

Electrical switching of spin and valley in spin-orbit coupled graphene multilayers

Shubhayu Chatterjee
Carnegie Mellon University

SPICE Workshop

Hybrid Correlated States and Dynamics in Quantum Materials

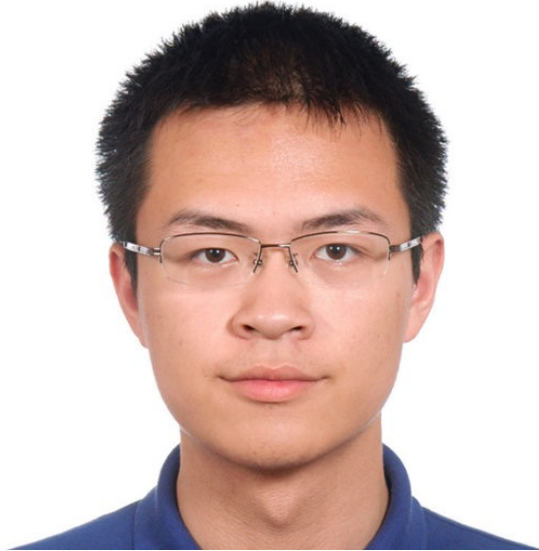
Ingelheim

May 14, 2024

$\langle P|Q|I \rangle$



In collaboration with:



Taige Wang
UC Berkeley



Marc Vila Tusell
UC Berkeley

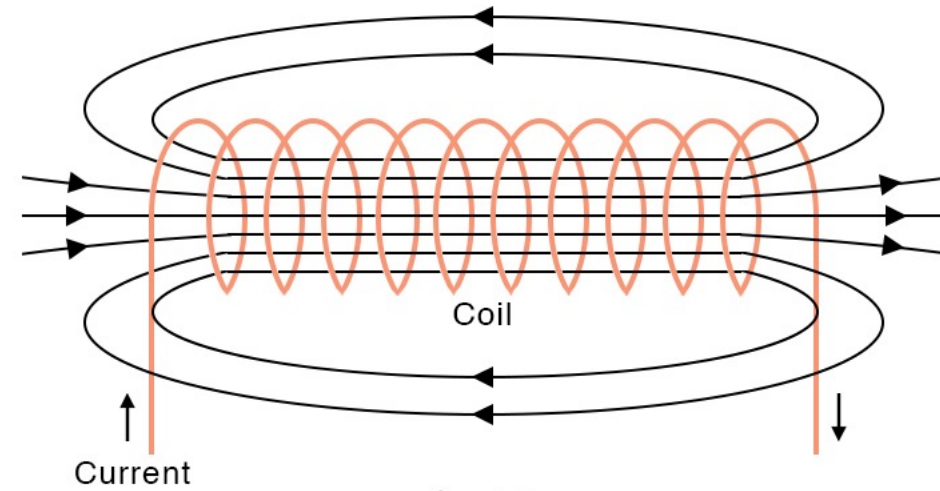


Mike Zaletel
UC Berkeley

T. Wang*, M. Vila Tusell*, M. P. Zaletel and SC, PRL **132**, 116504 (2024)

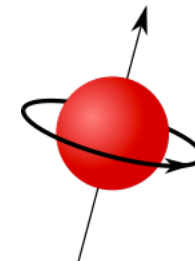
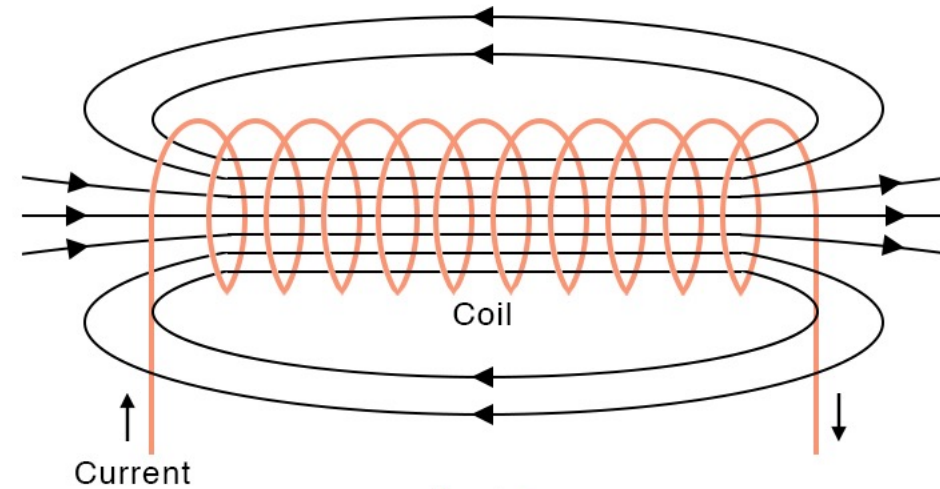
Electric field control of magnetism

- Magnetism in materials is typically controlled by magnetic fields, sourced by currents in a solenoid



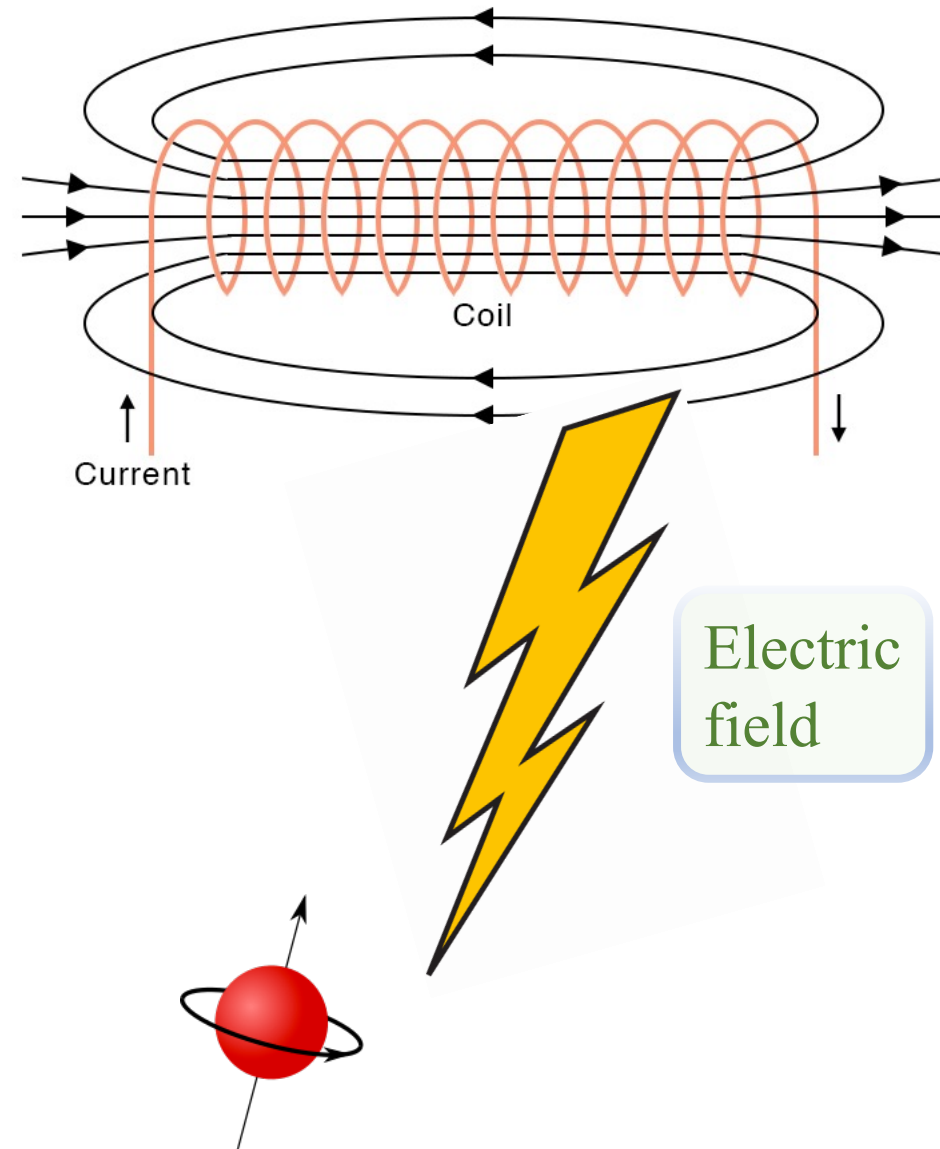
Electric field control of magnetism

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- Electrical control of magnetism – longstanding goal of the spintronics community
- Promise for energy-efficient next generation devices



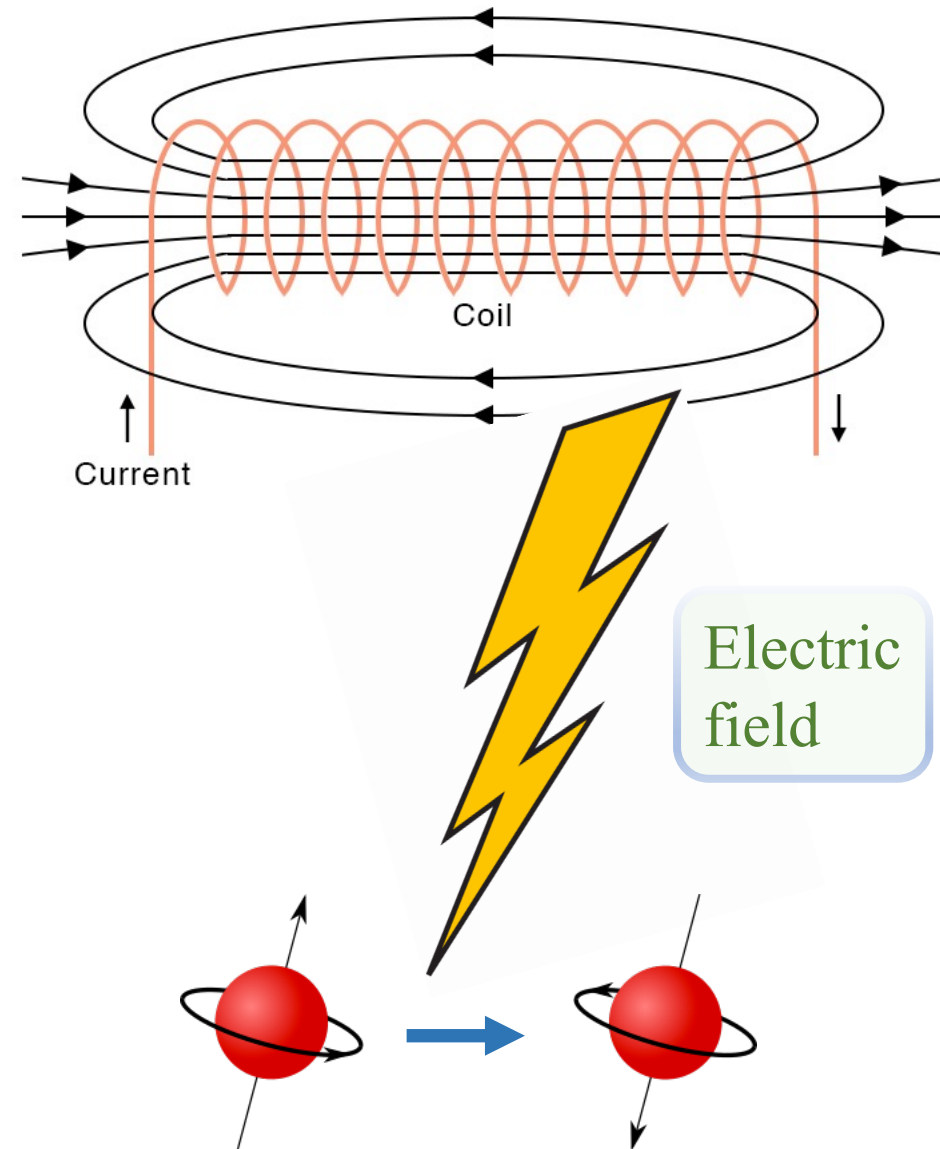
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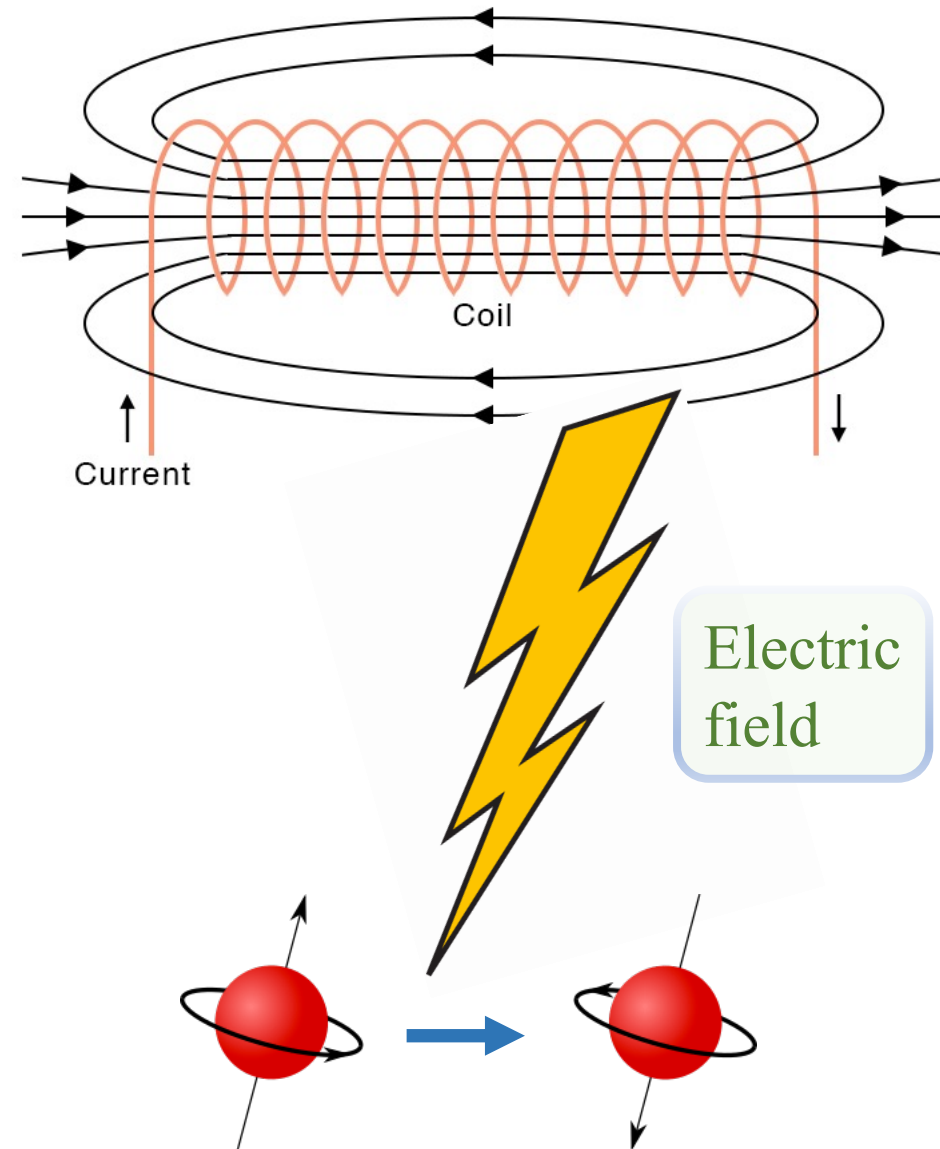
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Electric field control of magnetism

