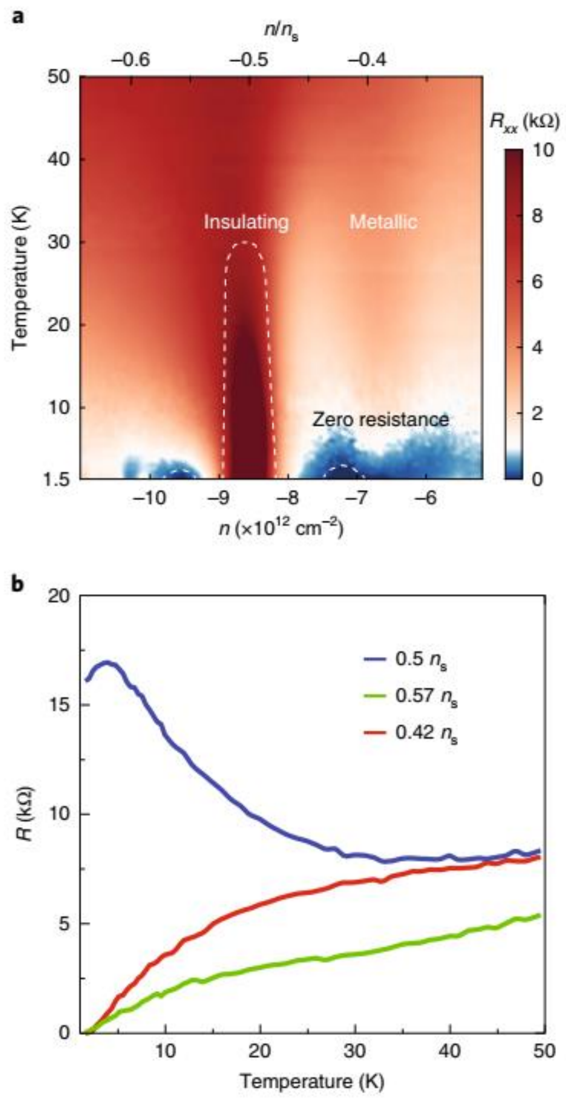


- *Only interlayer excitons show cross-circularly polarized PL*

TMDC-TMDC: correlated states

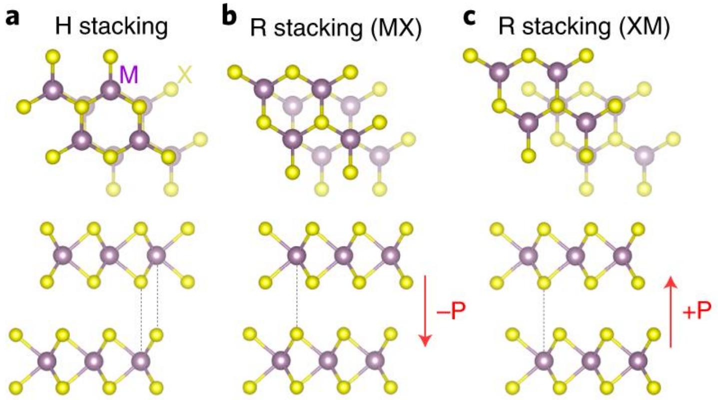
Nature Materials 19, 861 (2020)

