Visualizing designer quantum states in van der Waals heterostructures

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ACADEMY OF FINLAND

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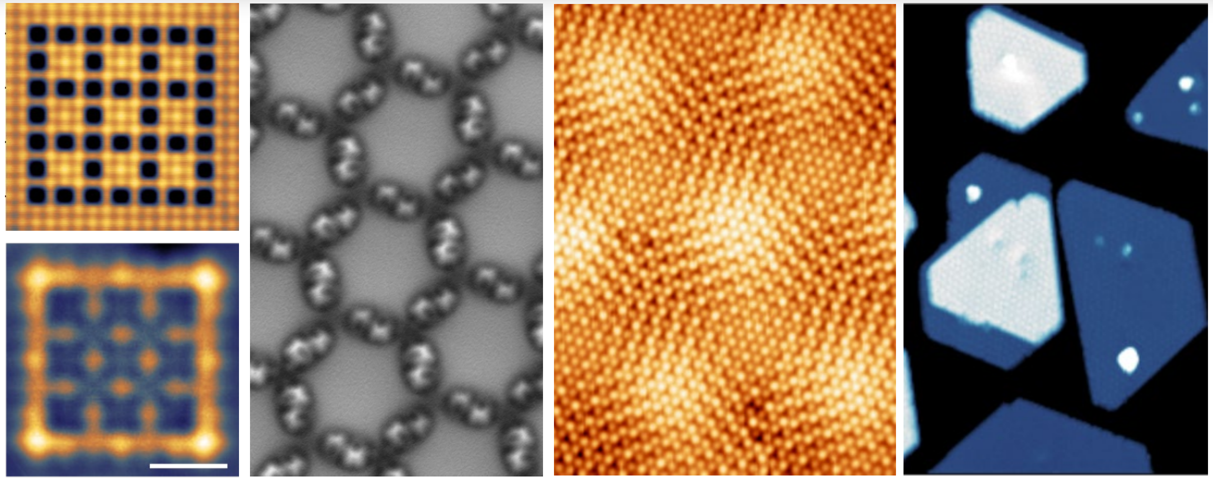
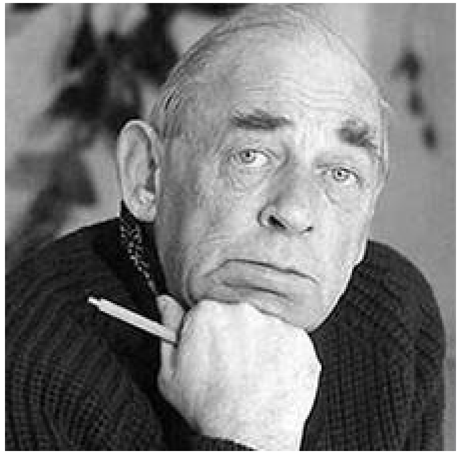


Where – what – who is Aalto?

Finnish designer and an architect, 1898 - 1976

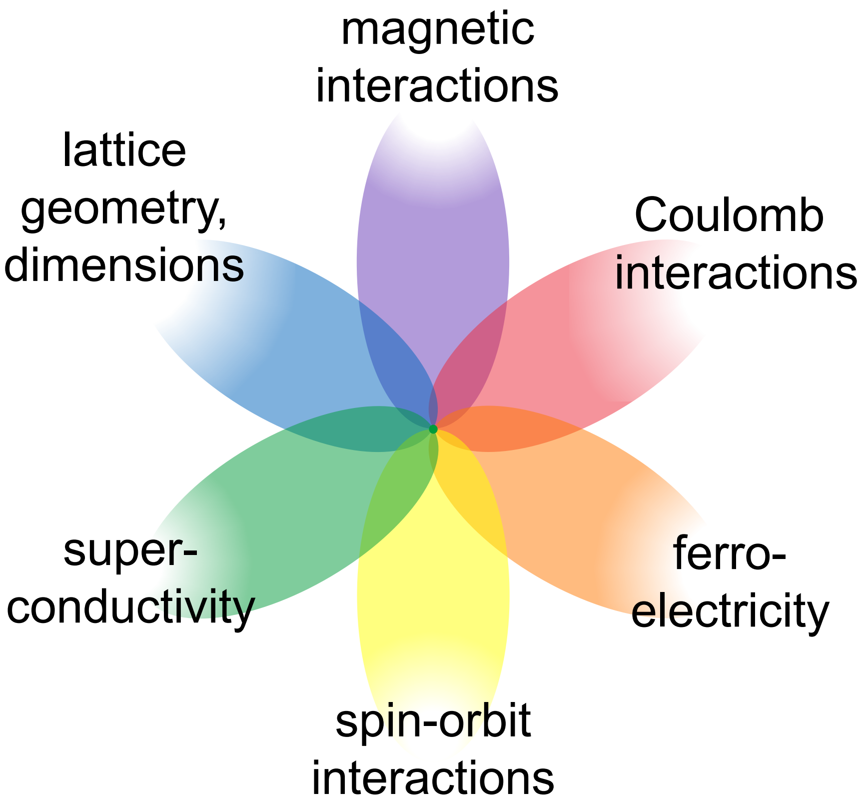


Greetings from Finland

Designer quantum materials

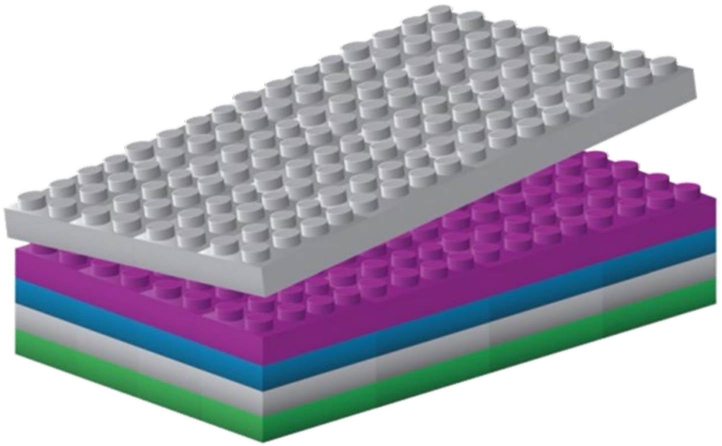
- Materials displaying quantum effects on macroscopic scale, complex emergent phenomena
- What are relevant parameters?
- Can be tuned in a single material
- Can be induced – proximitized – in a heterostructure

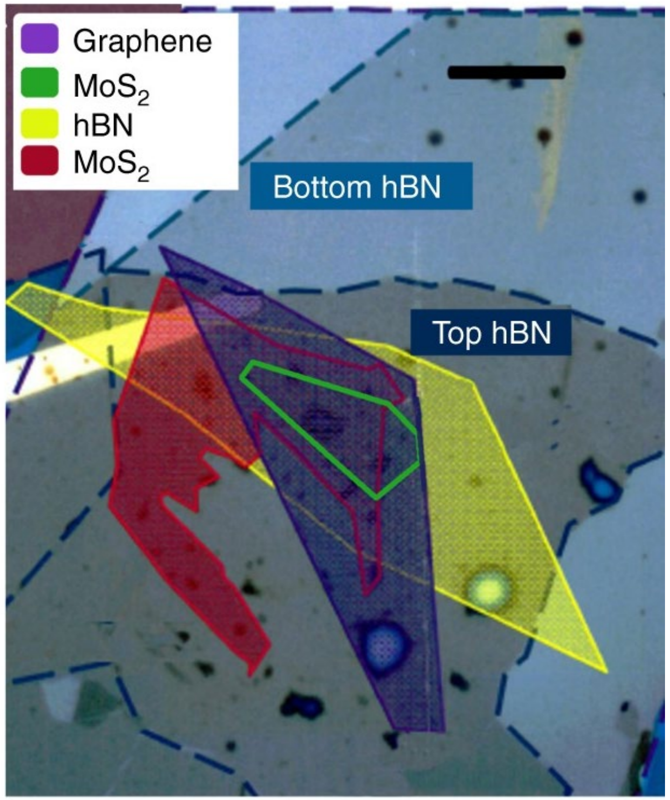
- What is the experimental platform?
  - **Two-dimensional materials**: metals, semiconductors, insulators, superconductors, magnets, charge-density wave materials, topological insulators, ferroelectrics, multiferroics...



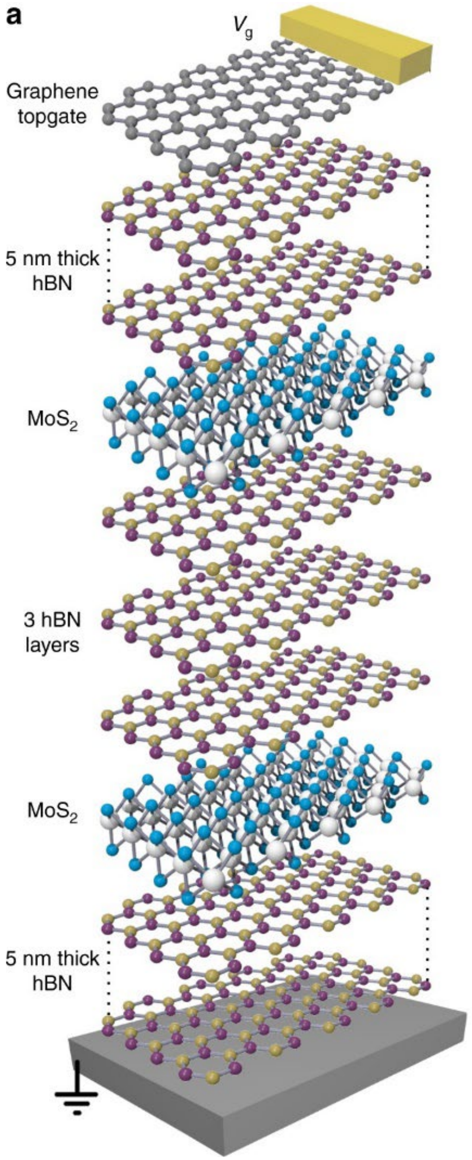
Playing lego with 2D materials?

- **van der Waals heterostructures**
  - Combine different material properties
  - Weak vdW bonding between the layers
  - Layers retain their intrinsic properties





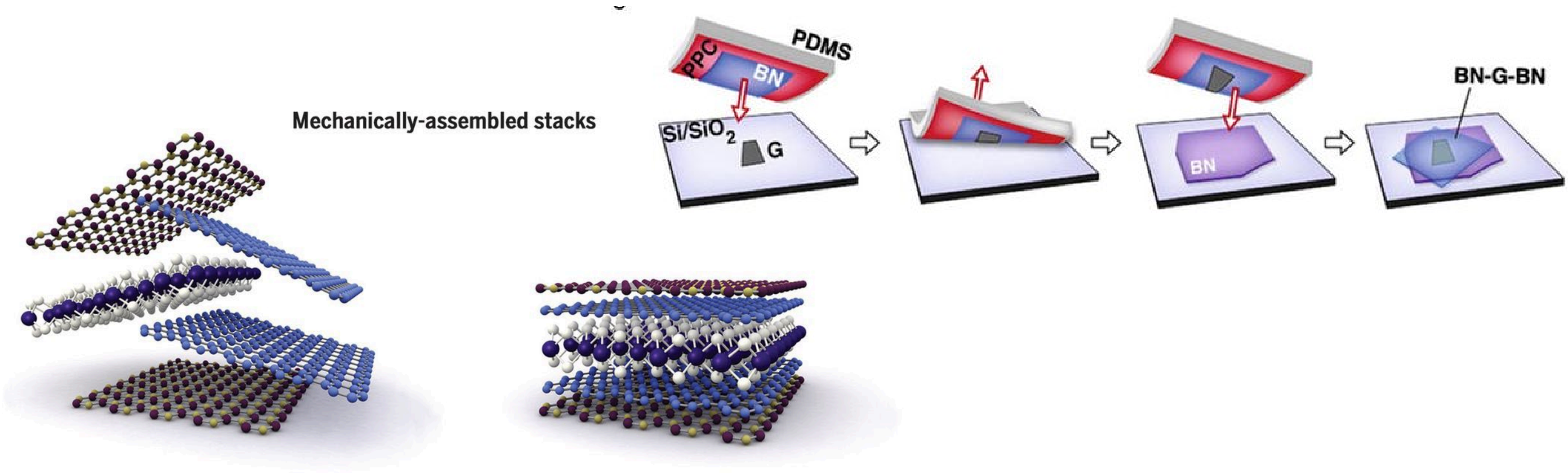
E. V. Calman et al. Nat. Commun. 9, 1895 (2018)



Reviews:
K.S. Novoselov et al. 2D materials and van der Waals heterostructures, Science 353, aac9439 (2016)
E.Y. Andrei et al. The marvels of moiré materials, Nat. Rev. Mater. 6, 201 (2021)
A. Castellanos-Gomez, X. Duan, Z. Fei, H. Rodriguez Gutierrez, Y. Huang, X. Huang, J. Quereda, Q. Qian, E. Sutter, P. Sutter, Van der Waals heterostructures, Nature Reviews Methods Primers 2, 58 (2022)

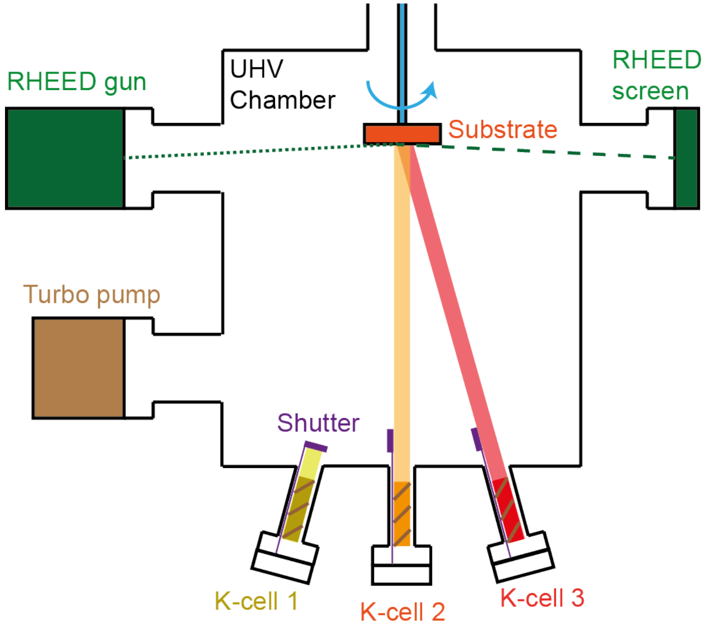
How to realize heterostructures?

- Mechanical exfoliation and transfer is the standard process in the field
- Pros: arbitrary layers in an arbitrary order, control of the twist angle between the layers
- Cons: air-sensitive materials difficult, sample size limited, possible contamination from the transfer steps

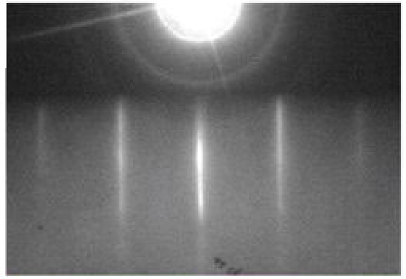


Growth my molecular beam epitaxy (MBE)

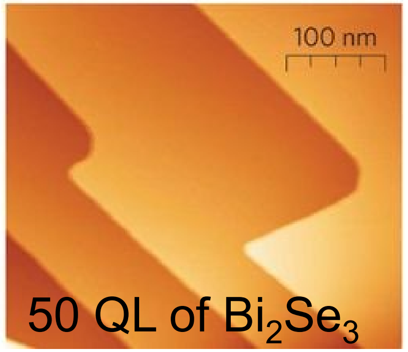
- Typically elemental precursors
  - E-beam evaporation of the metal
  - Direct evaporation of Se or Te (or using a cracker cell)
  - Sulfur from a compound source (decomposes upon heating)
- Follow growth in-situ with RHEED
- Can grow multilayer samples on different subtrates
- Cap (protect) the sample with Se or Te



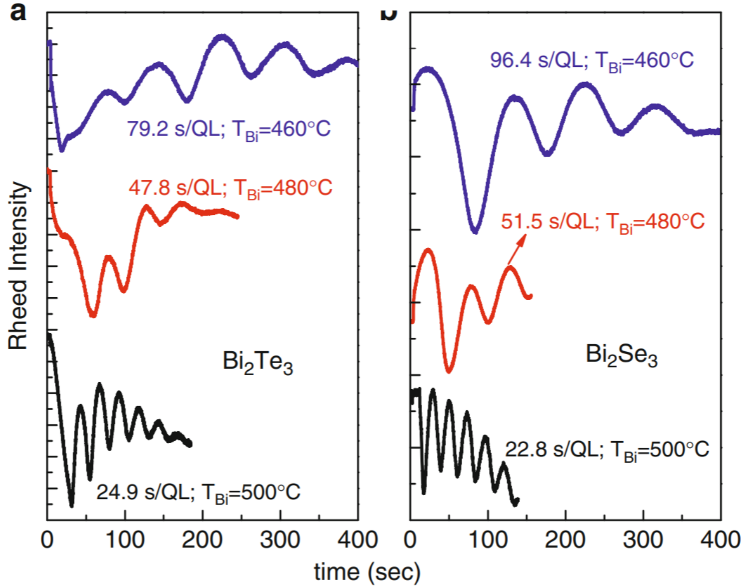
RHEED

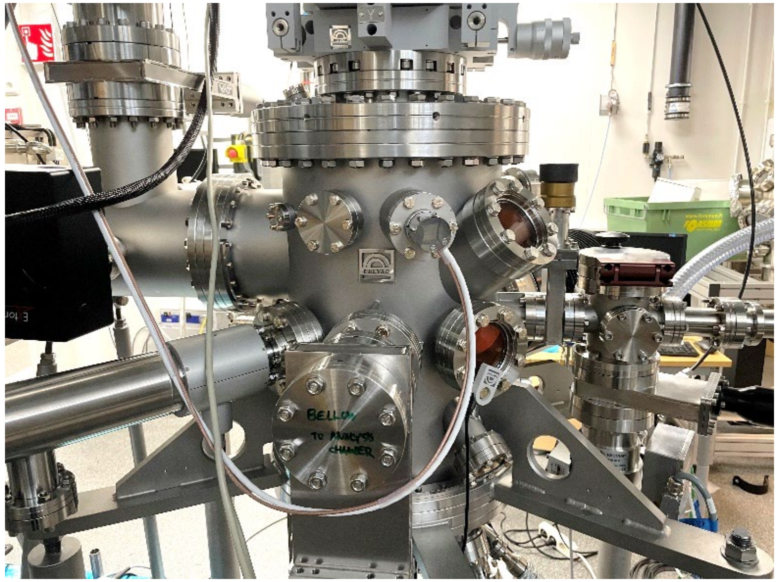


AFM



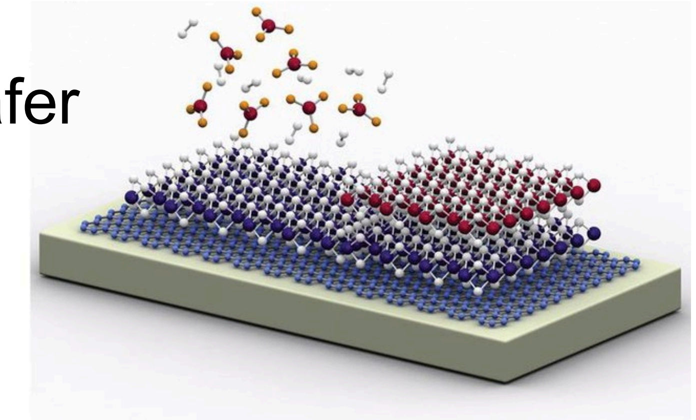
RHEED oscillations

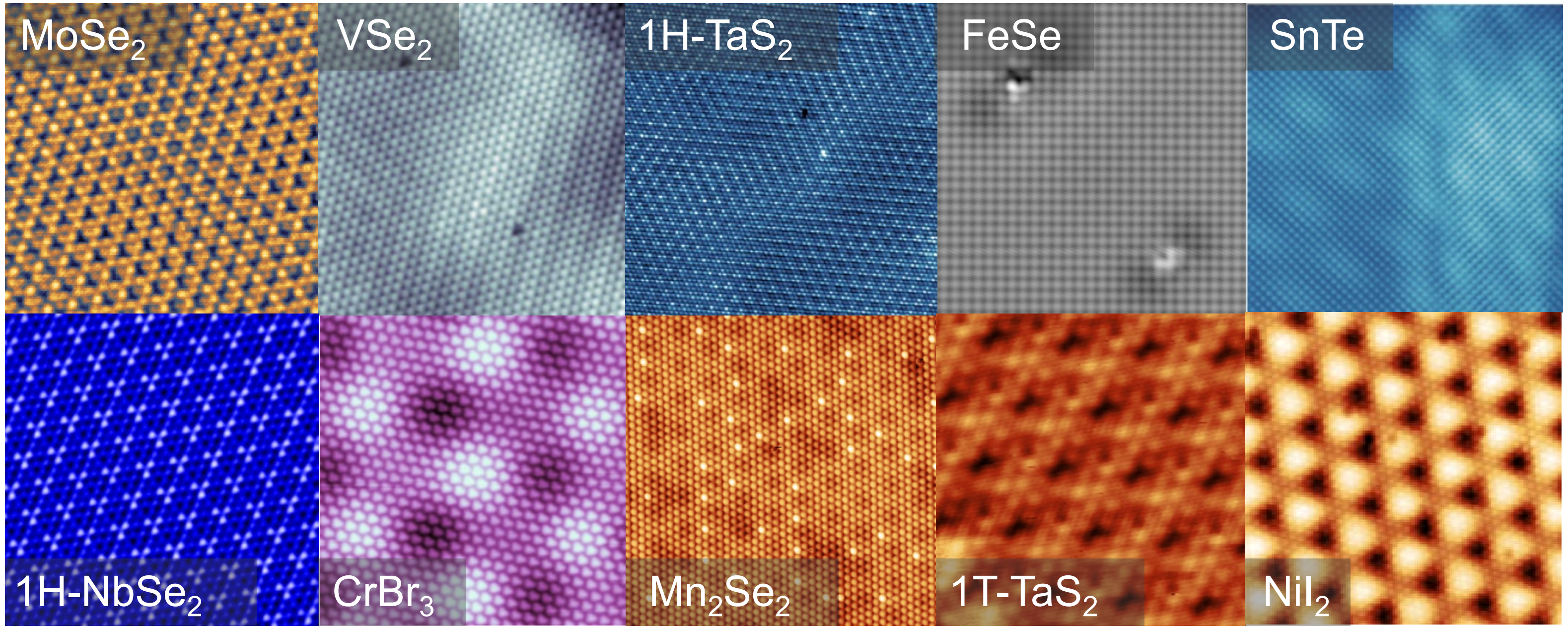




Growing heterostructures

- Direct growth on the desired substrates via MBE
- Pros: potentially ultraclean as everything is under UHV, wafer scale possible, no problems with air-sensitive materials
- Cons: not every material combination possible, usually a single orientation (twist), defect-free, pure monolayer samples require serious growth optimization