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- Electrical control of magnetism – longstanding goal of the spintronics community
- Promise for energy-efficient next generation devices
- Potential barrier – Magnetism originates in materials from a combination of Fermi statistics and repulsive Coulomb interaction
- Difficult to control *directly* via electric fields



2D materials: A Quantum Lego kit

- Atomically thin two dimensional materials have recently emerged as highly tunable and versatile platforms
- Distinct material properties from different stacking

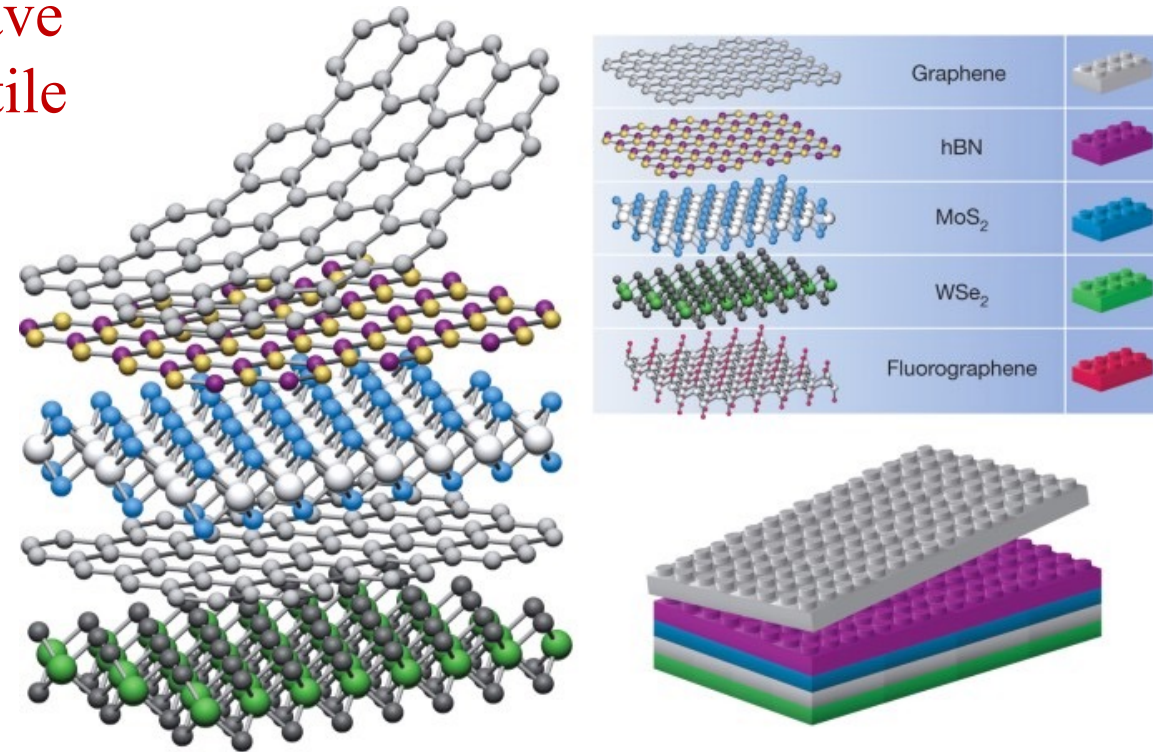


Figure: Geim, Grigorieva, Nature (2019)

2D materials: A Quantum Lego kit

- Atomically thin two dimensional materials have recently emerged as highly tunable and versatile platforms
 1. Vary carrier density via metallic gate
 2. Vary interaction strength by dielectric substrate

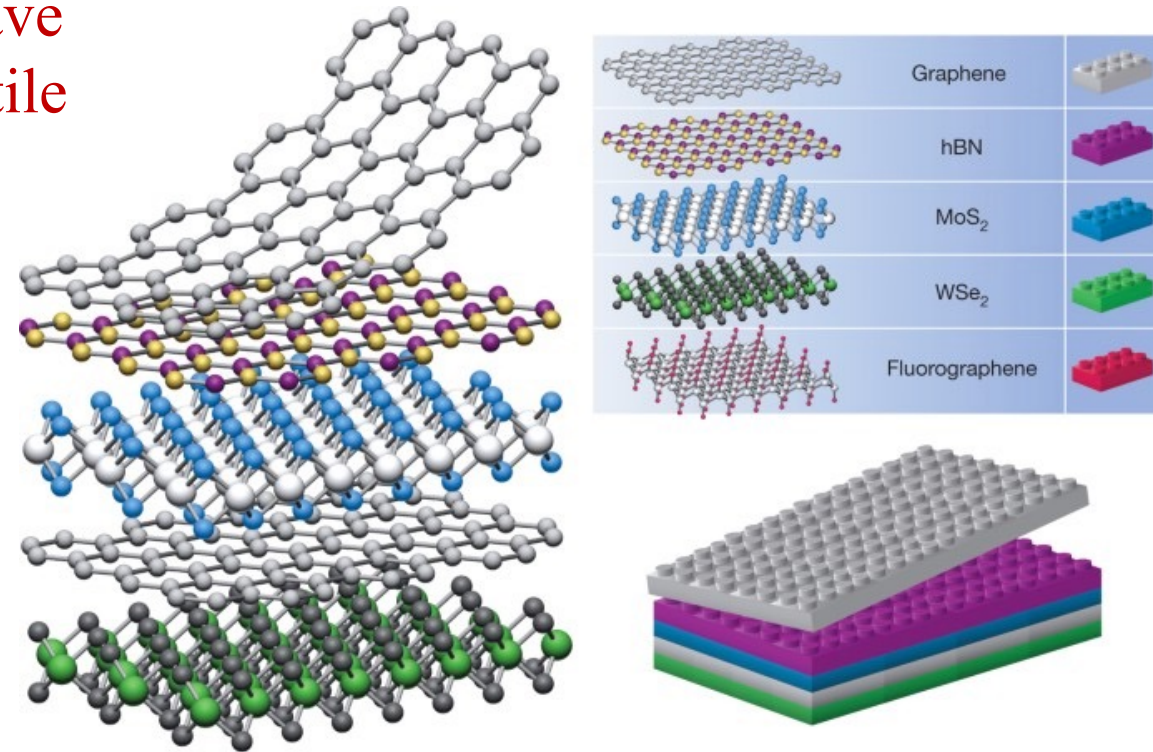


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2D materials: A Quantum Lego kit

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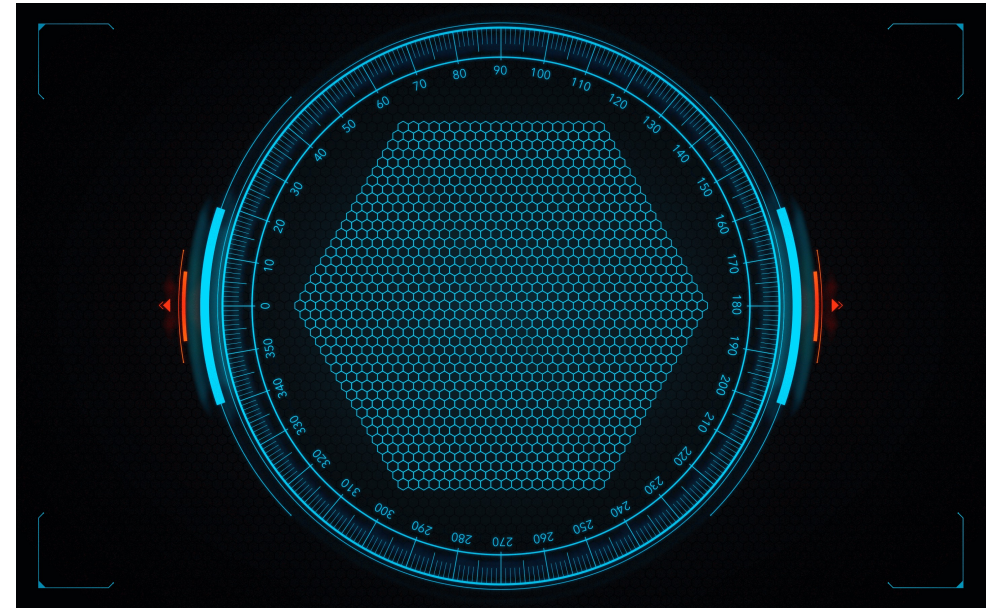


Figure: Quanta Magazine

2D materials: A Quantum Lego kit

- Atomically thin two dimensional materials have recently emerged as highly tunable and versatile platforms
 1. Vary carrier density via metallic gate
 2. Vary interaction strength by dielectric substrate
 3. *Twist angle* between layers – moiré materials (e.g., twisted bilayer graphene)
 4. Perpendicular **E** fields (*displacement fields*) to flatten bands (e.g., ABC trilayer graphene)

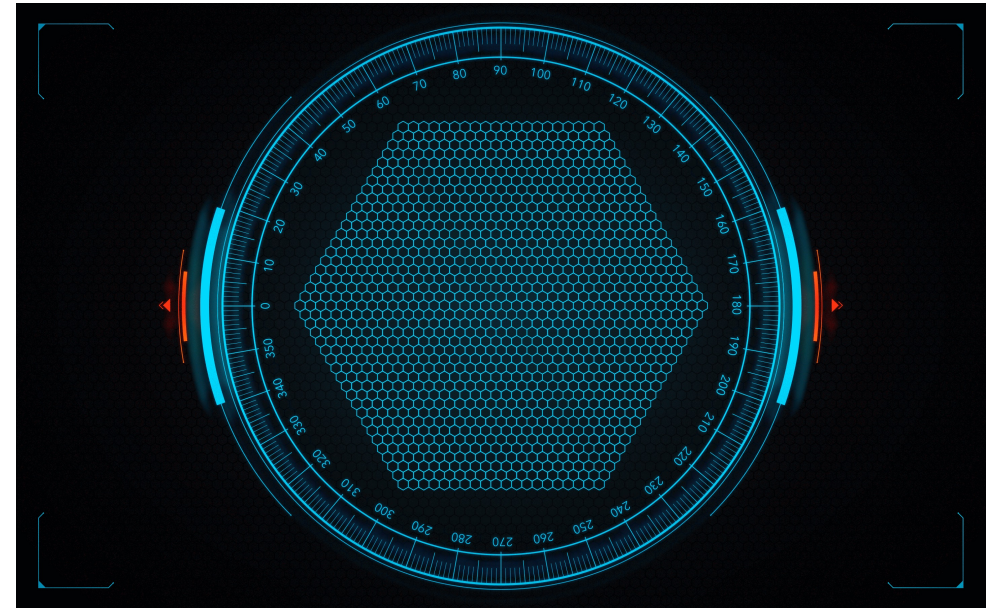


Figure: Quanta Magazine

Ferromagnets in Van der Waals materials

- Interaction is enhanced relative to bandwidth ($t/U \ll 1$)
- Different forms of *hybrid correlated states* present, e.g., orbital (Chern) ferromagnets, spin polarized ferromagnets

