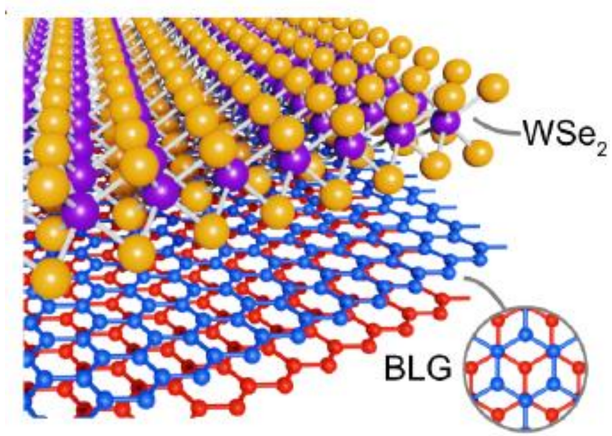
Rhombohedral graphene:

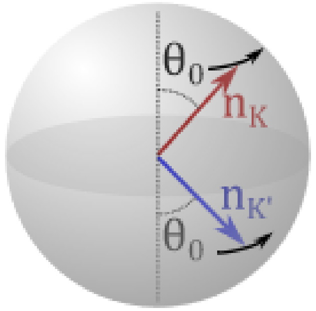
spin canting, collective modes and superconductivity

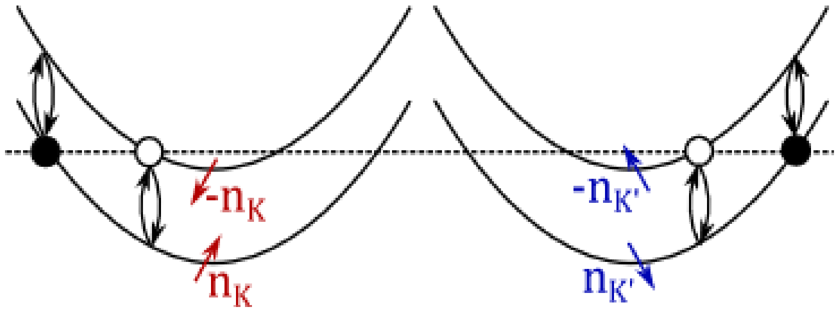


Étienne Lantagne-Hurtubise

KITP – Tunable 2D materials program

Sep 10, 2024





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GORDON AND BETTY MOORE FOUNDATION

Acknowledgments

Experiments:

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Stevan Nadj-Perge

Caitlin Patterson

Owen Sheekey

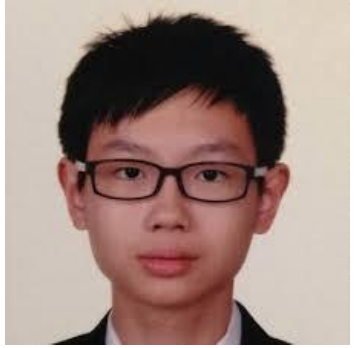
Trevor Arp

Ludwig Holleis

Youngjoon Choi

Andrea Young

Theory collaborators:



Jin Ming Koh
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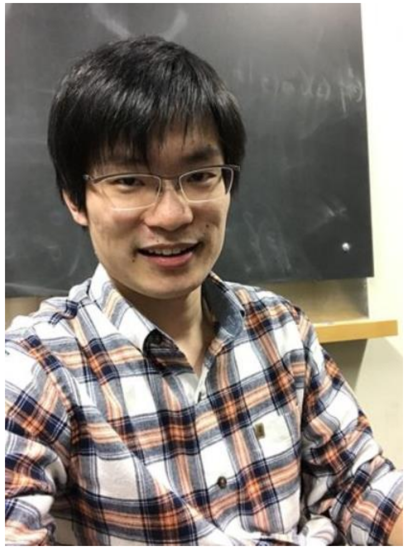
Zhiyu Dong

Alex Thomson (UC Davis)

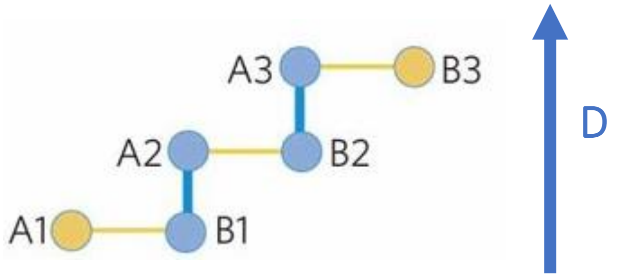
Jason Alicea



Rhombohedral graphene multilayers

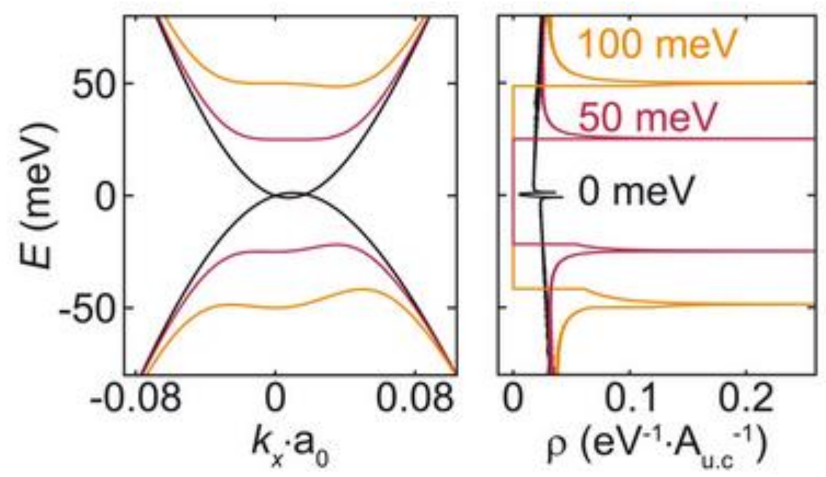
“ABC” stacking

Guinea, Castro Neto, & Peres, PRB **73**, 245426 (2006)



Perpendicular electric displacement field D **opens a gap** and **tunes the density of states**

- *In situ* tuning of doping level and correlation strength
- Extremely clean! Mean free paths ~10 µm



Bilayer graphene

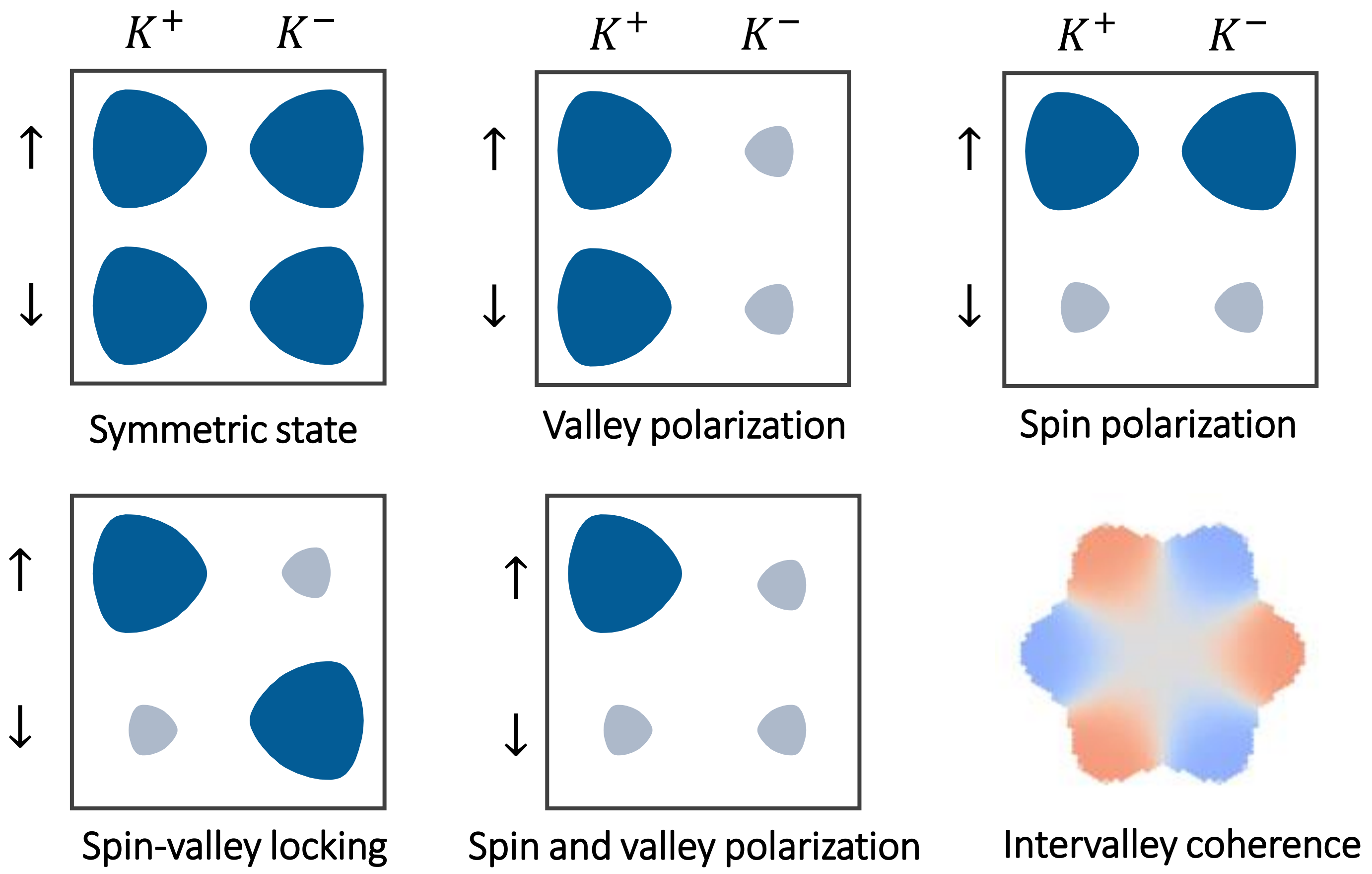
Zhou et al., Science (2022)

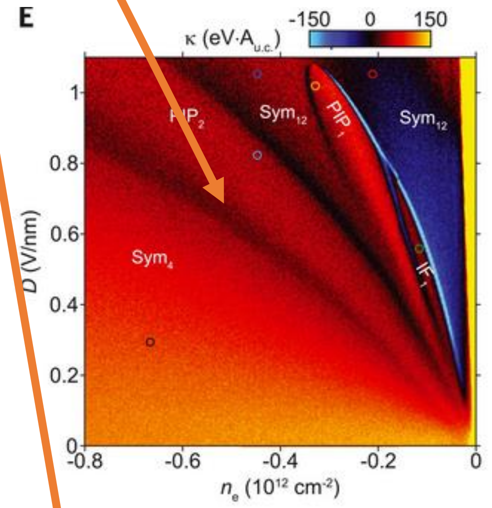
Flavor polarization

Symmetry breaking phase transitions between metals

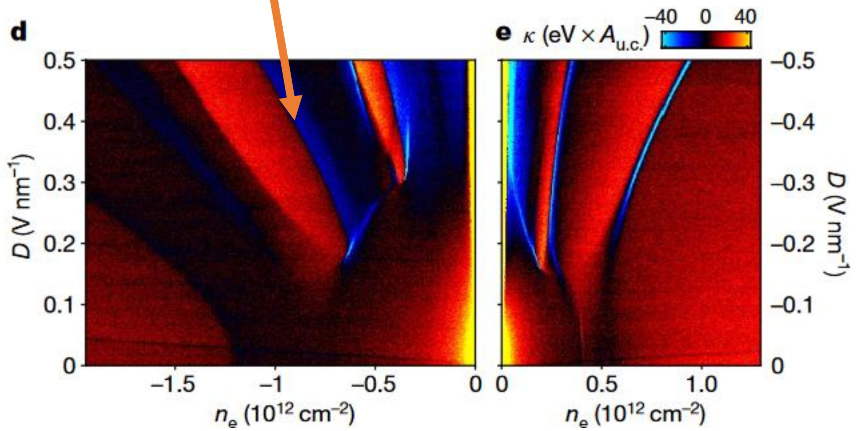
Four-fold degenerate band structure: valleys (**K, K’**) + spins (**↑,↓**)

Prone to **symmetry breaking** (Stoner-type) instabilities





Bilayers: Zhou et al, Science (2022)



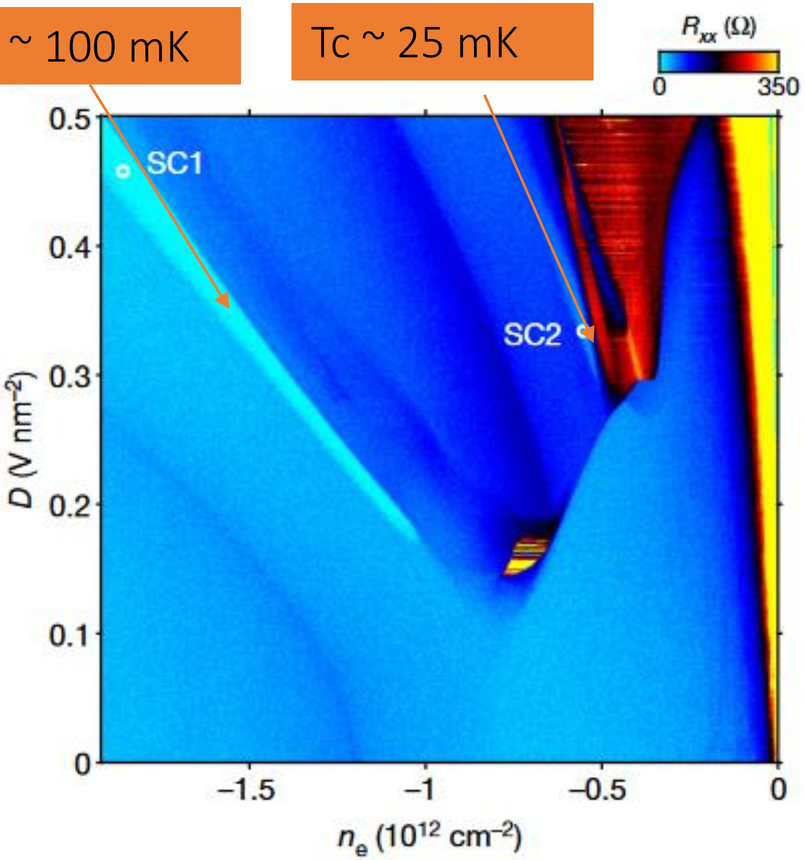
Trilayers: Zhou et al, Nature (2021)