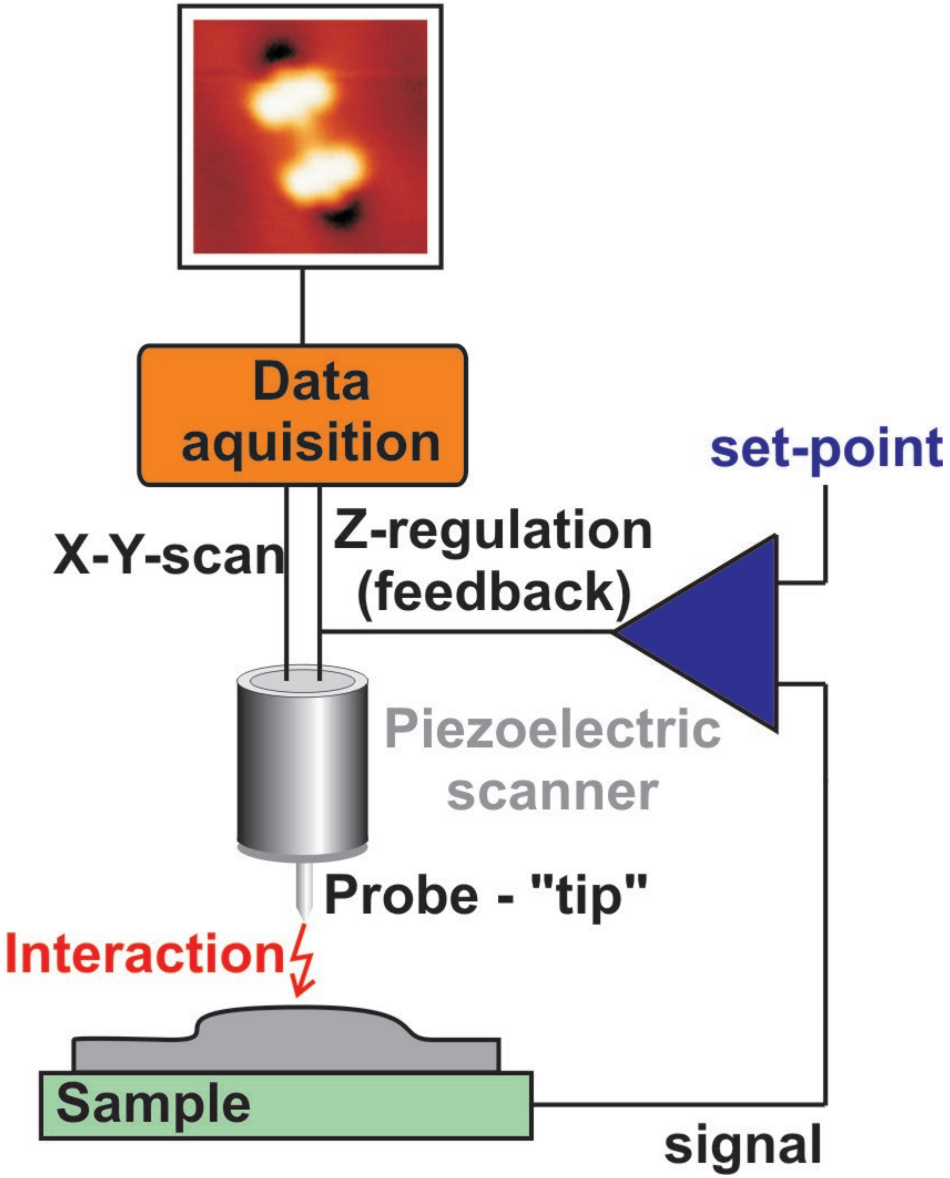




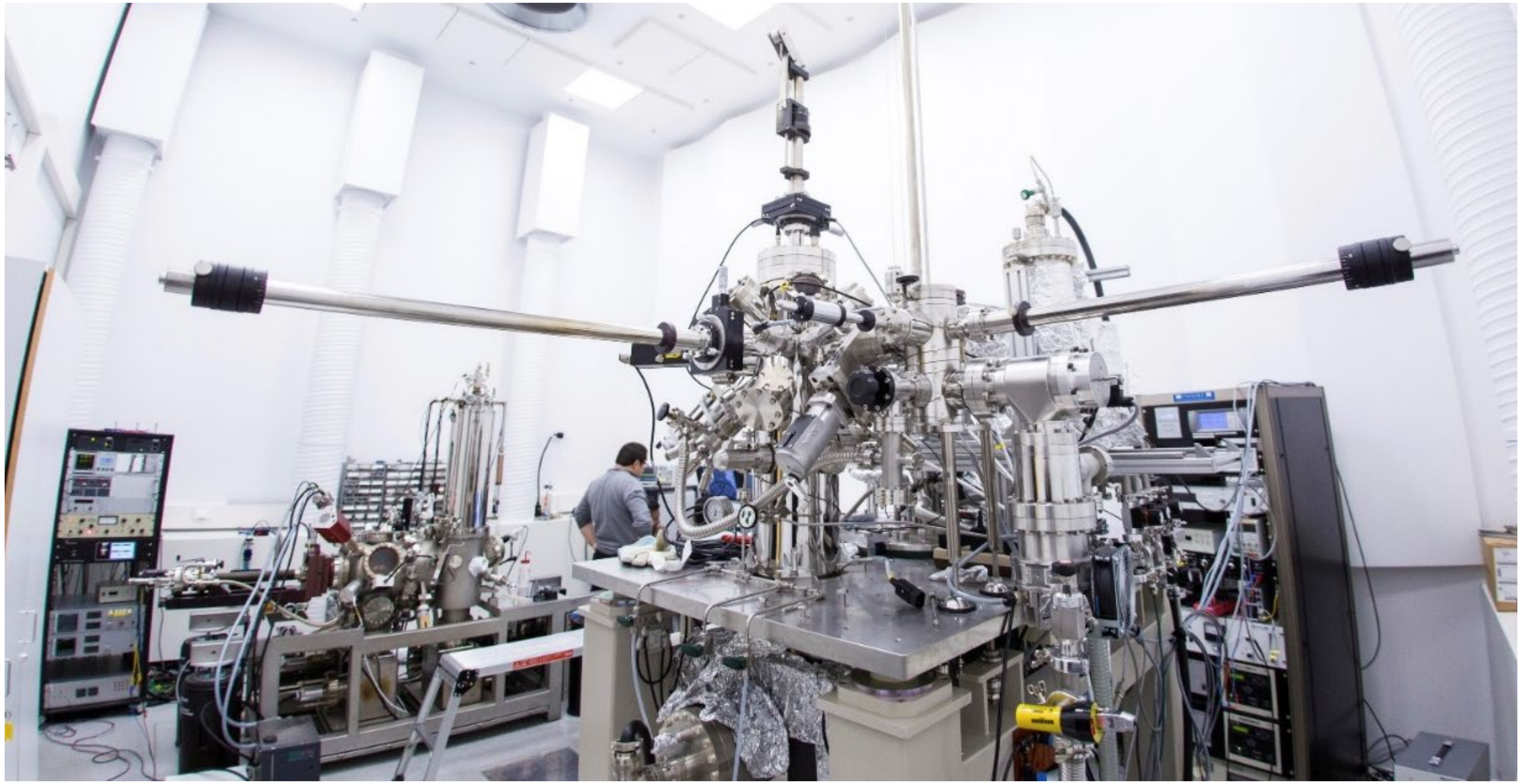
General principle of scanning probe microscopy

- Measure some interaction between a sharp probe and the sample
- **Scanning**: Move sample or tip while keeping this interaction constant (feedback) or while keeping height constant
- **Spectroscopy**: Change something (*z*, voltage, etc.) while keeping *x* and *y* constant (feedback off)

- Atomic-scale structural information of surfaces
- Energy-resolved local density of states (LDOS) with atomic spatial resolution



Scanning tunneling microscopy, experimentally

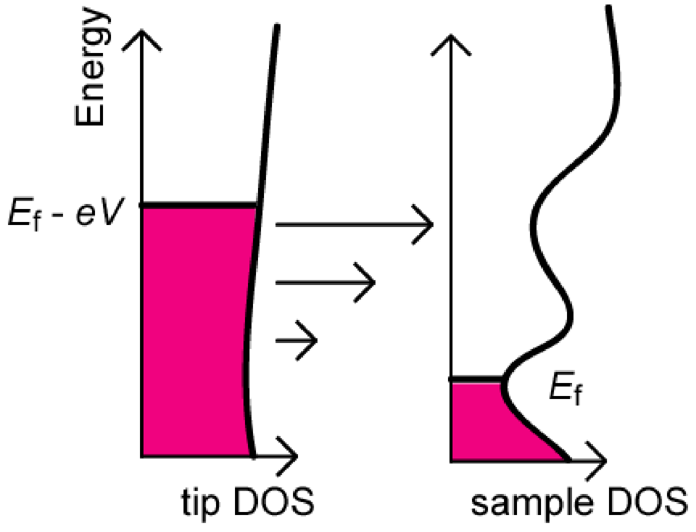


Typical performance metrics:

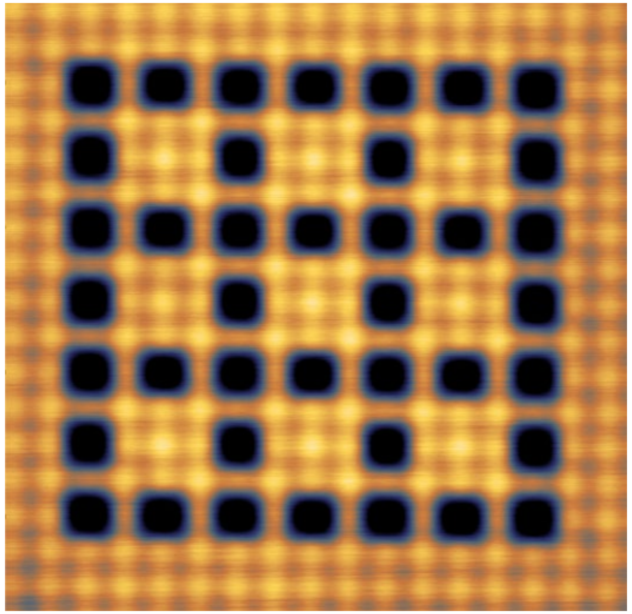
- z-noise < 1pm
- drift ~ 1 nm / day
- Hold-time at low temperatures: several days
- Can maintain the same atomic position for a month

- ultra-high vacuum, sample growth in the same UHV system
- low temperatures (T = 350mK – 4K)
- Careful vibration isolation etc. etc.
- Long experiments, lot of specific knowhow in experimental preparation

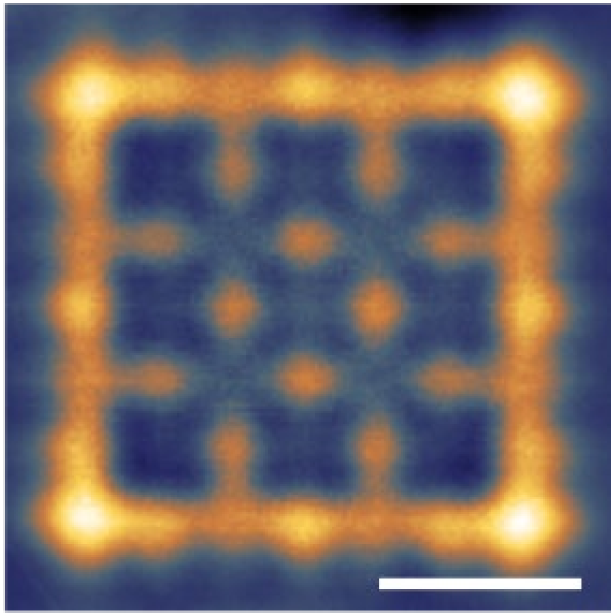
Scanning tunneling spectroscopy



geometry



local density of states



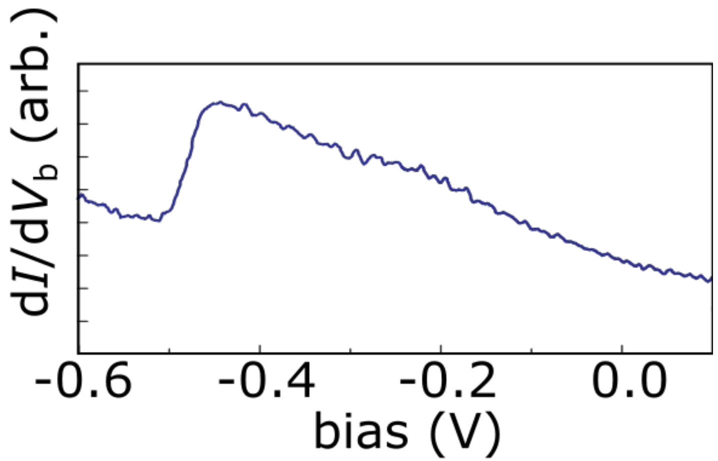
If ρ_{tip} and T energy independent
(at least close to the Fermi level):

$$\mathrm{d}I / \mathrm{d}V_\mathrm{b}(V_\mathrm{b}, x, y) \propto \rho(eV_\mathrm{b}, x, y) = \mathrm{LDOS}(eV_\mathrm{b}, x, y)$$

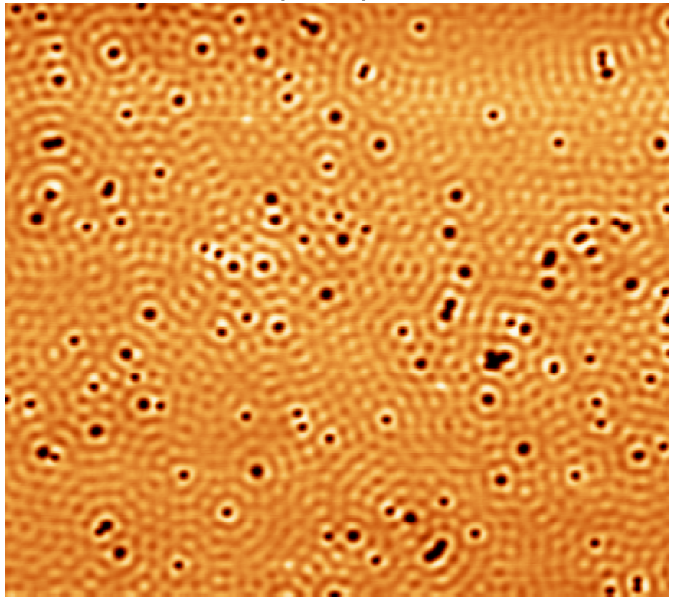
- STM maps the (integrated) **constant** density of states surface of the sample
- d*I*/d*V* spectroscopy gives the local density of states (spectral function for interacting systems)

STM and surface electronic structure

- We saw before (1D tight binding chain) that defects cause LDOS (and DOS) oscillations
- These can be used to get information on underlying band structure
- Example: two dimensional electron gas with parabolic dispersion
- $E = \frac{\hbar^2}{2m_{\text{eff}}}\mathbf{k}^2$
- Density of states: $\rho(E) = \text{const.}$

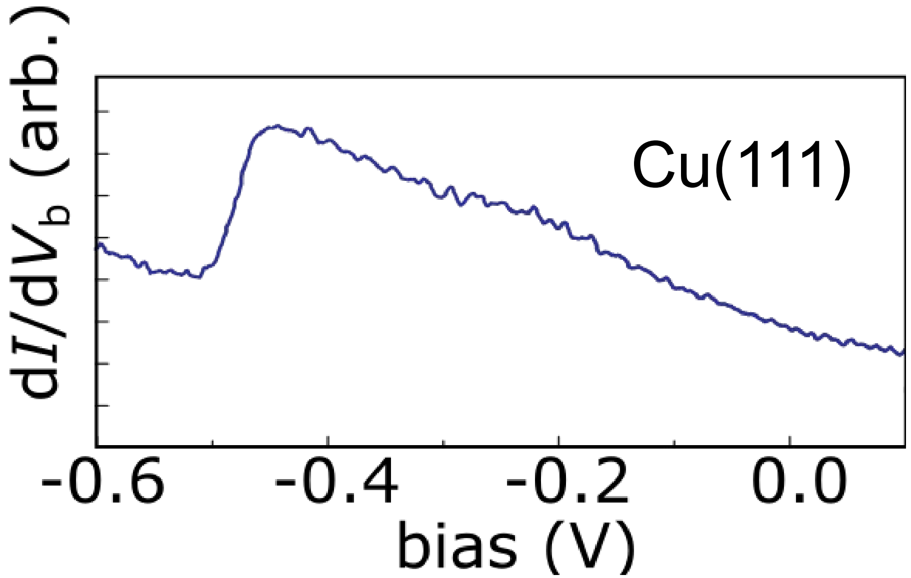


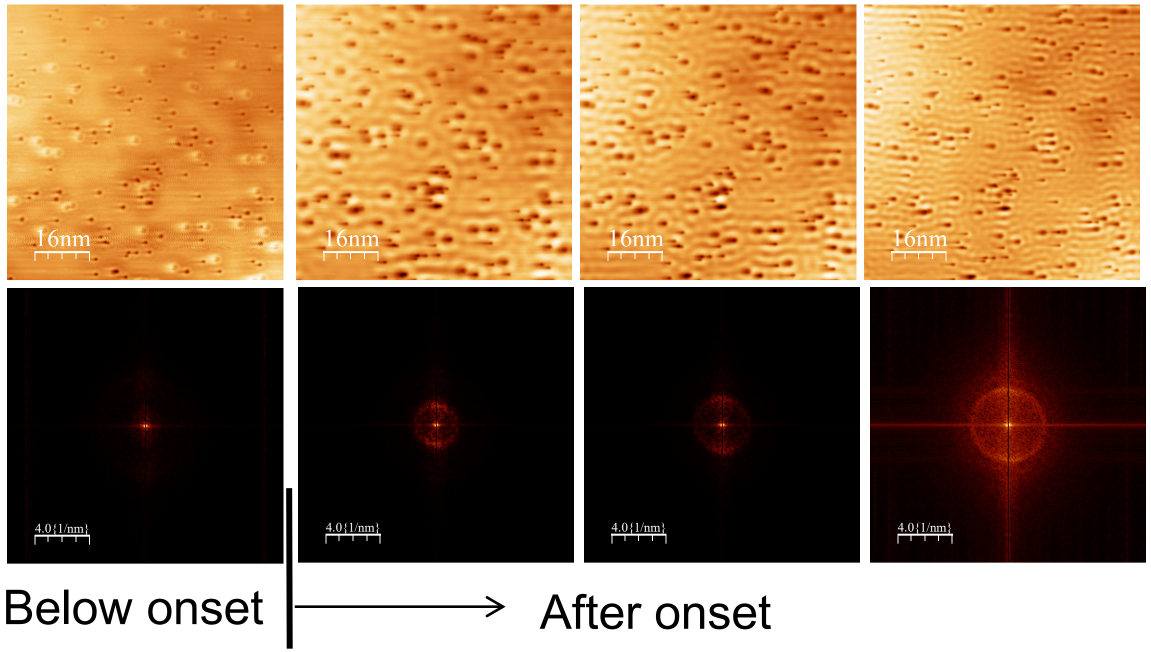
STM on Cu(111)