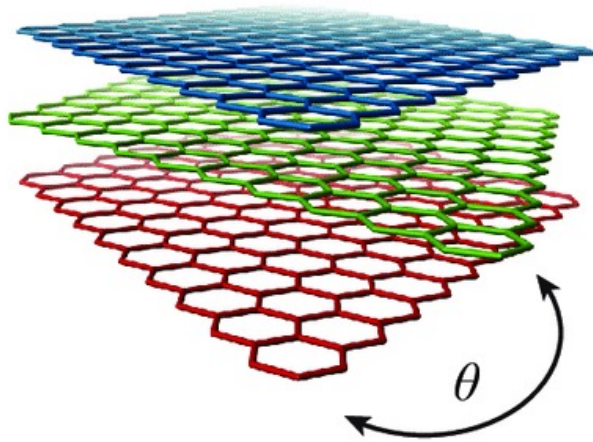
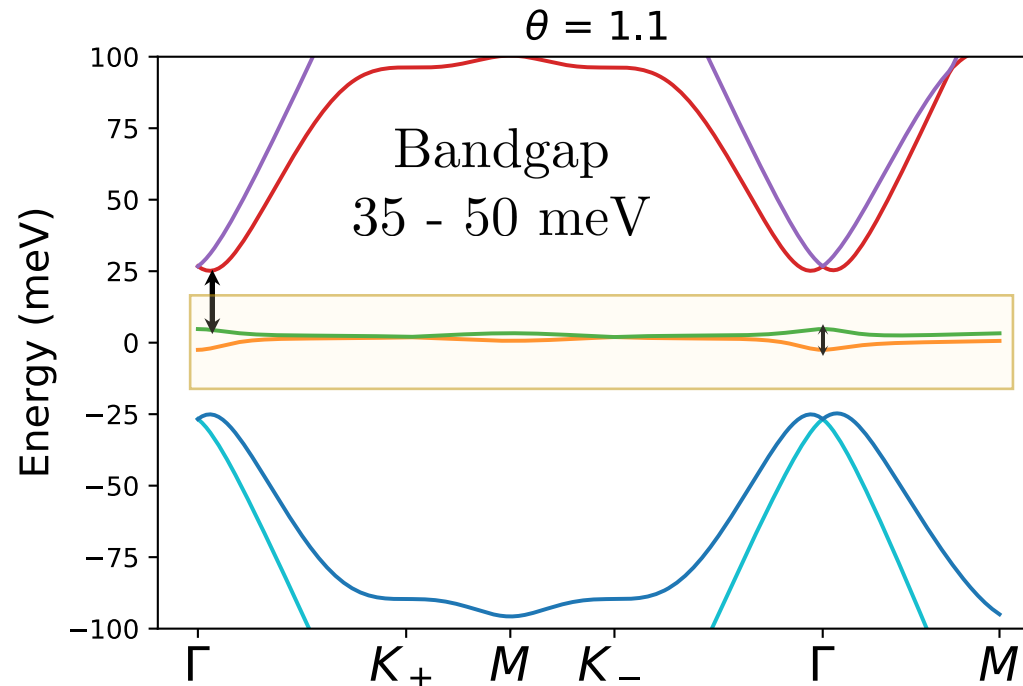


Figure: Khalaf *et al*, PRB 2019



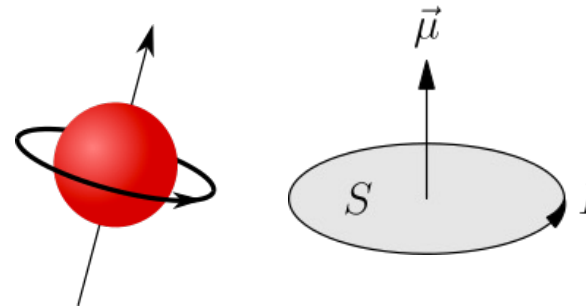
Cao *et al*, (Nature 2018)²
 Choi *et al*, Nat. Physics (2019)
 Sharpe *et al*, Science (2019)
 Arora *et al*, Nature (2020)
 Serlin *et al*, Science (2020)
 Park *et al*, Nature (2021)
 Hao *et al*, Science (2021)
 Choi *et al*, Nat. Physics (2021)
 Kim *et al*, Nature (2022)
 Many many others...

Bandwidth
 5 - 15 meV

Bistritzer, Macdonald, PNAS 2011,
 Los Santos *et al*, PRL 2007
 Tarnospolsky *et al*, PRL 2019
 Khalaf *et al*, PRB 2019
 Bernevig *et al*, PRB 2021 (TBG I-VI)

2D materials for electrical switches

- Can we exploit the tunability of Van der Waals heterostructures to design an electrical switch of magnetism?
- Can we *selectively* control spin and valley degrees of freedom?



2D materials for electrical switches

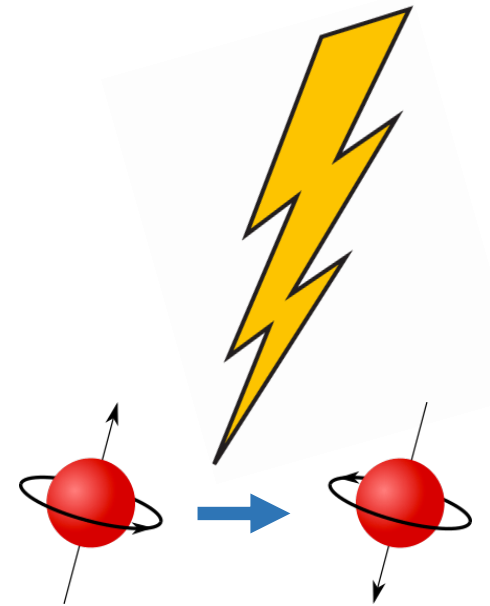
- Can we exploit the tunability of Van der Waals heterostructures to design an electrical switch of magnetism?
- Can we *selectively* control spin and valley degrees of freedom?
- This talk:

Propose a mechanism for selectively switching valley and/or spin *via reversing electric fields only* in chiral multilayer graphene

2D materials for electrical switches

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- This talk:

Propose a mechanism for selectively switching valley and/or spin *via reversing electric fields only* in chiral multilayer graphene



Outline

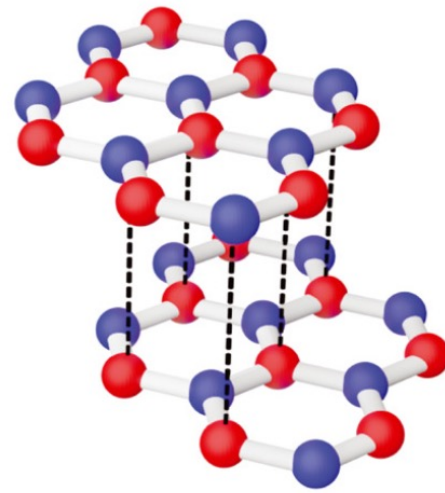
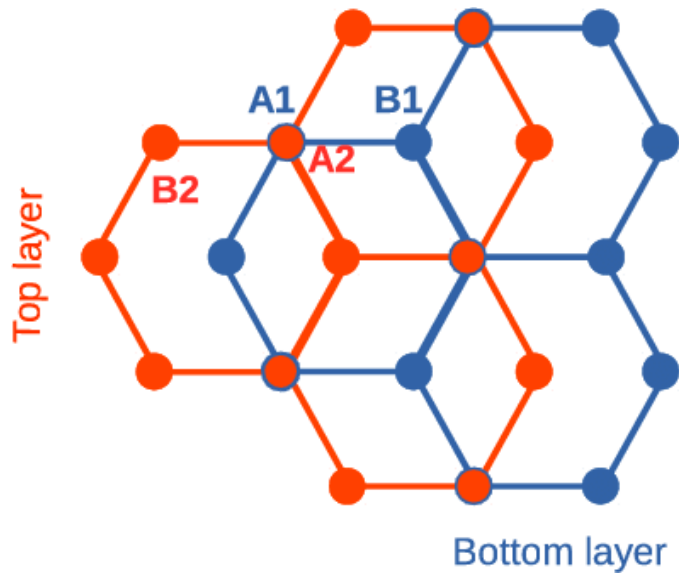
- Substrate induced spin-orbit coupling in chiral few-layered graphene
- Design principles of an electrical spin or valley switch
- Conclusion and outlook

Outline

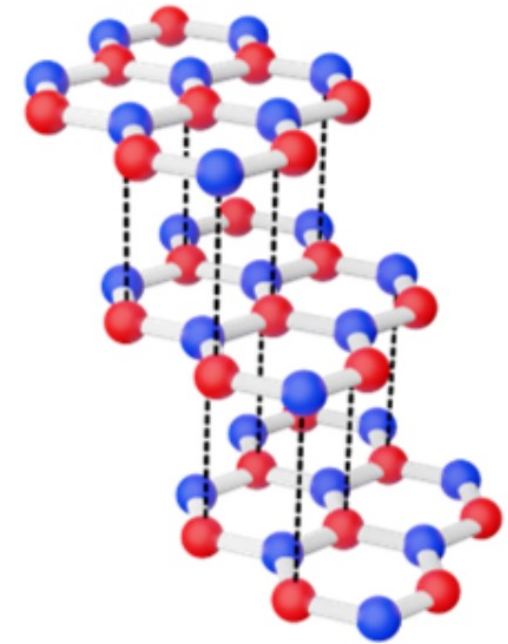
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Chiral multilayered graphene

- Chirally stacked graphene multilayers



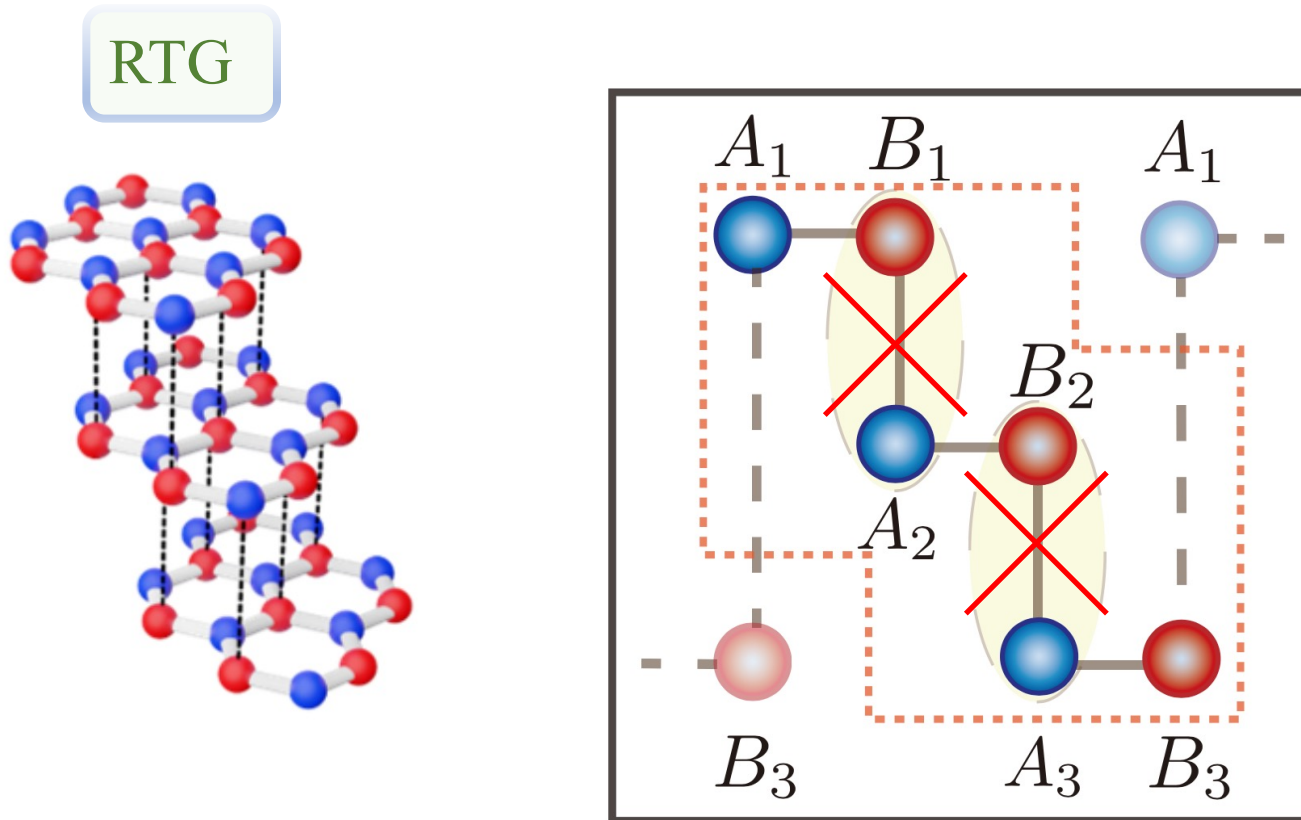
$N = 2$
Bernal bilayer graphene
(BBG)



$N = 3$
Rhombohedral trilayer
graphene (RTG)