Superconductivity in rhombohedral graphene

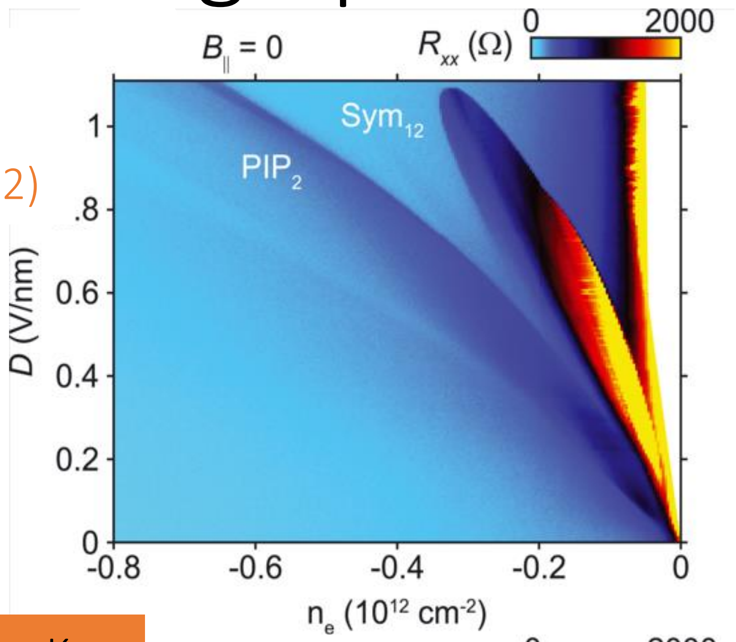
Trilayers:
Zhou et al., Nature (2021)

Tc ~ 100 mK

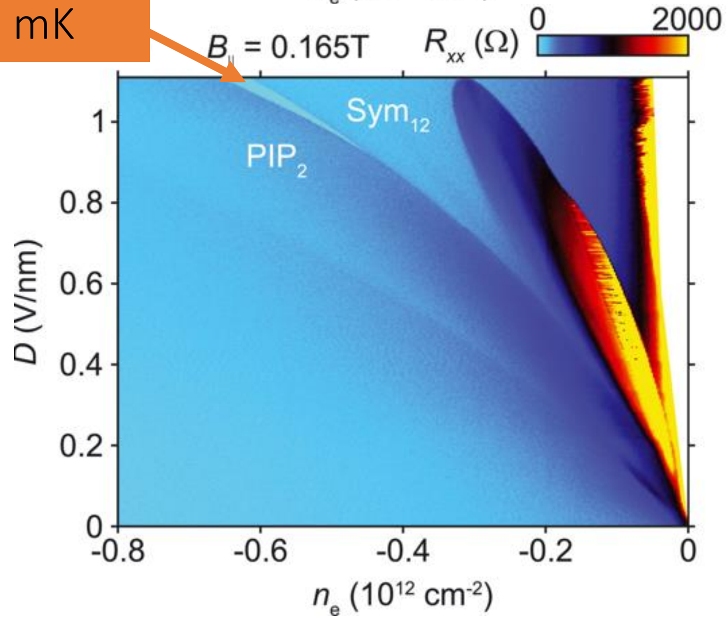
Tc ~ 25 mK



Bilayers:
Zhou et al, Science (2022)



Tc = 25 mK



SC induced by in-plane B field!

Pairing mechanism?

- Order parameter fluctuations close to phase transitions

  Chatterjee, Wang, Berg & Zaletel , Nat. Comm. **13**, 6013 (2022)
  Dong, Lee & Levitov, PNAS **120** (2023)
  Dong, Chubukov & Levitov, PRB **107,** 174512 (2023)
  Dong, Levitov & Chubukov, PRB **108**, 134503 (2023)

- Kohn-Luttinger (over-screened Coulomb interactions)

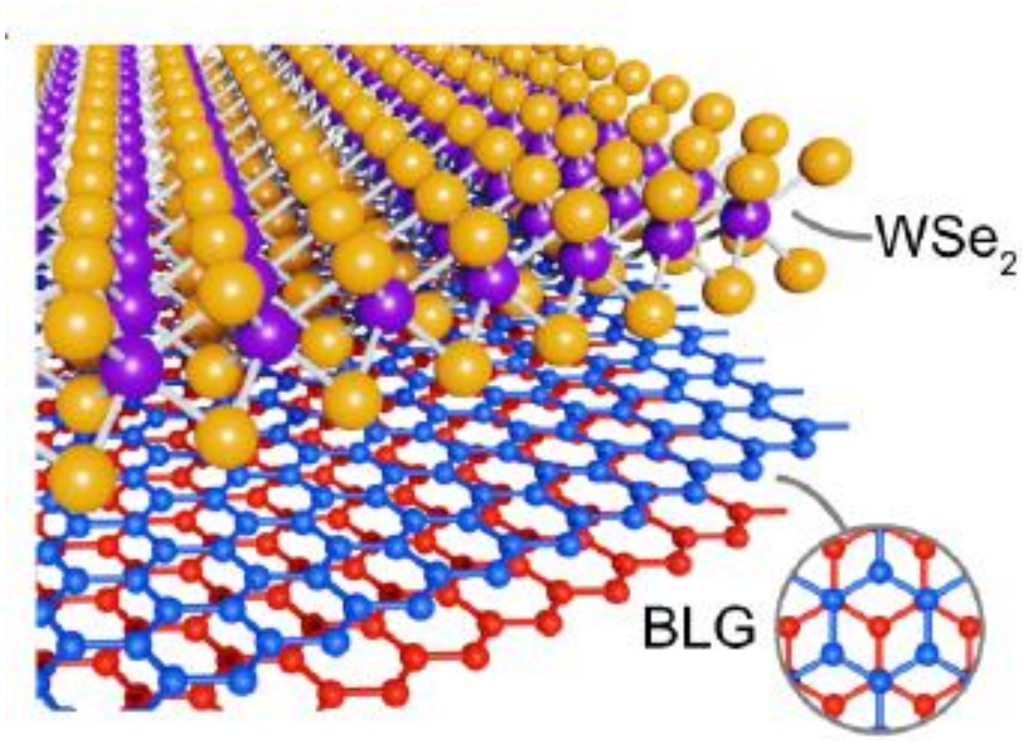
  Ghazaryan, Holder, Serbyn & Berg, PRL **127**, 247001 (2021)
  Szabó & Roy, PRB **105**, L081407 (2022)
  You & Vishwanath, PRB **105**, 134524 (2022)
  Cea, Pantaleón, Phong, Guinea, PRB **105**, 075432 (2022)
  Qin, Huang, Wolf, Wei, Blinov & MacDonald, PRL **130**, 146001 (2023)
  Li, Kuang, Jimeno-Pozo, Sainz-Cruz, Zhan, Yuan & Guinea, PRB **108,** 045404 (2023)
  Lu, Wang, Chatterjee & You, PRB **106**, 155115 (2022)
  Dai, Ma, Zhang, Guo & Ma, PRB **107**, 245106 (2023)
  Wagner, Kwan, Bultinck, Simon & Parameswaran, arXiv:2302.00682
  Son, Hsu & Kim, arXiv:2405.05442

- Phonon mediated

  Chou, Wu, Sau & Das Sarma, PRL **127**, 187001 (2021)
  Chou, Wu, Sau & Das Sarma, PRB **105**, L100503 (2022)
  Chou, Wu, Sau & Das Sarma, PRB **106**, 024507 (2022)
  Vinas Bostrom, Fischer, Profe, Zhang, Kennes & Rubio, npj Computational Materials **10** (2024).

And more…

1. Enhanced superconductivity in BLG-WSe2



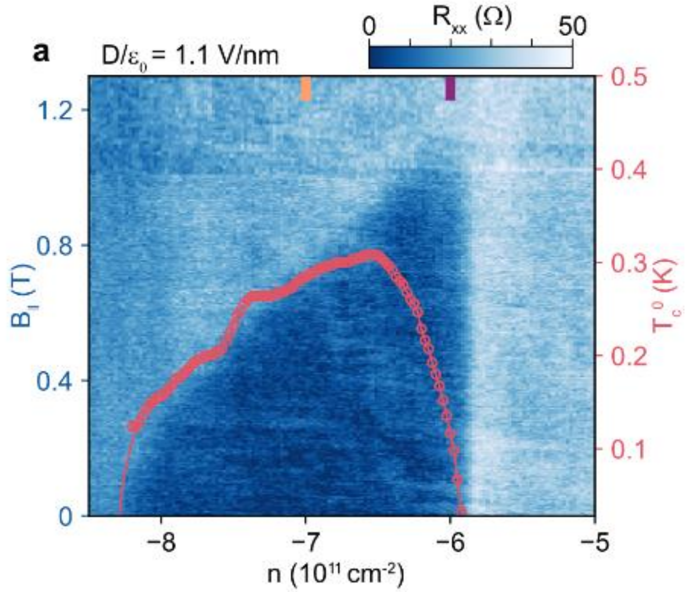
A surprise!

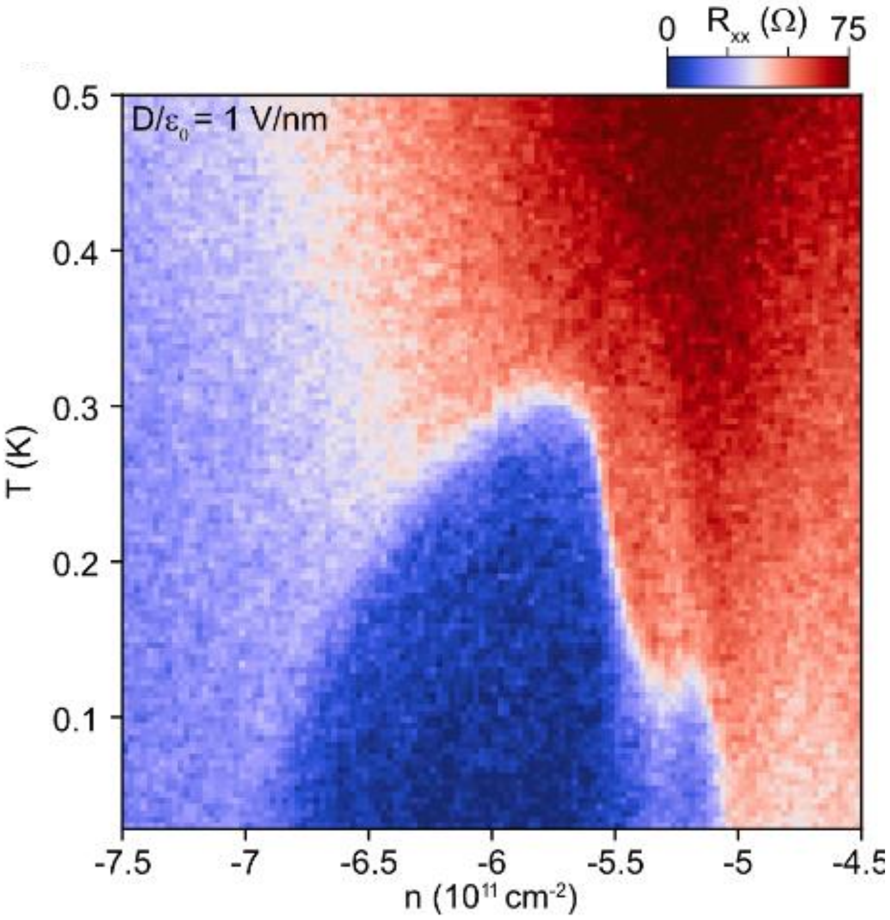
Zhang, Polski, Thomson, **ÉLH**, *et al.*
Nature **613**, 268-273 (2023)

Large SC region observed at **B = 0**

Only for D > 0: proximity to WSe_2 favors SC

Strongly **density-dependent** Pauli limit violation (singlet vs triplet like response)

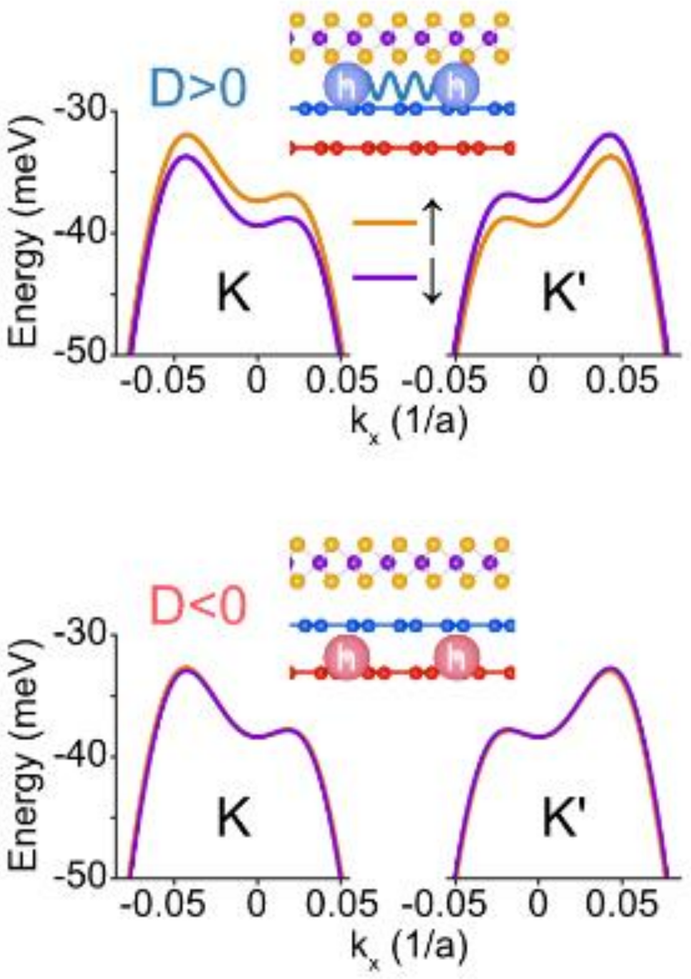




Tc = 300 mK

Pairing strongly enhanced!