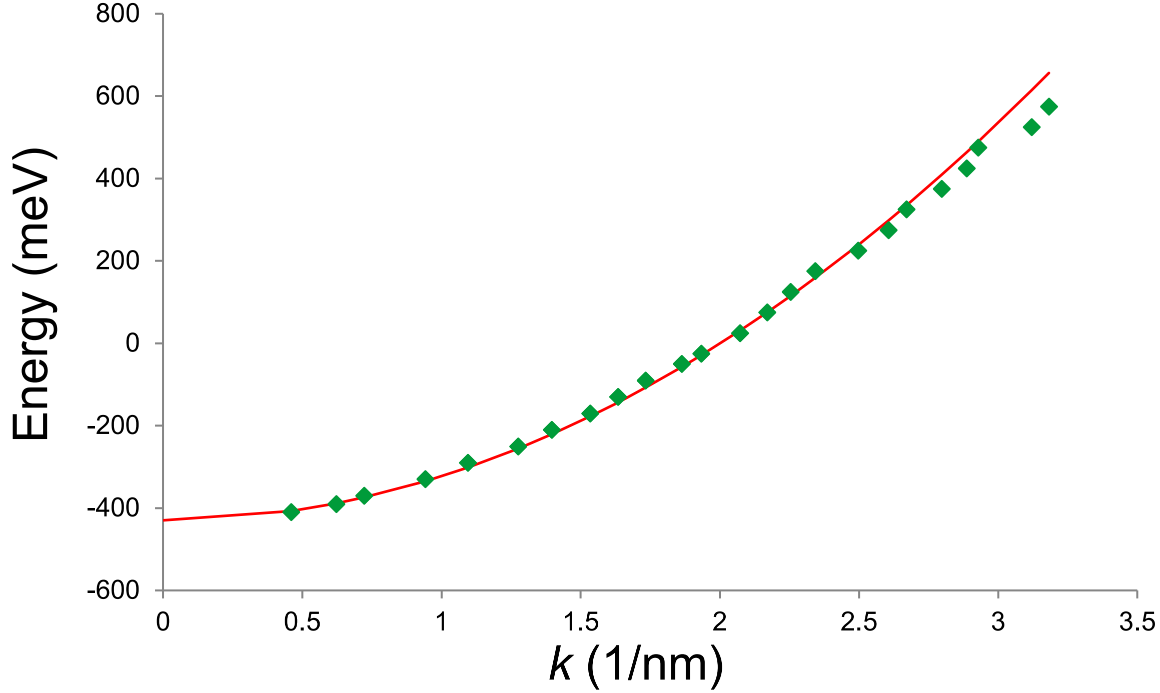


Quasi-particle interference (QPI)

- Measure dI/dV maps (LDOS oscillations) as a function of bias (energy)
- Fourier transform maps to extract different frequency components (**k** vectors)
- Plot energy (set by the bias) as a function of the **k**

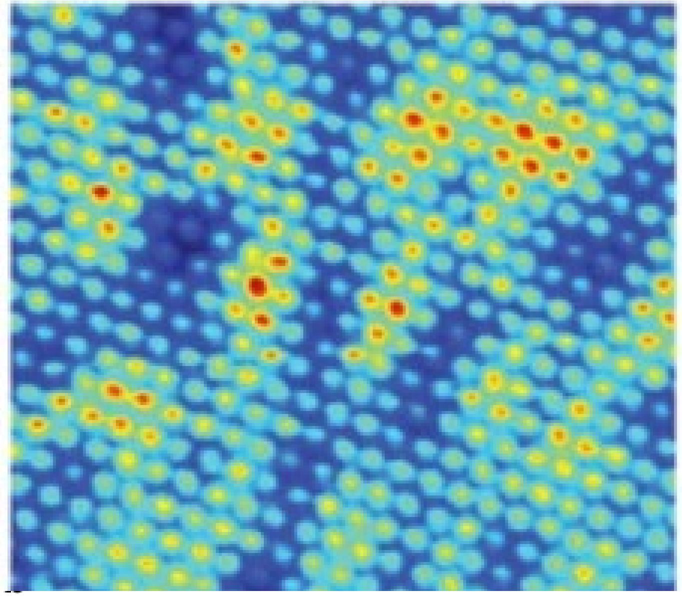


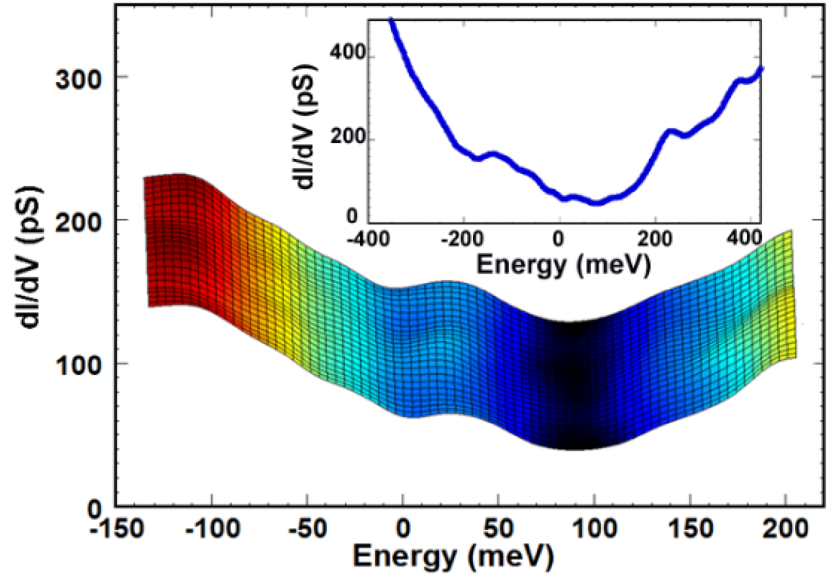




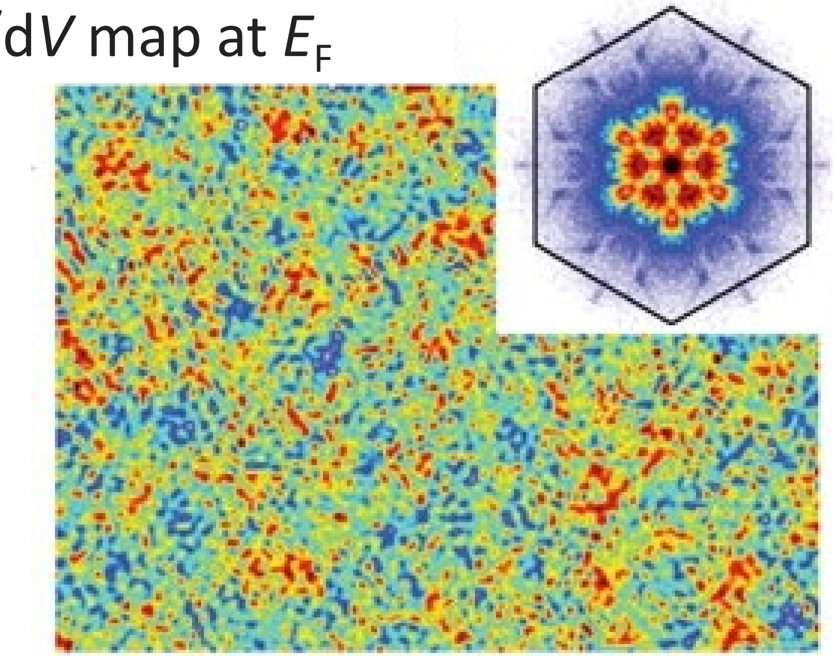
Spectroscopic imaging STM

$Bi_{0.92}Sb_{0.08}(111)$

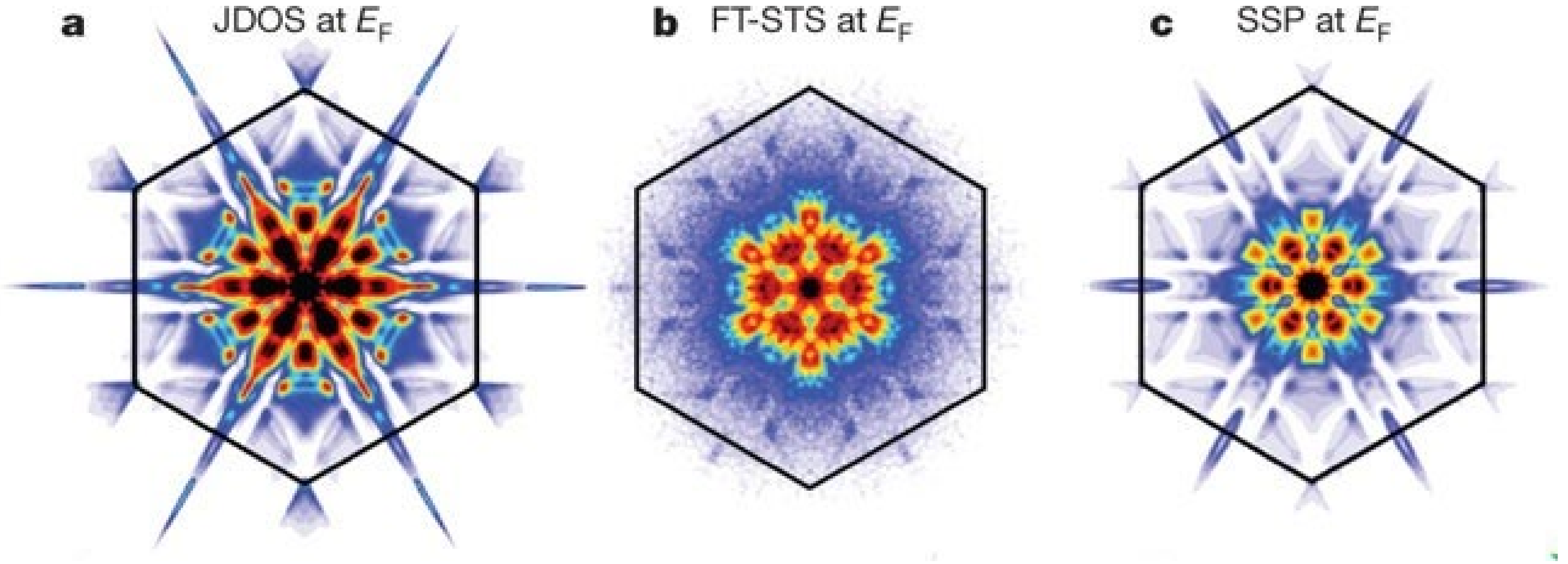




d*I*/d*V* map at E_F



P. Roushan (A. Yazdani group), Topological surface states protected from backscattering by chiral spin texture, Nature 460, 1106 (2009)



2D ferroelectric materials

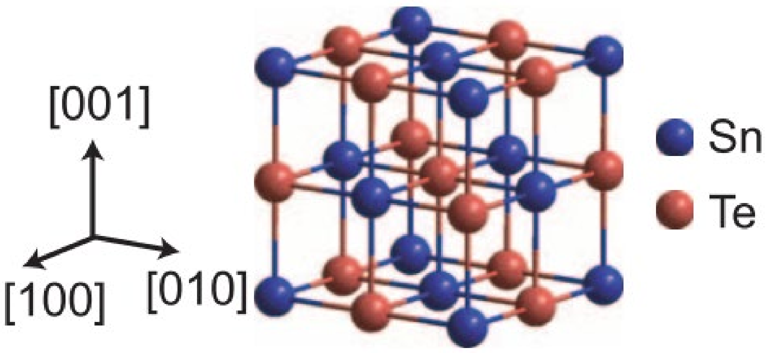
- Ferroelectric materials have an electric polarization in the bulk
- Not that uncommon – but does it exist in a monolayer?

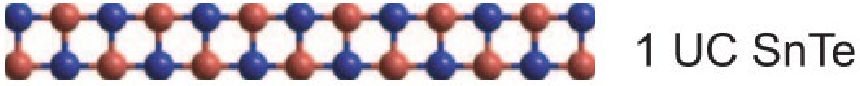
FERROELECTRICITY

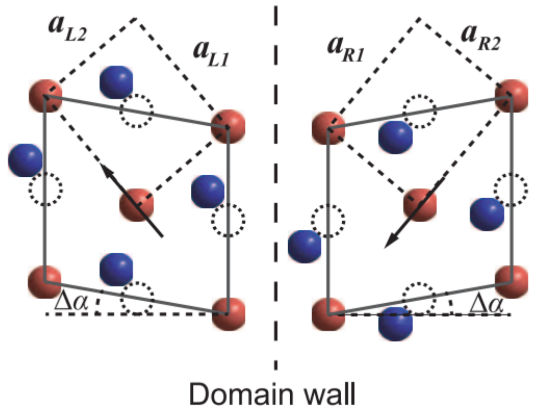
Science 353, 274 (2016)

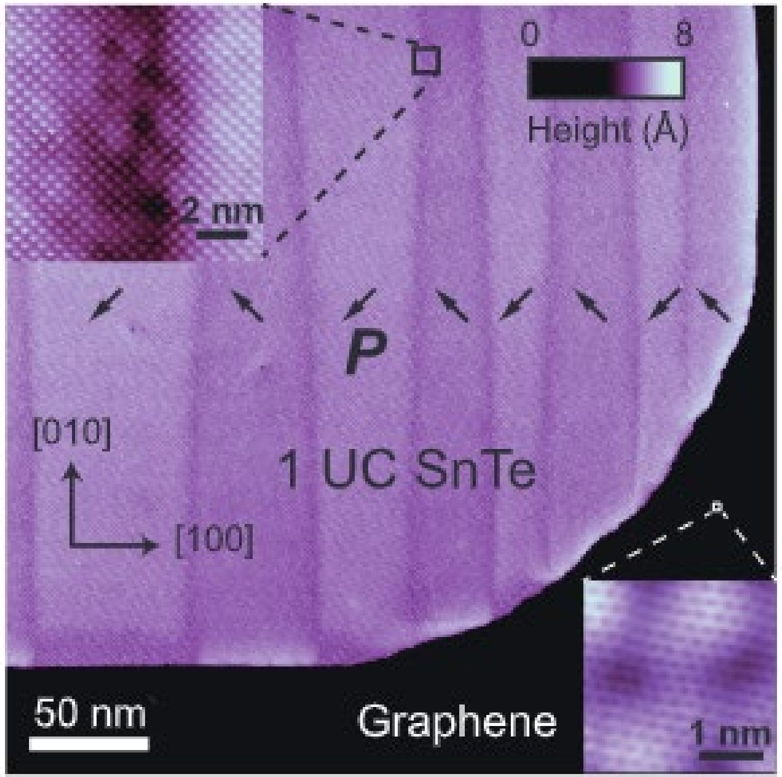
Discovery of robust in-plane ferroelectricity in atomic-thick SnTe

Kai Chang,[1,2]* Junwei Liu,[3,1,2]* Haicheng Lin,[1,2] Na Wang,[1,2] Kun Zhao,[1,2] Anmin Zhang,[4] Feng Jin,[4] Yong Zhong,[1,2] Xiaopeng Hu,[1,2] Wenhui Duan,[1,2] Qingming Zhang,[4,5] Liang Fu,[3] Qi-Kun Xue,[1,2] Xi Chen,[1,2]† Shuai-Hua Ji[1,2,6]†



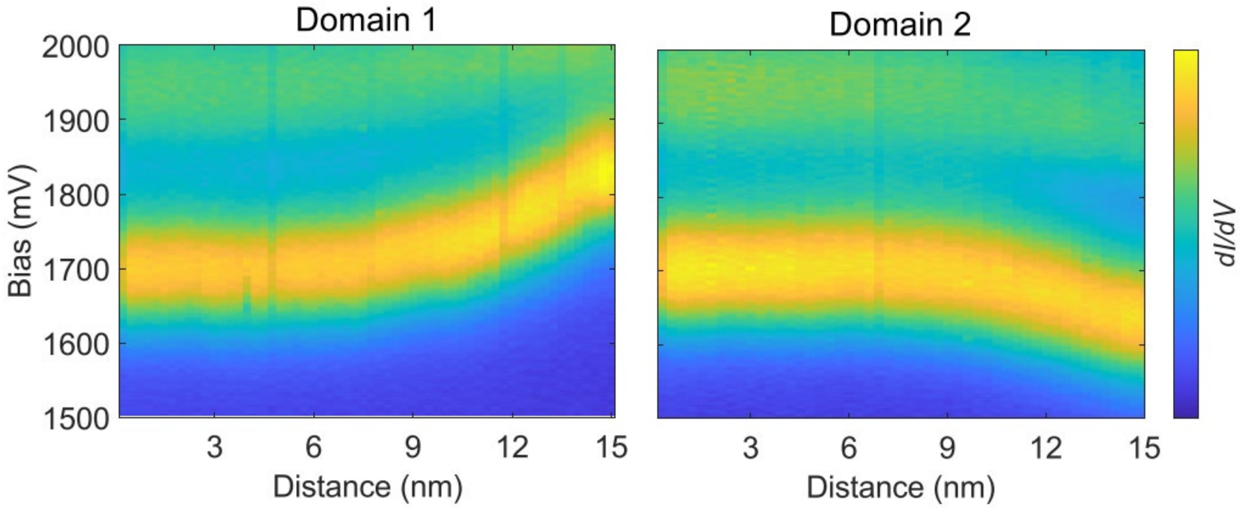


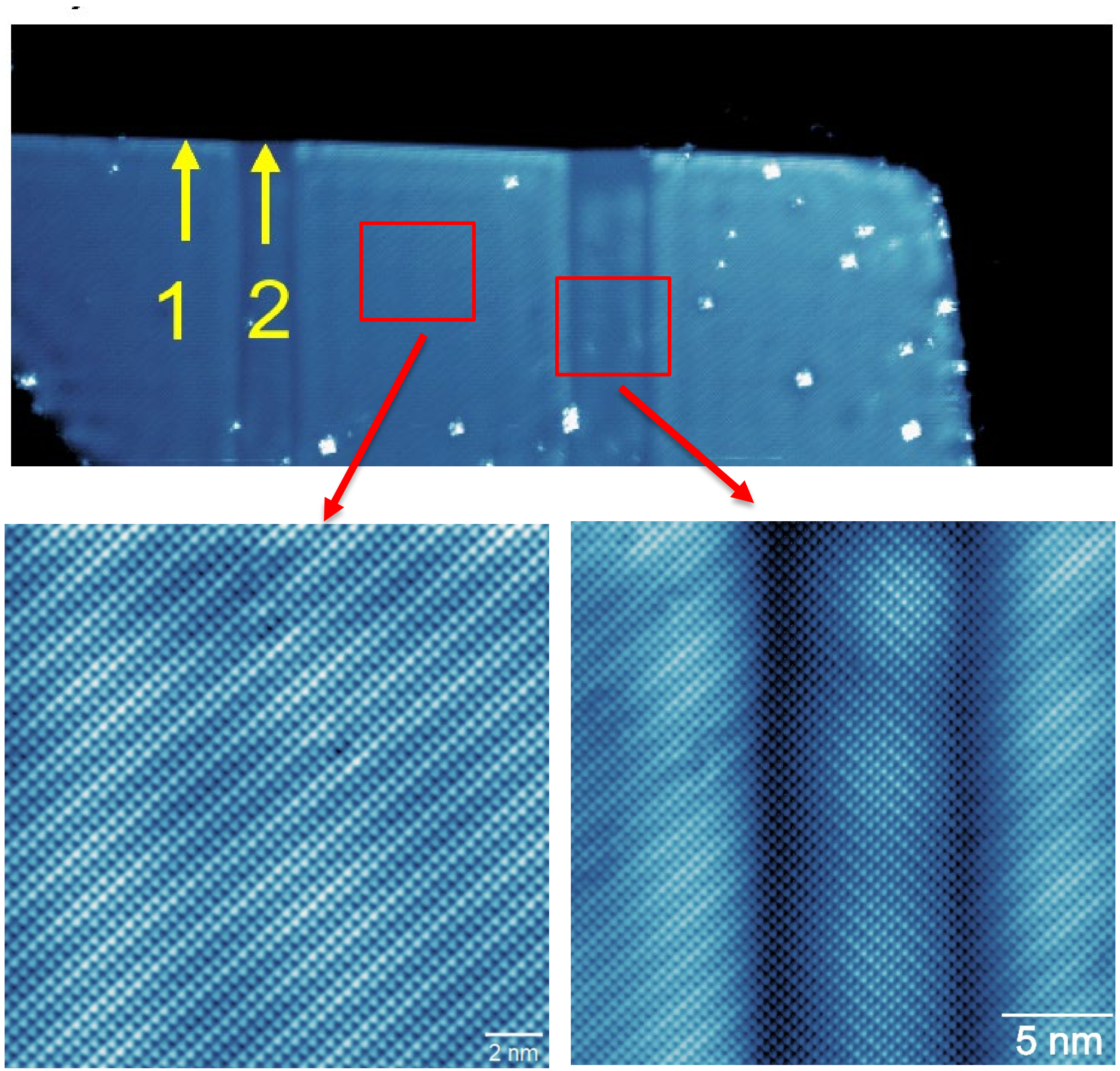




Visualizing ferroelectricity

- In the case of SnTe, atomic distortion can be directly measured
- Ferroelectric polarization causes shifts of the bands





M. Amini et al. Control of Molecular Orbital Ordering Using a van der Waals Monolayer Ferroelectric, Adv. Mater. 35, 2206456 (2023).