Chiral multilayered graphene

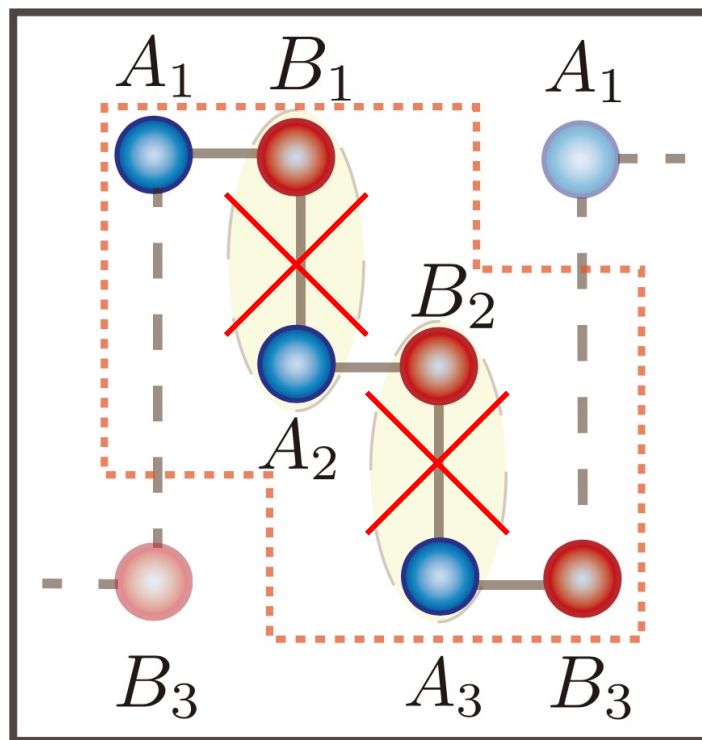
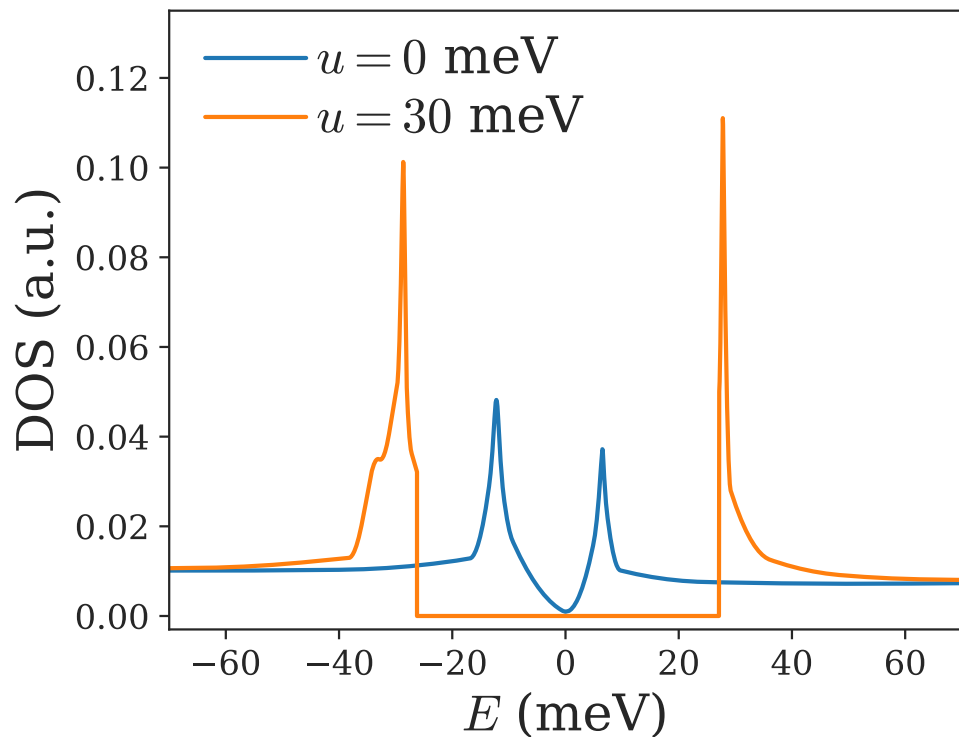
- Active sites for low-energy physics: A_1 and B_N



Chiral multilayered graphene

- Active sites for low-energy physics: A_1 and B_N
- Perpendicular displacement field D enhances DOS

$$H_{\text{eff}}(\tau, k) \approx \begin{pmatrix} u & \frac{(vk_-)^N}{t_{\perp}^{N-1}} \\ \frac{(vk_+)^N}{t_{\perp}^{N-1}} & -u \end{pmatrix}$$



Chiral multilayered graphene

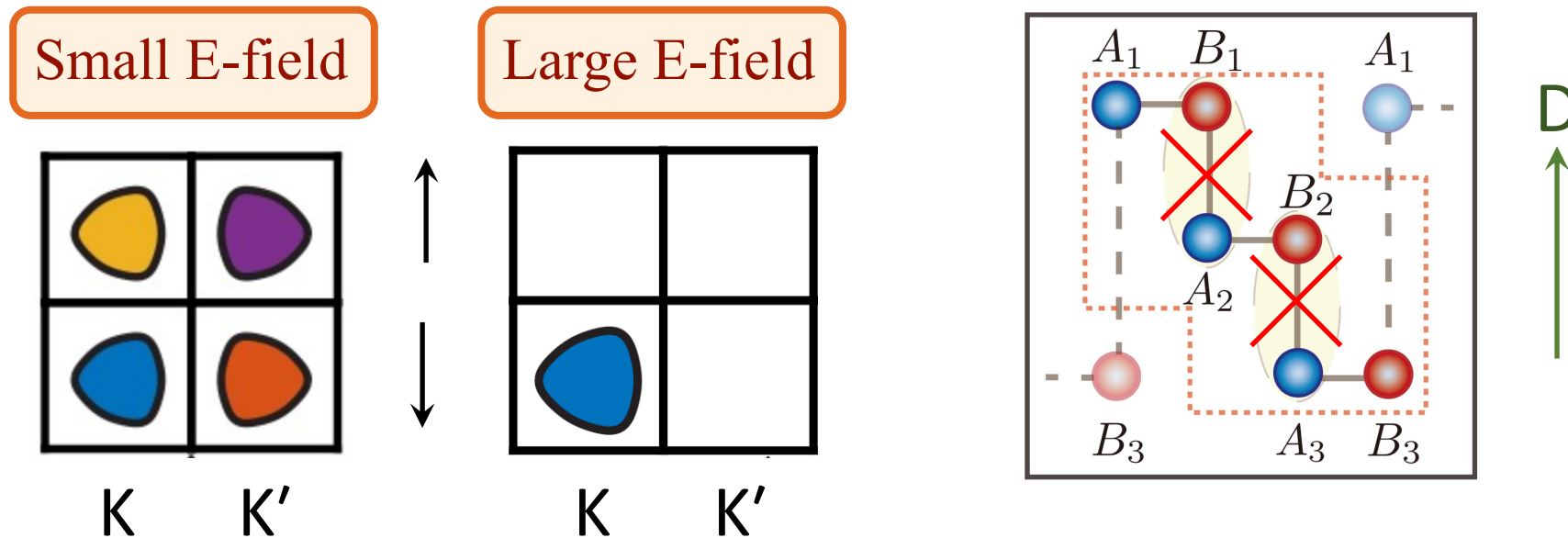
- Active sites for low-energy physics: A_1 and B_N
- Perpendicular displacement field D enhances DOS
- Enhanced DOS leads to interaction-driven flavor polarization

Zhou *et al*, Nature² (2021),
Science (2022)

Seiler *et al*, Nature (2022),
arXiv:2308.00827

Han, Lu *et al*, Science (2024)

Liu, Zheng *et al*, arXiv:2306.11042



Induced spin-orbit coupling

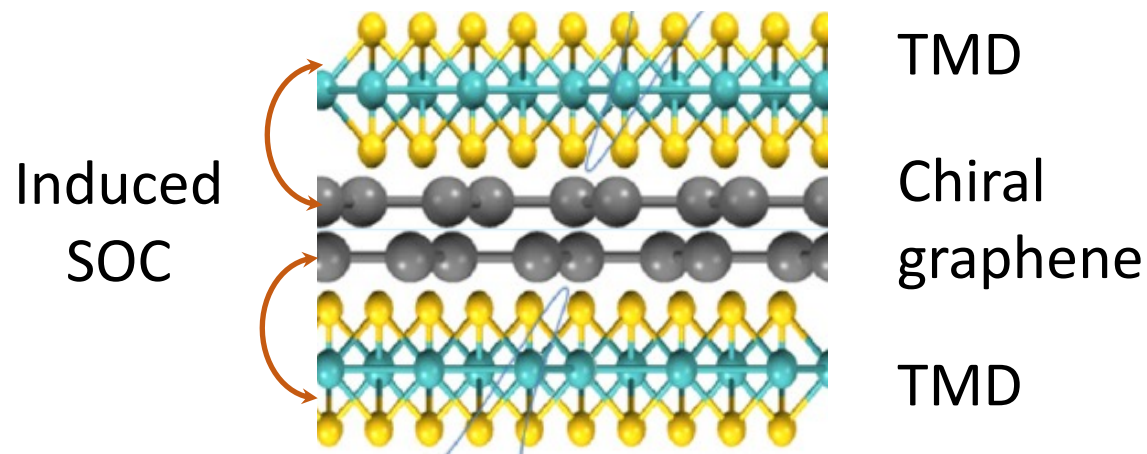
- Encapsulation by TMDs induces spin-orbit coupling (SOC) in chiral graphene
- On the adjacent monolayer – major contributions are:

$$H_{\text{Ising-SOC}} = \lambda_I \tau^z s^z$$

Reduces spin-rotation symmetry from SU(2) to U(1)

$$H_{\text{Rashba-SOC}} = \lambda_R (\tau^z \sigma^x s^y - \sigma^y s^x)$$

Breaks spin-rotation completely



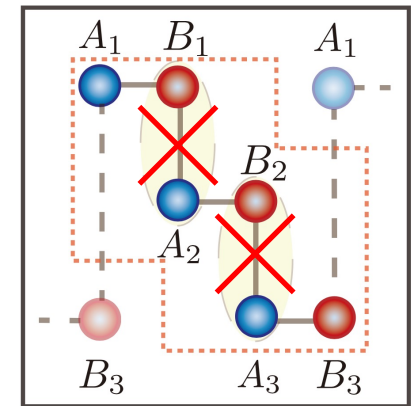
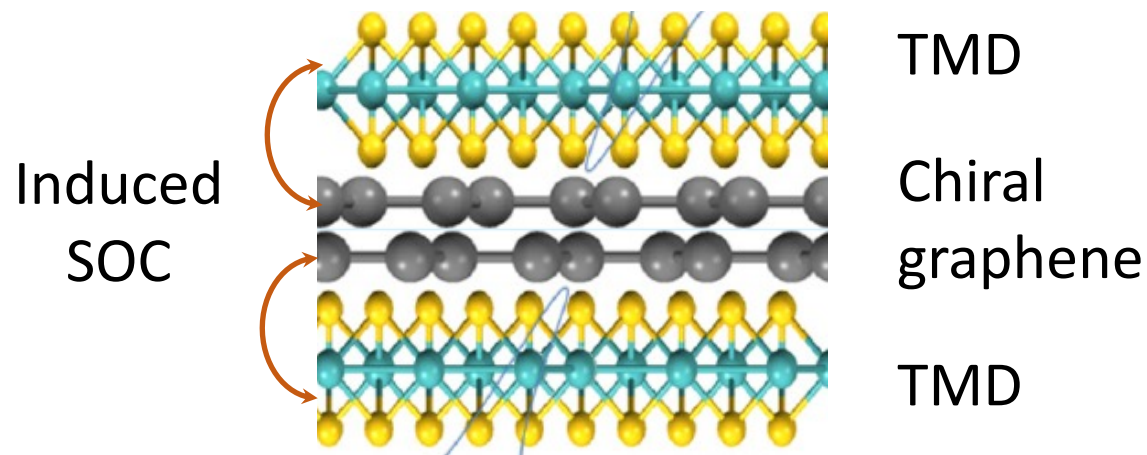
Gmitra, Fabian, PRB (2015)
Zollner *et al*, PRB (2022)

Induced spin-orbit coupling

- Encapsulation by TMDs induces spin-orbit coupling (SOC) in chiral graphene
- In chiral multilayers, the Rashba term is suppressed, only Ising SOC is important

$$H_{\text{Ising-SOC}} = \lambda_I \tau^z s^z$$

~~$$H_{\text{Rashba-SOC}} = \lambda_R (\tau^z \sigma^x s^y - \sigma^y s^x)$$~~

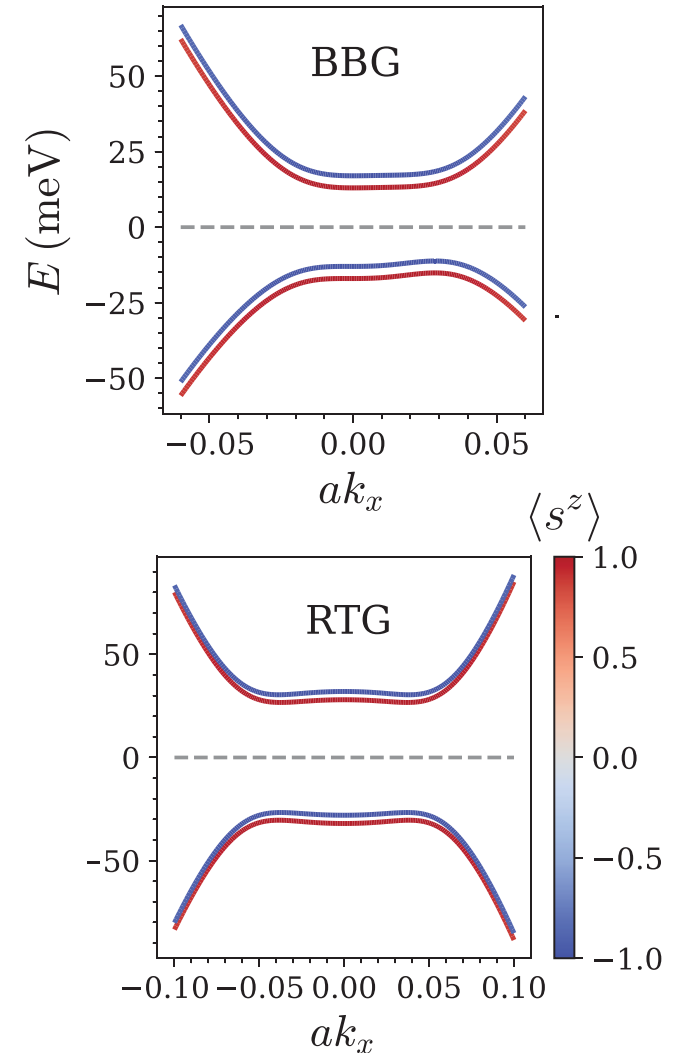
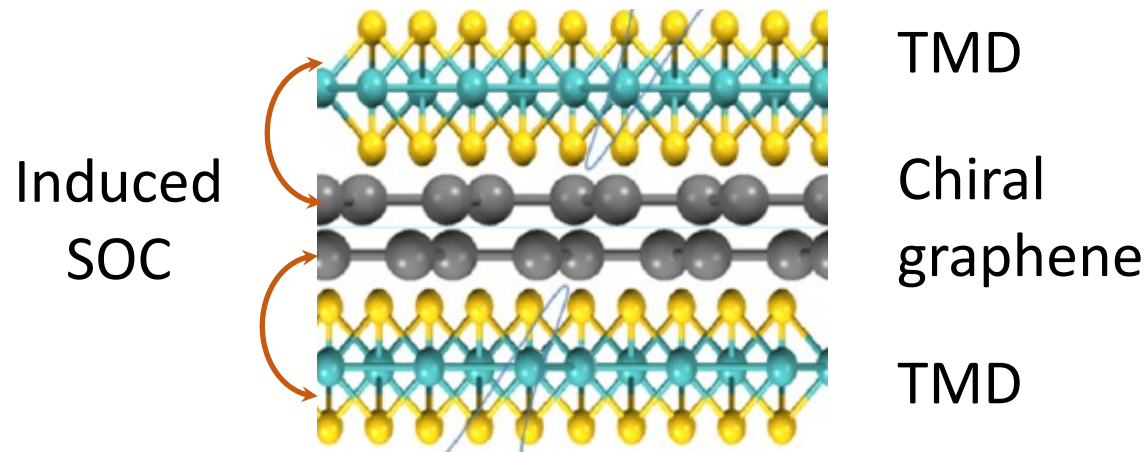