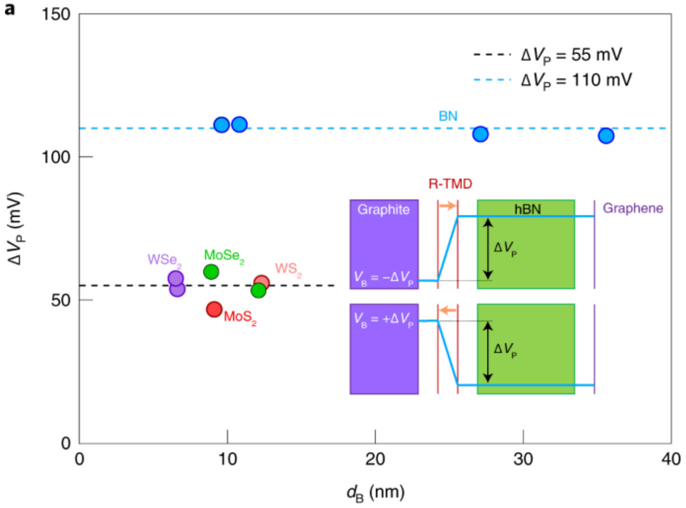




TMDC-TMDC: ferroelectricity

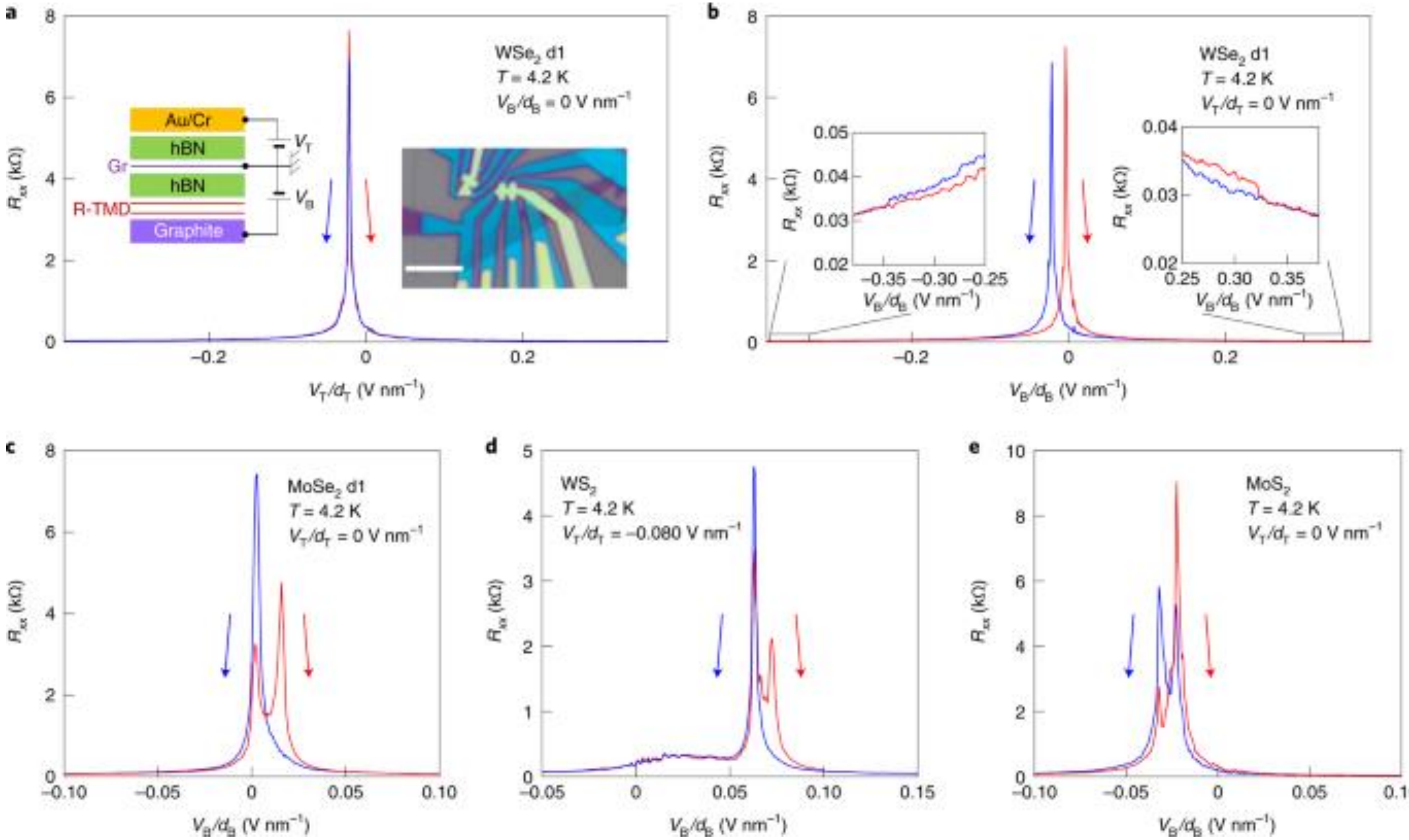
Nature Nanotech. 17, 367 (2022)



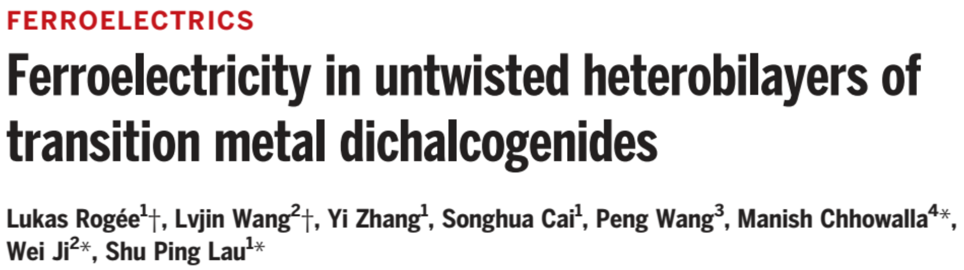


b

Material	BN	WSe_2	$MoSe_2$	WS_2	MoS_2
ΔV_P (mV), experiment	109 (2)	56 (3)	57 (5)	56	47
ΔV_P (mV), theory	100	56	66	66	64