2D materials toolbox: ferromagnets

- Discovery of ferromagnetism in van der Waals crystals
  - Exfoliated CrI_3 and $Cr_2Ge_2Te_6$
- Which ones can be (readily) grown as monolayers?
  - Several TMDs claimed to exhibit monolayer ferromagnetism
  - **VSe_2**: Bonilla et al. Nat. Nanotech. 13, 289 (2018), **$MnSe_2$**: O'Hara et al. Nano Lett. 18, 3125 (2018) – but they complicated magnetic properties
  - For VSe_2: J. Feng et al. Nano Lett. 18, 4493 (2018), P. Chen et al. Phys. Rev. Lett. 121, 196402 (2018), G. Duvjir et al. Nano lett. 18, 5432 (2018), W. Yu et al. Adv. Mater. 31, 1903779 (2019), S. Kezilebieke et al. Commun. Phys. 3, 116 (2020), and so on...
- $CrBr_3$ is an out-of-plane monolayer ferromagnet and can be directly grown

REVIEW

1 NOVEMBER 2018 | VOL 563 | NATURE | 47

Magnetism in two-dimensional van der Waals materials

Kenneth S. Burch[1], David Mandrus[2,3] & Je-Geun Park[4,5]*

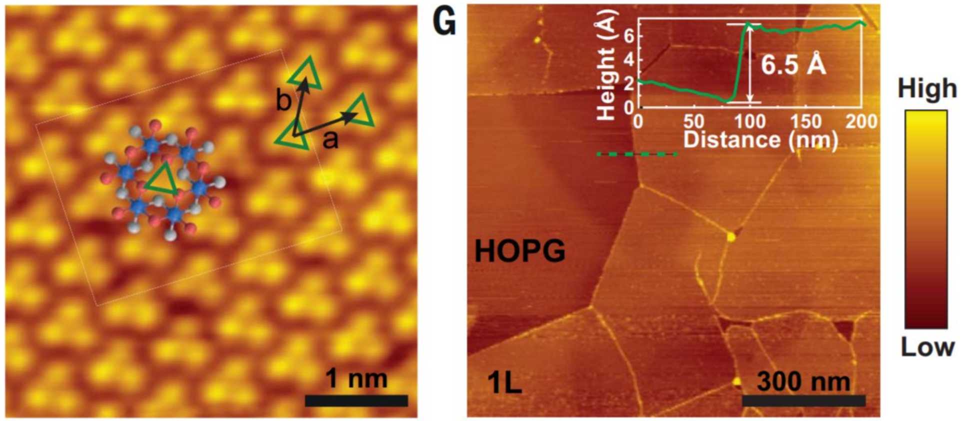
REPORT

Science **366**, 983–987 (2019)

2D MAGNETISM

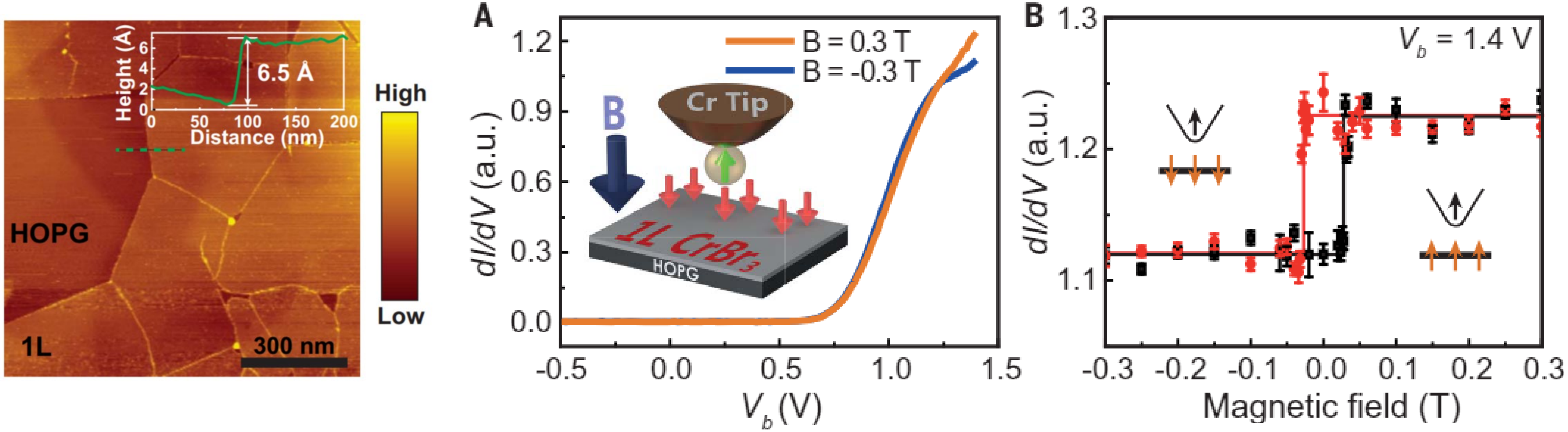
Direct observation of van der Waals stacking–dependent interlayer magnetism

Weijong Chen[1], Zeyuan Sun[1], Zhongjie Wang[1], Lehua Gu[1], Xiaodong Xu[2], Shiwei Wu[1,3]*, Chunlei Gao[1,3]*



How to detect magnetism with an STM?

- Spin-polarized tunneling – start with a magnetic tip which in the end gives contrast between different spin orientations on the surface

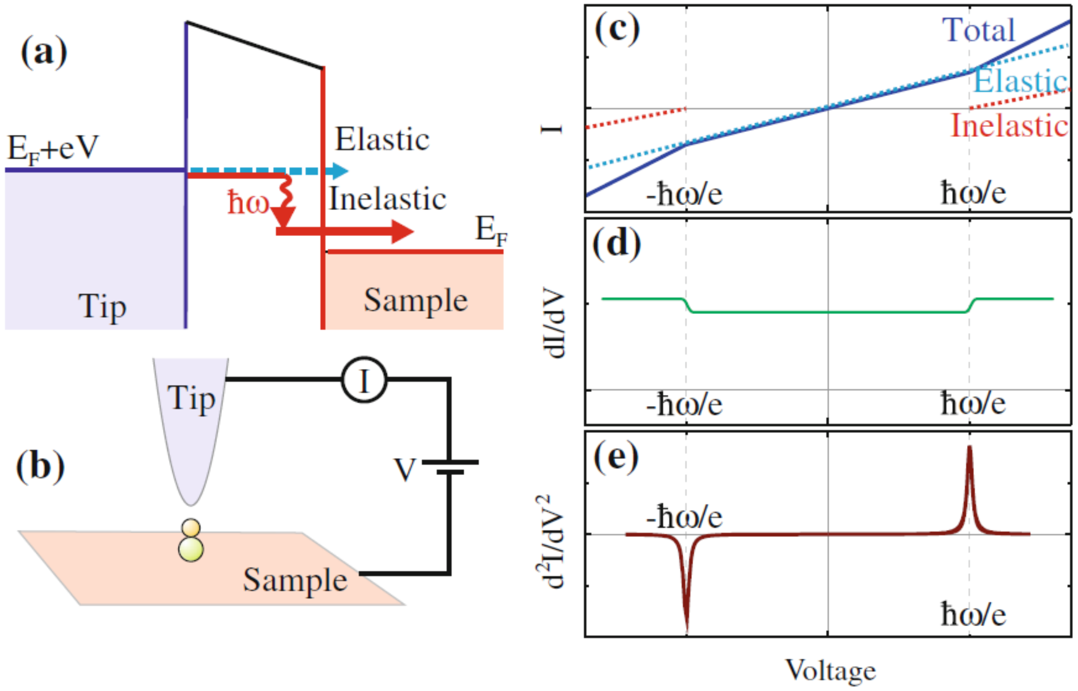


Science **366**, 983–987 (2019)

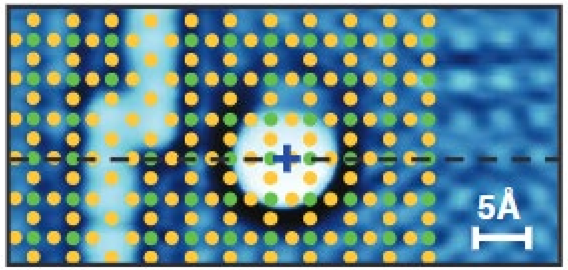
- In bilayers, stacking controls whether interlayer coupling is ferromagnetic or antiferromagnetic
- General review on spin-polarized STM: R. Wiesendanger, Rev. Mod. Phys. 81, 1495 (2009)

How to detect magnetism with an STM?

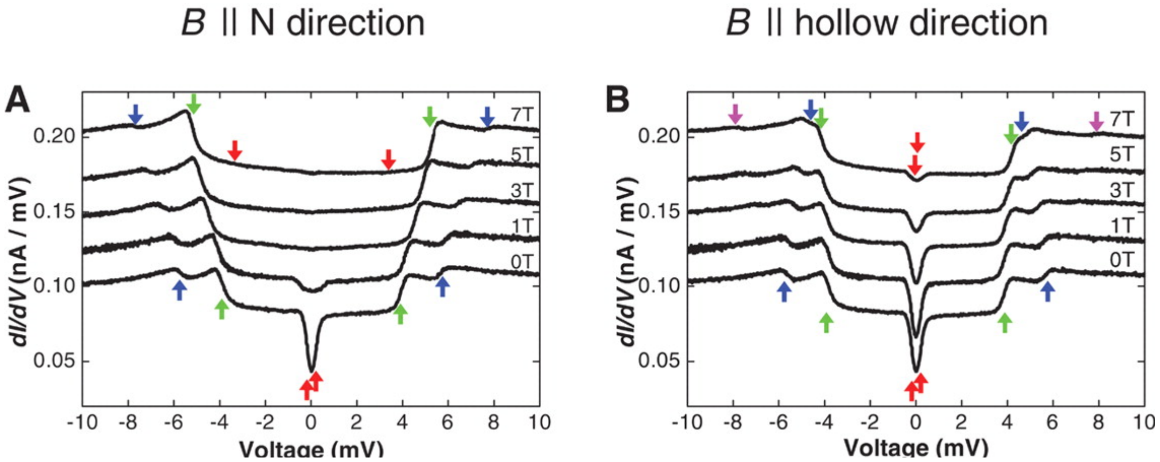
- Spin flip spectroscopy
- Inelastic tunneling giving rise to spin-flip transitions



Fe on Cu_2N on Cu(100)



C.F. Hirjibehedin, C.-Y. Lin, A,F. Otte, M. Ternes, C.P. Lutz, B.A. Jones, A.J. Heinrich, Science, 317, 1199 (2007).



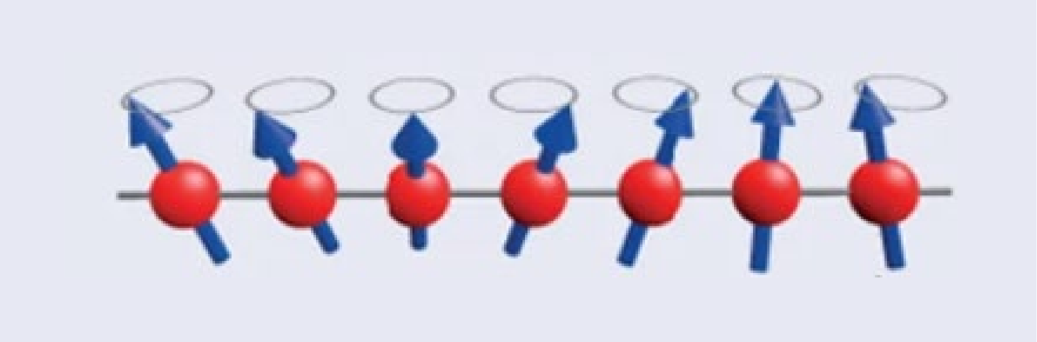
$$\hat{H} = g\mu_B \vec{B} \cdot \hat{\vec{S}} + D\hat{S}_z^2 + E(\hat{S}_x^2 - \hat{S}_y^2)$$

D (axial anisotropy) and *E* (transverse anisotropy)

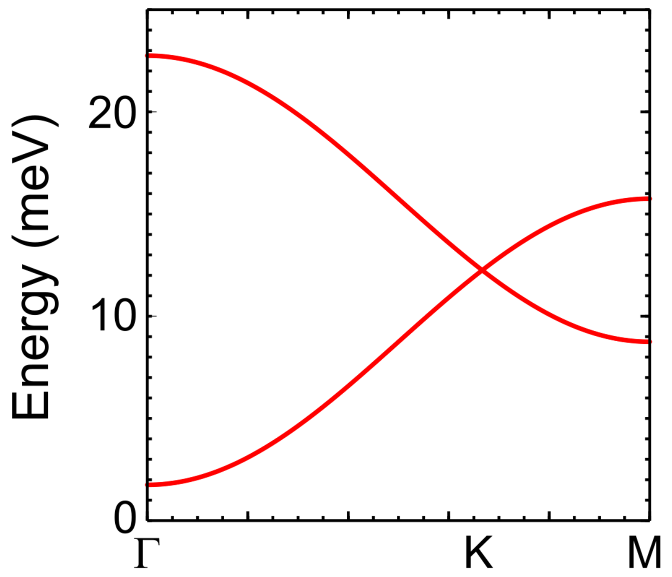
Review M. Ternes, Spin-excitations and correlations in scanning tunneling spectroscopy, New J. Phys. 17, 063016 (2015)

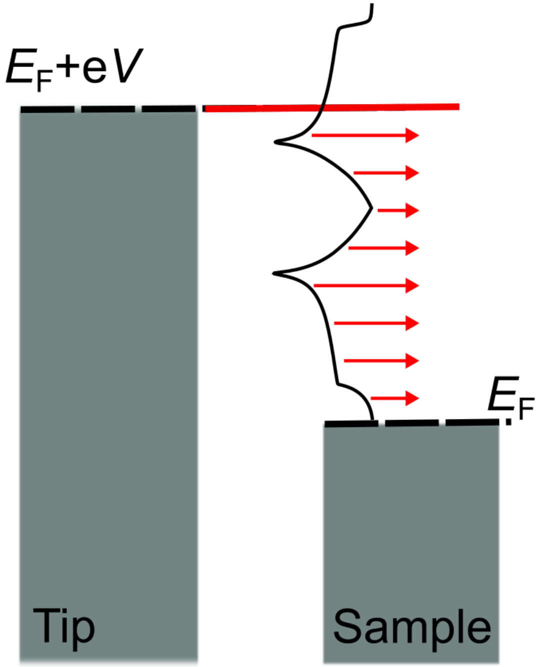
Magnons

- Elementary excitation in ferromagnets
- Can they be detected in STM
  - inelastic spectroscopy
  - similar to spin flips
  - d*I*/d*V* α integrated magnon DOS
  - d^2I/dV^2 α magnon DOS



CrBr$_3$ magnon bandstructure

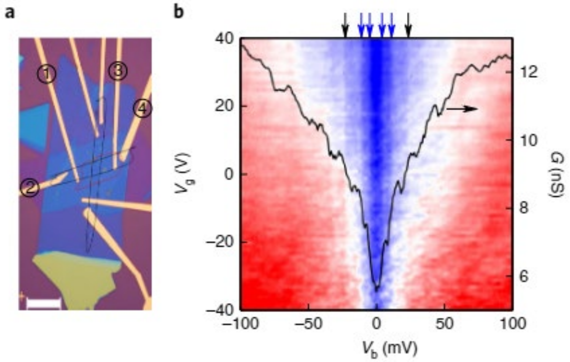




ARTICLES
https://doi.org/10.1038/s41928-018-0087-z
nature electronics

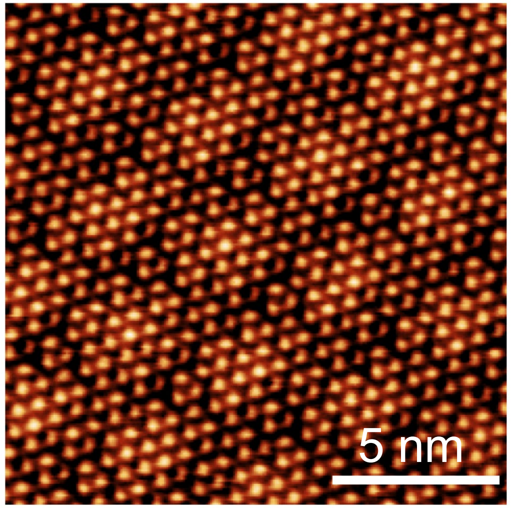
Magnon-assisted tunnelling in van der Waals heterostructures based on CrBr$_3$

D. Ghazaryan[1], M. T. Greenaway[2], Z. Wang[1], V. H. Guarochico-Moreira[1,3], I. J. Vera-Marun[1], J. Yin[1], Y. Liao[1], S. V. Morozov[4], O. Kristanovski[5], A. I. Lichtenstein[5], M. I. Katsnelson[6], F. Withers[7], A. Mishchenko[1,8], L. Eaves[1,9], A. K. Geim[1,8], K. S. Novoselov[1,8*] and A. Misra[1,10*]

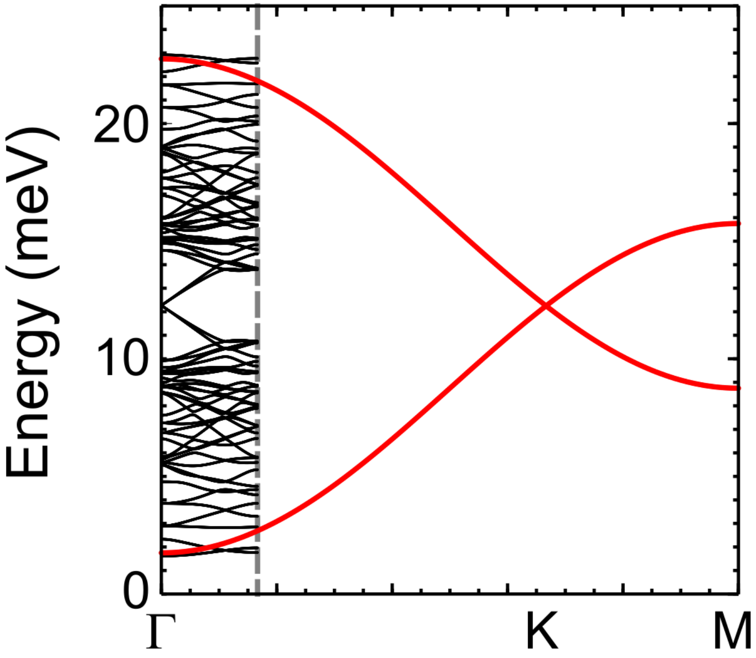


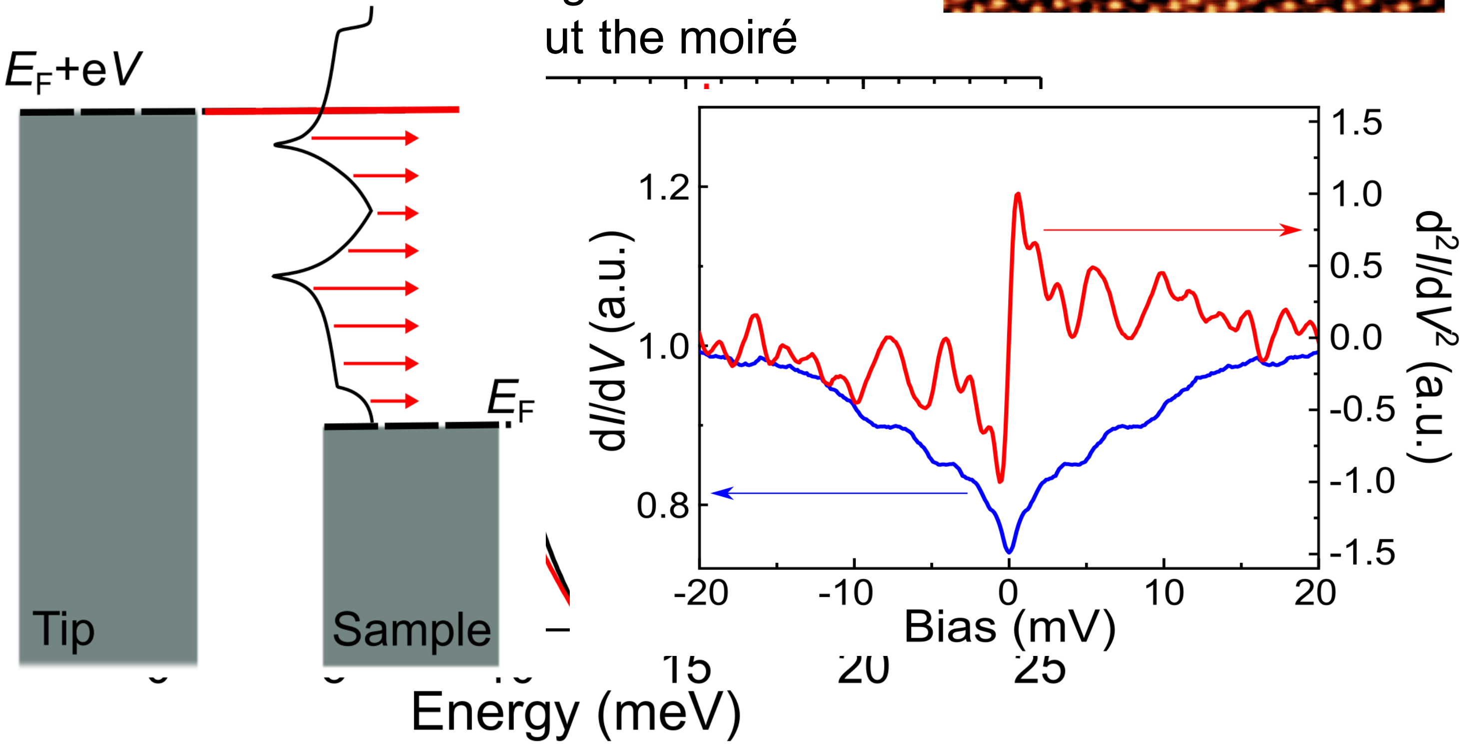
What is going on with the magnons

- d^2I/dV^2 proportional to the magnon DOS – doesn't look like that!
- Moiré pattern folds the magnon bandstructure
- Causes strong peaks in the magnon DOS