Spin-orbit coupling in BLG



Intrinsically weak (~ 50 μeV). Can be enhanced by proximity effect

- Two main contributions: Ising and Rasha SOC

$$\frac{\lambda_I}{2}\tau_z s_z \ , \quad \frac{\lambda_R}{2}(\tau_z\sigma_x s_y - \sigma_y s_x)$$

$$(\tau, \sigma, s) = (\mathrm{valley}, \mathrm{sublattice}, \mathrm{spin})$$

- Ising SOC couples asymmetrically with D field: “spin-orbit valve”

Gmitra and Fabian, PRL (2017)
Khoo, Morpurgo and Levitov, Nano Letters (2017)

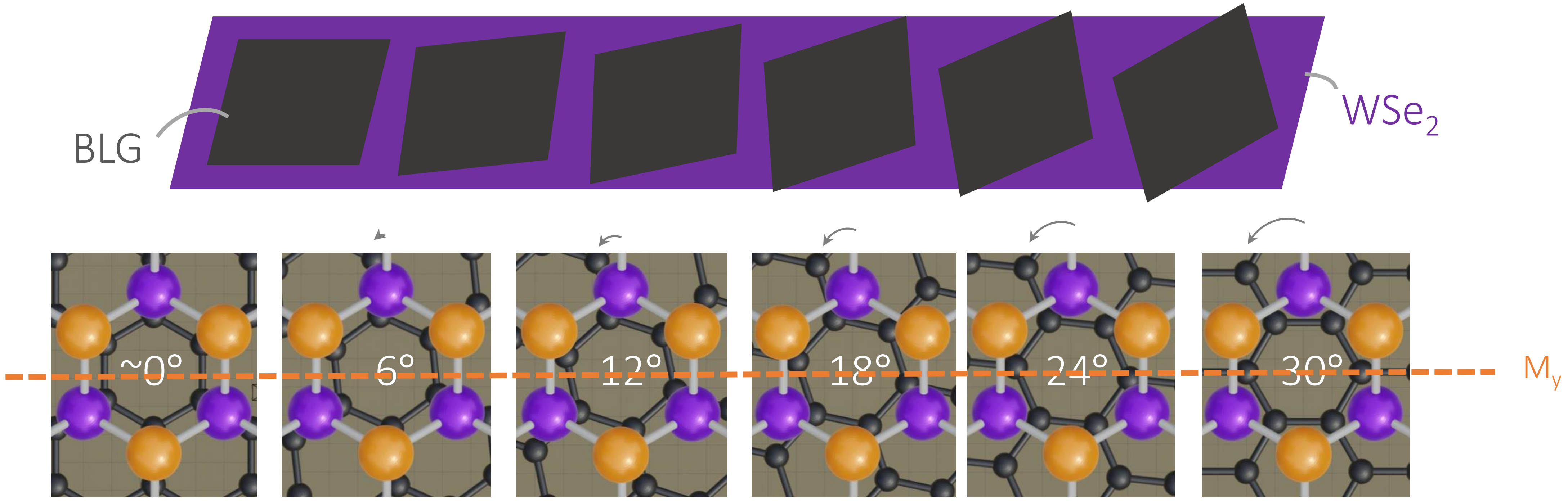
- Quantum Hall measurements:

$$\lambda_I \sim 0.6 - 1.6 \ \mathrm{meV}$$

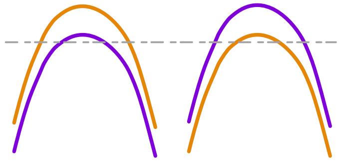
Tuning SOC with interfacial twist

Yiran Zhang *et al.*, arXiv:2408.10335

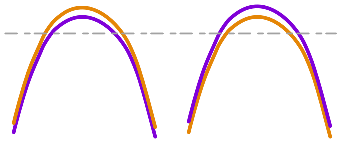
Tuning SOC with interfacial twist



large Ising



small Ising

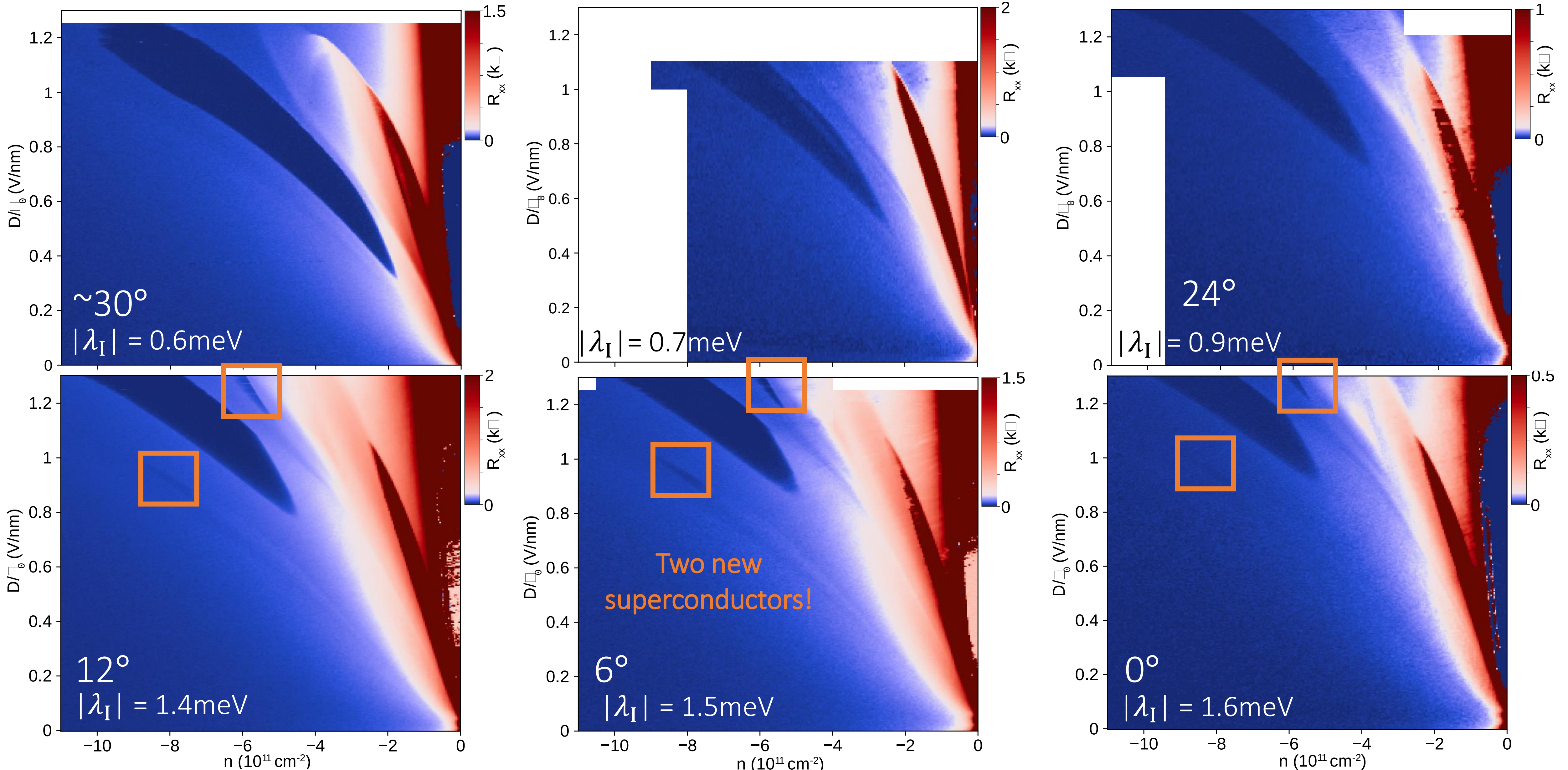


Ising SOC controlled by breaking M_y

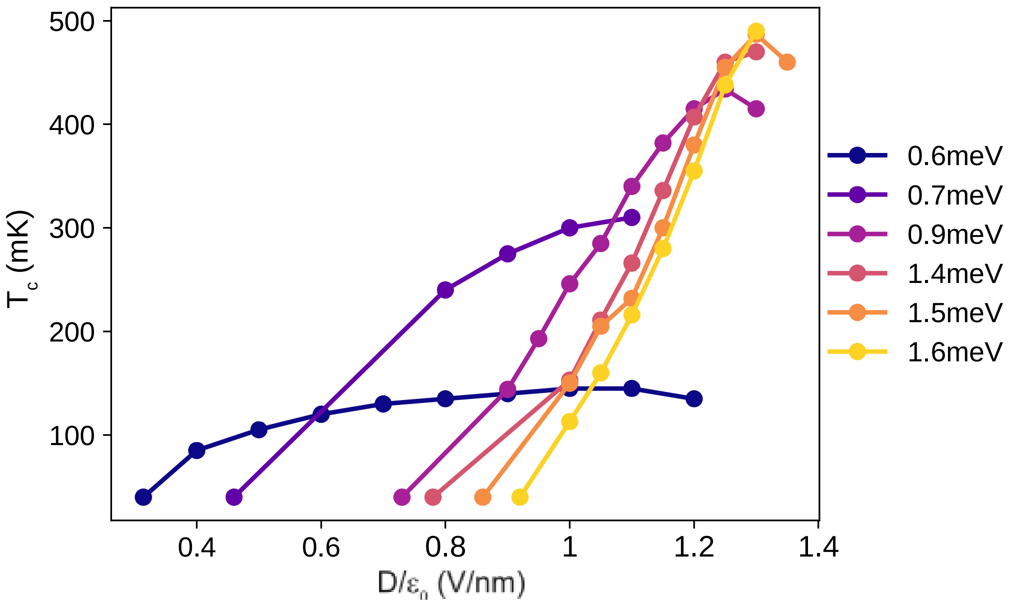
Li & Koshino, PRB **99**, 075438 (2019)
David *et al.*, PRB **100**, 085412 (2019)

Yiran Zhang *et al.*, arXiv:2408.10335

Phase diagrams for different Ising SOC



Trends (large SC pocket)



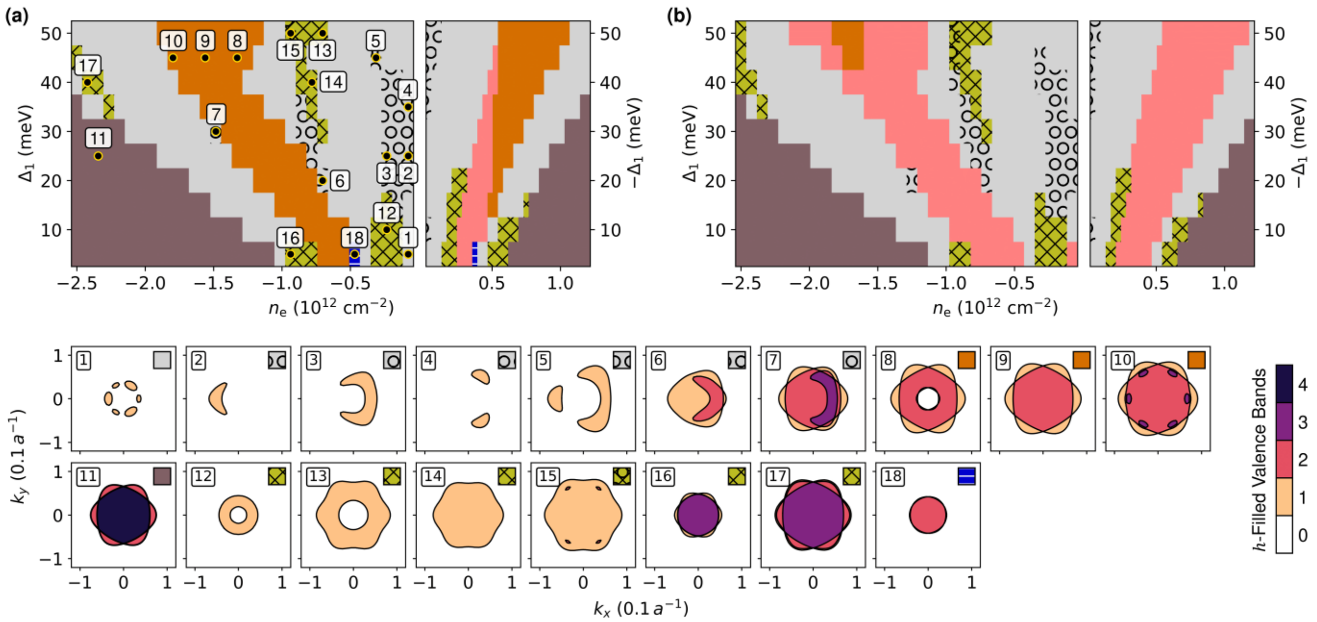
Increasing Ising SOC

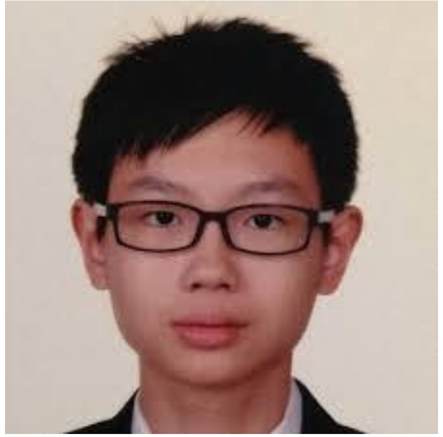
- Superconductivity is pushed to higher *D* field
  - Higher critical temperature

These opposing trends provide non-trivial constraints for theory!