TMDC-TMDC: ferroelectricity without twisting

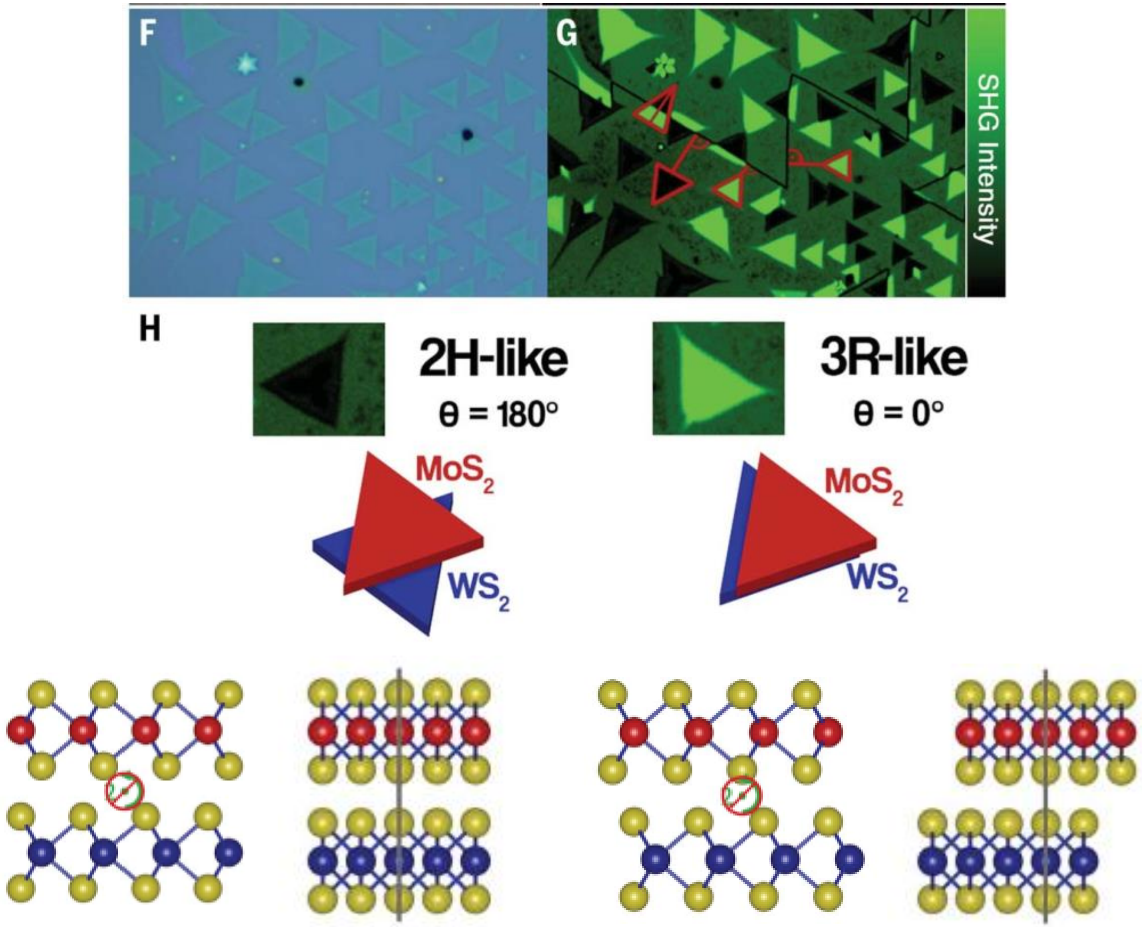


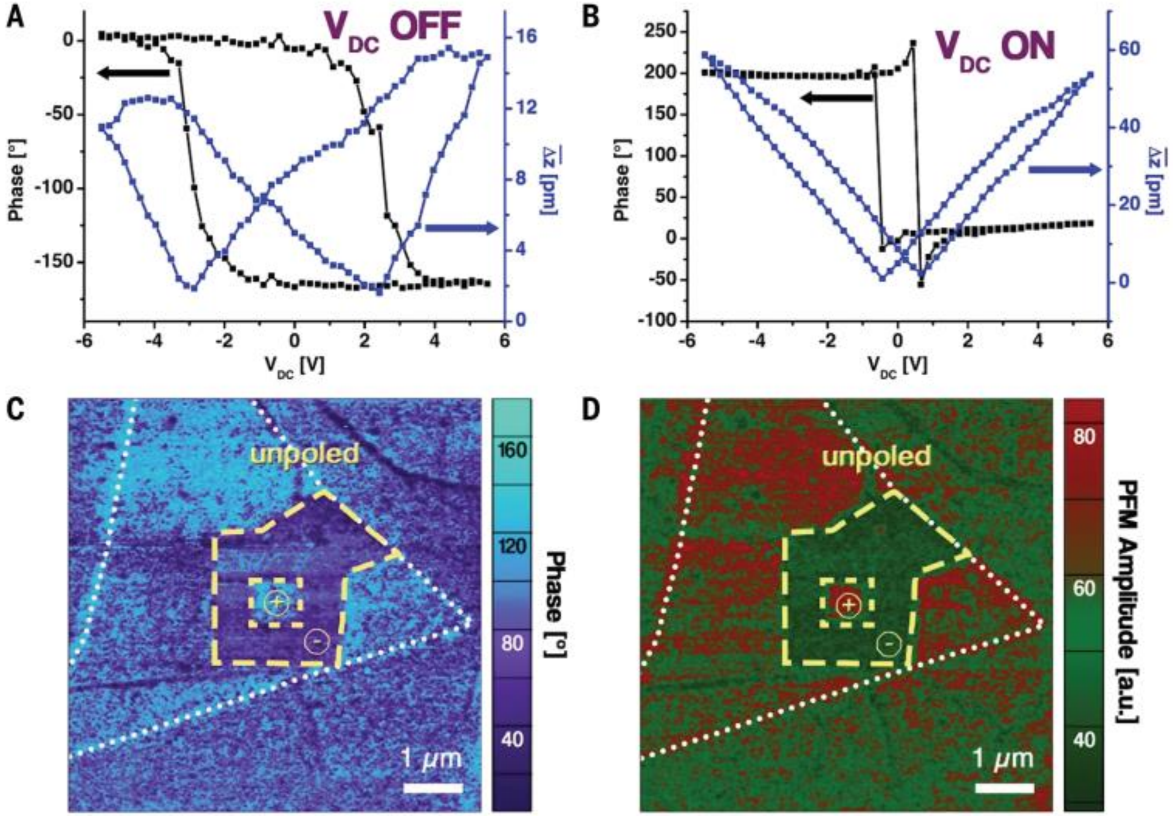
FERROELECTRICS

Ferroelectricity in untwisted heterobilayers of transition metal dichalcogenides