$CrBr_3$ magnon bandstructure with the moiré





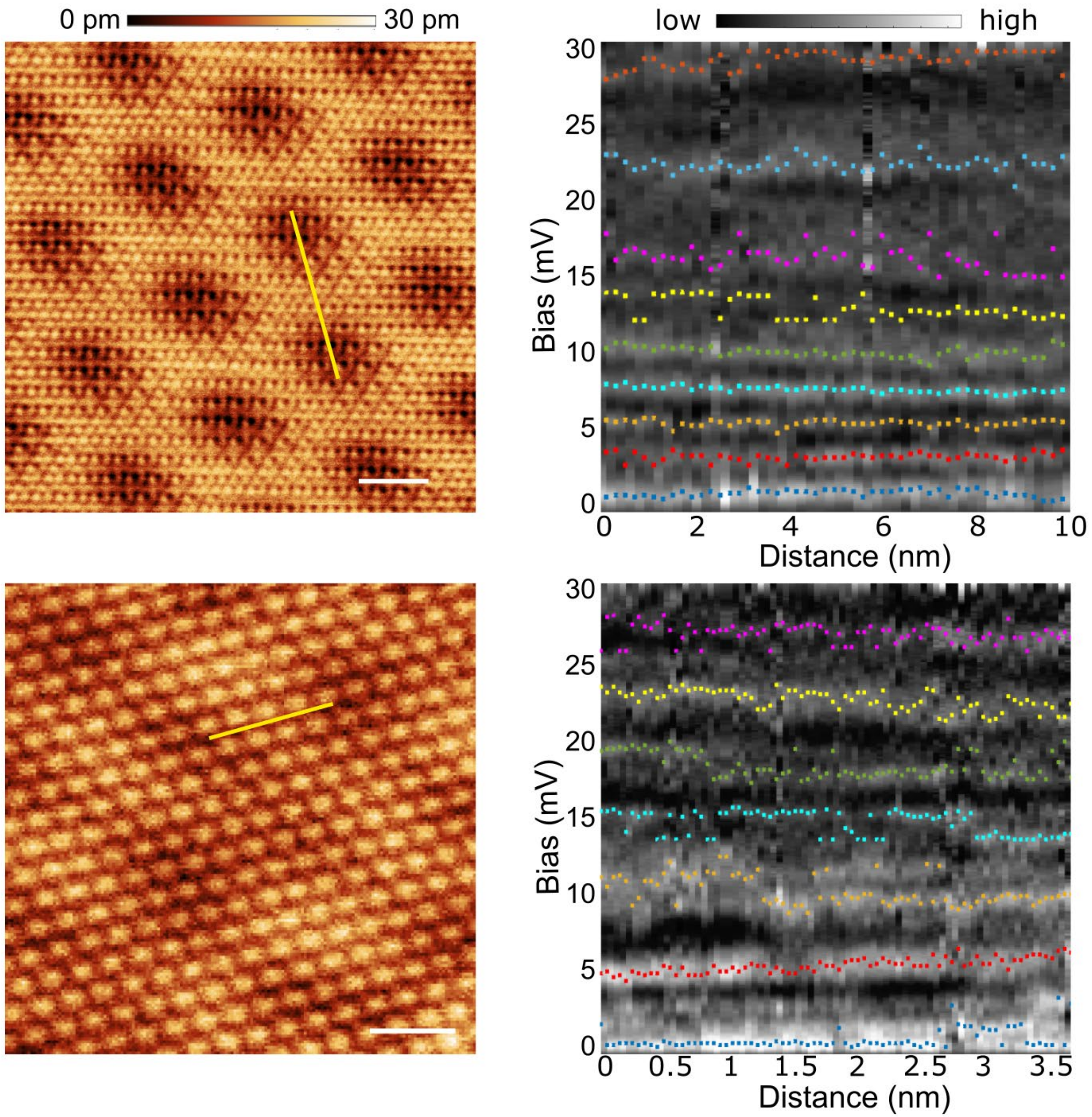
Moiré length dependence

7 nm
moiré

$L_M/a \approx 11$

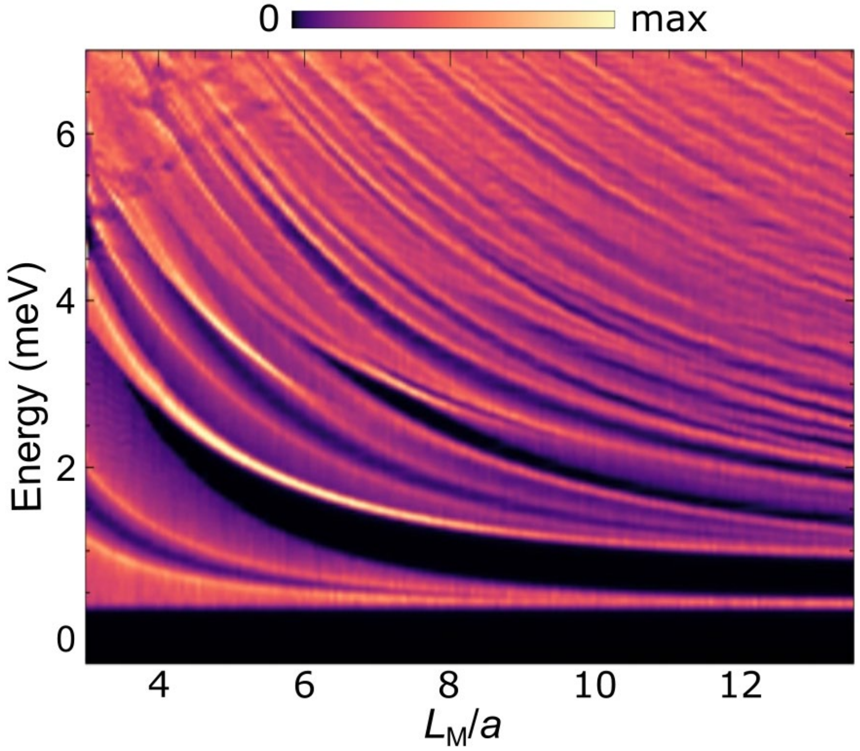
3.6 nm
moiré

$L_M/a \approx 5.6$



- Longer moiré period → more closely spaced van Hove singularities

Calculation:



S.C. Ganguli et al., "Visualization of moiré magnons in monolayer ferromagnet", Nano Lett. 23, 3412 (2023)

2D materials toolbox: monolayer multiferroics

- Multiferroic materials: coupling between ferroelectric and ferromagnetic orders
- Are there monolayer materials where this happens?
- **NiI_2** – but how does it work?

NANO LETTERS Nano Lett. 21, 5126 (2021)
pubs.acs.org/NanoLett Letter

Possible Persistence of Multiferroic Order down to Bilayer Limit of van der Waals Material NiI_2

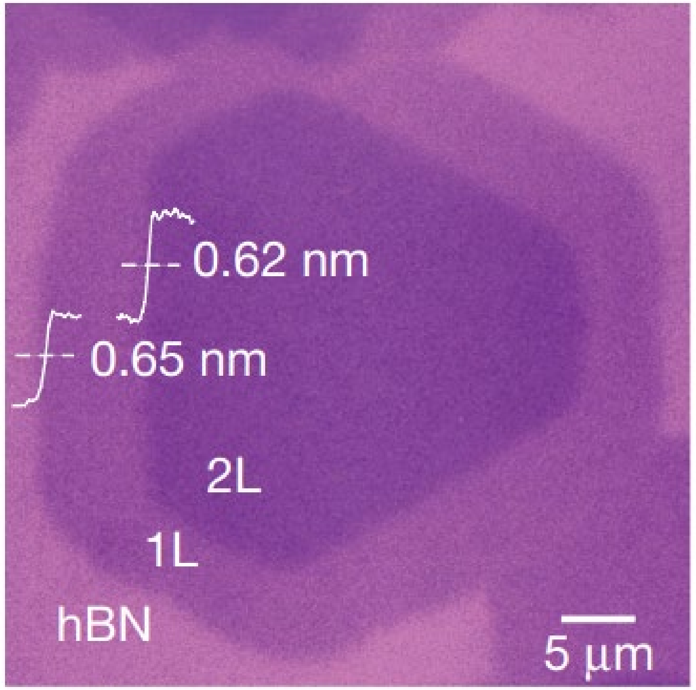
Hwiin Ju,[∇] Youjin Lee,[∇] Kwang-Tak Kim, In Hyeok Choi, Chang Jae Roh, Suhan Son, Pyeongjae Park, Jae Ha Kim, Taek Sun Jung, Jae Hoon Kim, Kee Hoon Kim, Je-Geun Park,[*] and Jong Seok Lee[*]

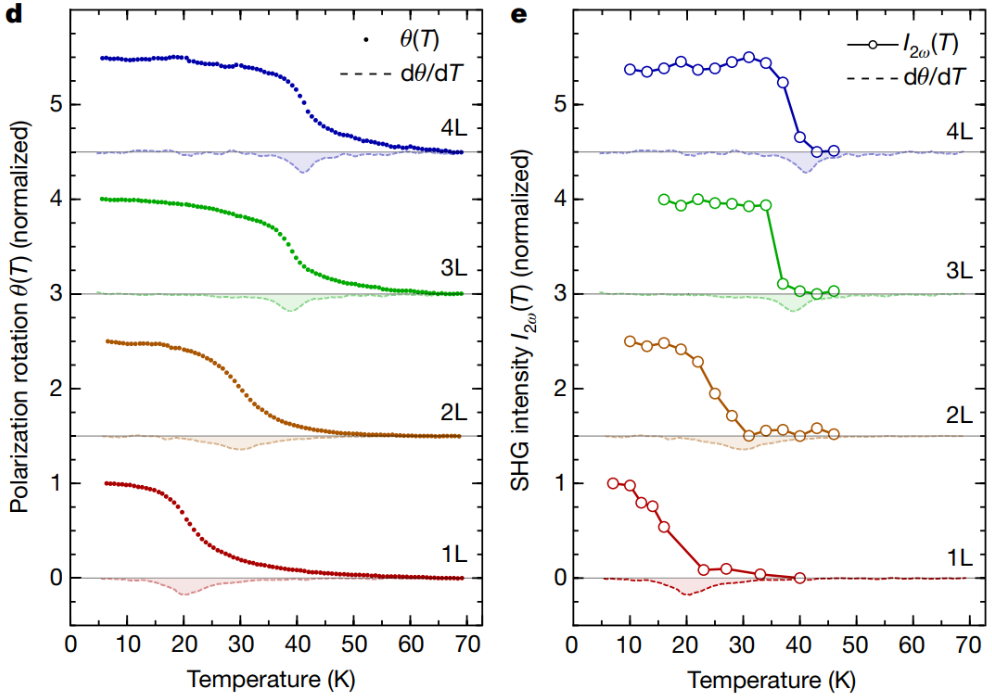
Article

Evidence for a single-layer van der Waals multiferroic

Nature 602, 601 (2022)

Qian Song[1,2,9], Connor A. Occhialini[1,9], Emre Ergeçen[1,9], Batyr Ilyas[1,9], Danila Amoroso[3,4], Paolo Barone[5], Jesse Kapeghian[6], Kenji Watanabe[7], Takashi Taniguchi[8], Antia S. Botana[6], Silvia Picozzi[3], Nuh Gedik[1] & Riccardo Comin[1✉]

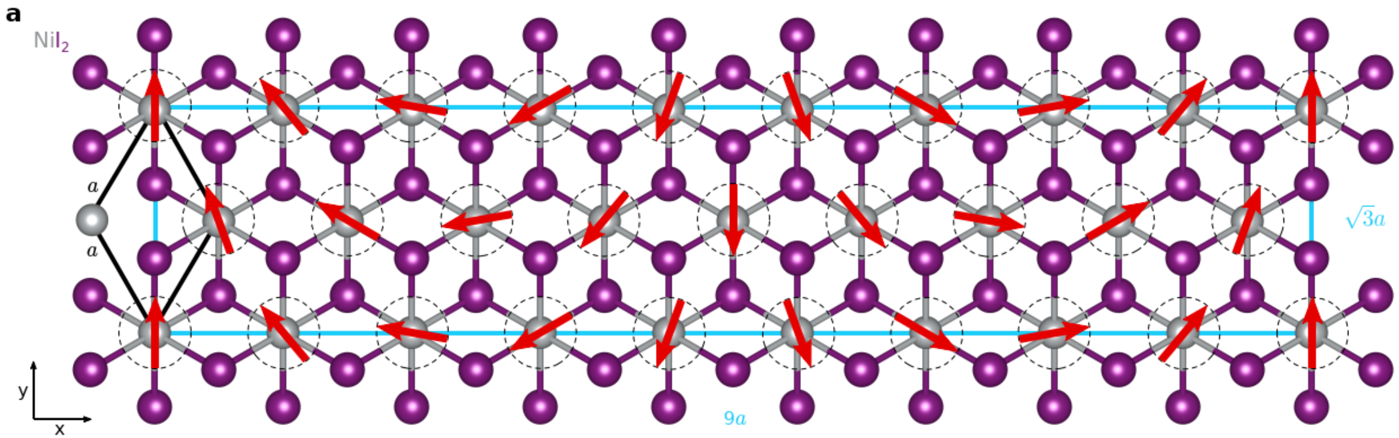




How does it work?

- Spin spiral resulting from competition between ferromagnetic nearest neighbor and antiferromagnetic third neighbor magnetic exchange

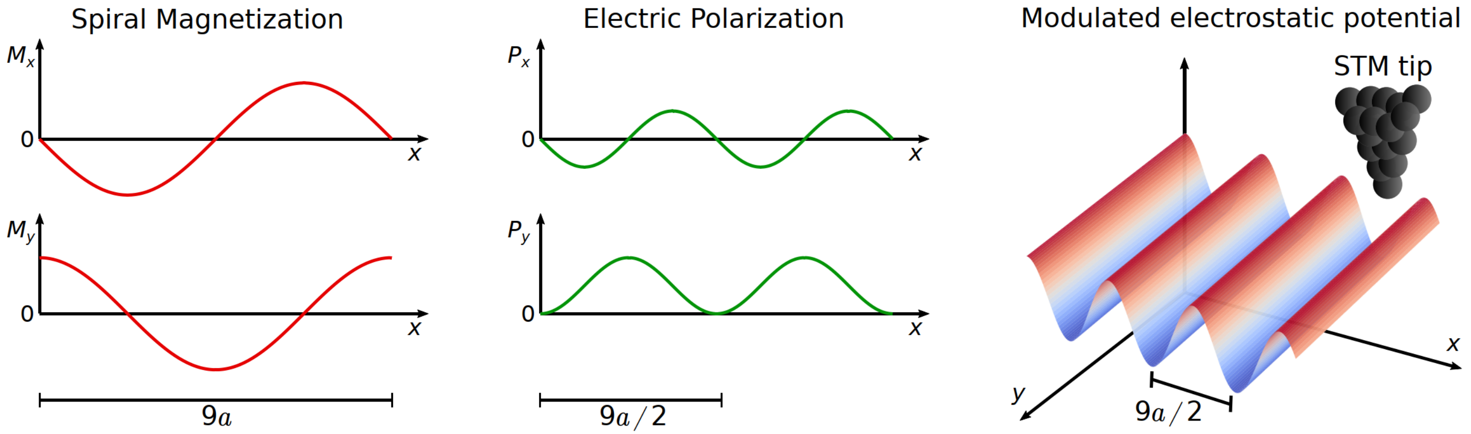
A.O. Fumega, J.L. Lado, Microscopic origin of multiferroic order in monolayer NiI_2, 2D Mater. 9, 025010 (2022)



- Spin spiral + spin-orbit interactions → ferroelectric polarization

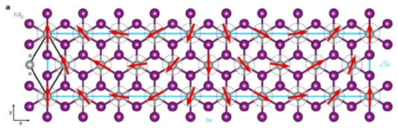
$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{M^2}$$

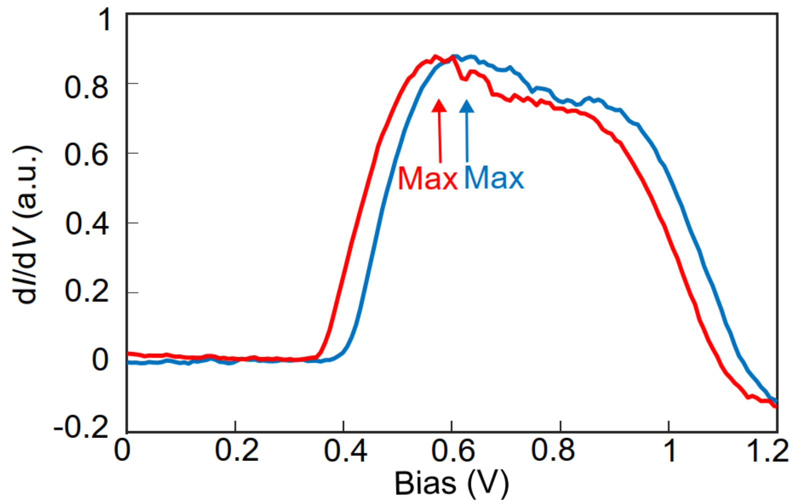
J. Hu, "Microscopic origin of magnetoelectric coupling in noncollinear multiferroics," Phys. Rev. Lett. 100, 077202 (2008).

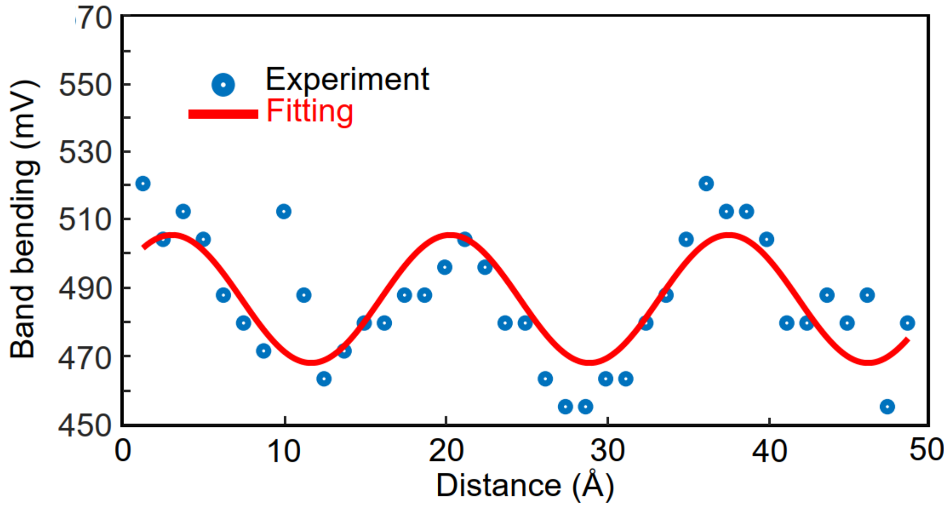


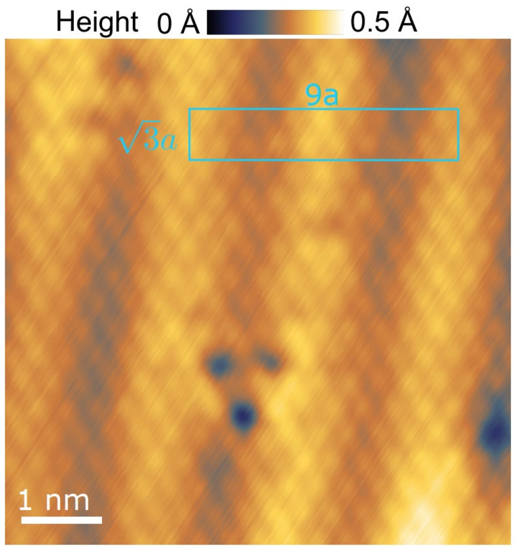
NiI_2 characterization

- Insulating gap in spectroscopy
- Moiré pattern (lattice mismatch between NiI_2 and HOPG substrate)
- Stripe pattern when imaging the conduction band
  - Matches half the spin spiral unit cell
- Periodic modulation of the conduction band on-set







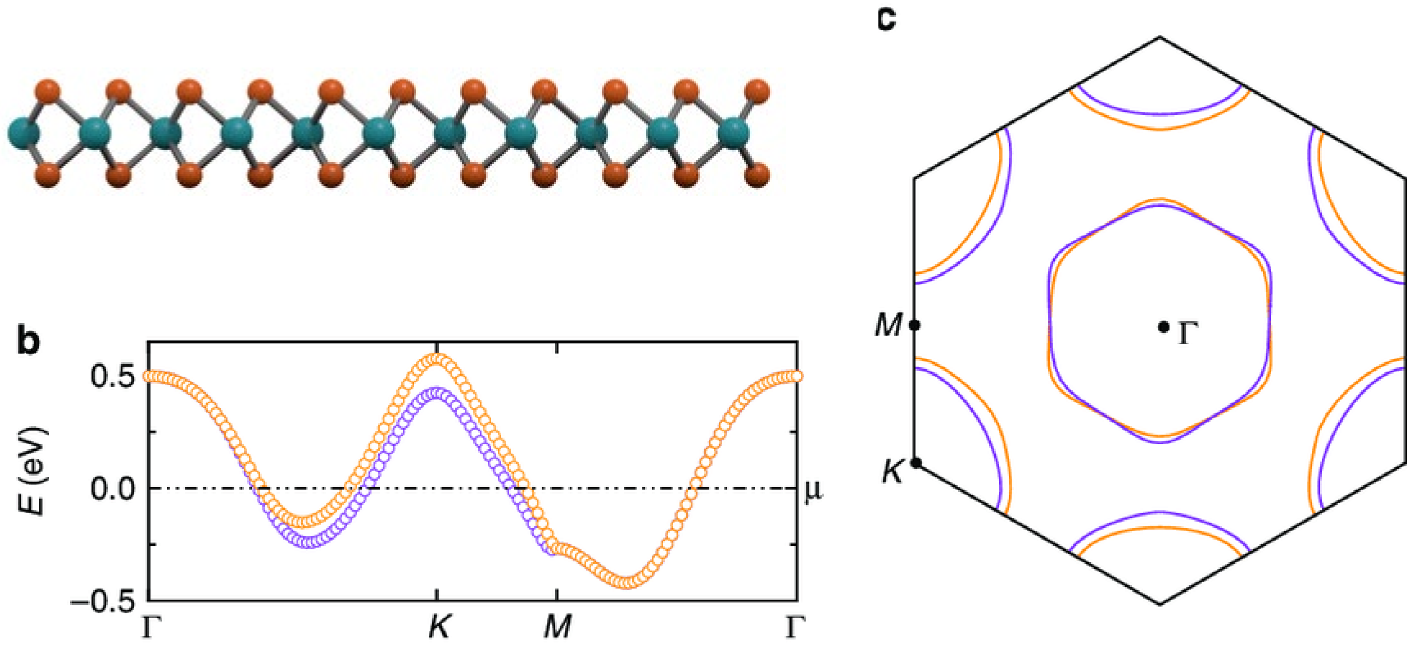


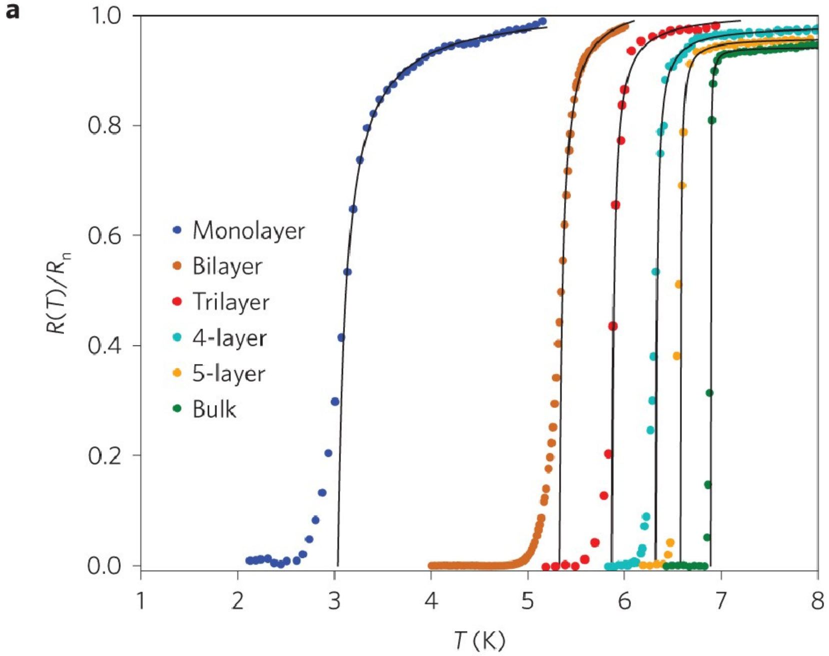
→ Polarization $P \sim 10^{-12}$ C/m

Mohammad Amini, Adolfo O. Fumega et al. "Atomic-Scale Visualization of Multiferroicity in Monolayer NiI_2", Adv. Mater. (2024) 10.1002/adma.202311342