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Induced spin-orbit coupling

- Encapsulation by TMDs induces spin-orbit coupling (SOC) in chiral graphene
- Effective Hamiltonian in (A_1, B_N) basis:

$$H_{\text{eff}}(\tau, k) \approx \begin{pmatrix} u + \lambda_I^{\text{top}} s^z \tau^z & \frac{(vk_-)^N}{t_{\perp}^{N-1}} \\ \frac{(vk_+)^N}{t_{\perp}^{N-1}} & -u + \lambda_I^{\text{bot}} s^z \tau^z \end{pmatrix}$$



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- If TMDs are *aligned*, we expect the same sign of Ising SOC on the top and bottom layers

$$\lambda_I^{\text{top}} = \lambda_I^{\text{bot}}$$

Induced spin-orbit coupling

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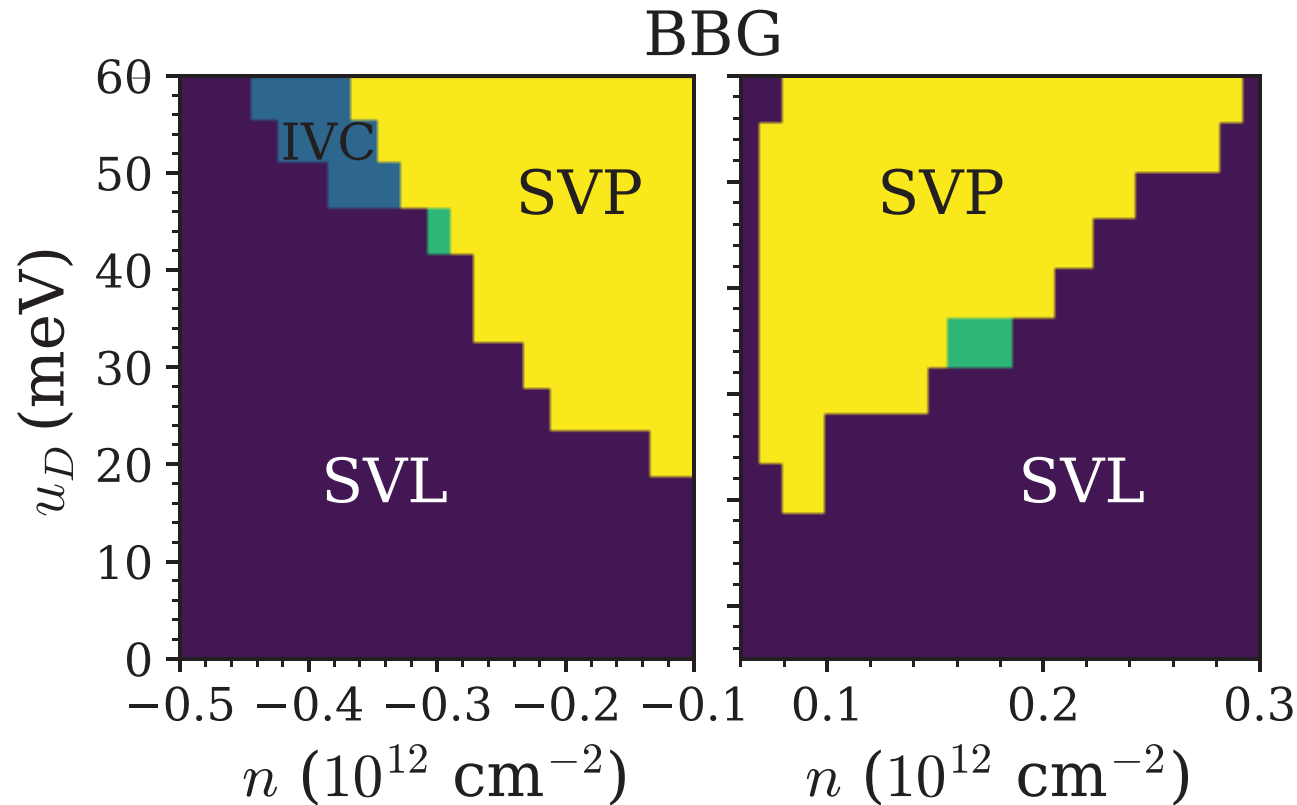
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- If TMDs are *anti-aligned*, inversion symmetry is preserved (at $u = 0$)
- Inversion *requires* opposite signs of Ising SOC on top and bottom layers

$$\lambda_I^{\text{top}} = -\lambda_I^{\text{bot}}$$

Interactions cause flavor polarization

- Requires finite D in BBG/RTG, seen at $D = 0$ in rhombohedral pentalayer graphene (RPG)



Experiments:

Zhang *et al*, Nature (2023)

Seiler et al, arXiv:2403.17140

BBG on WSe_2

Han *et al*, Science (2024)

RPG on WS_2

Theory:

Wang, SC *et al*, PRL (2024)

Koh *et al*, PRB (2024)

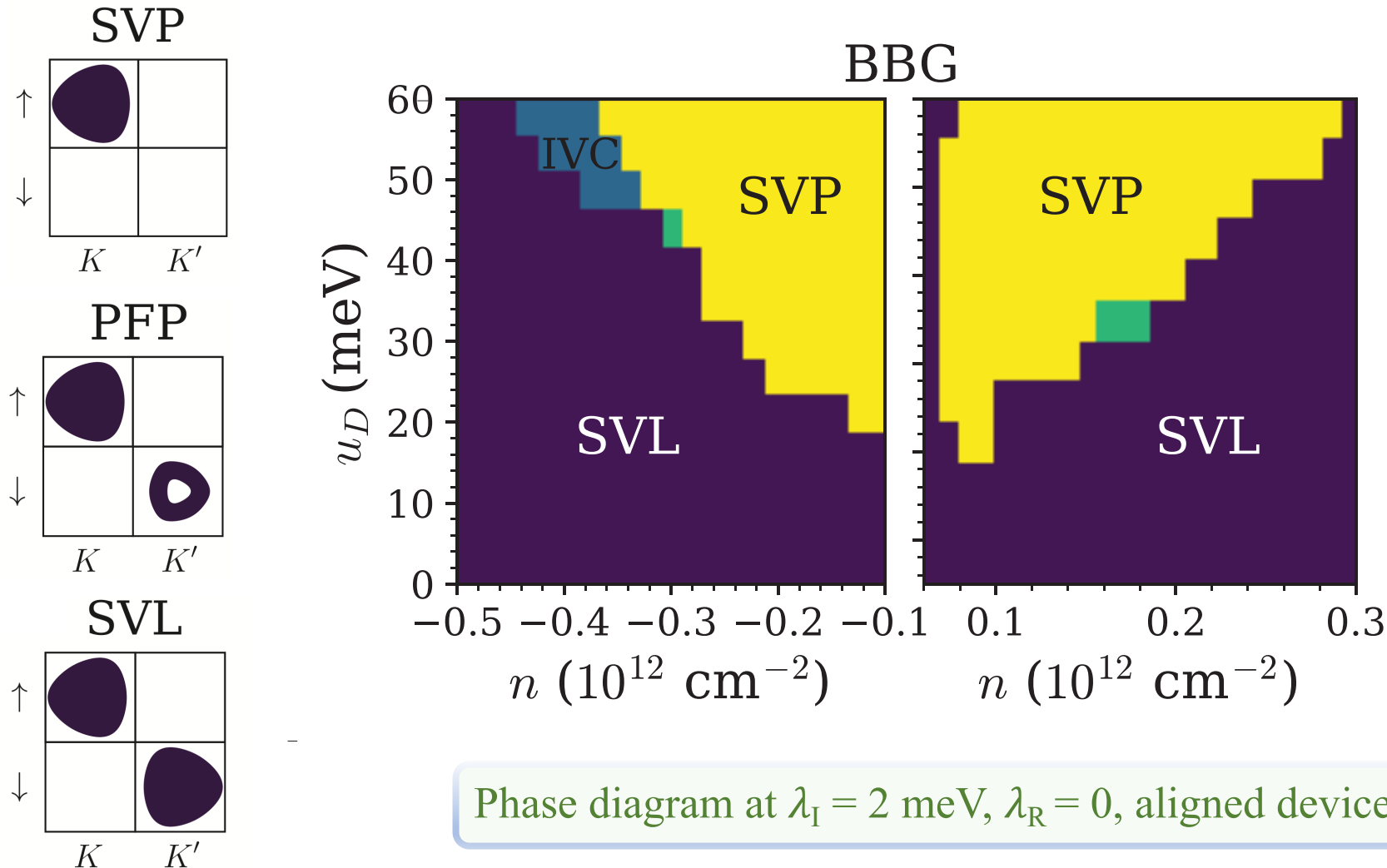
Zhumagulov *et al*, PRL (2024),

Others...

Phase diagram at $\lambda_I = 2 \text{ meV}$, $\lambda_R = 0$, aligned device

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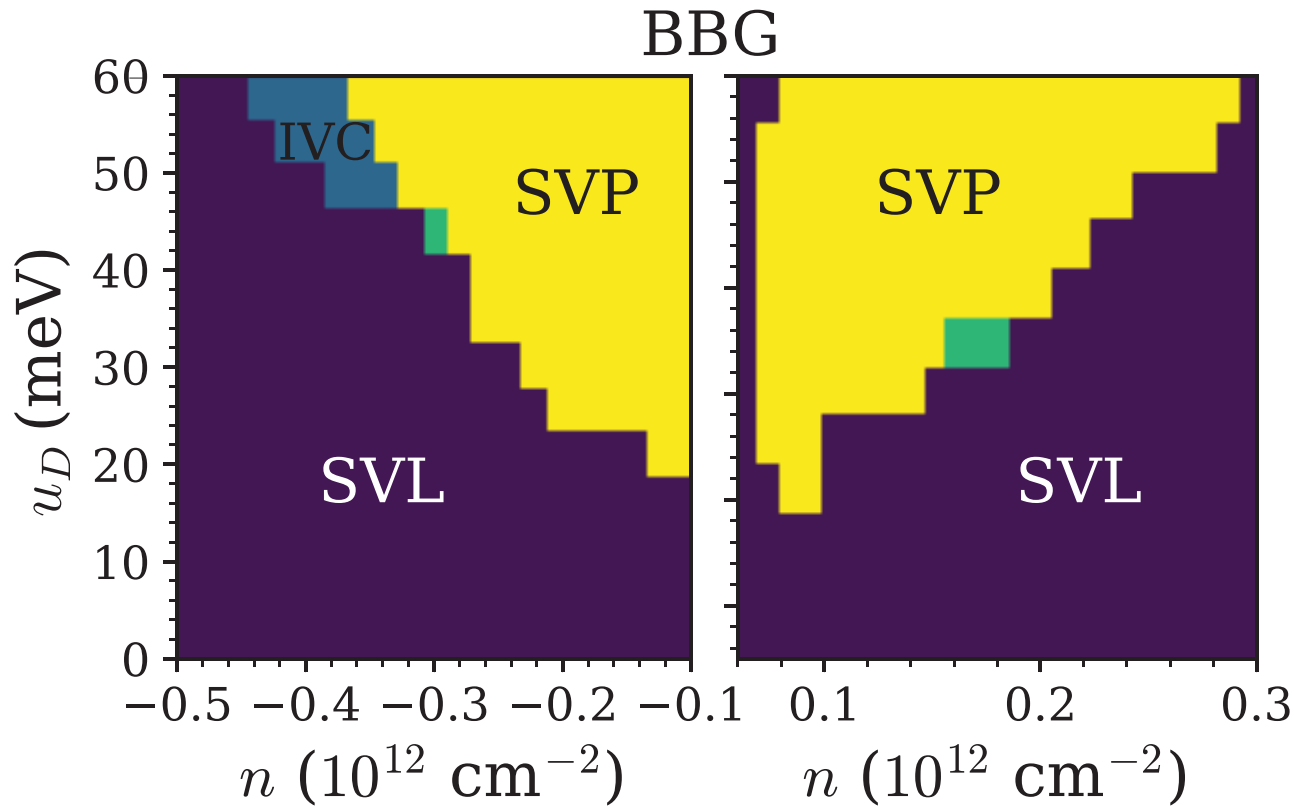
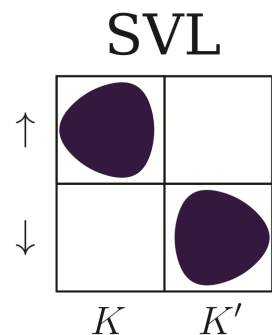
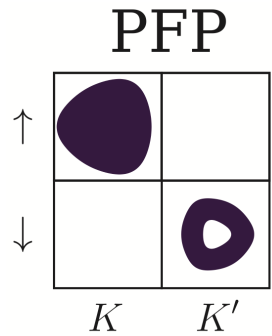
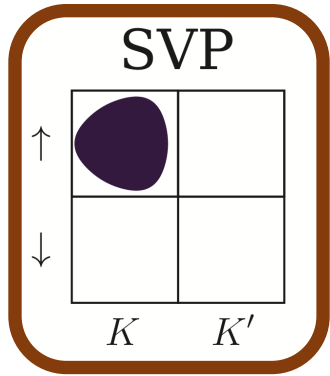
RPG on WS_2