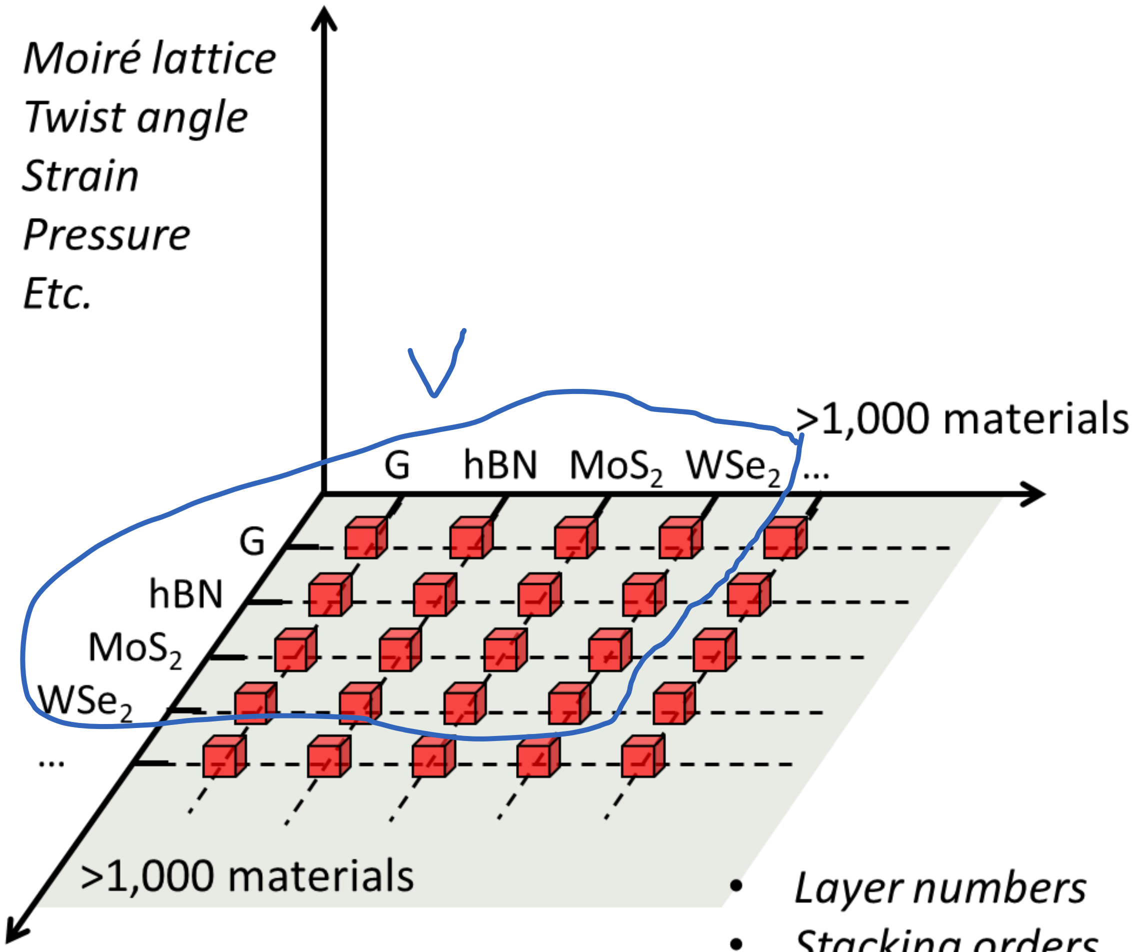
Lukas Rogée[1]†, Lvjin Wang[2]†, Yi Zhang[1], Songhua Cai[1], Peng Wang[3], Manish Chhowalla[4]*, Wei Ji[2]*, Shu Ping Lau[1]*

- No twisting
- CVD grown





What we have learnt so far.