Monolayer superconductors

- 2H-$NbSe_2$ is a superconductor down to monolayer limit
- Ising-type spin-orbit coupling locks spins out of plane
- in-plane critical field greatly exceed the Pauli paramagnetic limit





nature physics
LETTERS
PUBLISHED ONLINE: 9 NOVEMBER 2015 | DOI: 10.1038/NPHYS3538

Ising pairing in superconducting $NbSe_2$ atomic layers

Xiaoxiang Xi[1†], Zefang Wang[1†], Weiwei Zhao[1], Ju-Hyun Park[2], Kam Tuen Law[3], Helmuth Berger[4], László Forró[4], Jie Shan[1*] and Kin Fai Mak[1*]

Superconductivity in monolayer 1H-$NbSe_2$

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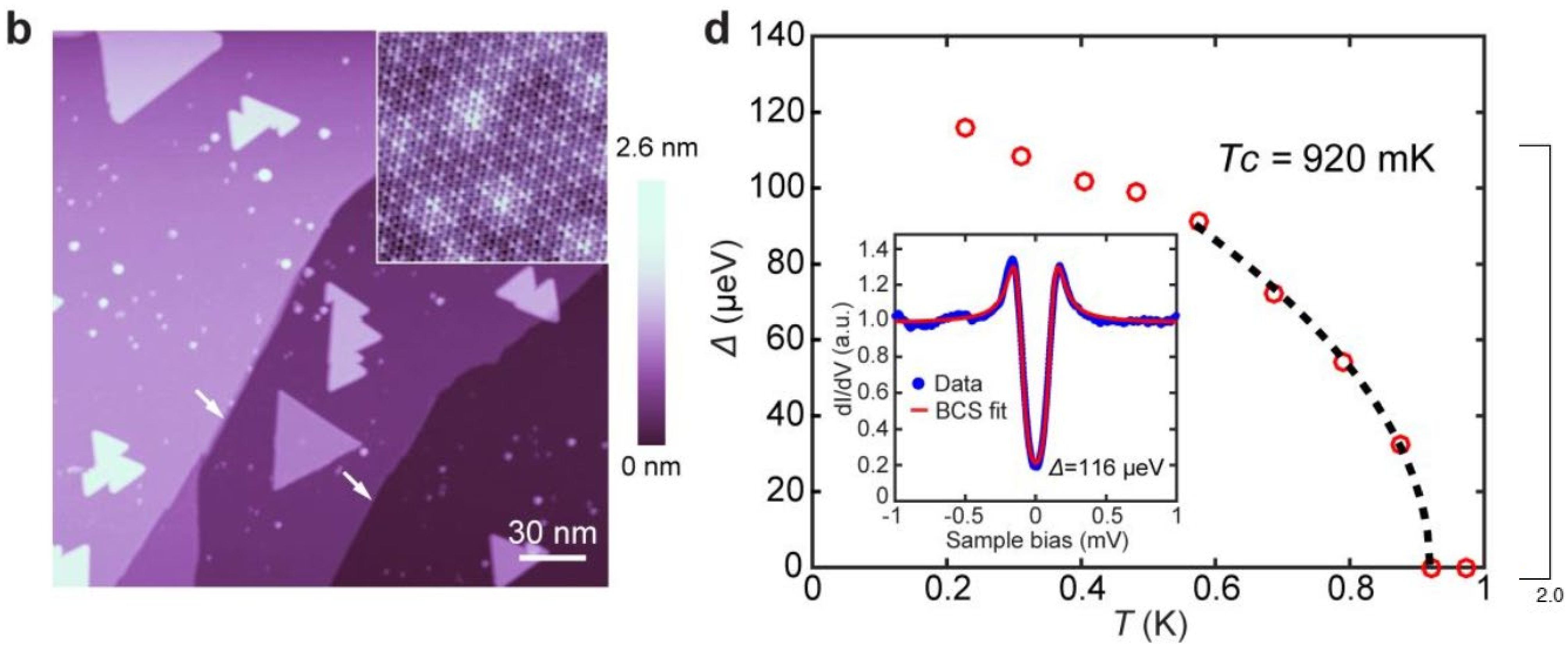
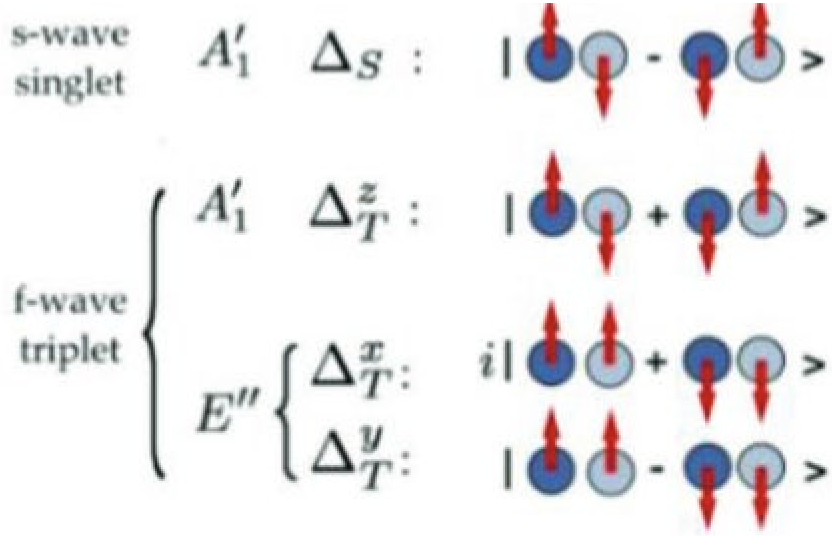
- l

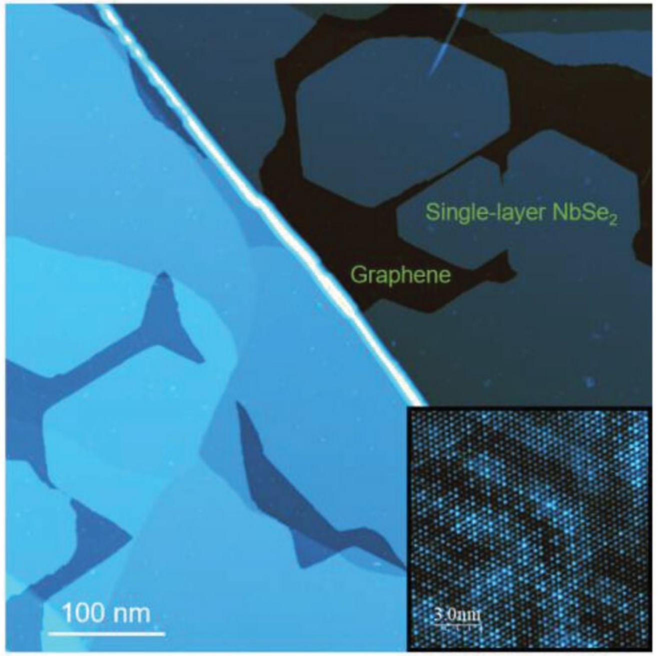


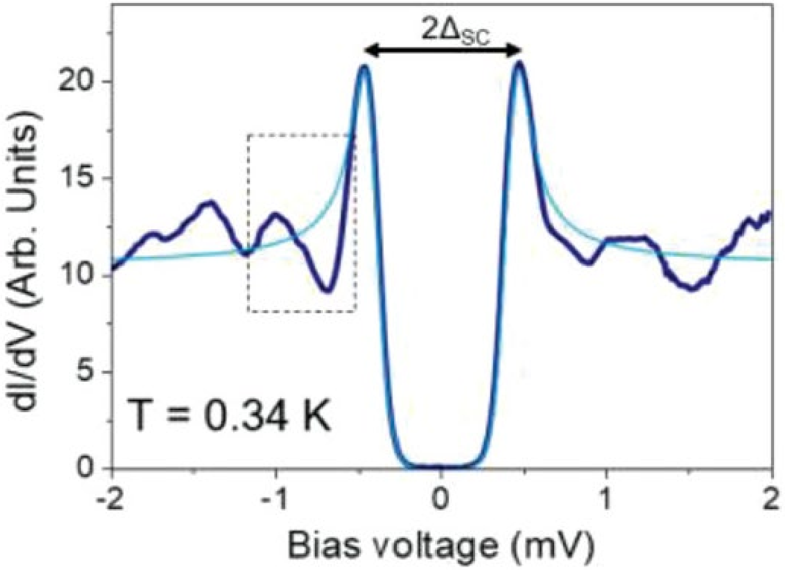
- K. Zhao et al. (Shuai-Hua Ji group), Disorder-induced multifractal superconductivity in monolayer niobium dichalcogenides, Nat. Phys. 15, 904 (2019)
- C. Rubio-Verdú et al. (M. Ugeda group), Visualization of Multifractal Superconductivity in a Two-Dimensional Transition Metal Dichalcogenide in the Weak-Disorder Regime, Nano Lett. 20, 5111 (2020)

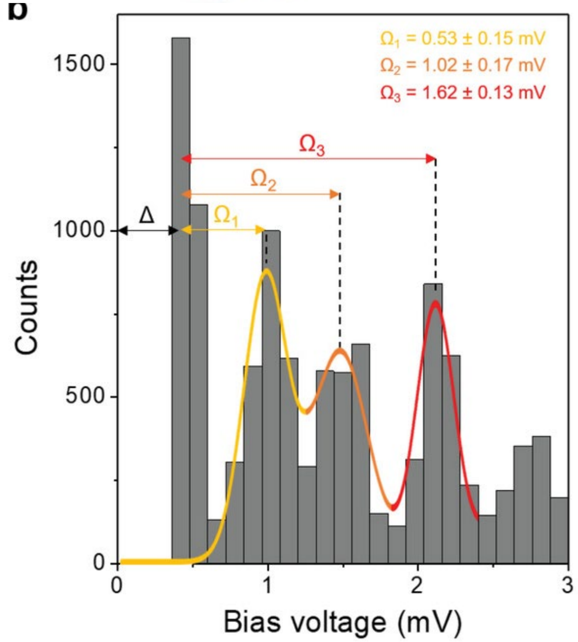
Order parameter in $NbSe_2$

- Equally spaced dip-peak features beyond the superconducting gap
- Competing order parameters
- Leggett mode – fluctuation towards a proximate spin-triplet f-wave state







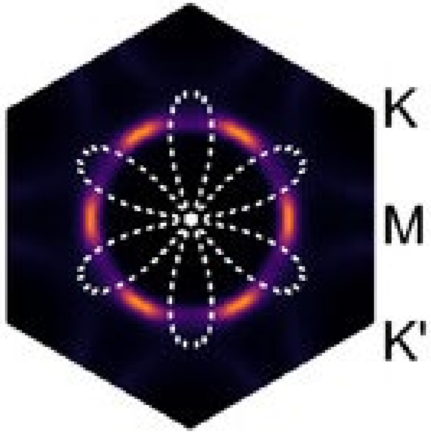


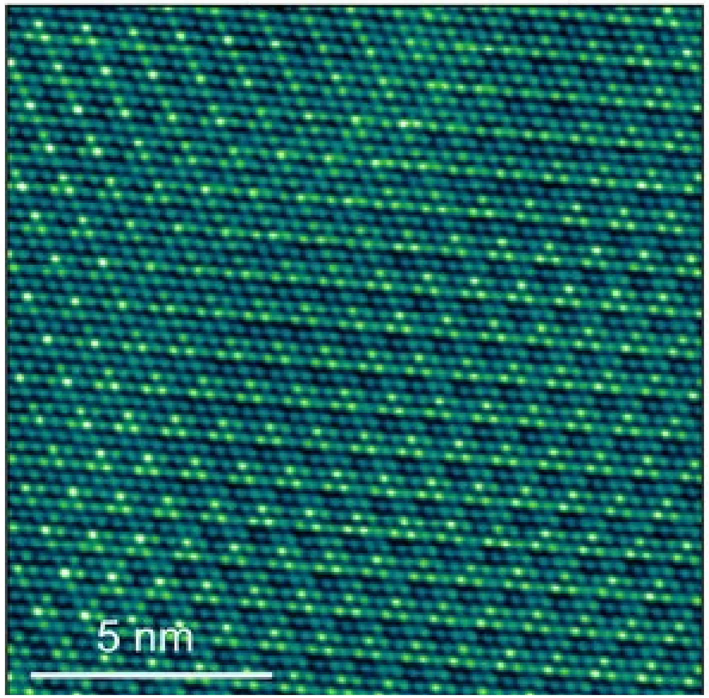
W. Wan et al. (Ugeda group), Adv. Mater. 34, 2206078 (2022)

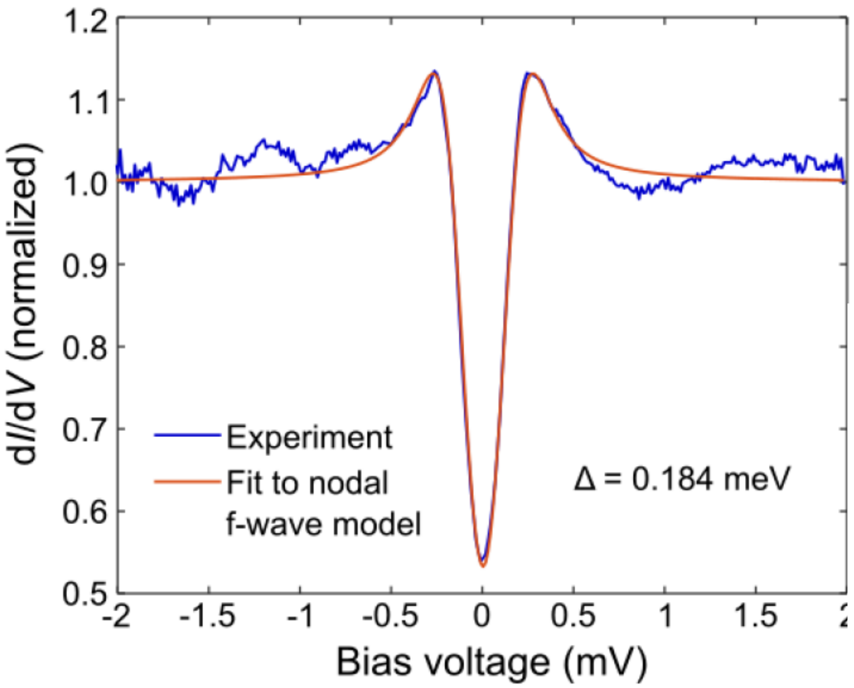
Nodal superconductivity in TMDs?

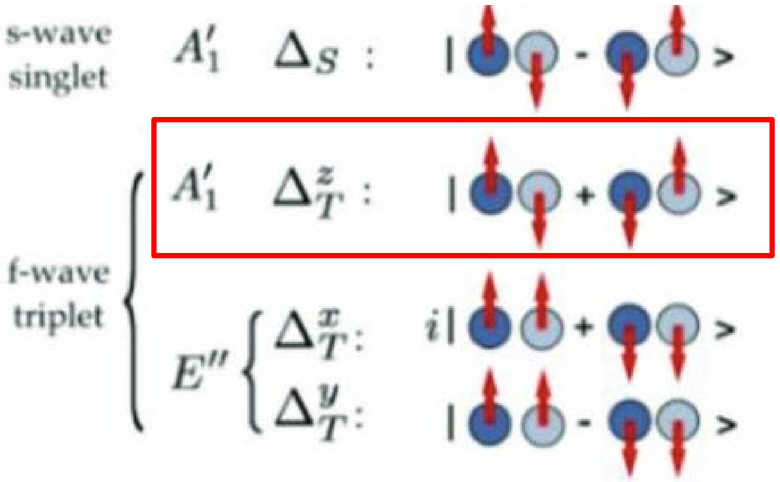
- Superconductivity in 1H-TaS_2: nodal superconductivity with f-wave pairing
- Experiments say: nodal SC
- Theory says: it is most consistent with f-wave pairing

Fermi surface with f-wave SC





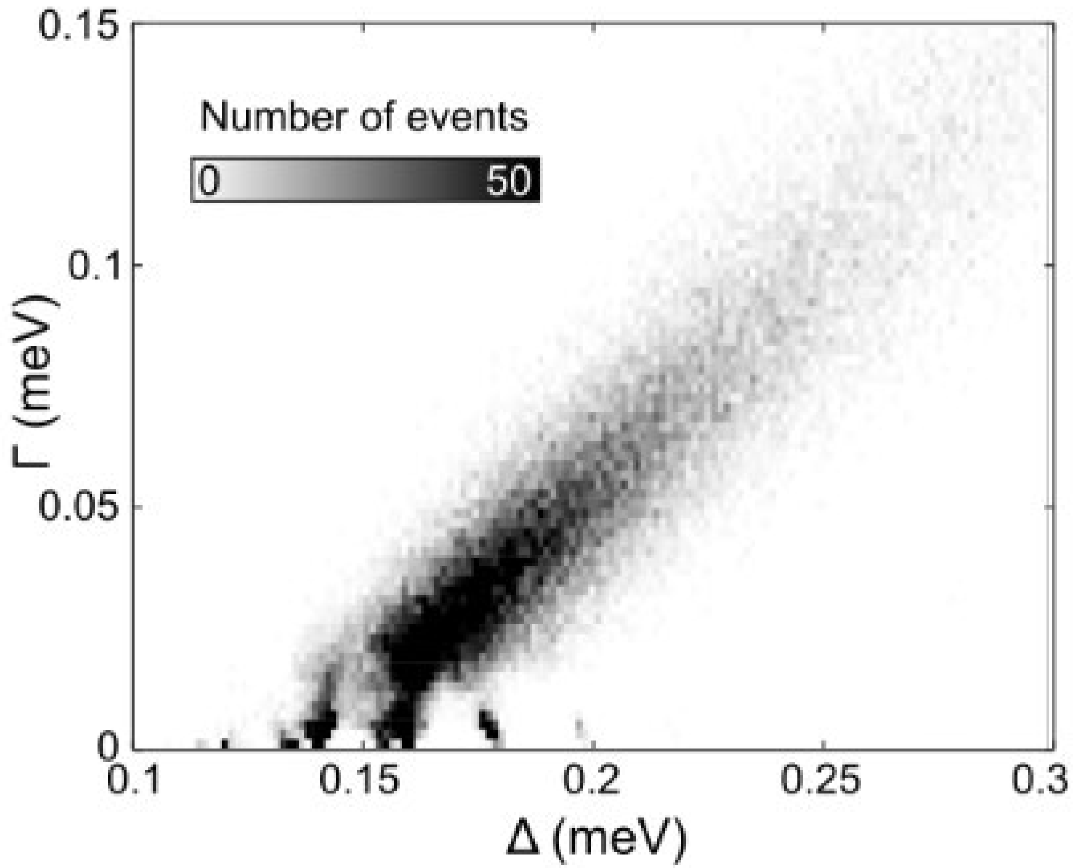




V. Vaňo, S.G. Ganguli et al. Evidence of nodal f-wave superconductivity in monolayer 1H-TaS_2 with hidden order fluctuations, Adv. Mater. 35, 2305409 (2023)

How to check nodal vs. other gaps?

$$\frac{\mathrm{d}I}{\mathrm{d}V}(eV) = \int_{-\infty}^{\infty} \rho(E) \left(-\frac{\partial f(E - eV, T_{\mathrm{eff}})}{\partial V} \right) \mathrm{d}E$$



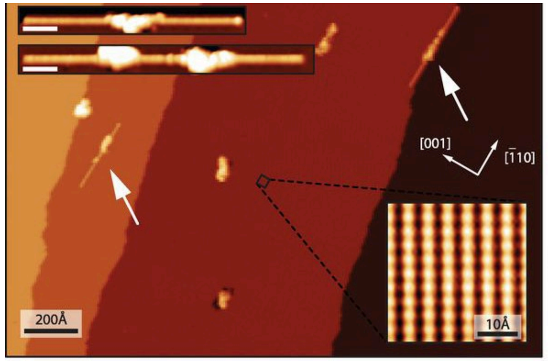
s-wave BCS model: $\rho(E) = \left| \Re \left[\frac{E}{\sqrt{E^2 - \Delta^2}} \right] \right|$

s-wave Dynes model: $\rho(E) = \left| \Re \left[\frac{E - i\Gamma}{\sqrt{(E - i\Gamma)^2 - \Delta^2}} \right] \right|$