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Phase diagram at $\lambda_I = 2$ meV, $\lambda_R = 0$, aligned device

Experiments:
Zhang *et al*, Nature (2023)

BBG on WSe₂

Han *et al*, Science (2024)

RPG on WS₂

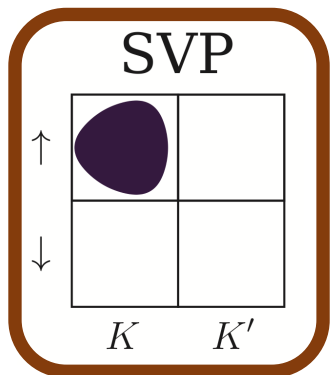
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Interactions cause flavor polarization

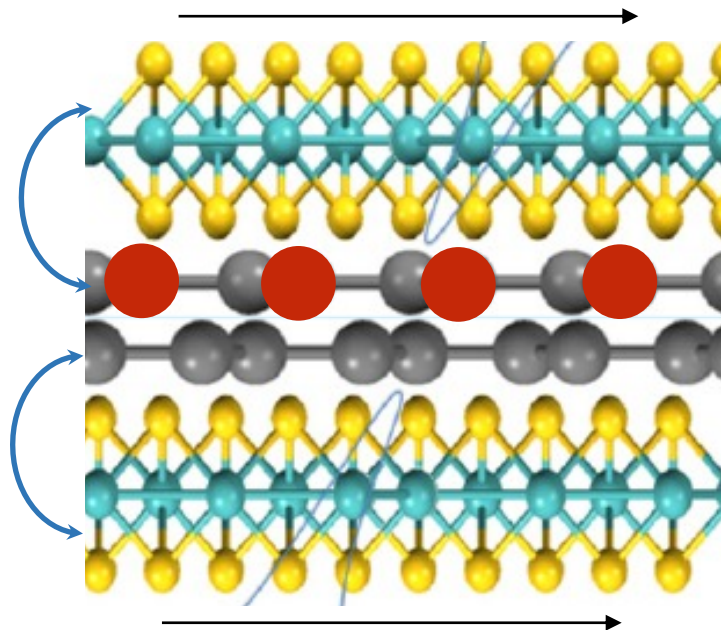
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- Fully isospin polarized phases thus have $\tau^z s^z = 1$ (say)

Experiments:
Zhang *et al*, Nature (2023)

BBG on WSe_2



$+\lambda$
Induced
Ising SOC
 $+\lambda$



TMD

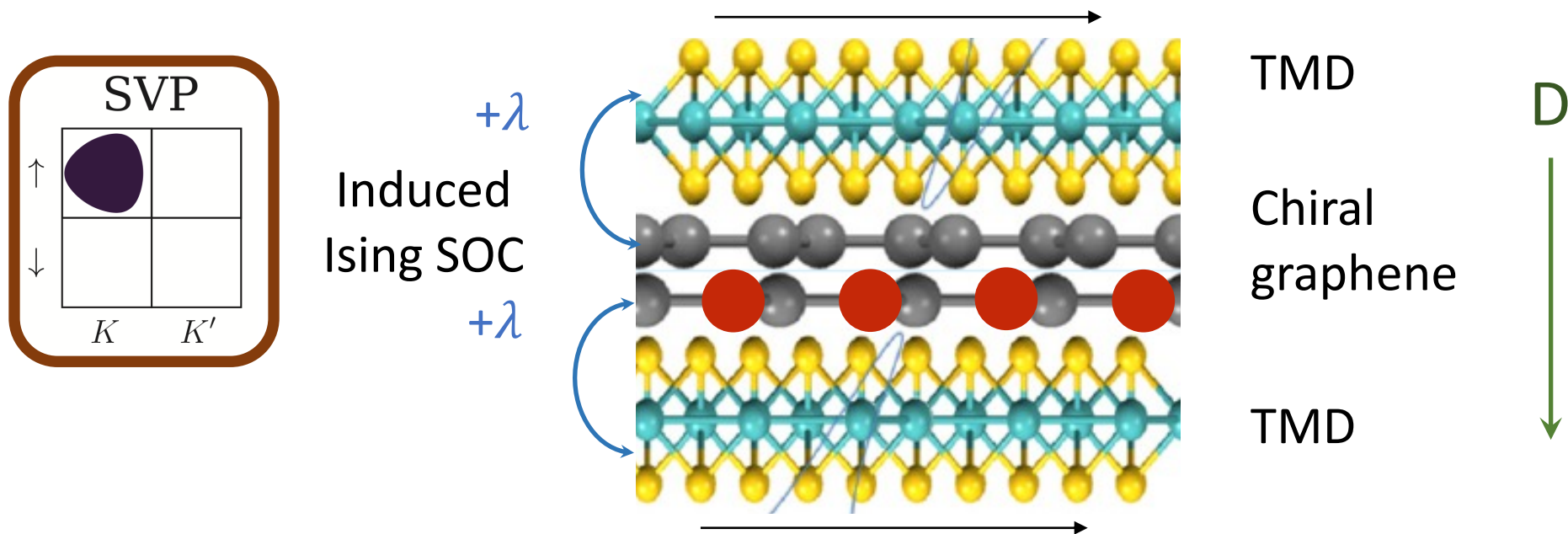
Chiral
graphene

TMD

D
 \uparrow

Interactions cause flavor polarization

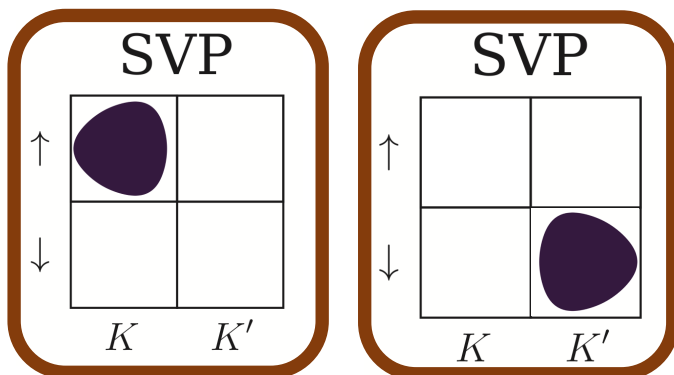
- D fixes whether the low-energy electrons (or holes) lie on the top or bottom layer
- Fully isospin polarized phases thus have $\tau^z s^z = 1$ (say)
- Flipping D moves electrons to opposite layer, preserves $\tau^z s^z = 1$ in aligned device



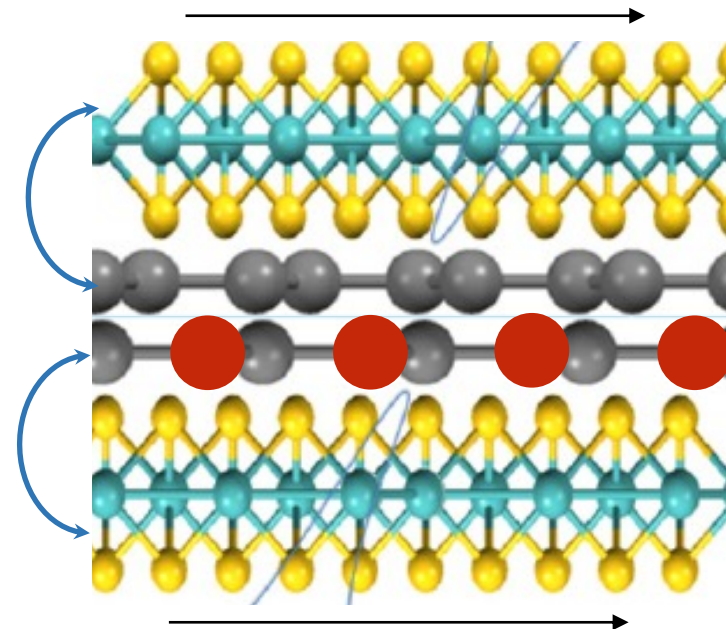
Interactions cause flavor polarization

- D fixes whether the low-energy electrons (or holes) lie on the top or bottom layer
- Reversing D also flips the orbital magnetic moment (arising from Berry phase effects) in each valley
- Fully isospin polarized phase with $\tau^z s^z = 1$ (or -1) still allows for two possibilities for a given direction of the electric field:

1. $\tau^z = 1, s^z = 1$ (K, \uparrow)
2. $\tau^z = -1, s^z = -1$ (K', \downarrow)



$+ \lambda$
Induced
Ising SOC
 $+ \lambda$



TMD

Chiral
graphene

TMD

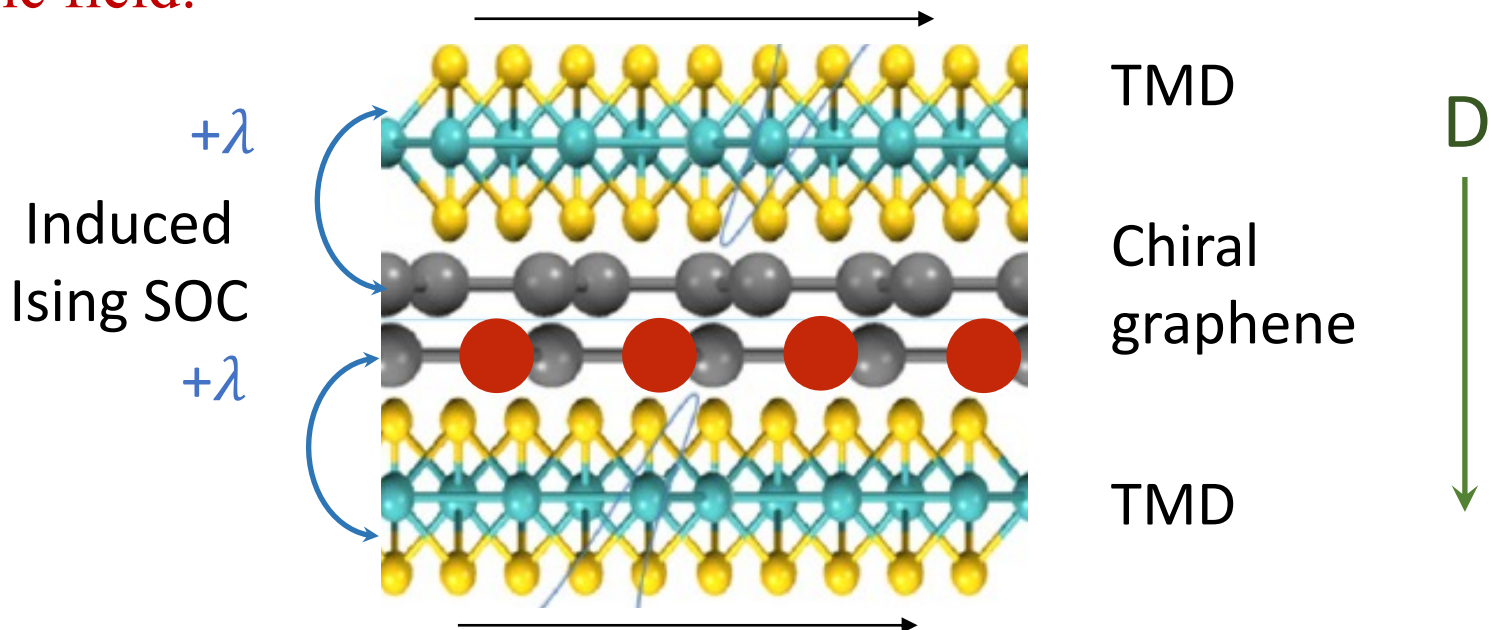
D

Interactions cause flavor polarization

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- Reversing D also flips the orbital magnetic moment (arising from Berry phase effects) in each valley
- Fully isospin polarized phase with $\tau^z s^z = 1$ (or -1) still allows for two possibilities for a given direction of the electric field:

1. $\tau^z = 1, s^z = 1$ (K, \uparrow)
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Would like to flip valley and spin on demand

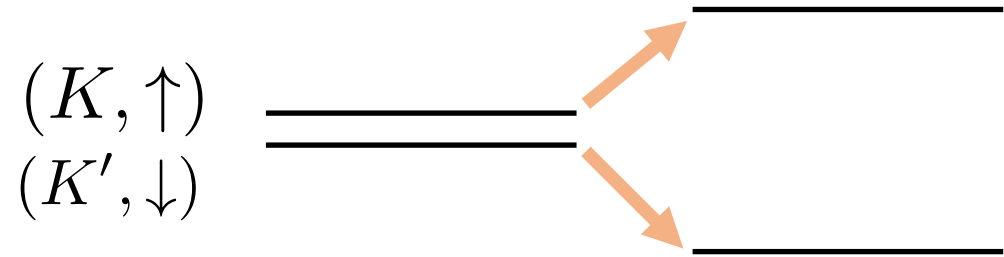


Outline

- Substrate induced spin-orbit coupling in chiral few-layered graphene
- Design principles of an electrical spin or valley switch
- Conclusion and outlook

Resolution: Static magnetic field

- Need to split degeneracy between (K, \uparrow) and (K', \downarrow)
- Solution: add a constant perpendicular magnetic field B