- Understand the parent normal state
- Pairing mechanism?

2. Phase diagrams of spin-orbit coupled graphene multilayers



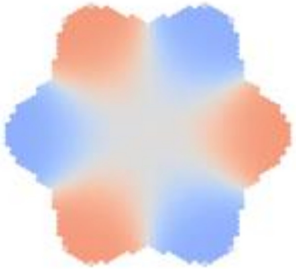


Jin Ming Koh

Trilayers: Koh, Alicea and **ÉLH**, Phys. Rev. B **109**, 035113 (2024)
Bilayers: Koh, Thomson, Alicea and **ÉLH**, arXiv:2407.09612

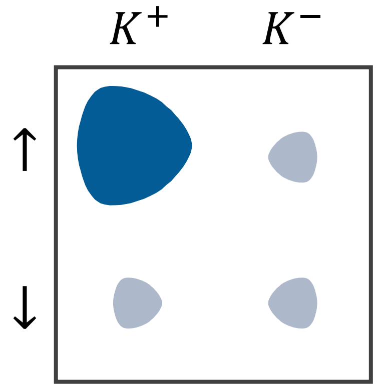
See also:
Xie & Das Sarma, PRB **107**, L201119 (2023)
Zhumagulov, Kochan & Fabian, PRB **110**, 045427 (2024)
Wang, Vila, Zaletel & Chatterjee, PRL **132**, 116504 (2024)
Zhumagulov, Kochan & Fabian, PRL **132**, 186401 (2024)

Bilayer graphene phase diagram

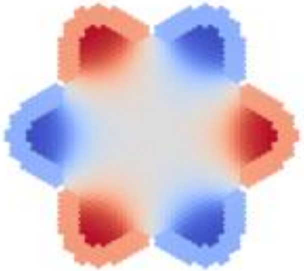


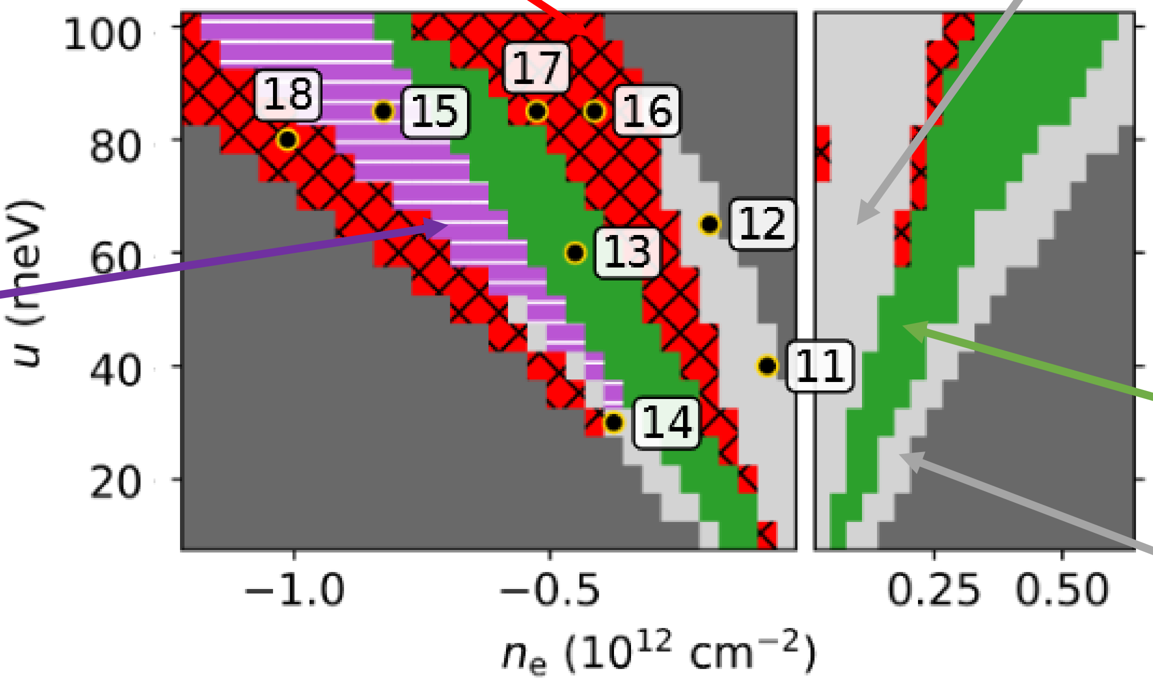
Spin-polarized, quarter-metal IVC

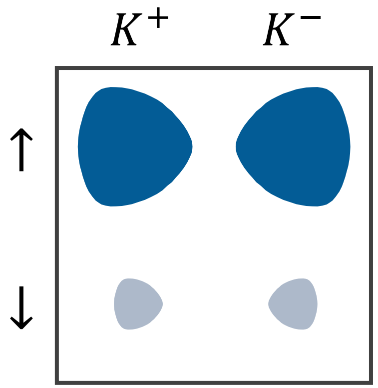
Stoner quarter metals



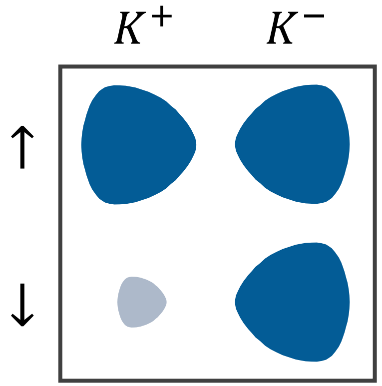
Half-metal IVC







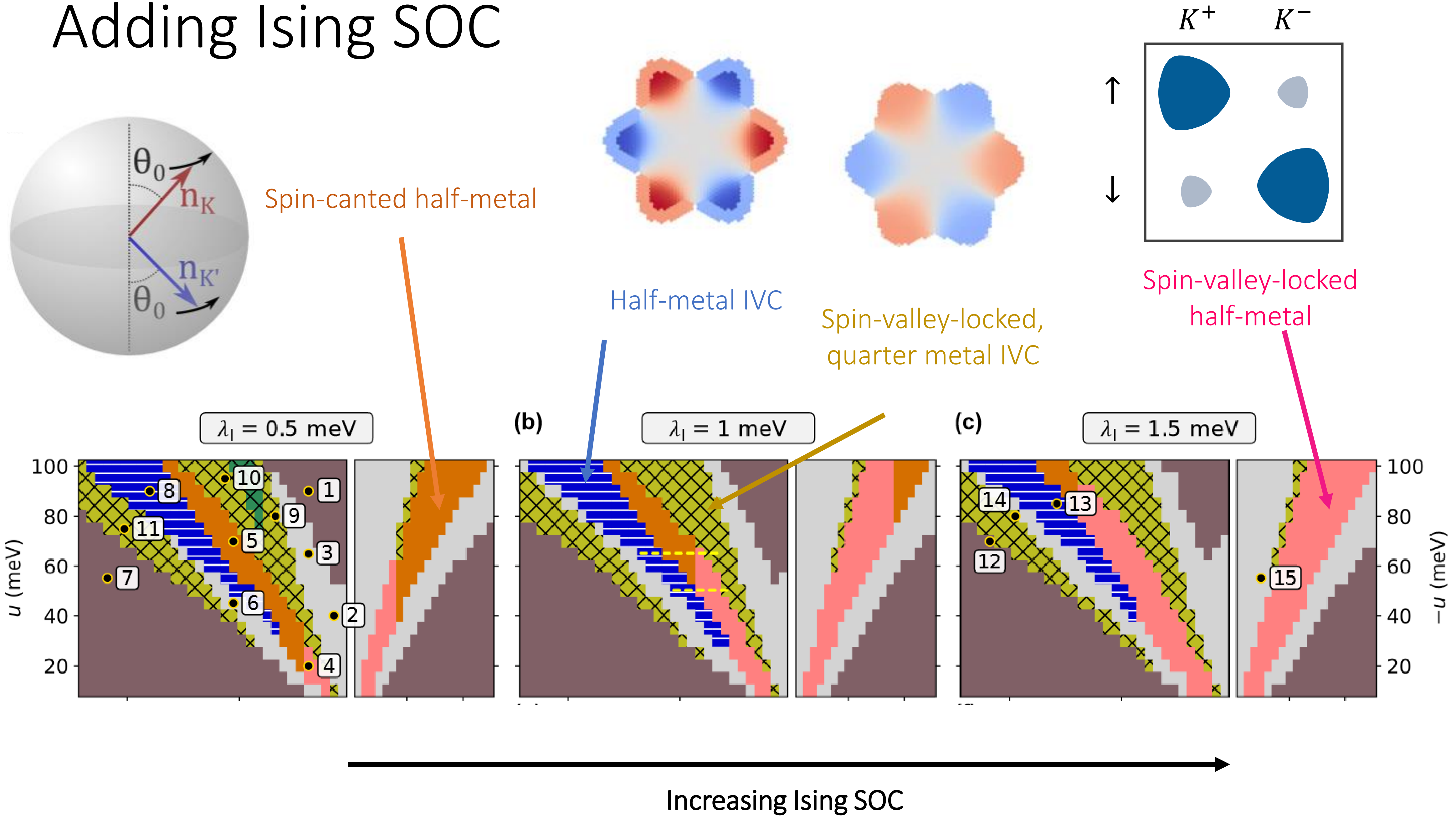
Spin polarized half-metal



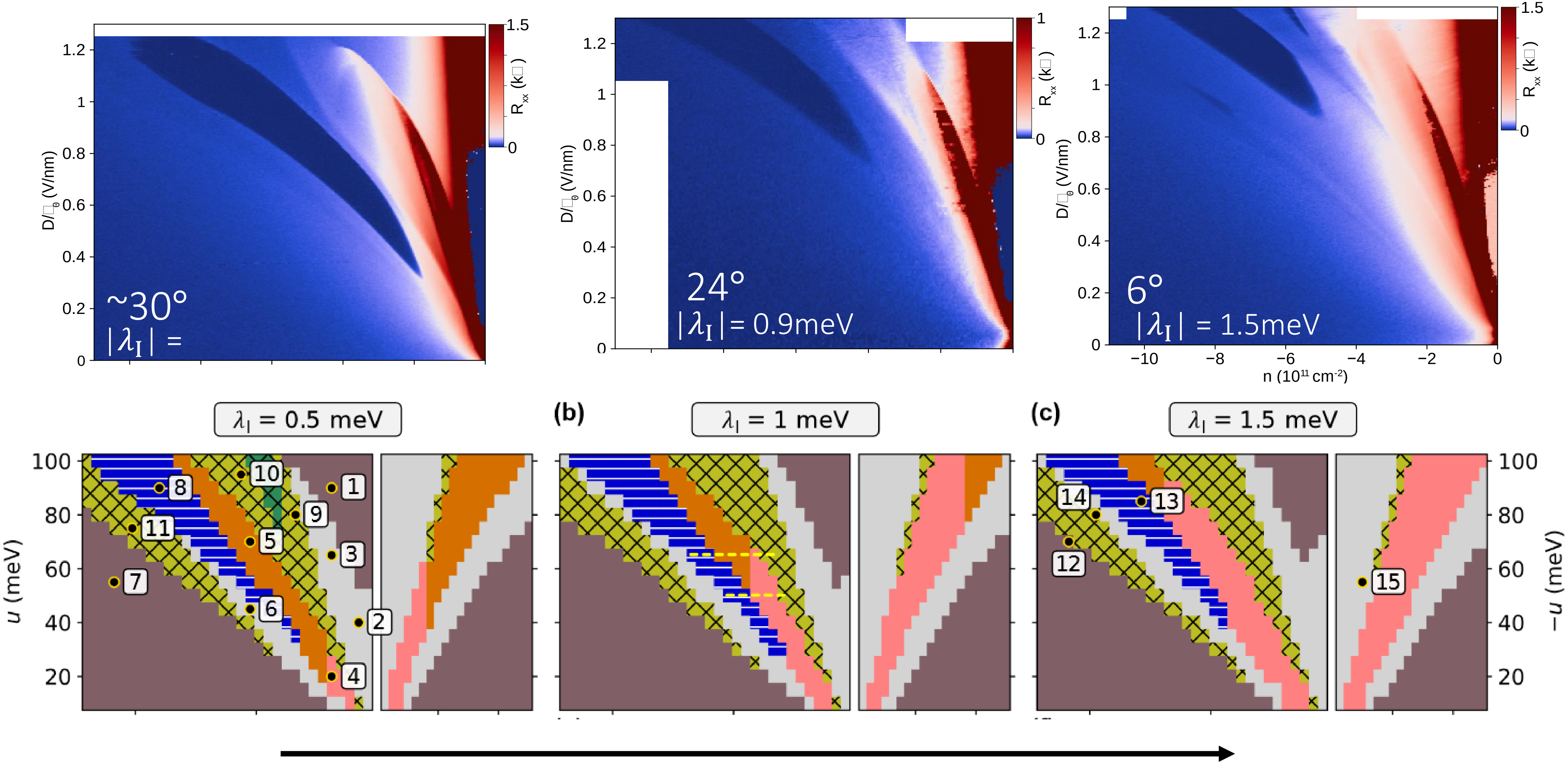
Only capturing **commensurate** IVC orders

Includes (ferromagnetic) Hund's coupling but no spin-orbit

Adding Ising SOC



Comparing with experiments



Increasing Ising SOC

Identifying the normal state

Hartree-Fock suggests two candidate half-metal states suppressed by Ising SOC:

- **Spin-canted phase:** breaks U(1) spin rotations along the z-axis
- **Inter-valley coherent state**: breaks U(1) valley conservation