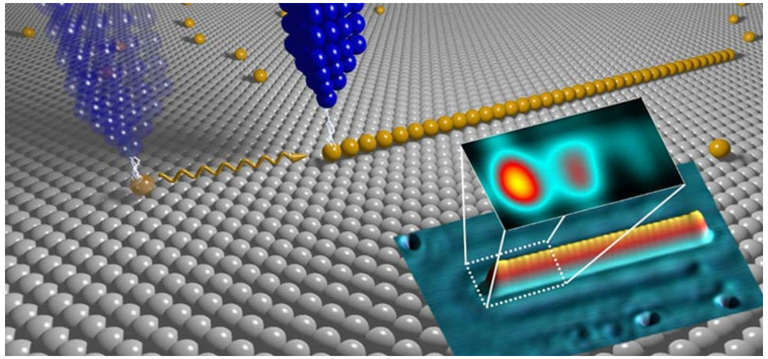
nodal f-wave model: $\rho(E) = \int_0^{2\pi} \left| \Re \left[\frac{E}{\sqrt{E^2 - (\Delta \cos(\theta/3))^2}} \right] \right| d\theta$

Topological superconductivity

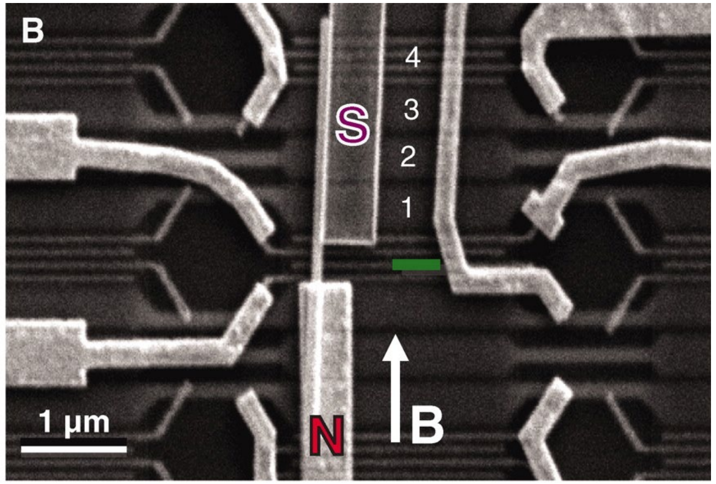
- New phase of matter
  - Rare in naturally occurring materials
- Majorana modes at the boundaries
  - Can be used to construct topologically protected qubits?
- Engineered in nanowire devices or atomic-scale systems



Yazdani group, Science 346, 602 (2014)



Wiesendanger group, e.g. Nat. Phys. 17, 943 (2021), Nat. Nano (2022)

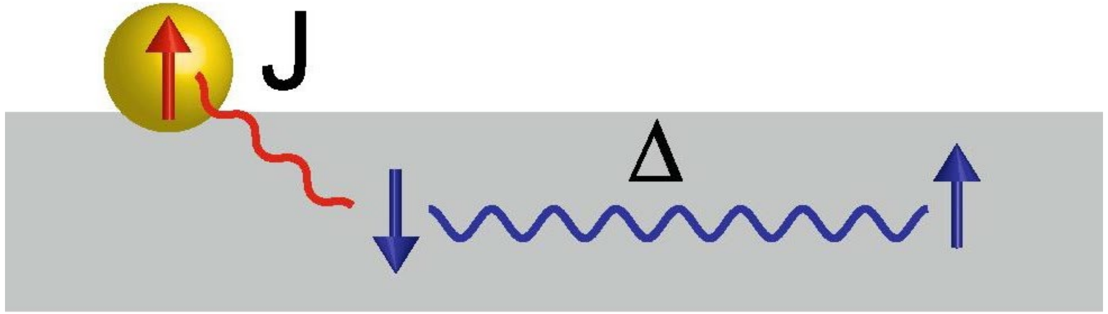


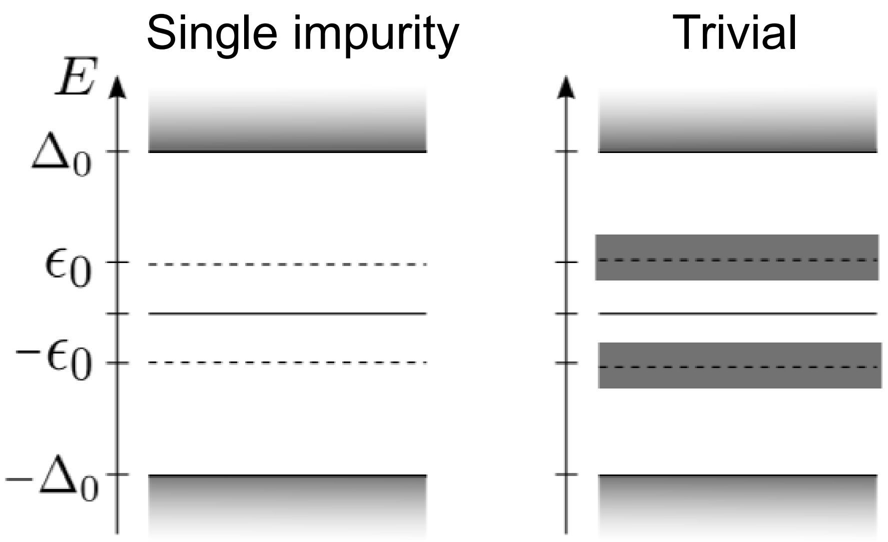
More opportunities with 2D heterostructures?

Review: K. Flensberg, F. von Oppen, A. Stern, Engineered platforms for topological superconductivity and Majorana zero modes, Nat. Rev. Mater. 6, 944 (2021)

How to think about this?

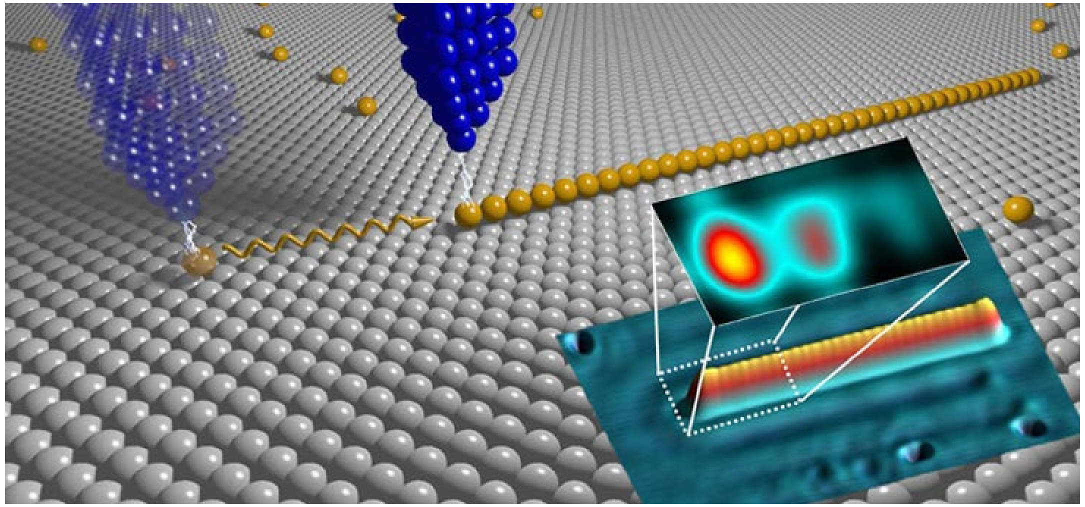
- Single spins give rise to Yu-Shiba-Rusinov bound states
- Neighboring YSR states can hybridize and form coupled states
  - D.-J. Choi et al. Colloquium: Atomic spin chains on surfaces, Rev. Mod. Phys. 91, 041001 (2019)
- Larger assemblies – YSR bands and possibly topological superconductivity
  - Relevant parameters – *J*, magnetic texture, spin-orbit interactions etc.





Atomic scale chains for topological superconductivity

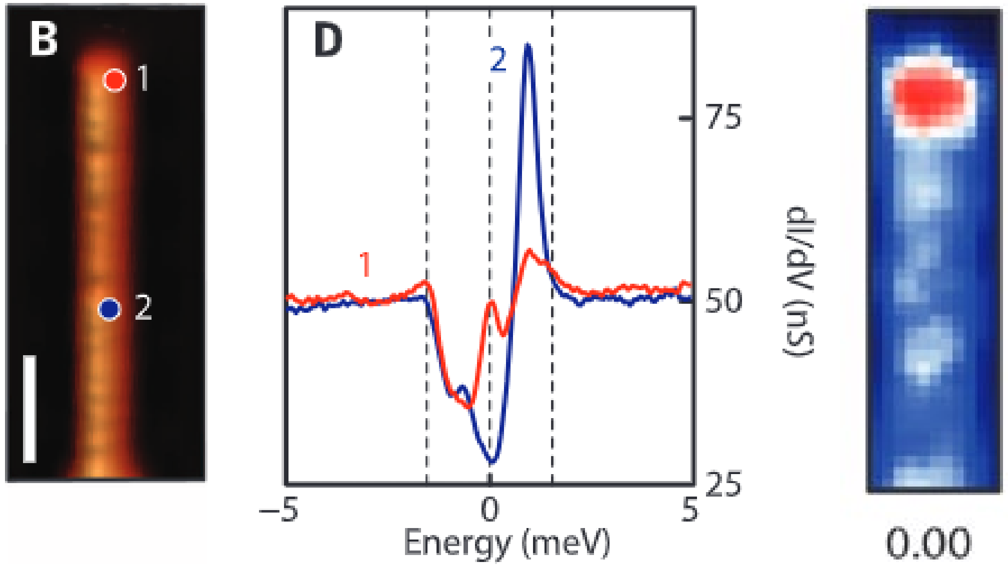
- Systems can be realized via "self-assembly", e.g. iron atoms on Pb(100)
- More elegant (?) and controlled – use atom manipulation



Wiesendanger group, e.g.
Nat. Phys. 17, 943 (2021),
Nat. Nano. 17, 384 (2022)

Observation of Majorana fermions in ferromagnetic atomic chains on a superconductor Science 346, 602 (2014)

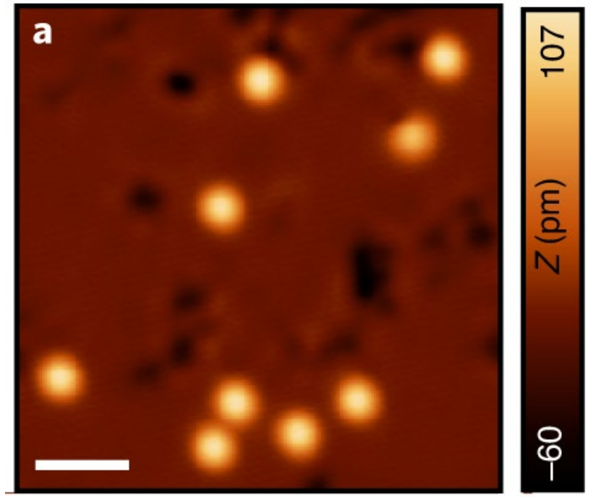
Stevan Nadj-Perge,[1*] Ilya K. Drozdov,[1*] Jian Li,[1*] Hua Chen,[2*] Sangjun Jeon,[1] Jungpil Seo,[1] Allan H. MacDonald,[2] B. Andrei Bernevig,[1] Ali Yazdani[1]†

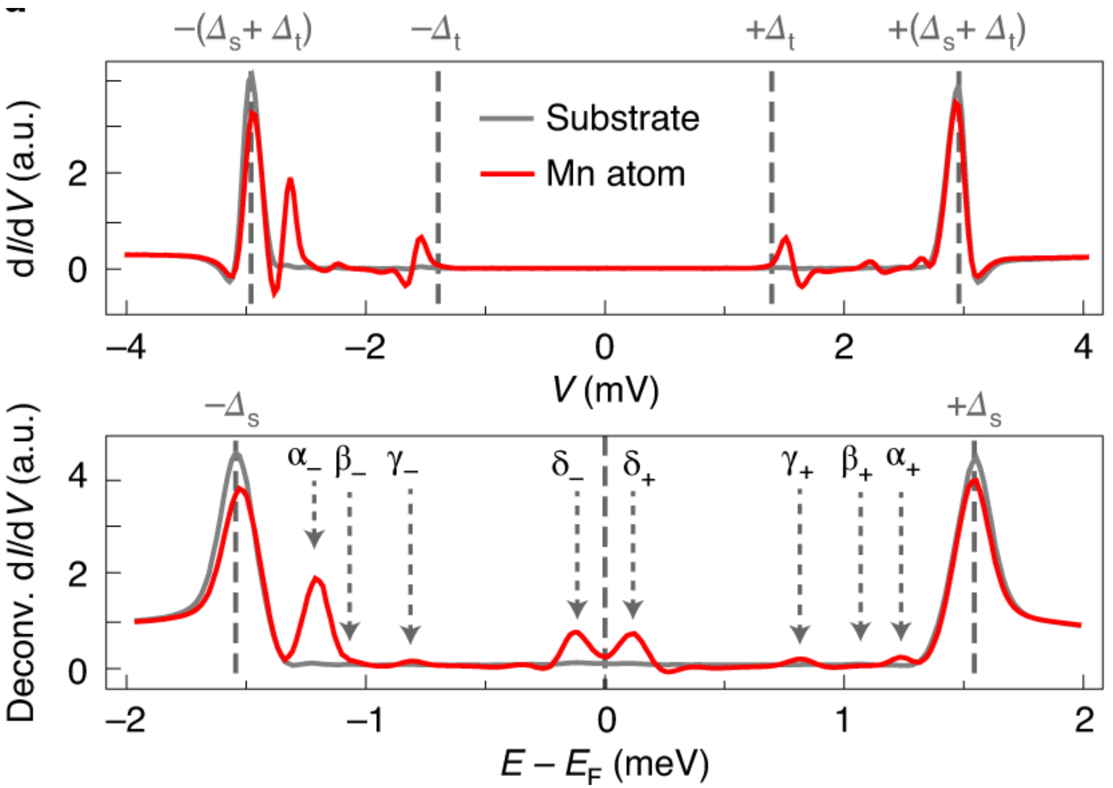


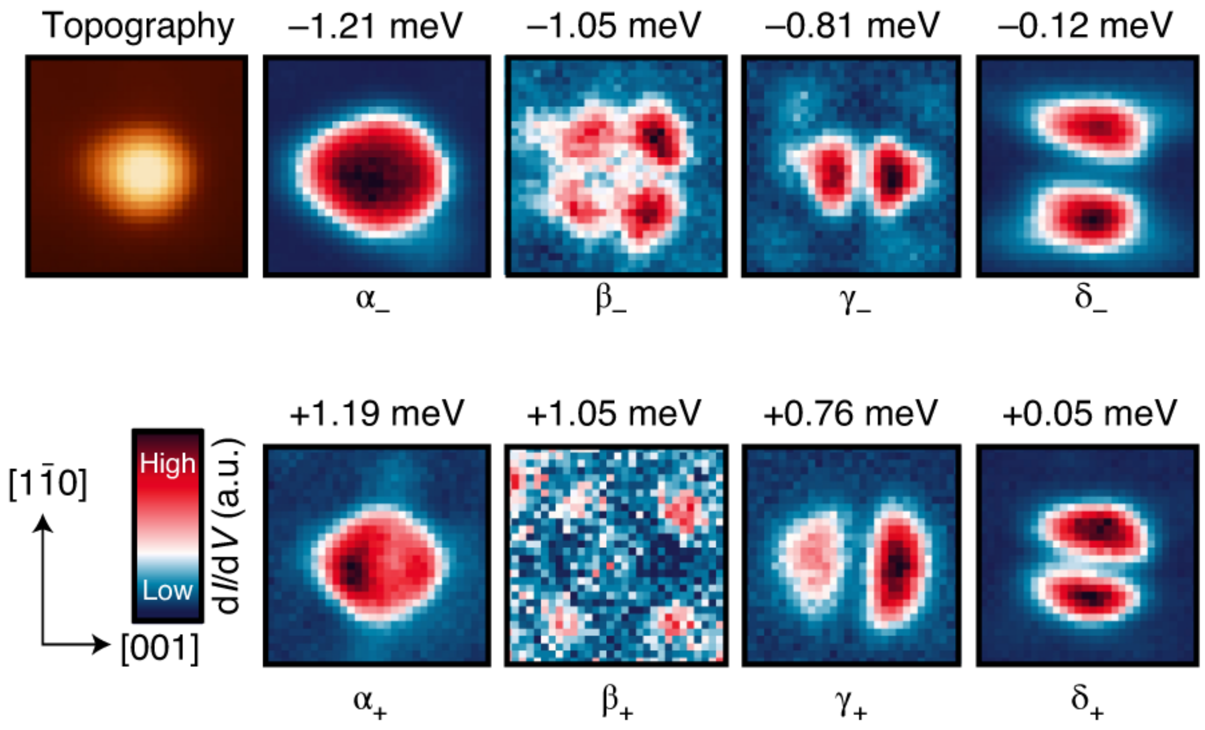
Single atom: Mn on Nb(110)

- Single manganese atoms on Nb(110) give rise to several YSR states
- Each with different wavefunction symmetries

L. Schneider et al. (Wiesendanger and Wiebe groups), Topological Shiba bands in artificial spin chains on superconductors, Nat. Phys. 17, 943 (2021).

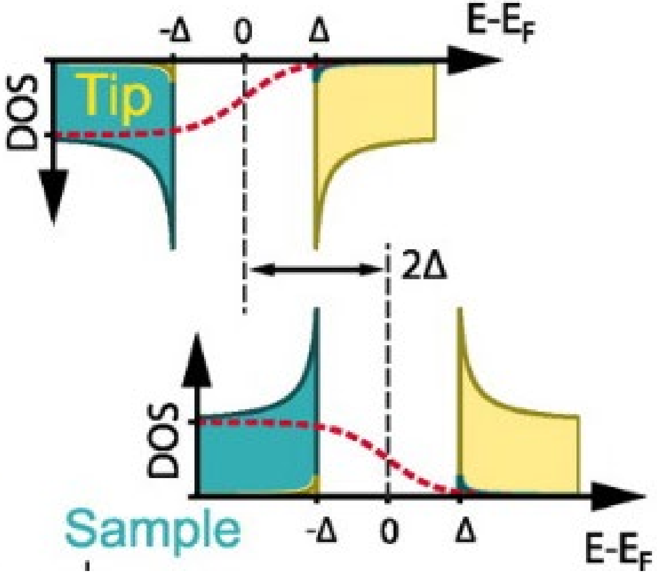






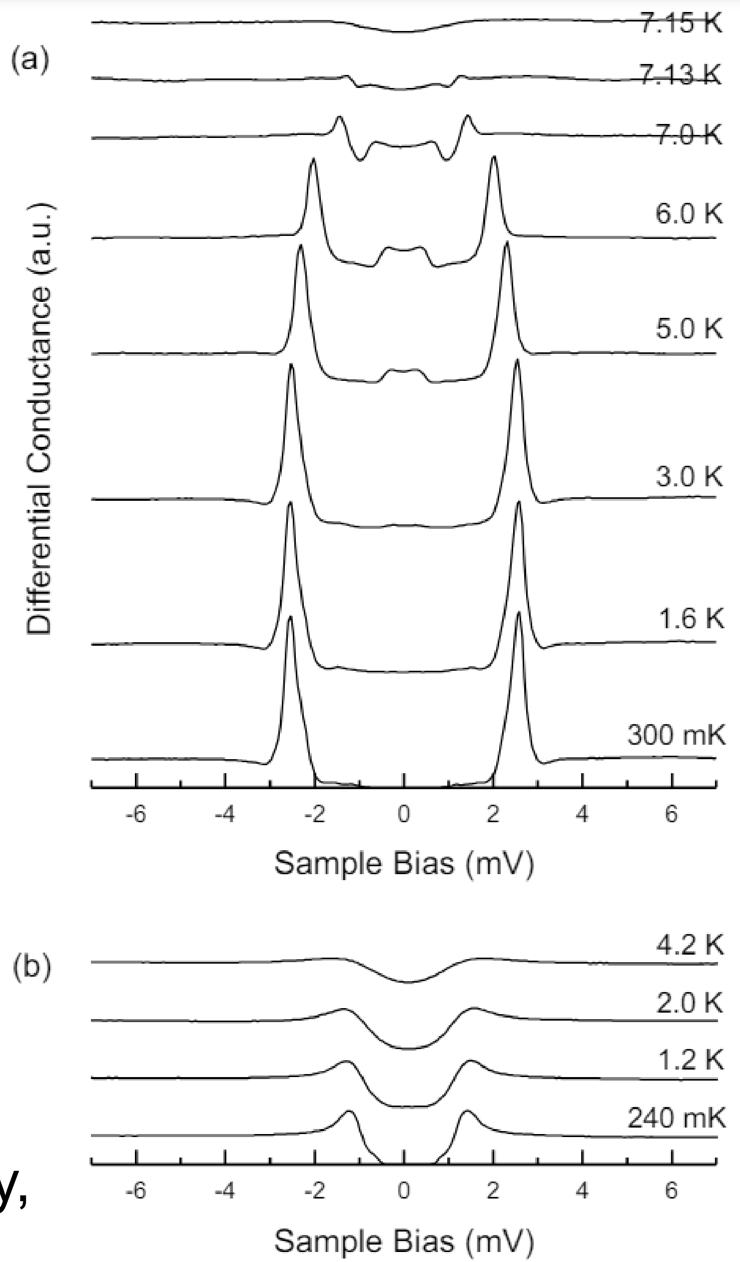
STM with a superconducting tip

- Superconducting tip convolves the substrate DOS with the coherence peaks
- Enhances energy resolution
- Standard "trick" in this type of STM experiments
- (also possible to look at the Josephson current, but that is another story)