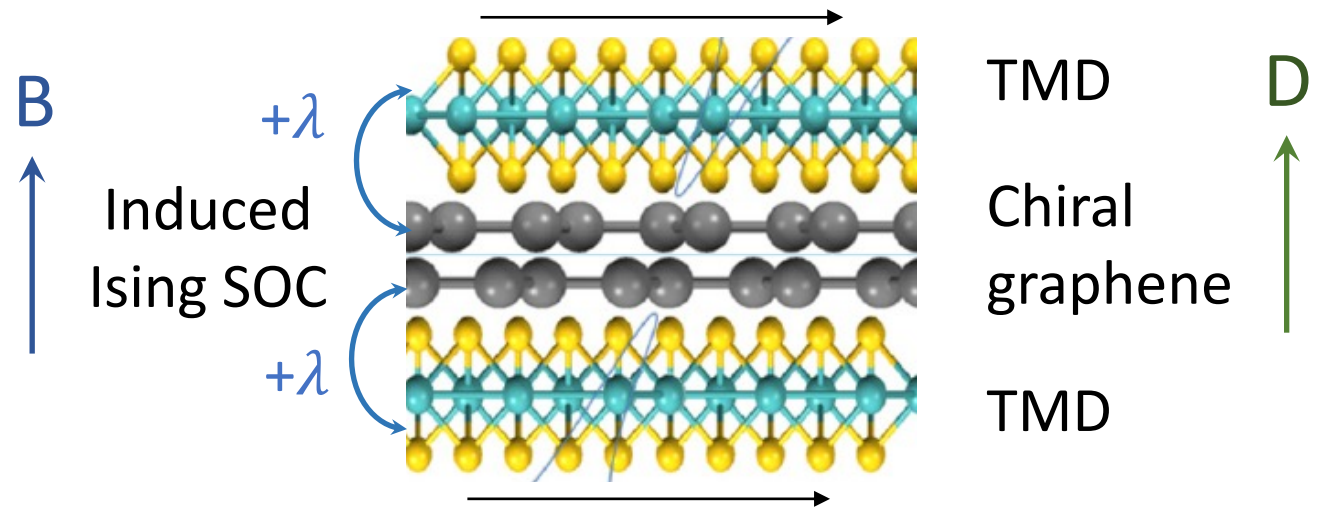


- Couples to both orbital and spin magnetic moment:

$$H_B = -(g_s s^z + g_v \tau^z) B$$

$$g_v = |g_v| \text{sign}(D)$$

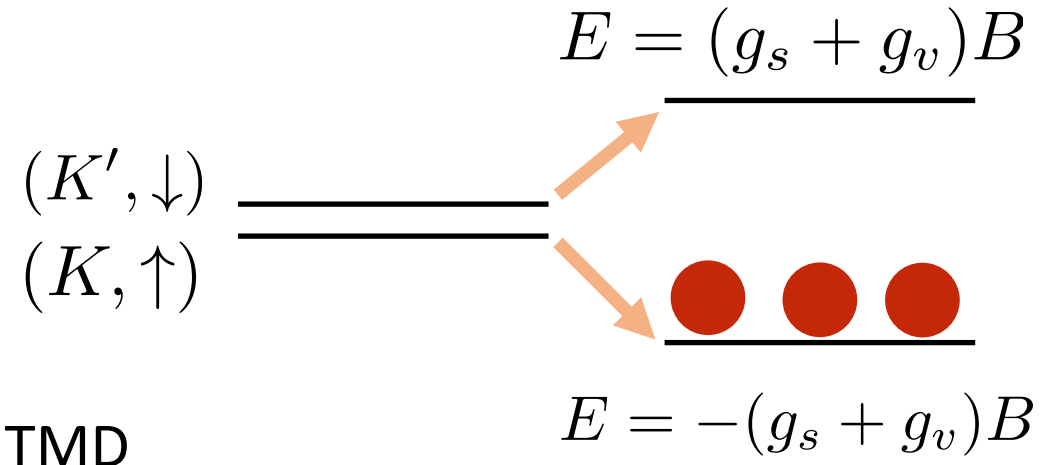
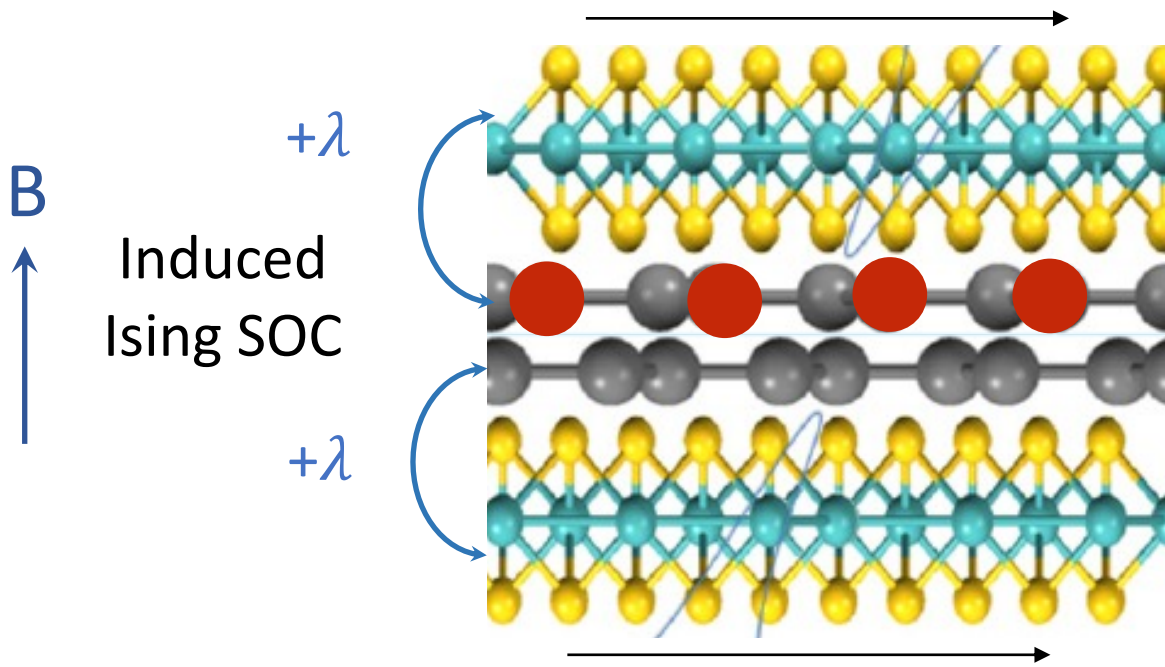
- If $g_s < g_v$, then orbital magnetization is pinned by B , hence valley and spin are both flipped



Resolution: Static magnetic field

Assume g_s, g_v are positive

- (K, \uparrow) is occupied when D points up



TMD
Chiral graphene
TMD

D
↑

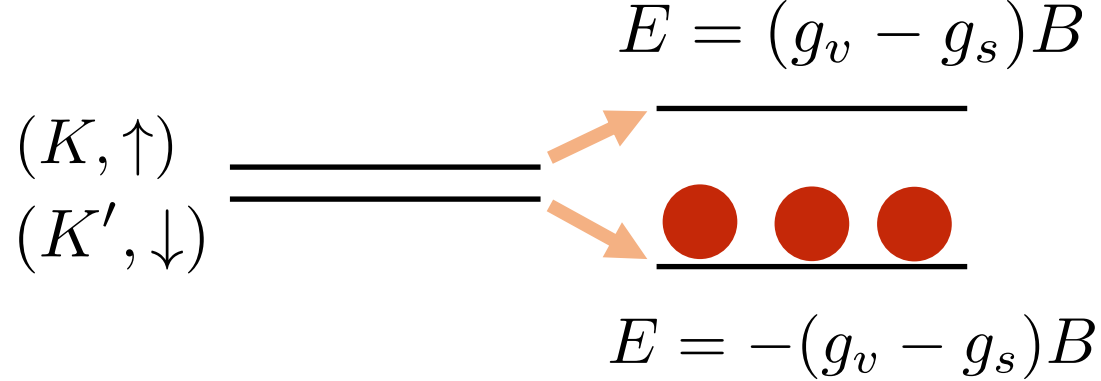
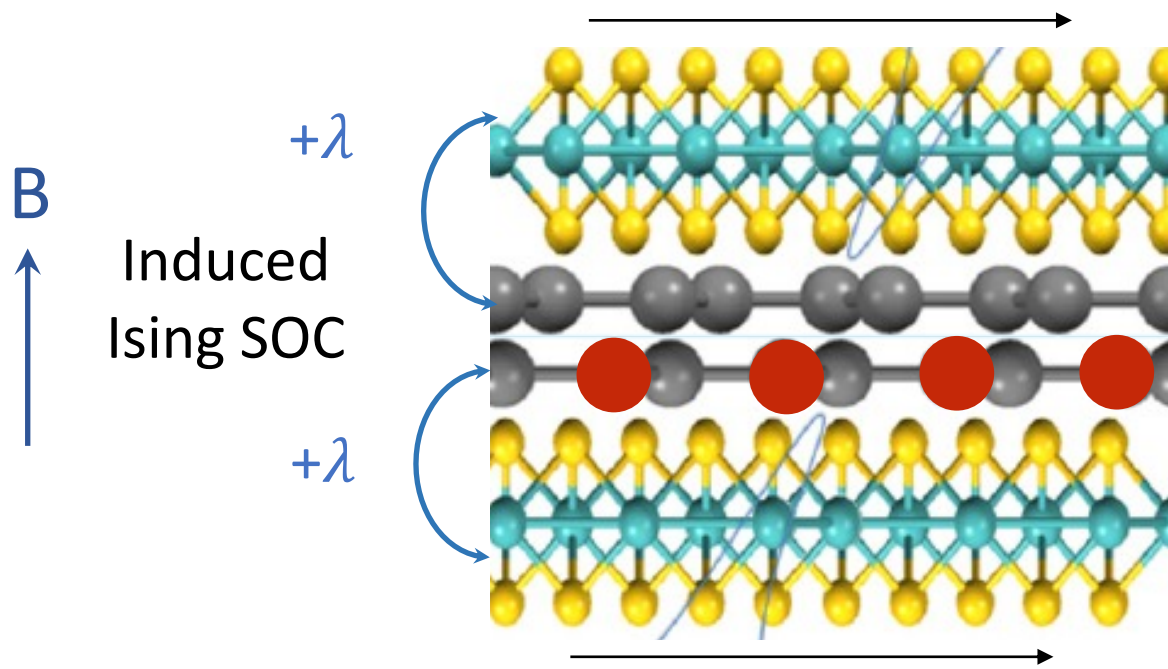
$$H_B = -(g_s s^z + g_v \tau^z) B$$

$$g_v = |g_v| \text{sign}(D)$$

Resolution: Static magnetic field

If $g_s < g_v$, orbital moment remains fixed, i.e., valley is flipped

- (K', \downarrow) is occupied when D is reversed



TMD
 Chiral graphene
 TMD

D

↓

Electric field induced
spin + valley switch

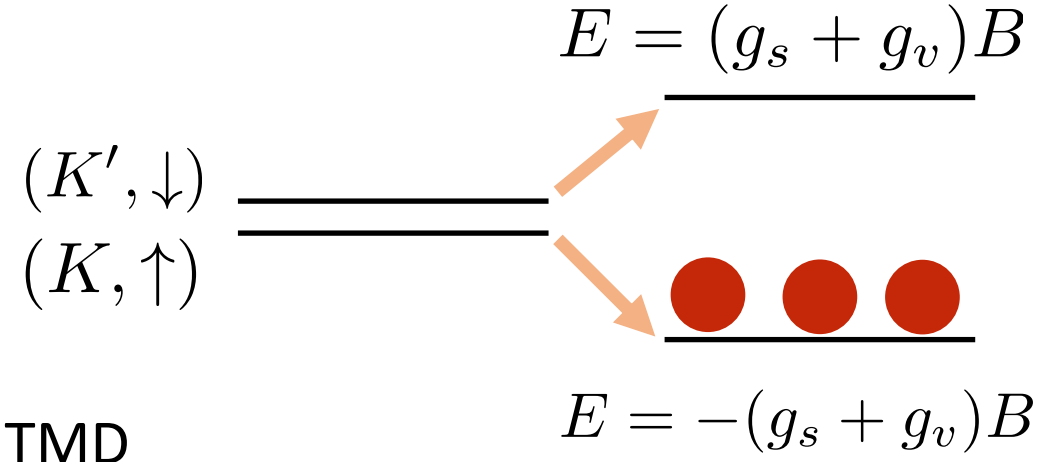
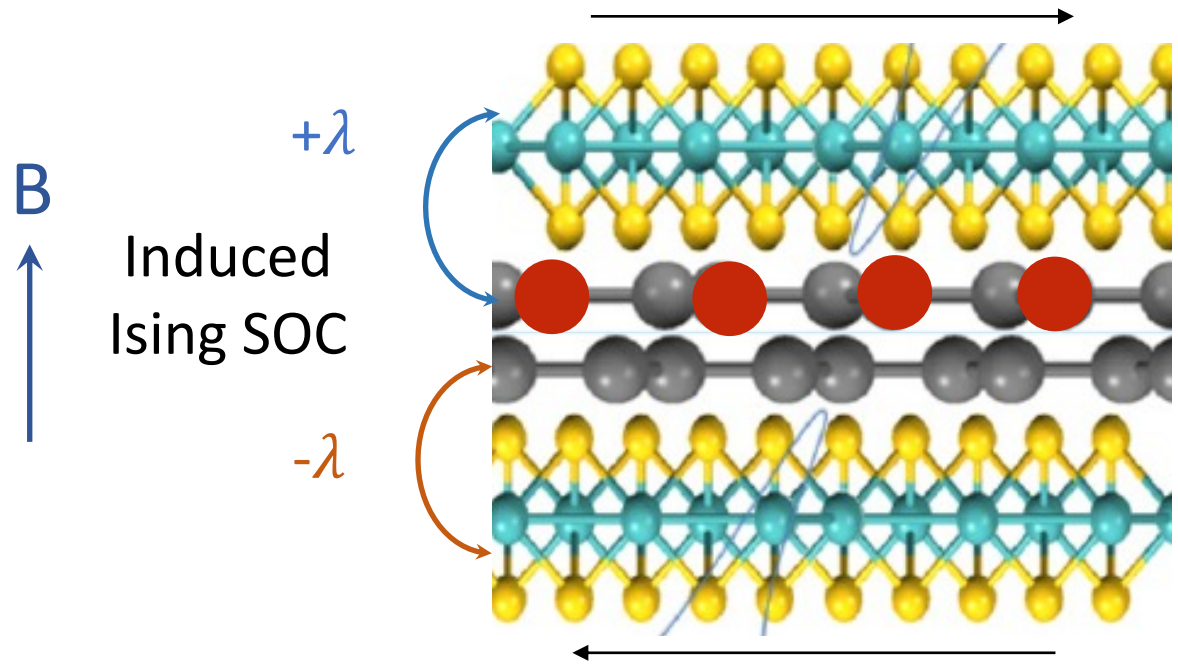
$$H_B = -(g_s s^z + g_v \tau^z) B$$