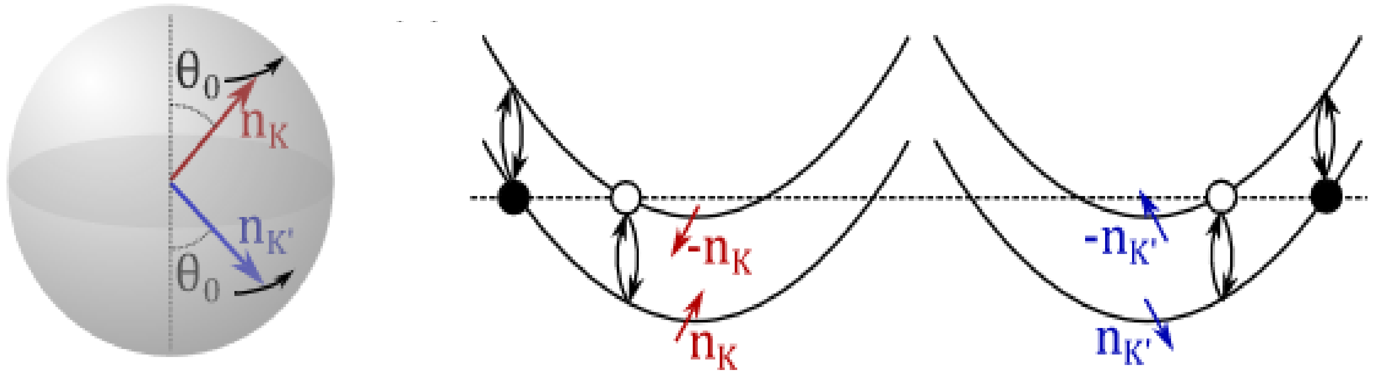
Low-energy collective modes (Goldstone magnons) could mediate Cooper pairing

Dong, **ÉLH** & Alicea, arXiv:2406.17036

Similar ideas for IVC fluctuations

Chatterjee et al, Nature Communications (2022)
You and Vishwanath, PRB (2022)
Dong, Lee, Levitov, PNAS (2023)
Vituri, Xiao, Pareek, Holder, Berg, arXiv:2408.10309

3. Pairing from magnons?



Dong, **ÉLH** and Alicea, arXiv:2406.17036

+ Collaboration with Andrea Young's group
C. Patterson, O. Sheekey, T. Arp, L. Holleis, J.M. Koh, … ,
ÉLH, J. Alicea and A. Young, arXiv:2408.10190

Spin canting order

C. Patterson, O. Sheekey, T. Arp, L. Holleis, J.M. Koh, … , **ÉLH**, J. Alicea and A. Young
arXiv:2408.10190

Competition between Ising SOC and Hund's coupling

Spin polarization in each valley

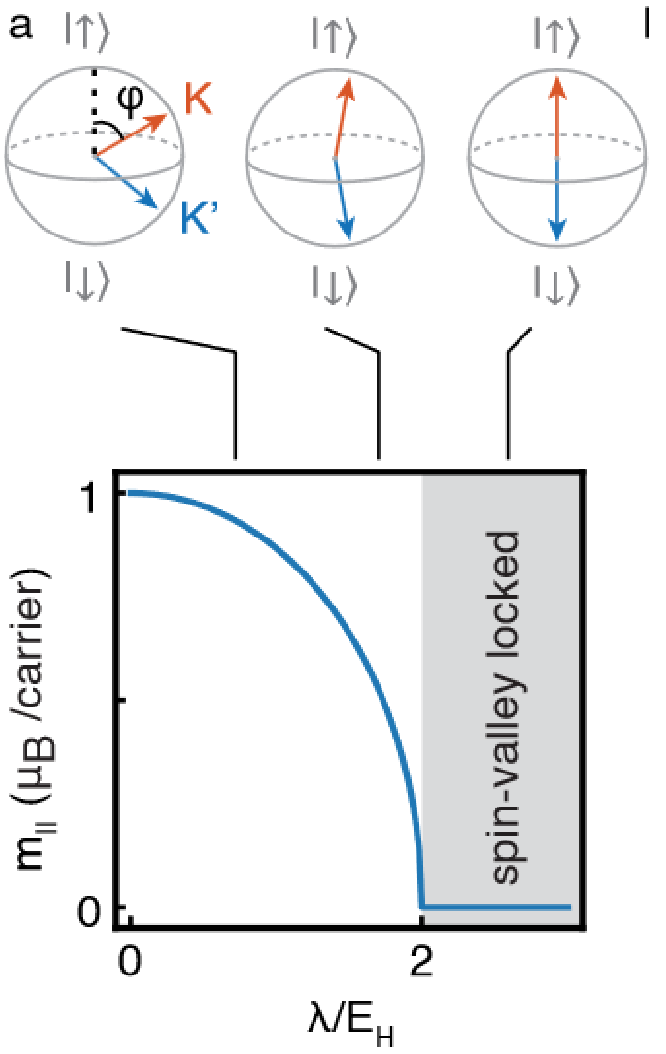
$$\mathcal{F} = \frac{\kappa}{2}(\mathbf{n}_K^2 + \mathbf{n}_{K'}^2) - J\mathbf{n}_K \cdot \mathbf{n}_{K'} + \frac{\lambda}{2}(n_K^z - n_{K'}^z) + \ldots$$

Controls onset of Stoner ferromagnetism (Coulomb + kinetic energy)

Ferromagnetic Hund's coupling arises from lattice-scale electron-electron repulsion

Two valley-balanced solutions: spin-valley locked & spin canted

Spin canting breaks continuous U(1) spin rotations
-> Goldstone modes

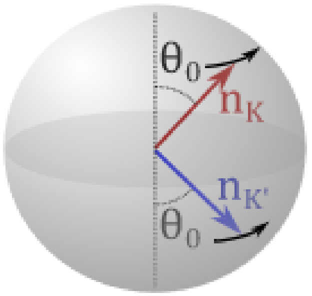


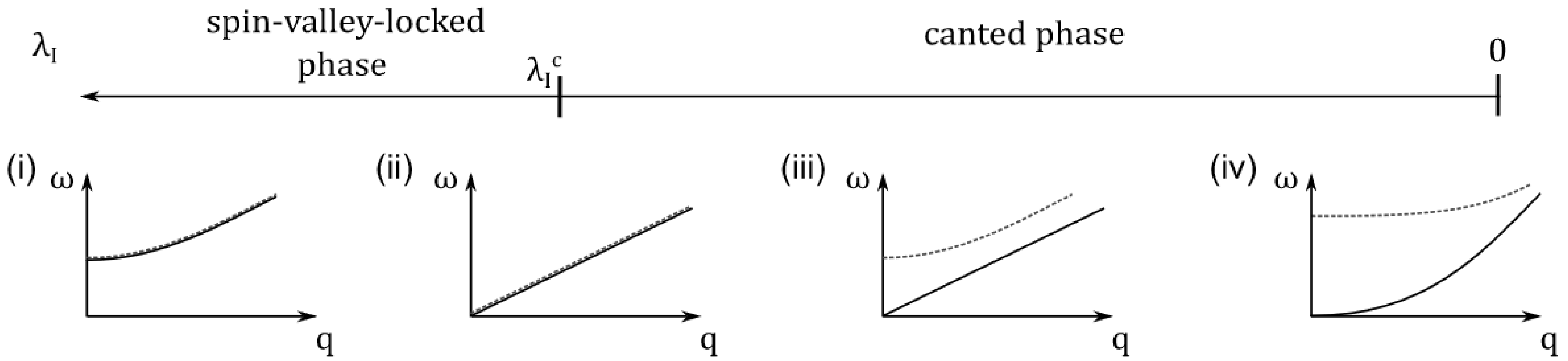
$$E_H = J|\mathbf{n}_{K,K'}|$$

Low-energy magnons

Goldstone mode: valley-symmetric azimuthal fluctuations

Critical mode: valley-antisymmetric polar fluctuations





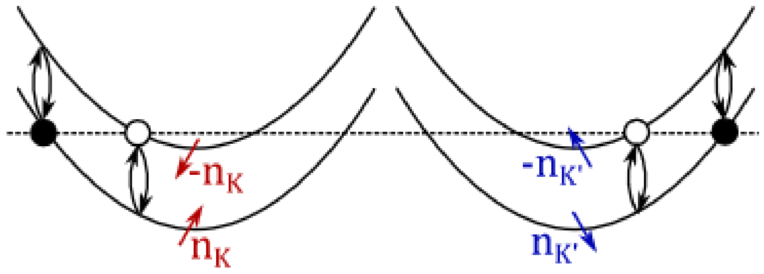
Spin-valley-locked (symmetric) phase

Critical point: two gapless modes

Spin-canted phase, U(1) ferromagnet

Spin polarized phase, SU(2) ferromagnet

Magnon mediated interactions



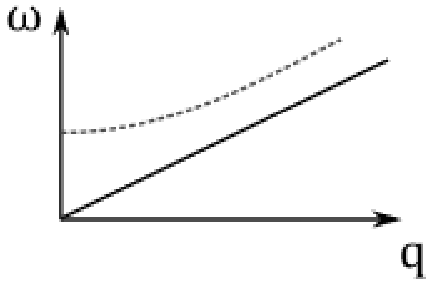
- Spin-flipping processes
- Qualitatively different scaling in two regimes:

band indices

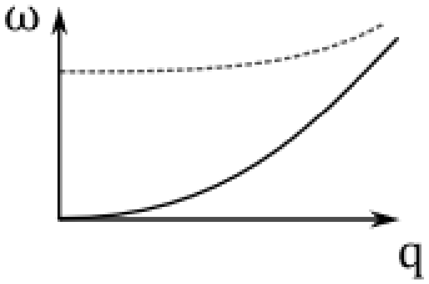
$$g_{\xi_1,\xi_2}^{-+,+-}(\nu,\mathbf{q}) \sim -\frac{\cos^2\theta_0}{2z_s\kappa}\frac{1}{\nu^2 + c_s^2\mathbf{q}^2}$$

valley indices

$$g_{\xi_1,\xi_2}^{-+,+-}(\nu,\mathbf{q}) \sim -\frac{1}{-i\gamma\nu + D\mathbf{q}^2}$$

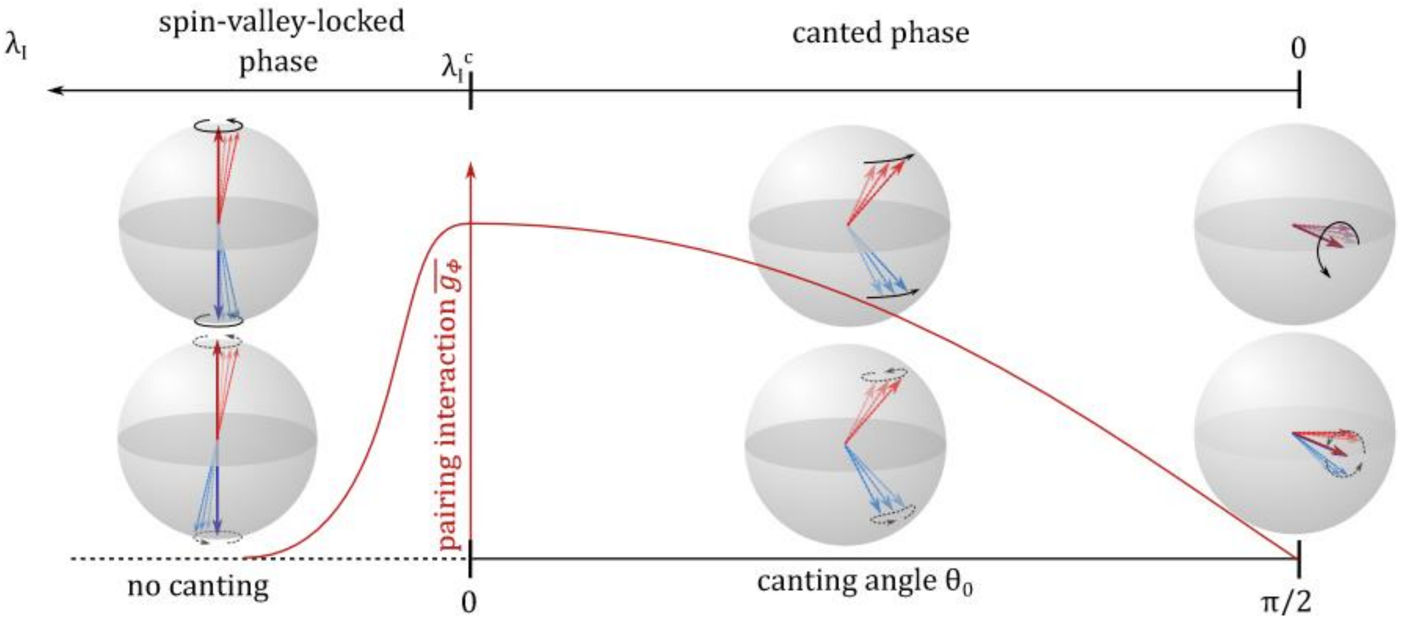


Spin-canted phase,
U(1) ferromagnet



Spin polarized phase,
SU(2) ferromagnet

Pairing from magnons



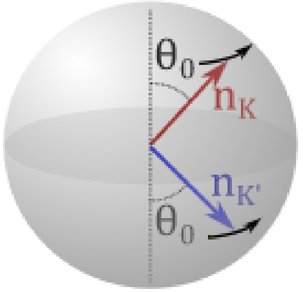
Leading-order diagrams:

Pairing interaction (integrated over the Fermi surface):