- *Layer numbers*
- *Stacking orders*
- *Dimensions*

- Graphene-TMDC
  - Weak vdW interaction allows to induce strong SOC in graphene without affecting most of the graphene’s properties

- TMDC-TMDC
  - Can create artificial semiconductors
  - Moiré excitons
  - Ferroelectricity

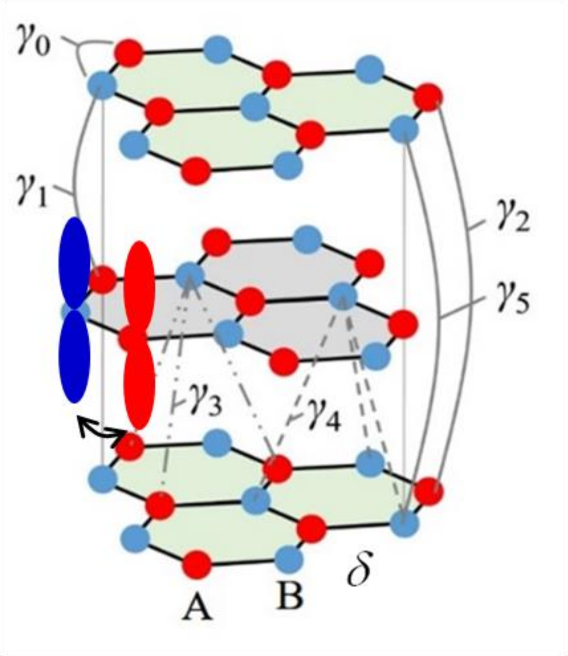
    - 2+2 TMDCs
    - Twisted BLG + TMDC
    - TMDC-TMDC without moiré
    - And more….

- *Carriers move between atomic orbitals:* ***atomic registry/potential is important***

Summary

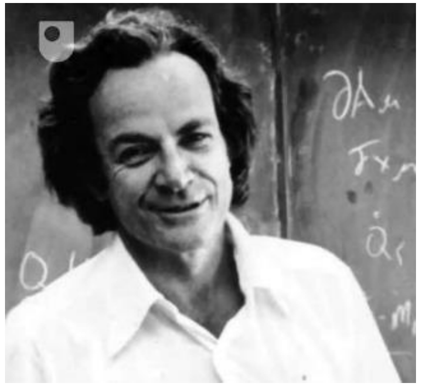
- *What are these layers?*
- *Why are they so special?*
- *What are the fundamental mechanisms that create new properties?*

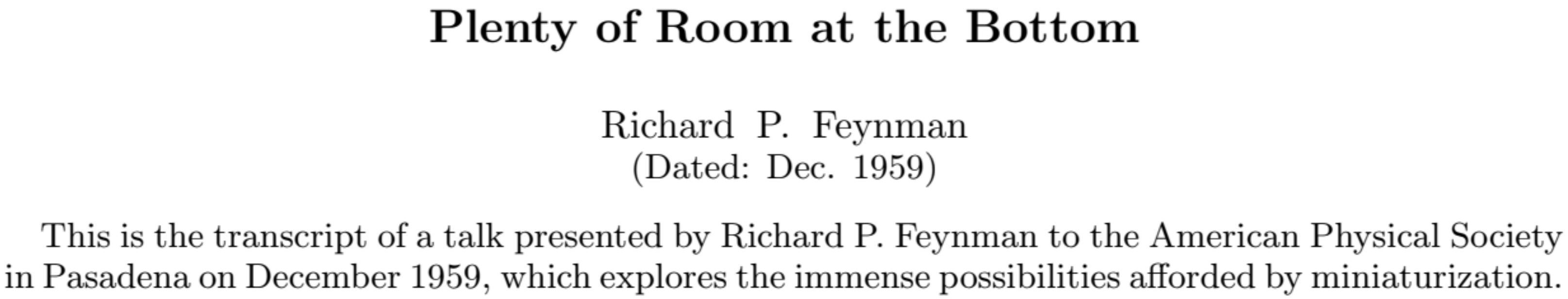
van der Waals coupled layered materials → defect-free monolayers



- *Carriers move between atomic orbitals:* ***atomic registry/potential is important (chemical composition)***
- *Carriers move at the atomically flat surface:* ***highly sensitive to the environment***
- *Surface is defect free in principle:* ***no dangling bonds and strong vdW interactions***