$$g_v = |g_v| \text{sign}(D)$$

Device with anti-aligned TMDs

Assume g_s, g_v are positive

- (K, \uparrow) is occupied when D points up



TMD
 Chiral graphene
 TMD

D

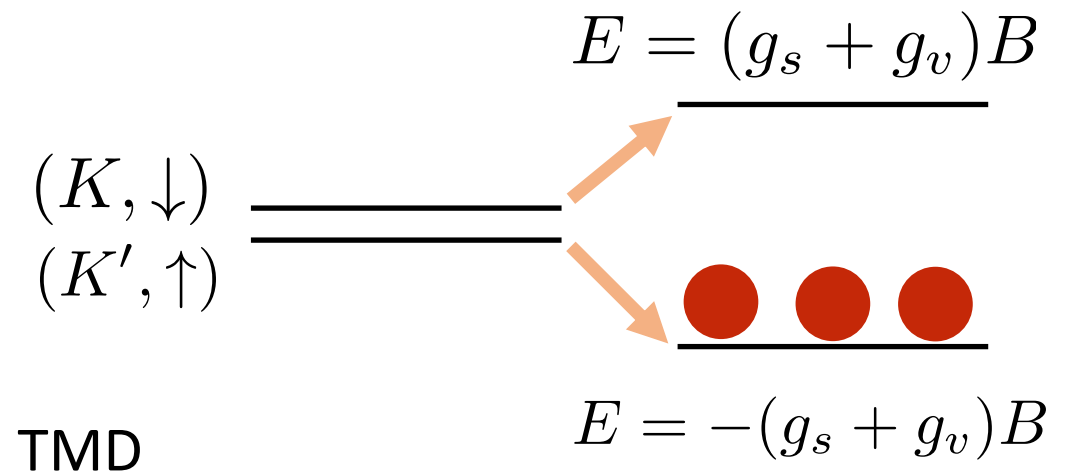
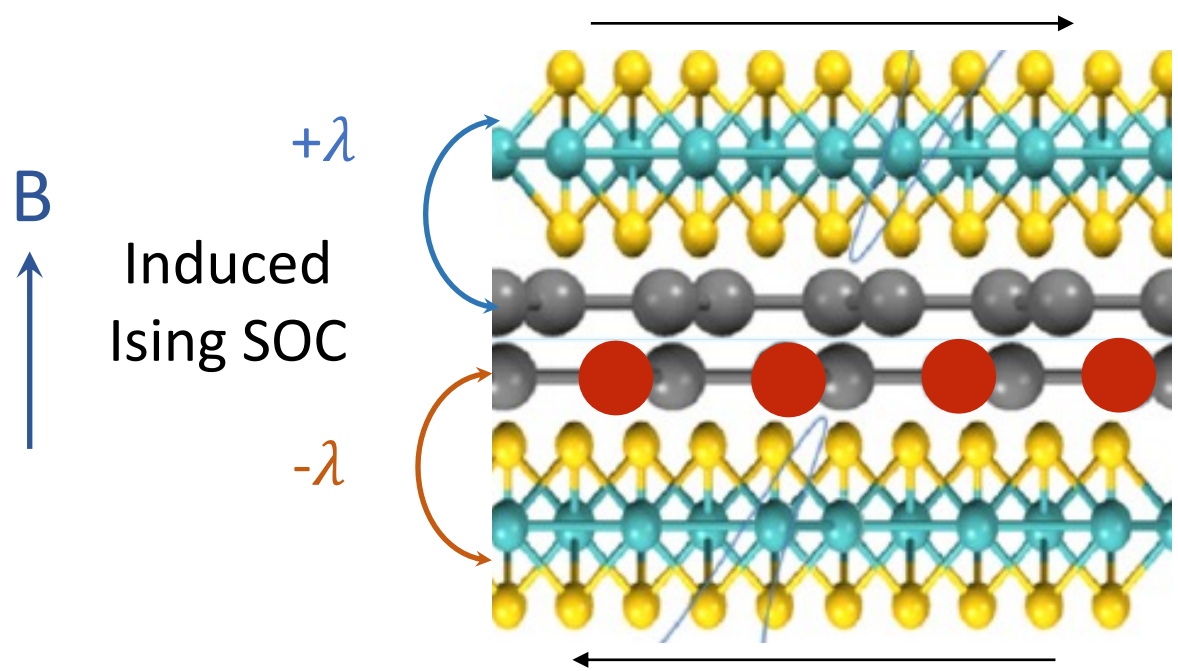
$$H_B = -(g_s s^z + g_v \tau^z) B$$

$$g_v = |g_v| \text{sign}(D)$$

Device with anti-aligned TMDs

When D is reversed, low-lying manifold is different due to opposite sign of SOC

- (K', \uparrow) is occupied when D is reversed



TMD
Chiral graphene
TMD

D

↓

Electric field induced
valley switch

$$H_B = -(g_s s^z + g_v \tau^z) B$$

$$g_v = |g_v| \text{sign}(D)$$

Typical g_v/g_s is large in chiral graphene

- Finally, can control ratio of g_v to g_s by tuning D or no. of layers N
- Large $D \rightarrow$ localized orbitals \rightarrow small Berry curvature \rightarrow small g_v
- Large $N \rightarrow$ larger Berry curvature \rightarrow large g_v

Niu *et al*, RMP (2010)

Thonhauser, Int. J. Mod. Phys (2011)

$$M = \frac{e}{\hbar} \int_{\mathbf{k}} n_F(\varepsilon_{n,\mathbf{k}}) \Omega_{n,\mathbf{k}} \quad \text{if p-h symmetric}$$

Typically, $g_v \approx 15-30$

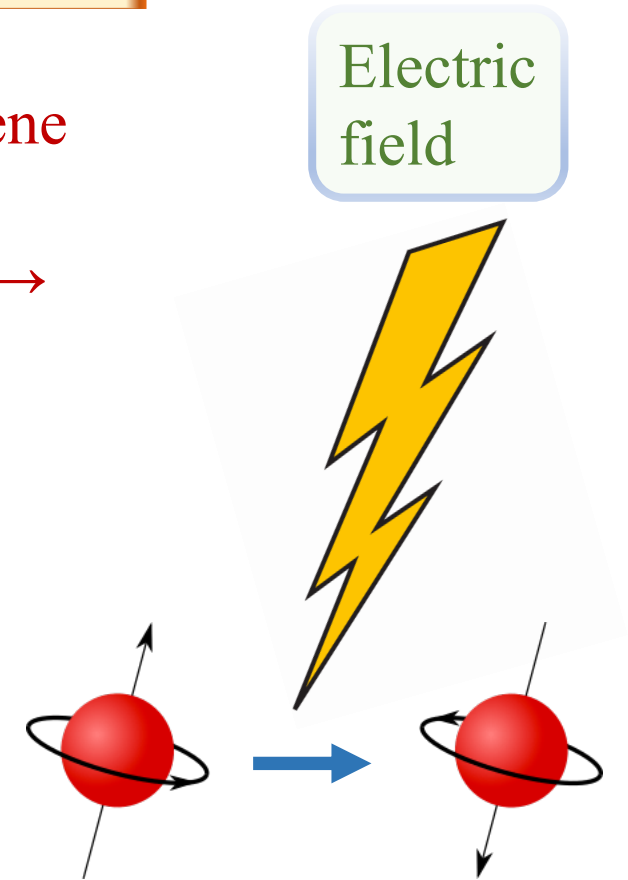
- Verified numerically by considering contributions from all bands that $g_v \gg g_s$
- Flipping only spin, without reversing valley, may be obtained by reversing chemical potential in the TMD aligned device

Outline

- Substrate induced spin-orbit coupling in chiral few-layered graphene
- Design principles of an electrical spin or valley switch
- Conclusion and outlook

Conclusions

- Discussed the effects of induced SOC in chirally stacked graphene
- Showed that induced SOC + electric field induced correlations \rightarrow selective spin and/or valley switch



Conclusions

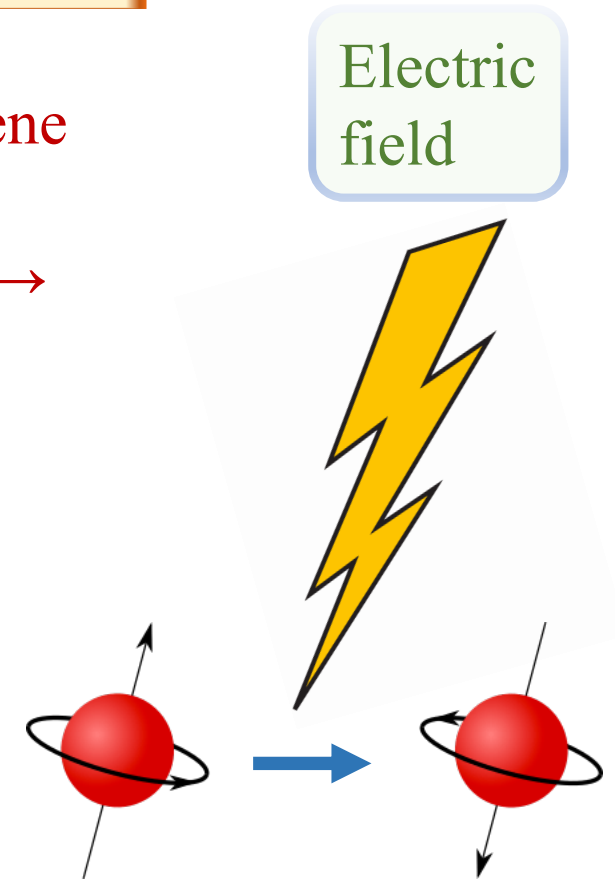
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Open questions:

1. How stable are these phases to thermal fluctuations?
2. Can one use correlations and induced SOC to create a stable magnetic memory?
3. Similar physics in moiré TMDs for higher tunability?

Han et al, Science (2024)

Abouelkomsan, Bergholtz and SC, arXiv:2210.14918

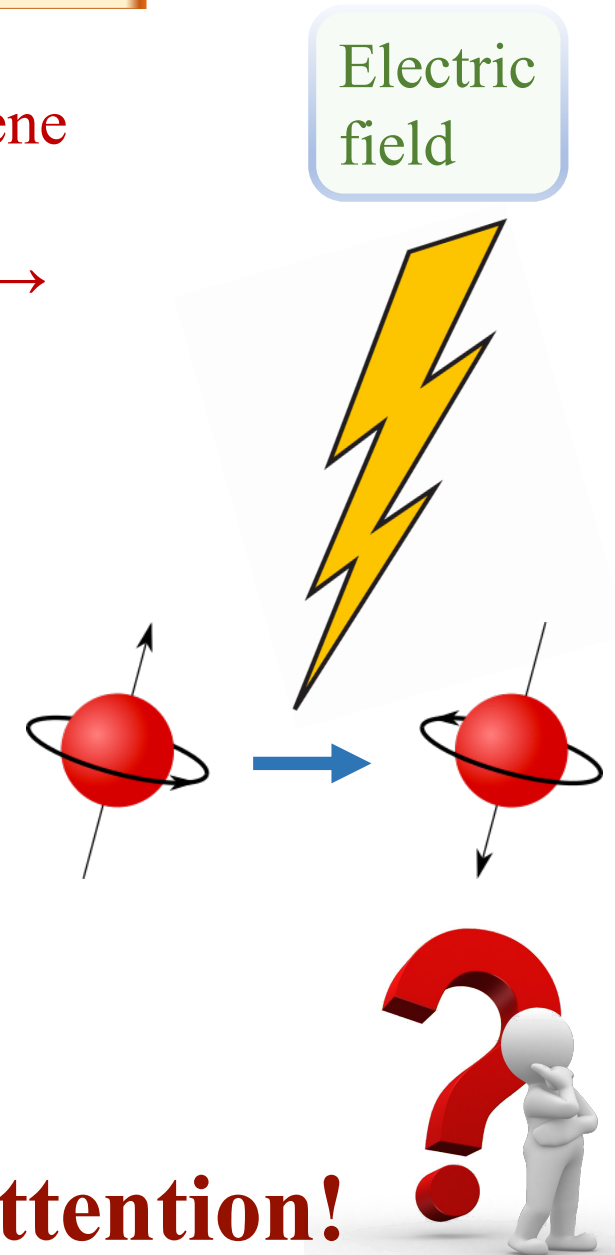


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Thank you for your attention!