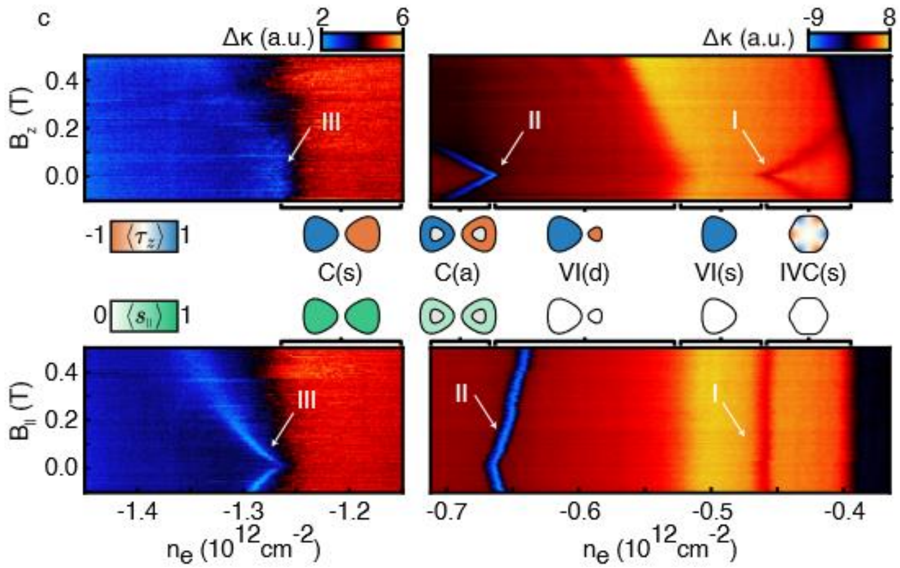
Sign refers to majority/minority Fermi surfaces

$$\overline{g^{(2)}_{\pm}}(\nu) \sim \frac{\cos\theta_0}{k_F^{\pm}} \ln\frac{|\nu|}{\Lambda}$$

- Crucially depends on SOC: vanishes in the FM limit, $\theta_0 \to \pi/2$
- Low-frequency divergence: strong retardation effects
- Is enhanced for small Fermi pockets



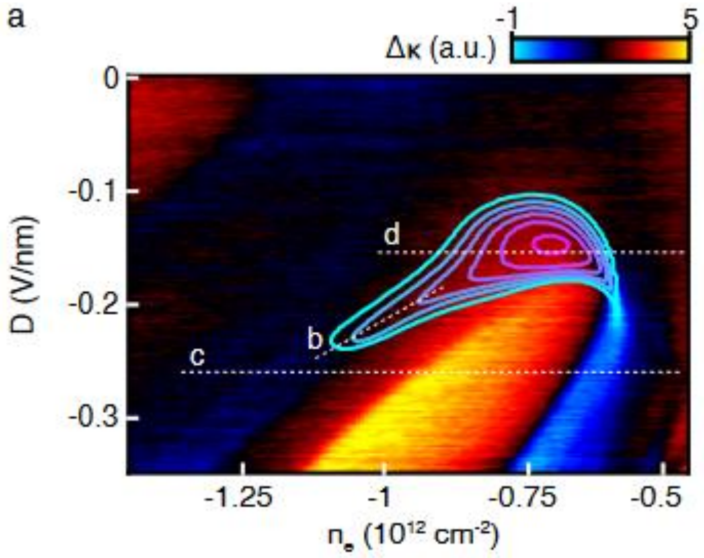
Pairing from magnons

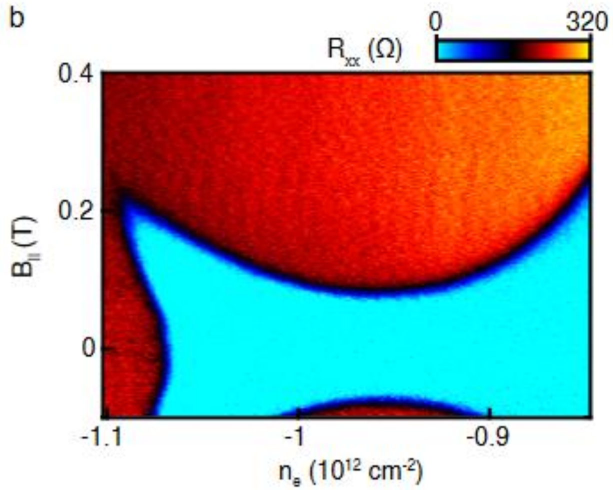


C. Patterson, O. Sheekey, T. Arp, L. Holleis, J.M. Koh, … , **ÉLH**, J. Alicea and A. Young arXiv:2408.10190

Our theory predicts that the normal state hosting SC should host a **spontaneously-generated, in-plane magnetic moment**

- Could be resolved in magnetic scanning probes (SQUID on tip) or with magneto-transport/compressibility
- Collaboration with Andrea Young's group on trilayers/WSe2





Outlook

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GORDON AND BETTY MOORE FOUNDATION

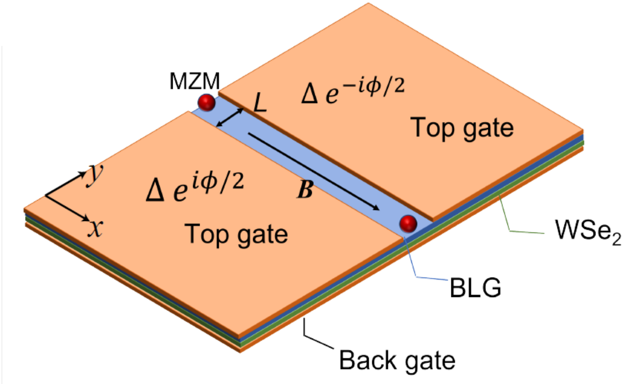
Rhombohedral graphene: exciting interplay of correlations, SOC and superconductivity deep in the clean limit

- Tunable and rich phase diagram. Control over microscopic parameters (e.g. band dispersion and induced SOC) highly constrains theory
- Multiple unconventional superconductors, possibly exotic pairing mechanisms
- **How to experimentally access collective modes and probe their (spin/valley) structure?**
- **Can constrain pairing mechanisms?** (e.g. gapping out magnon modes due to applied magnetic field. Analog for IVC fluctuations?)
- Investigate **dynamical** and/or **beyond mean-field** methods…

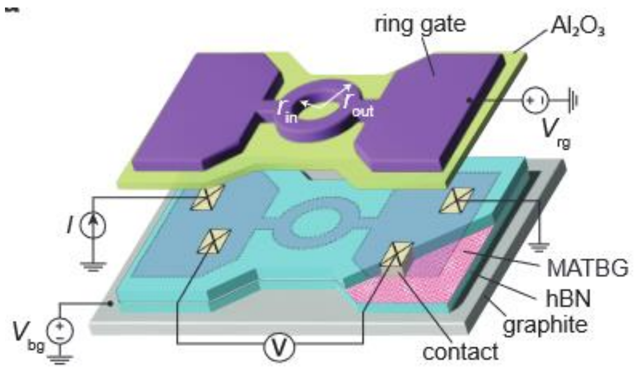
Vituri, Xiao, Pareek, Holder, Berg, arXiv:2408.10309
Wolf, Wei, Zhou, Huang, arXiv:2408.15884

c.f. discussion by Agnes Valenti last week

- **Gate-defined** architectures for quantum devices?



Xie, **ÉLH,** Young, Nadj-Perge & Alicea, Phys. Rev. Lett. **131**, 146601 (2024)



Iwakiri *et al.*, Nat. Comms. **15**, 390 **(2024)**

Extras

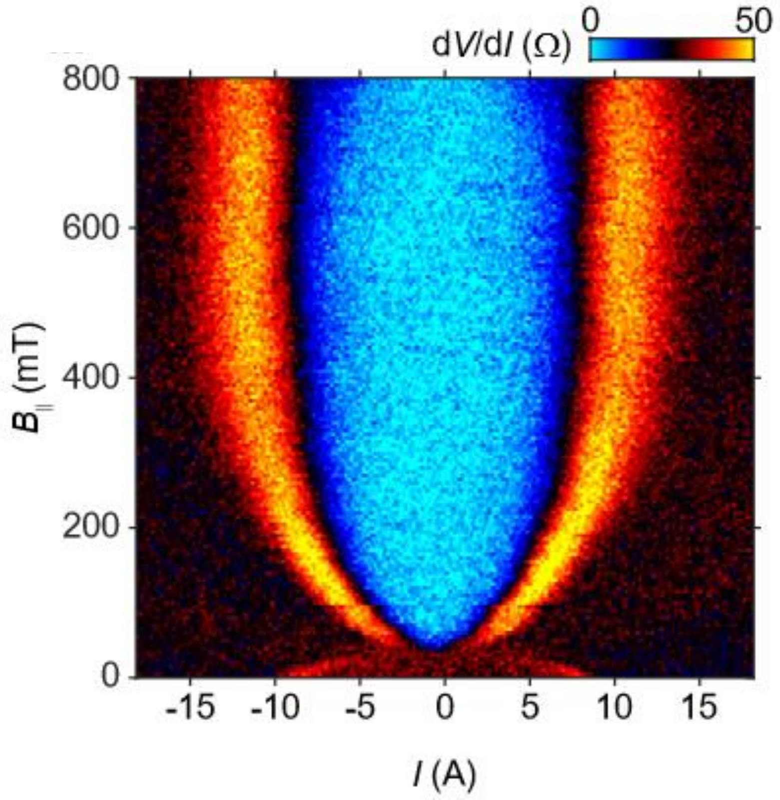
SC induced by a magnetic field?

Large (> 20) violation of Pauli limit:

$$B_p = \frac{\Delta}{\sqrt{2}\mu_B} = \frac{1.76 k_B T_c}{\sqrt{2}\mu_B} \approx \ 40 \text{ mT}$$

Ginzburg-Landau coherence length $\xi_{\text{GL}} \ll l$ Electronic mean free path

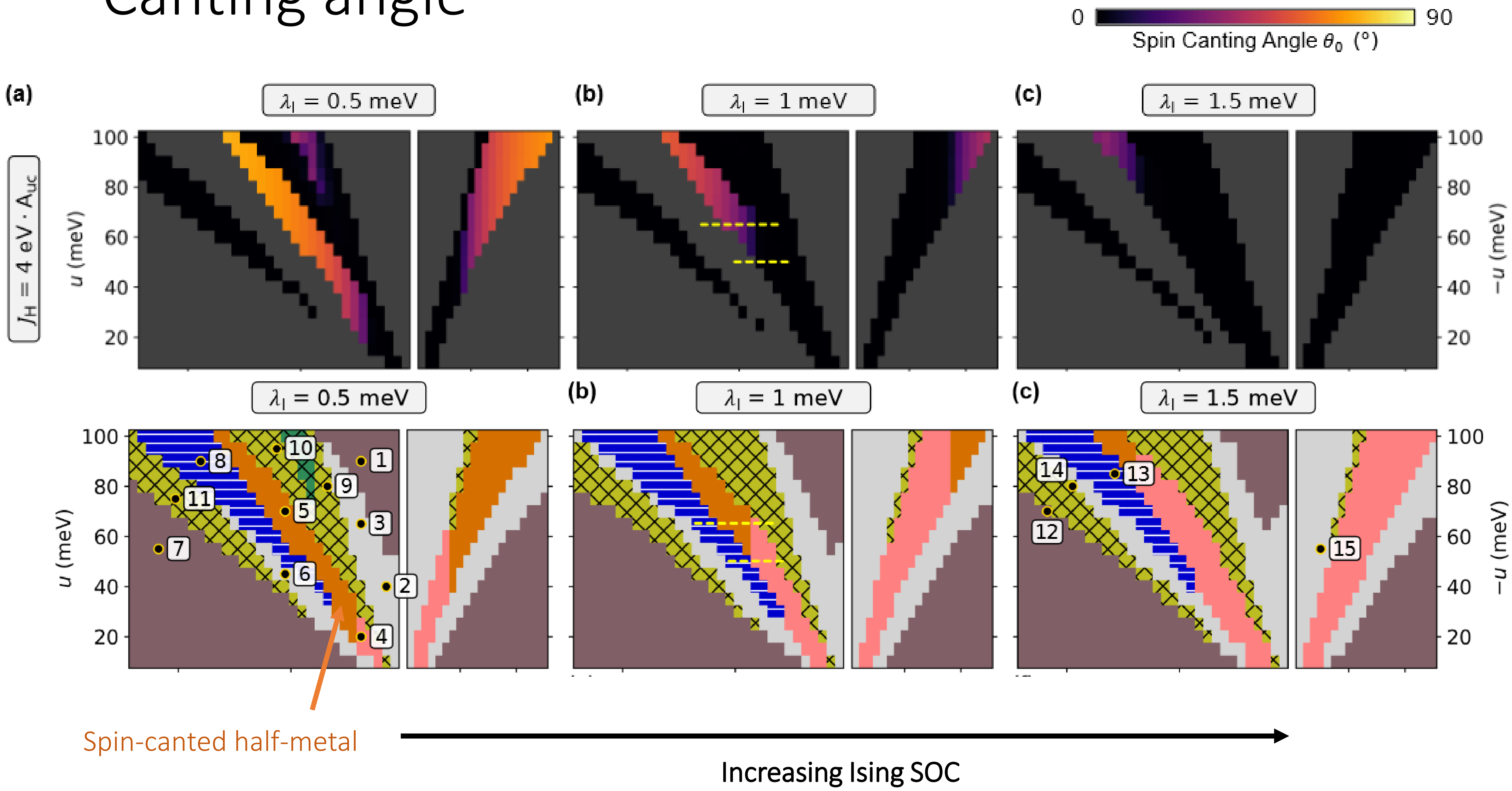
$$\xi_{\text{GL}} \sim 250 \text{ nm} \qquad l \sim 10 \ \mu\text{m}$$