Key questions to answer

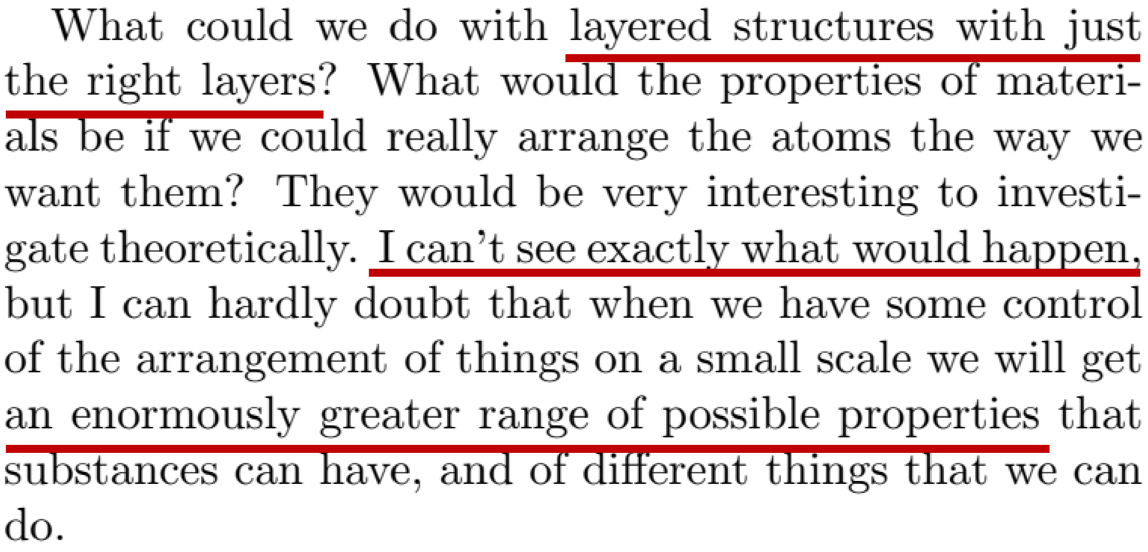




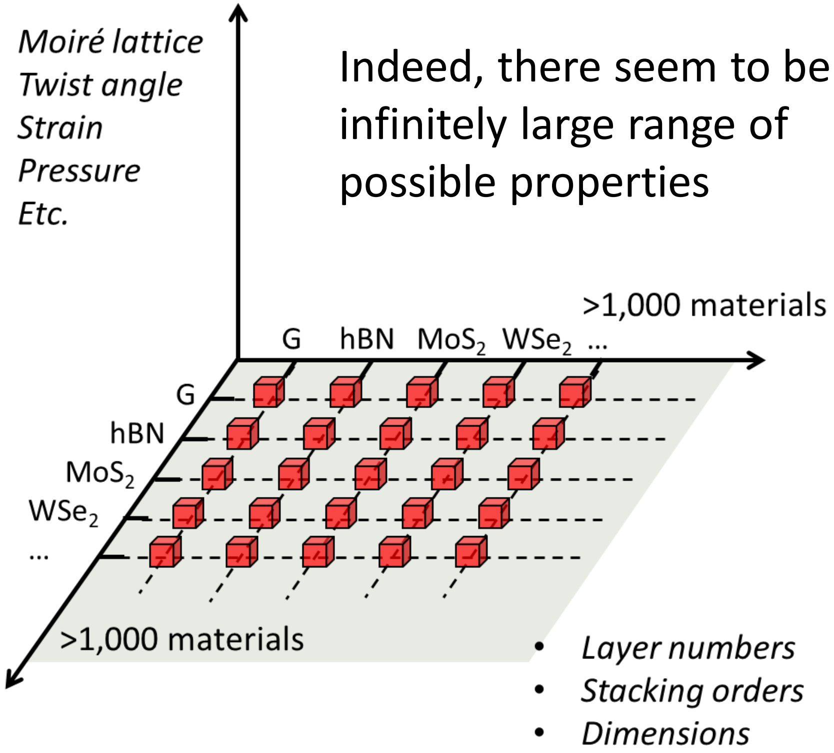
Plenty of Room at the Bottom

Richard P. Feynman
(Dated: Dec. 1959)

This is the transcript of a talk presented by Richard P. Feynman to the American Physical Society in Pasadena on December 1959, which explores the immense possibilities afforded by miniaturization.



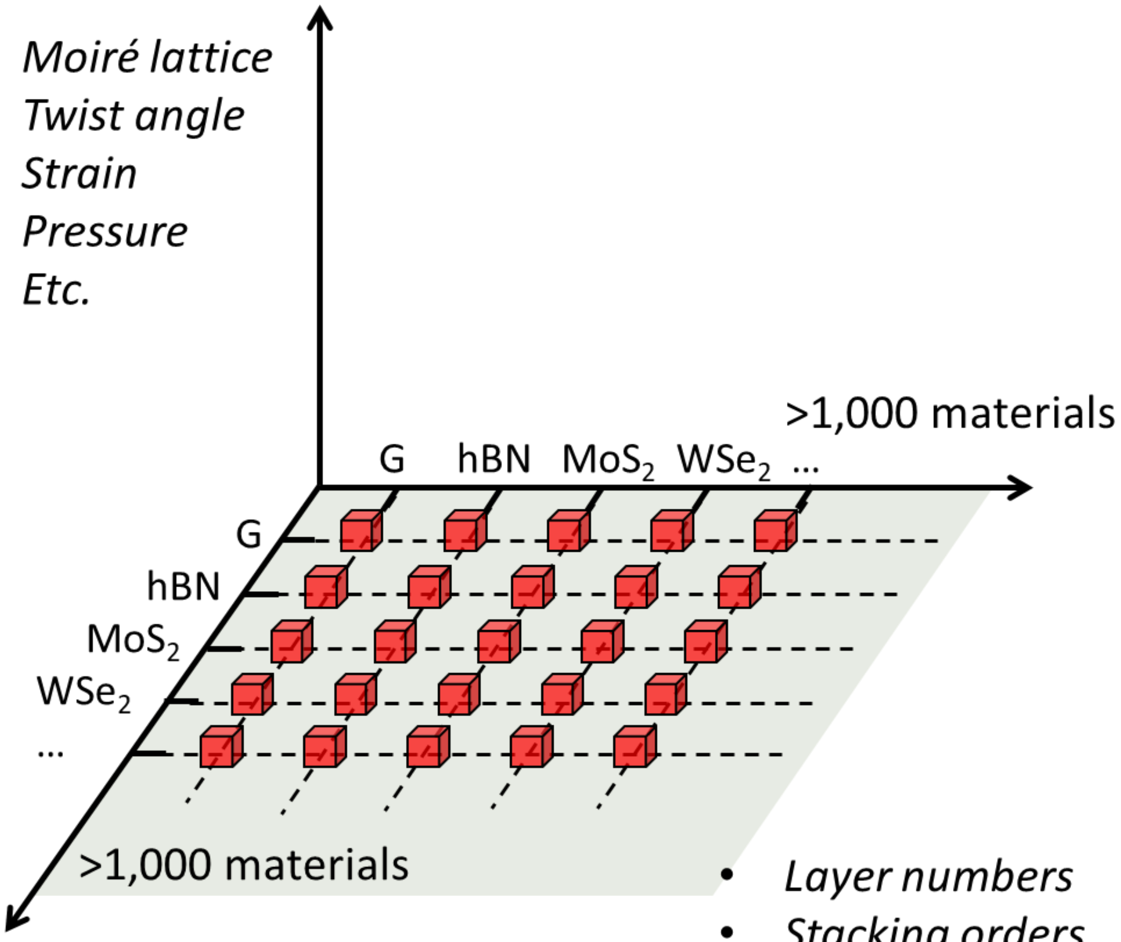
What could we do with layered structures with just the right layers? What would the properties of materials be if we could really arrange the atoms the way we want them? They would be very interesting to investigate theoretically. I can't see exactly what would happen, but I can hardly doubt that when we have some control of the arrangement of things on a small scale we will get an enormously greater range of possible properties that substances can have, and of different things that we can do.



Indeed, there seem to be infinitely large range of possible properties

https://web.pa.msu.edu/people/yang/RFeynman_plentySpace.pdf

New challenges?



- *Layer numbers*
- *Stacking orders*
- *Dimensions*

- *Developments in computational studies*
  - *Too many possible combinations*
  - *Large lattice size in moiré structures*

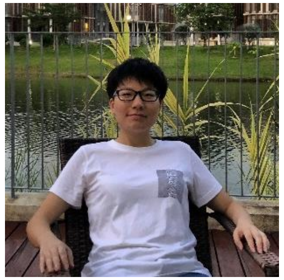
- *Developments of in-situ manipulation methods*
  - *Changing moiré potential continuously after the structure has been made*
  - *In-situ strain control*

- *Developments of new measurement techniques to detect internal degrees of freedom*
  - *Spin, valley, and layer quantum numbers*
  - *Rotation angle, local strain, etc.*

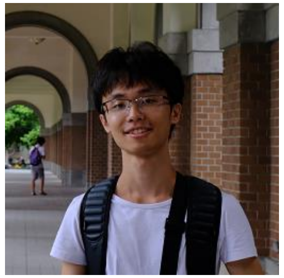
- *Condensation of quasiparticles in vdW heterostructures*

Thank you

Qing Rao



Tianyu Zhang



Dr. Hongxia Xue

Yueyang Wang

Xinyu Wang

- *AoE on 2D materials*
- *MoST National Key R&D Program*
- *GRF and ECS grants*
- *Start-up and Various Seed Funds from HKU*

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Graphene-hBN heterostructures: moiré structure

$$\mathcal{H}_{\text{G-hBN}} = \begin{pmatrix} H_{\text{G}} & U^{\dagger} \\ U & H_{\text{hBN}} \end{pmatrix}, \qquad (12)$$

$(A1, B1, A2, B2)$

A1, B1: Graphene

A2, B2: hBN

with

$$U = \begin{pmatrix} U_{A_2A_1} & U_{A_2B_1} \\ U_{B_2A_1} & U_{B_2B_1} \end{pmatrix} = u_0\left[\begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} + \begin{pmatrix} 1 & \omega^{-\xi} \\ \omega^{\xi} & 1 \end{pmatrix} e^{i\xi \mathbf{G}_1^{\text{M}}\cdot\mathbf{r}} + \begin{pmatrix} 1 & \omega^{\xi} \\ \omega^{-\xi} & 1 \end{pmatrix} e^{i\xi(\mathbf{G}_1^{\text{M}}+\mathbf{G}_2^{\text{M}})\cdot\mathbf{r}}\right], \qquad (13)$$