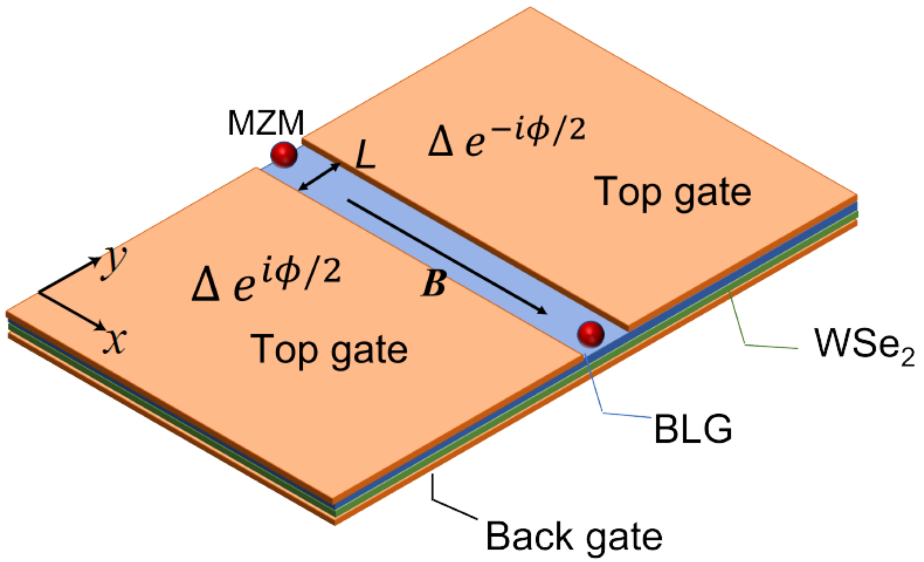
Anderson theorem: singlet superconductors are stable against T-preserving disorder

P. W. Anderson, Theory of dirty superconductors, J. Phys. Chem. Solids. **11**, 26–30 (1959)

Canting angle



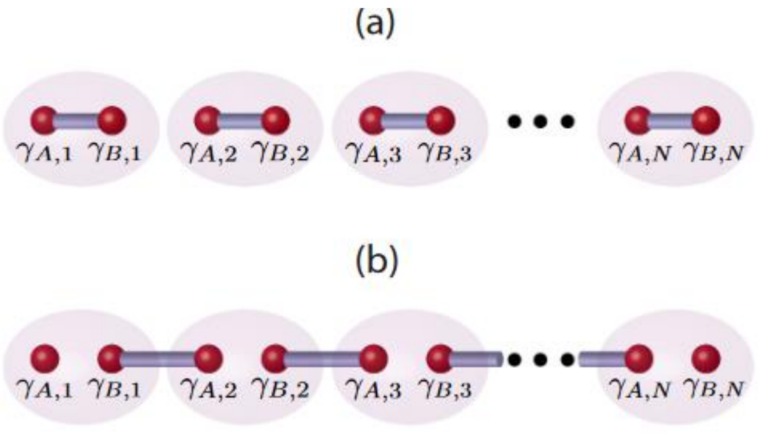
4. Gate-defined topological Josephson junctions



Xie, **ÉLH,** Young, Nadj-Perge & Alicea, Phys. Rev. Lett. **131**, 146601 (2024)

Topological SC and the Kitaev model

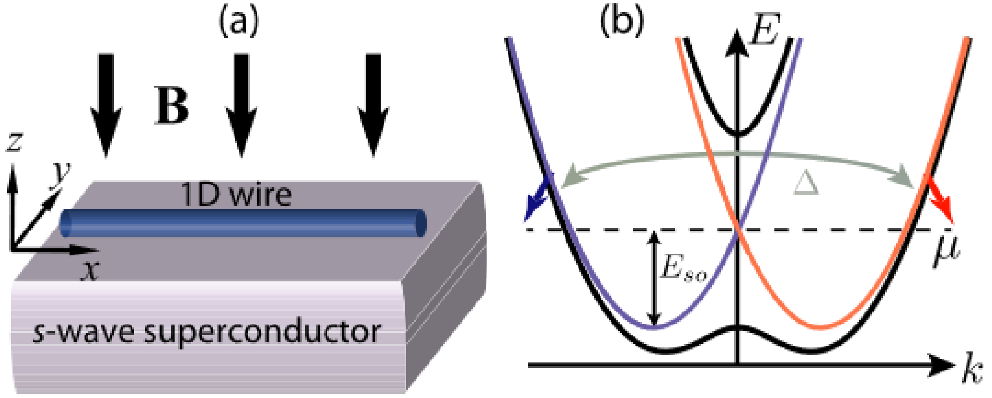
- Cooper pairing in a 1D chain of **spinless fermions** can lead to topological SC with unpaired **Majorana zero-modes**
  Kitaev (2001)



Alicea, Rep. Prog. Phys (2012)

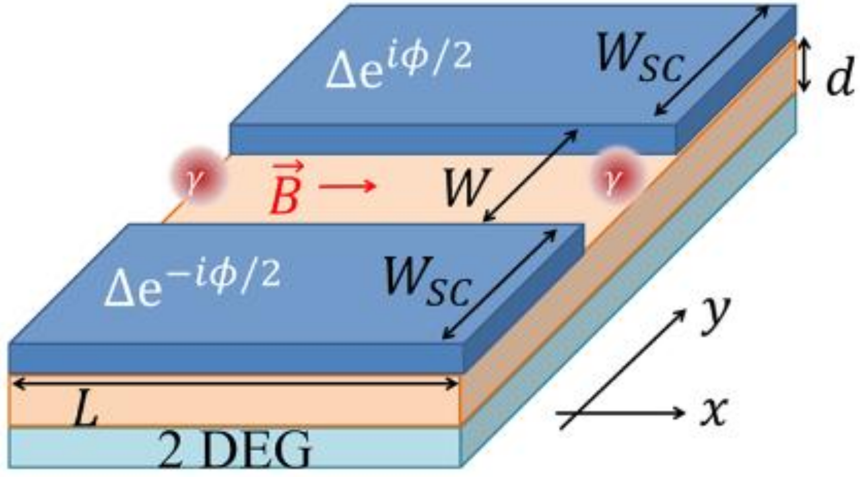
- Various theoretical proposals to create effectively spinless fermions

- Proximitized spin-orbit coupled nanowires at the heart of a multi-million $ effort!
  Oreg, Refael and von Oppen, PRL (2010)
  Lutchyn, Sau and das Sarma, PRL (2010)



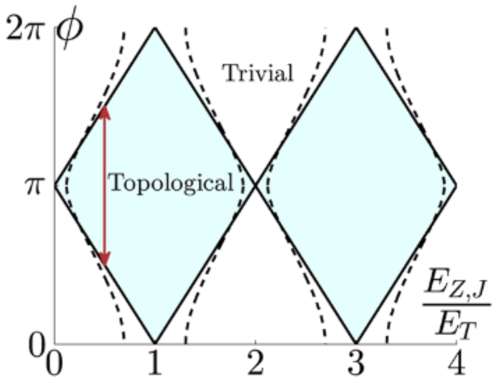
Microsoft

Topological SC in planar Josephson junctions



- SOC in 2D electron gas
- Proximity-induced SC
- In-plane Zeeman field
- **Phase difference**

Pientka, Keselman, Berg, Yacoby, Stern & Halperin, PRX (2017)
Hell, Leijnse & Flensberg, PRL (2017)



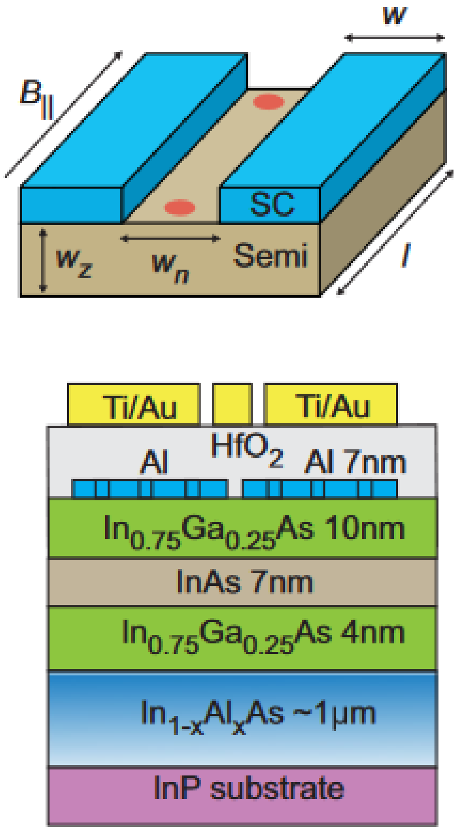
Topological SC in planar Josephson junctions

Current experiments rely on **complex heterostructures**:

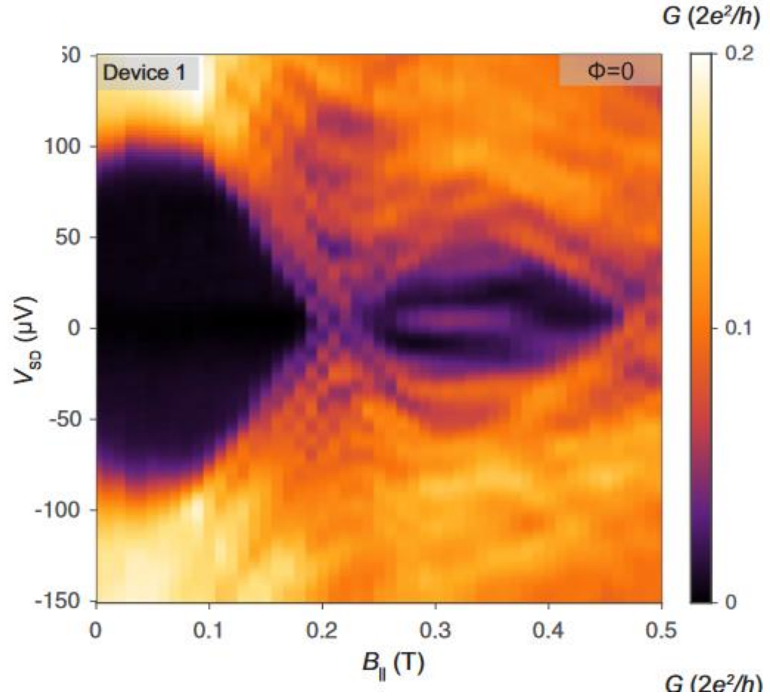
- SC film (Al or NbTiN)
- 2DEG (HgTe, InAs or InSb quantum wells)

Ren *et al.*, Nature **569**, 93 (2019)
Fornieri *et al.*, Nature **569**, 89 (2019)
Ke *et al.*, Nat. Commun. **10**, 3764 (2019)
Dartiailh *et al.*, PRL **126**, 036802 (2021)
Banerjee *et al.*, PRB **107**, 245304 (2023)

Can we realize this physics in an **intrinsic** junction?



Banerjee *et al.*,
PRB **107**, 245304 (2023)

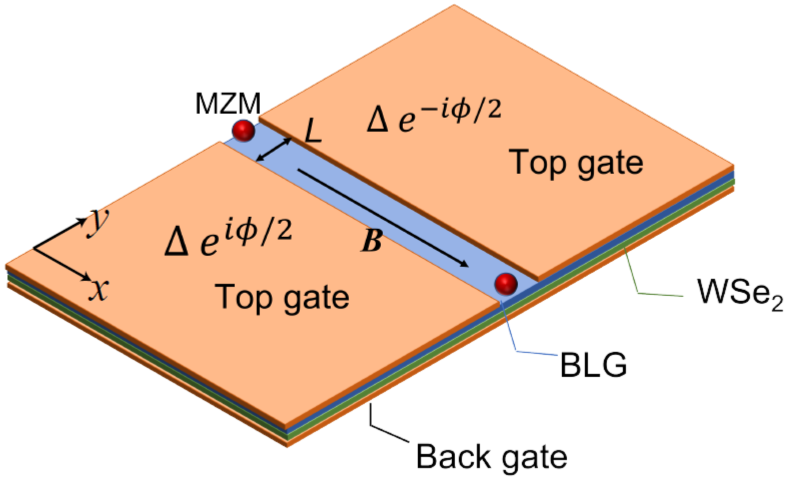


Tunneling conductance at the junction ends reveals gap closing followed by "zero-bias" features

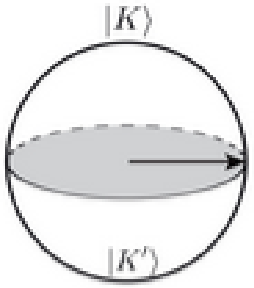
Topological SC in planar Josephson junctions

Superconductivity in BLG+WSe2 is **gate tunable**

- Intrinsic JJ by patterning with gates!
- Induced SOC + Zeeman field lifts the spin degeneracy
- Graphene entails an additional headache: the valley



Valley degeneracy can be lifted by interactions: inter-valley coherence



Potential payoffs:
- No material interfaces
- (Almost) no orbital coupling of **B**