$$\omega = e^{2\pi i/3}$$

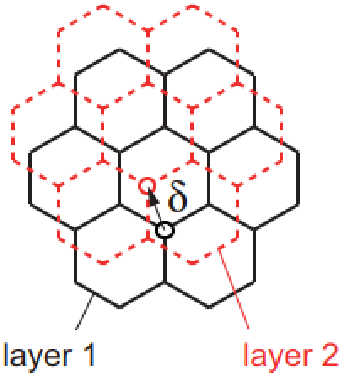
$$\xi = \pm 1$$

$$H_{\text{G}} \approx -\hbar v \mathbf{k}\cdot\boldsymbol{\sigma}_{\xi},$$

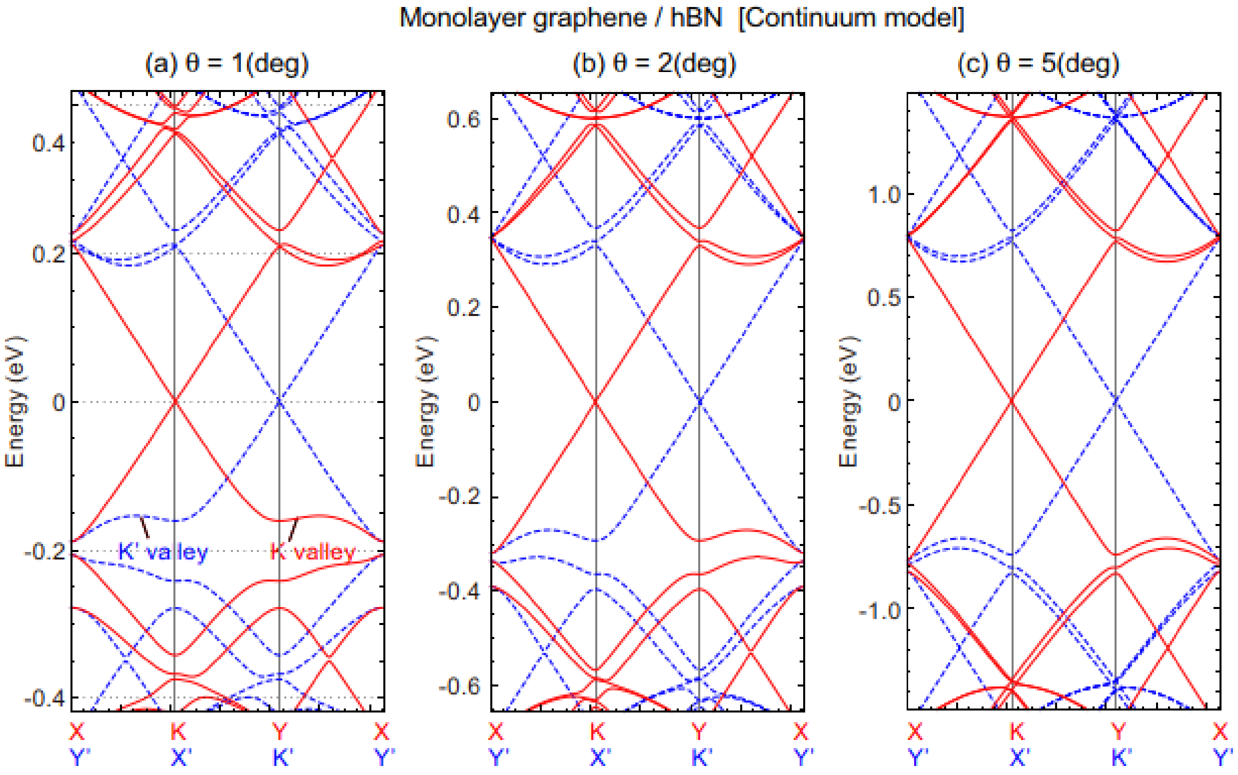
$$H_{\text{hBN}} \approx \begin{pmatrix} V_{\text{N}} & 0 \\ 0 & V_{\text{B}} \end{pmatrix}.$$

$$H_{G-hBN} = H_G + V_{hBN}$$

$$V_{\text{hBN}} = V^{\text{eff}}(\mathbf{r}) + M^{\text{eff}}(\mathbf{r})\sigma_z + ev\mathbf{A}^{\text{eff}}(\mathbf{r})\cdot\boldsymbol{\sigma}_{\xi}.$$



$$U_{A_2A_1}(\mathbf{k},\boldsymbol{\delta}) \equiv \langle \mathbf{k}, A_2|H|\mathbf{k}, A_1\rangle = u(\mathbf{k},\boldsymbol{\delta}),$$
$$U_{B_2B_1}(\mathbf{k},\boldsymbol{\delta}) \equiv \langle \mathbf{k}, B_2|H|\mathbf{k}, B_1\rangle = u(\mathbf{k},\boldsymbol{\delta}),$$
$$U_{B_2A_1}(\mathbf{k},\boldsymbol{\delta}) \equiv \langle \mathbf{k}, B_2|H|\mathbf{k}, A_1\rangle = u(\mathbf{k},\boldsymbol{\delta}-\boldsymbol{\tau}_1),$$
$$U_{A_2B_1}(\mathbf{k},\boldsymbol{\delta}) \equiv \langle \mathbf{k}, A_2|H|\mathbf{k}, B_1\rangle = u(\mathbf{k},\boldsymbol{\delta}+\boldsymbol{\tau}_1),$$



PRB 90, 155406 (2014)