Minimal model

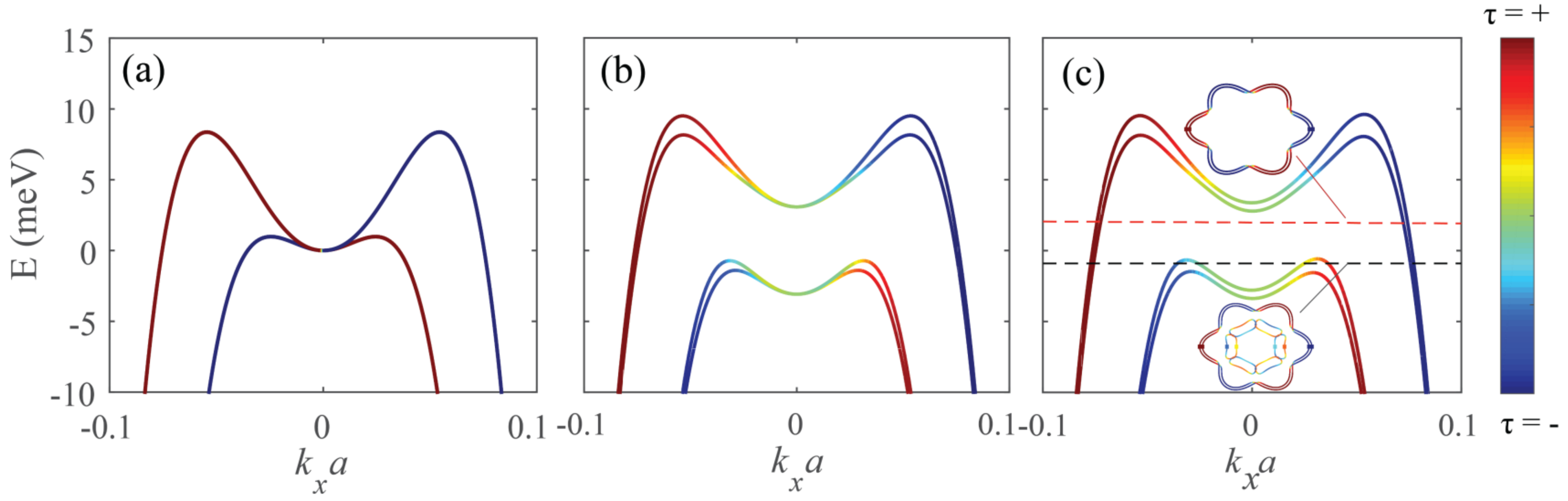
$$H(\boldsymbol{k}) = h_0(\boldsymbol{k}) + \beta_I \tau_z s_z + \alpha_R (k_x s_y - k_y s_x) + \lambda_0 \tau_x + \boldsymbol{h} \cdot \boldsymbol{s}$$

Zeaman coupling to in-plane field

Dispersion of BLG

Induced Ising and Rashba SOC

Inter-valley coherence



BLG dispersion

Adding IVC and SOC

Applying Zeeman field

Topological SC relies on inter-valley coherence
Can be turned into a diagnostic tool

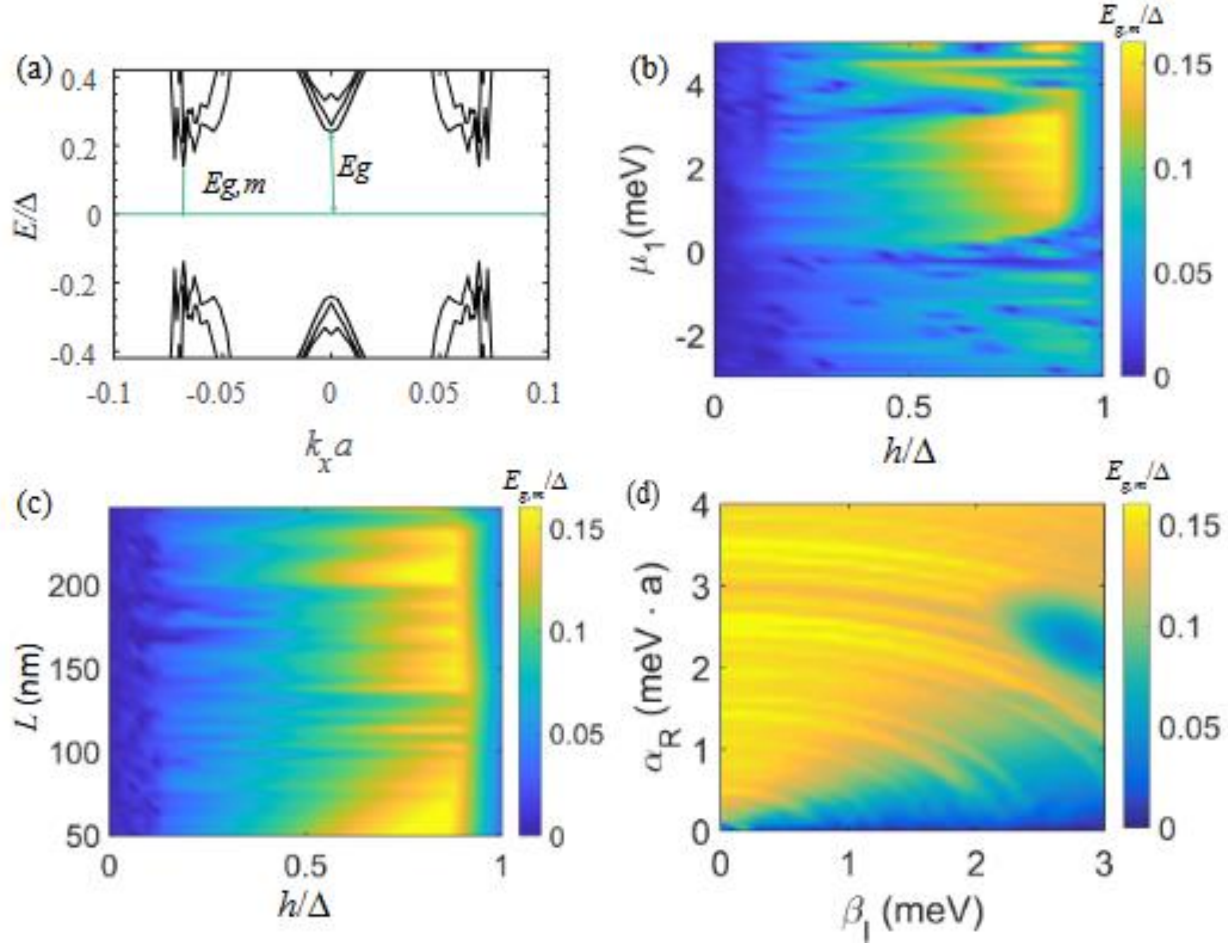
Optimizing the minimal topological gap

Minimal gap across the junction generically occurs for non-zero k_x

Optimal regime:

- Doping level without "polluting" Fermi pockets
- Short junction limit
- Effective SOC in low-energy bands dominated by Rashba contribution

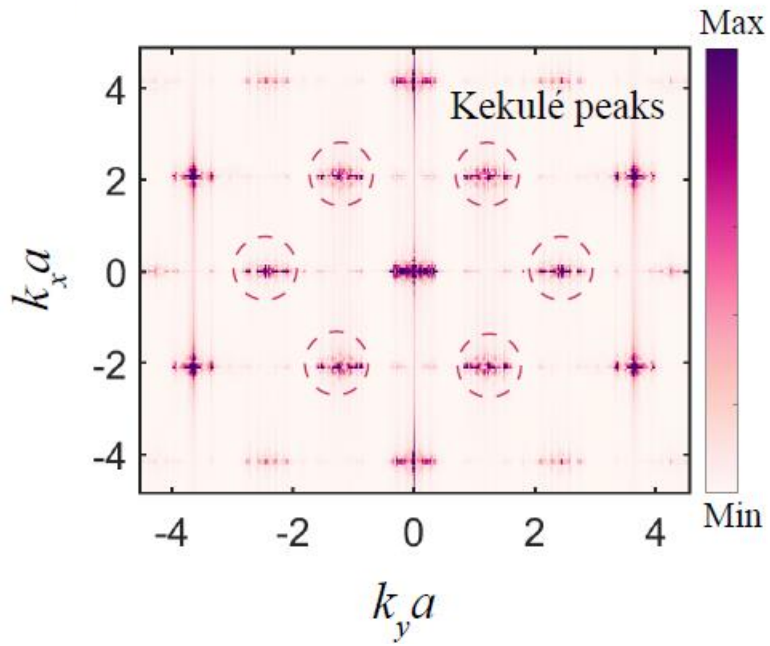
$$\tilde{\alpha}_R = \frac{\lambda_0}{\sqrt{\lambda_0^2 + \beta_I^2}} \alpha_R \quad , \quad \tilde{\boldsymbol{h}} = \frac{\lambda_0}{\sqrt{\lambda_0^2 + \beta_I^2}} \boldsymbol{h}$$



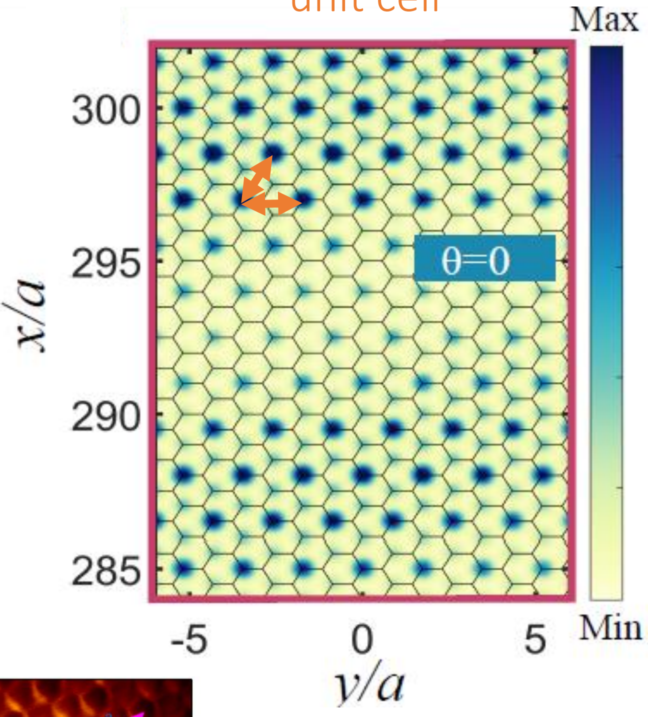
Identifying IVC order by tunneling

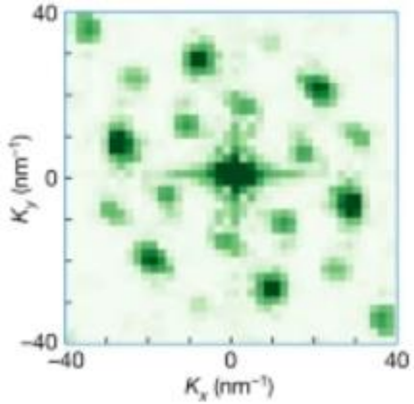
Majorana wavefunctions inherit Kekulé pattern:

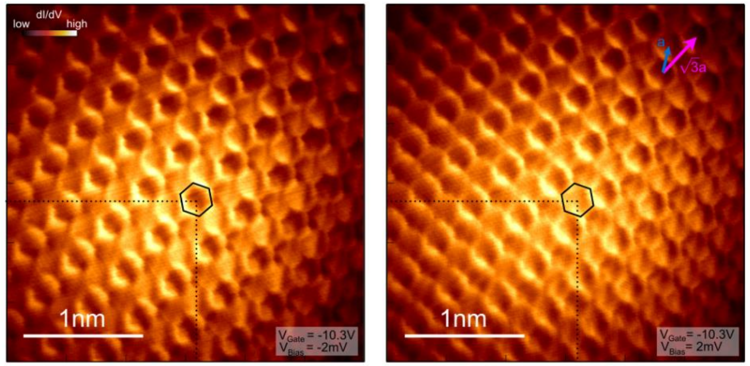
√3 x √3
enlargement of
unit cell



Fourier transform





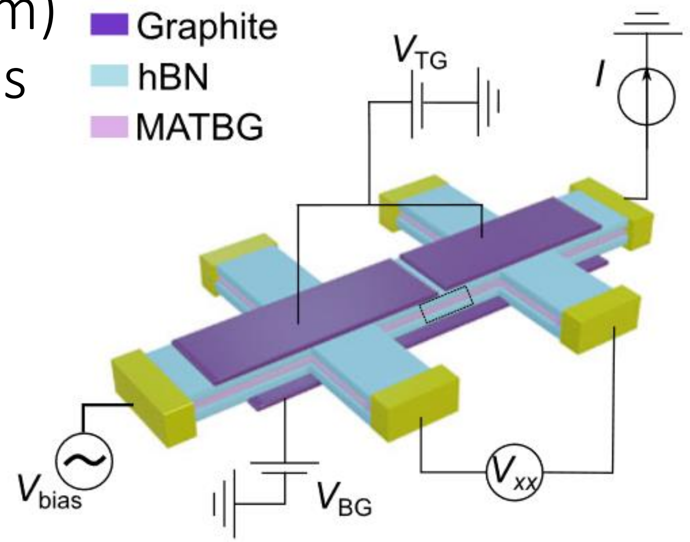


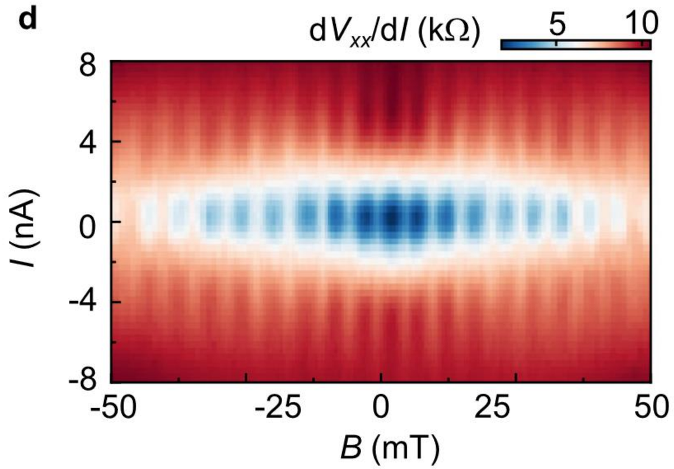
Kim, Choi, **ÉLH** *et al.*, Nature **623**, 942 (2023)

Gate-defined experiments probing phase coherence

- Gate-defined Josephson junctions (~100 nm) can image Fraunhofer interference patterns

de Vries *et al.*, Nat. Nanotech. **16**, 760 (2021)
Rodan-Legrain *et al.*, Nat. Nanotech. **16**, 769 (2021)
Díez-Mérida *et al.*, Nat. Comm. **14**, 2396, (2023)





Díez-Mérida *et al.*, Nat. Comm. 14, 2396, (2023)

- Little-Parks experiment with gate-defined superconducting loops (~1 µm)

Iwakiri *et al.*, arXiv:2308.07400