Niobium tip
$NbSe_2$ substrate

Normal metal tip
$NbSe_2$ substrate

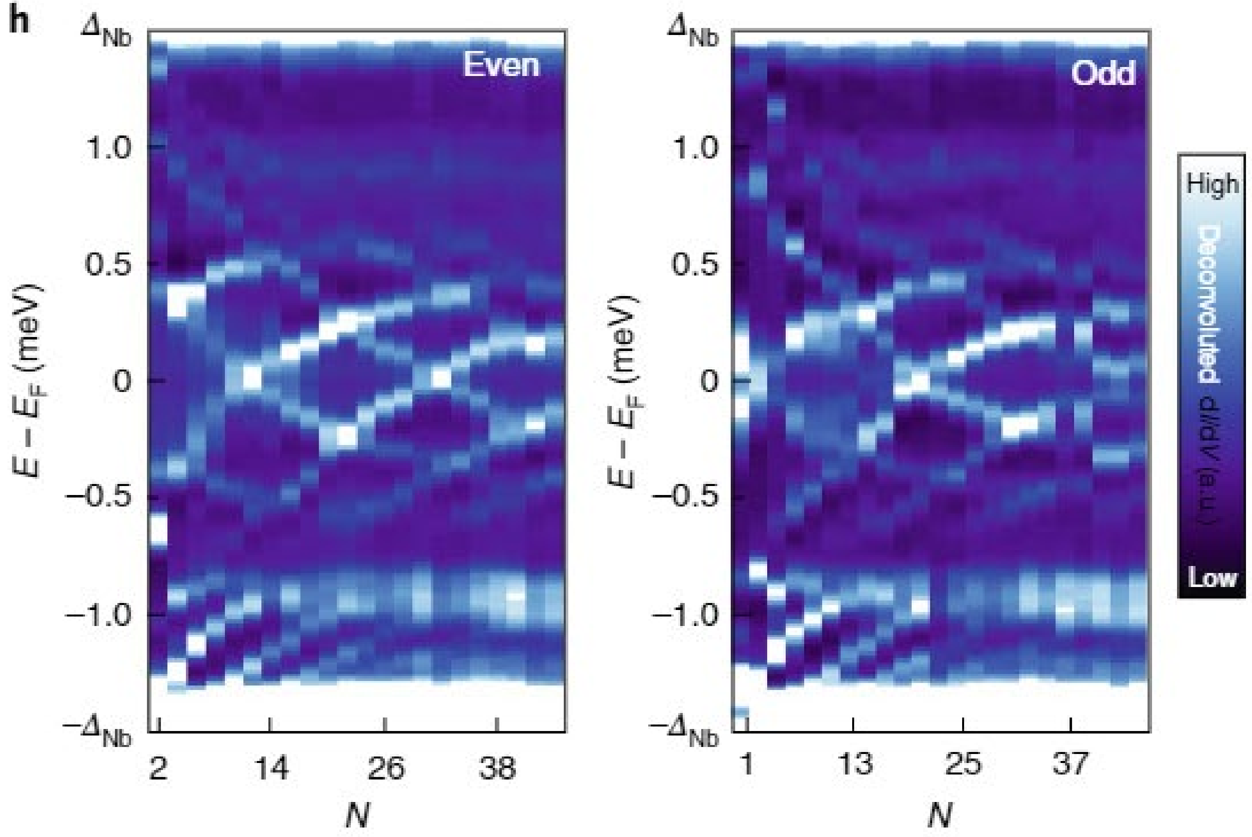


E.W. Hudson, PhD thesis (UC Berkeley, group of J.C. Séamus Davis), 1999.

Chains

- Chain directions has a big effect – [110] direction gives end states

- Odd – even effect in the end state energies
- Additionally, smooth variation
- Overlap between the end modes ("precursor Majorana modes")
- System is topological, but topological gap is small → Majoranas are extended
- Need other system parameters, e.g. larger spin-orbit coupling



L. Schneider et al. (Wiesendanger and Wiebe groups), Precursors of Majorana modes and their length-dependent energy oscillations probed at both ends of atomic Shiba chains, Nat. Nanotechnol. 17, 384 (2022).

Topological superconductivity – ”continuum”

Quantum cooking topological superconductivity - standard recipe

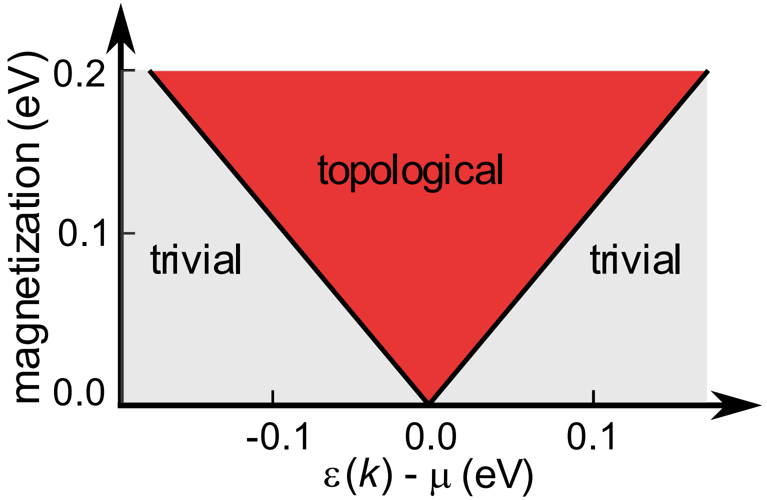
1. Take a metal
2. Add spin-orbit interactions (Rashba type, strength α)
3. Add magnetization *M* (perpendicular to spin-orbit)
4. Add superconductivity Δ (and realize a spinless SC)

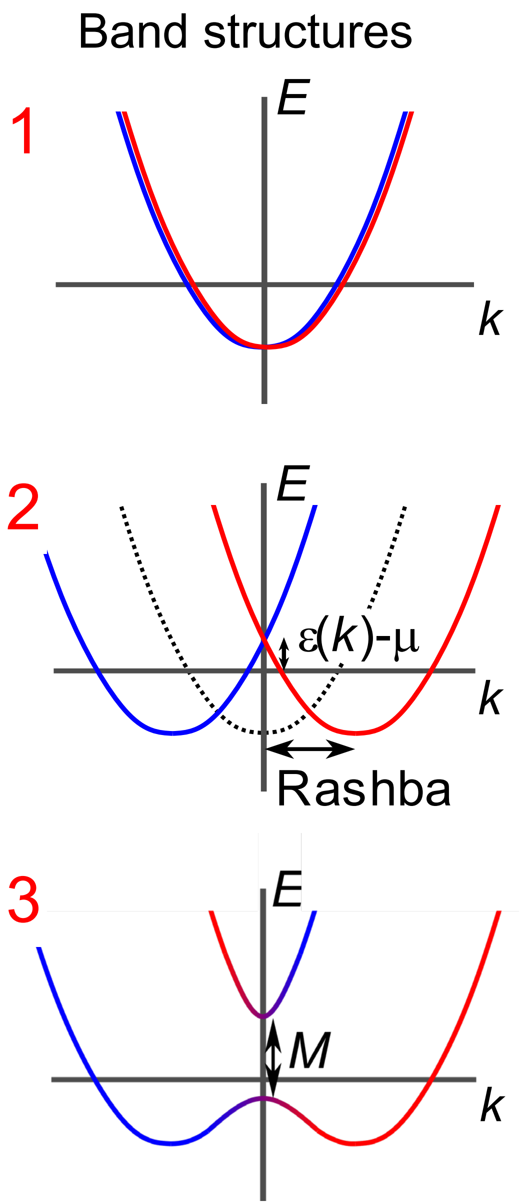
- Condition for the topological phase

$$|\epsilon(\vec{k}_0) - \mu| \leq M$$

- The topological gap is

$$\Delta_t = \frac{\alpha k_F}{[(\alpha k_F)^2 + M^2]^{1/2}} \Delta$$





Jay D. Sau et al. Phys. Rev. Lett. **104**, 040502 (2010)

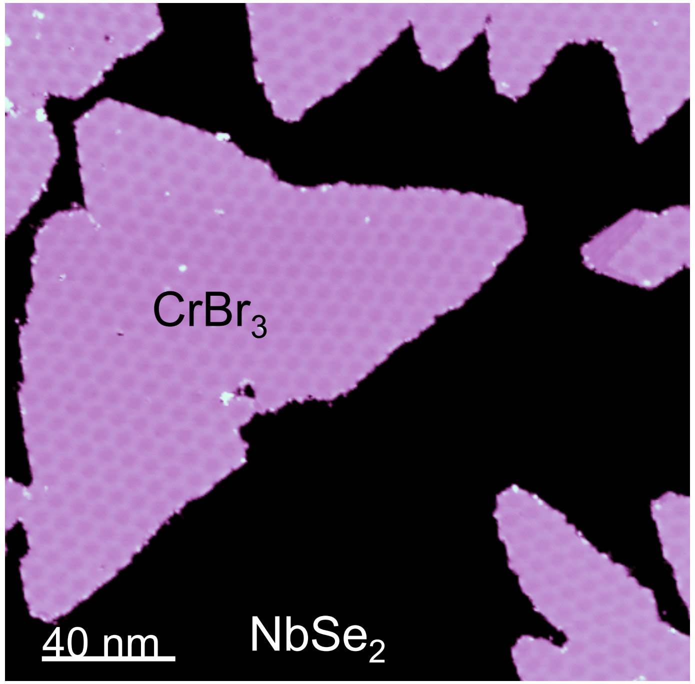
Which materials? $CrBr_3$ growth on $NbSe_2$

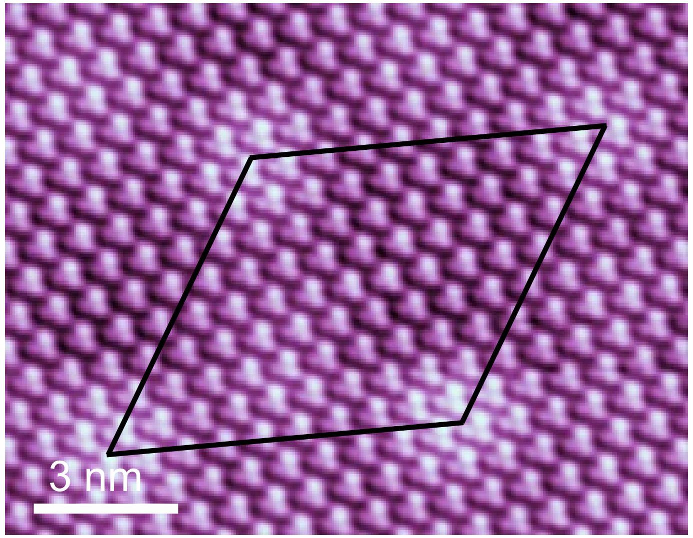
- $NbSe_2$ substrate
  - Well-known vdW superconductor, Tc ~ 7K
  - Preparation: cleave in vacuum
- $CrBr_3$ is an out of plane ferromagnetic insulator

- Layers interact only weakly and can retain their intrinsic properties
- Proximity effects allow properties to ”leak” to neighboring layers
- $NbSe_2$ substrate + $CrBr_3$ (out-of-plane ferromagnetic insulator)

⇒ **$CrBr_3$ magnetizes the top $NbSe_2$ layer**

⇒ **NbSe$_2$ under CrBr$_3$ is a topological superconductor**

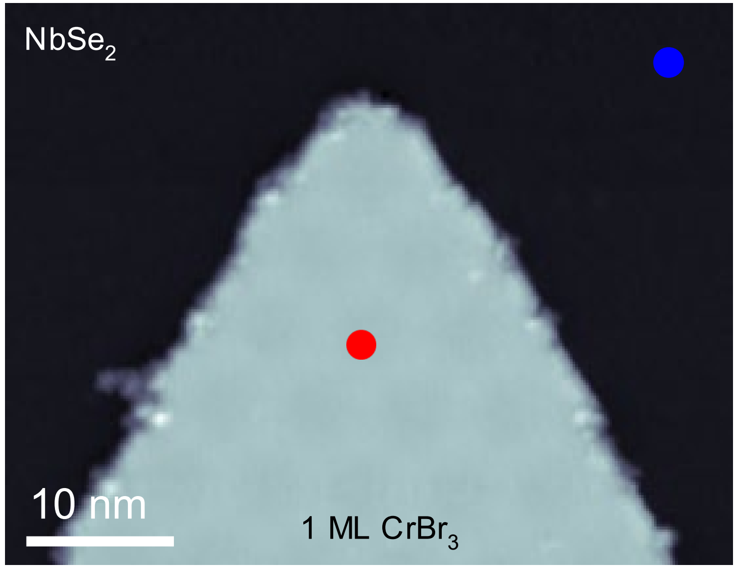




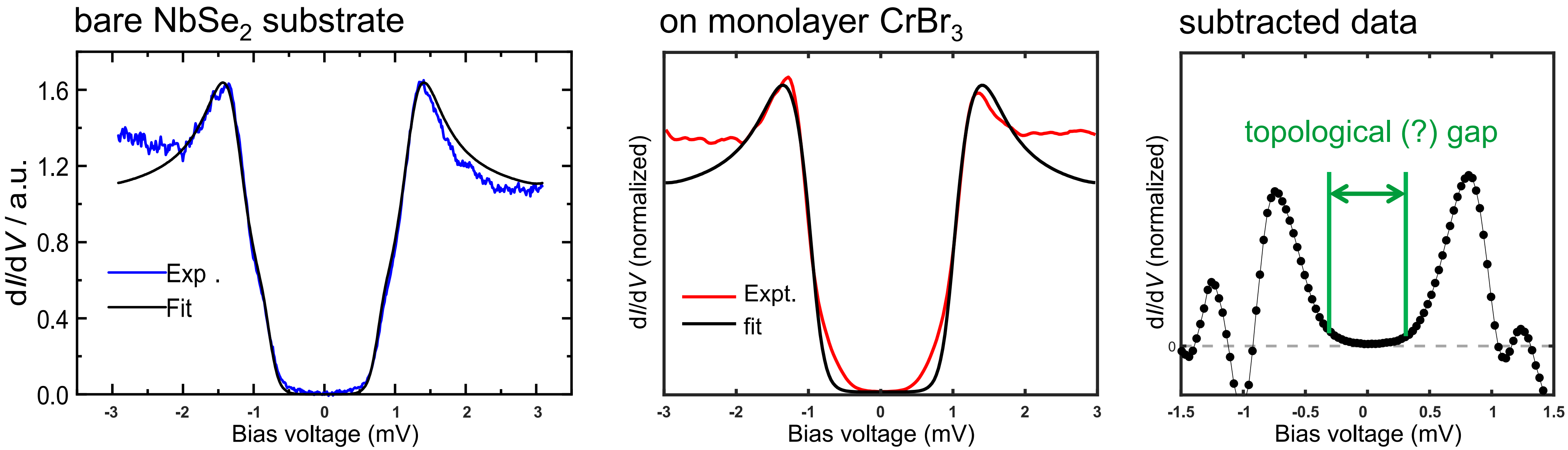
S. Kezilebieke et al. ”Electronic and Magnetic Characterization of Epitaxial $CrBr_3$ Monolayers on a Superconducting Substrate” Adv. Mater. 33, 2006850 (2021).

Low-bias spectroscopy

- Combining the required ingredients for topological superconductivity
- On the island, there is signal inside the SC gap: formation of Shiba bands due to the magnetic layer
- Gap between the Shiba bands



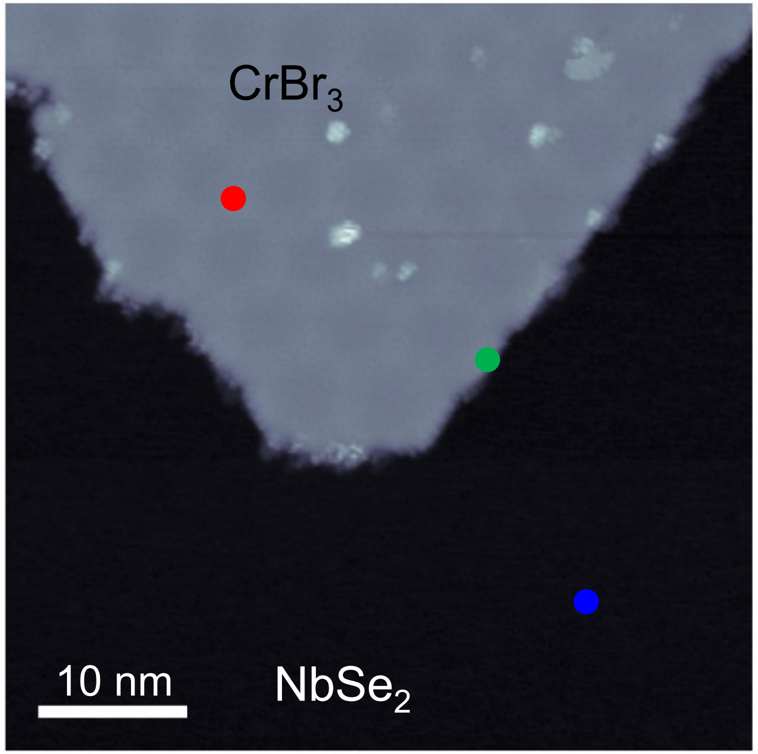
STM experiments at T = 350 mK

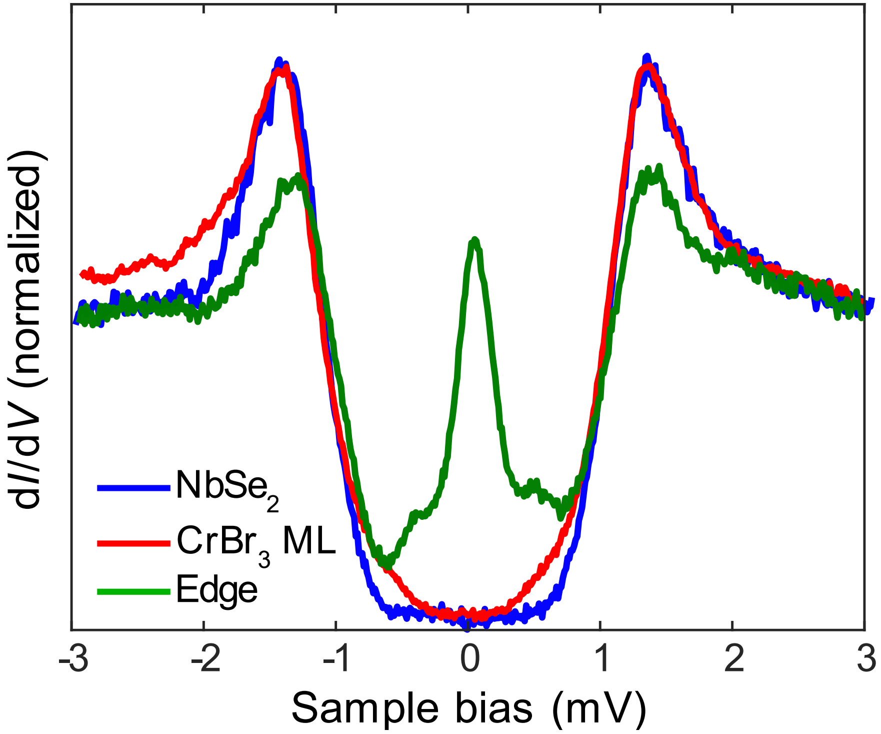


Are there Majorana modes?

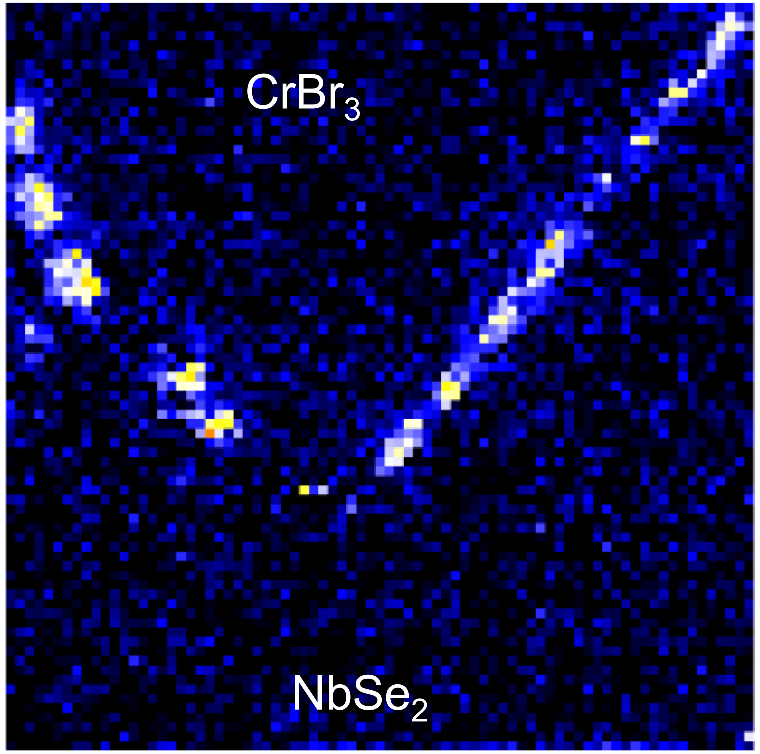
- Distinct zero bias signature at the island edges
- Spatial mapping: localized edge modes

STM image:





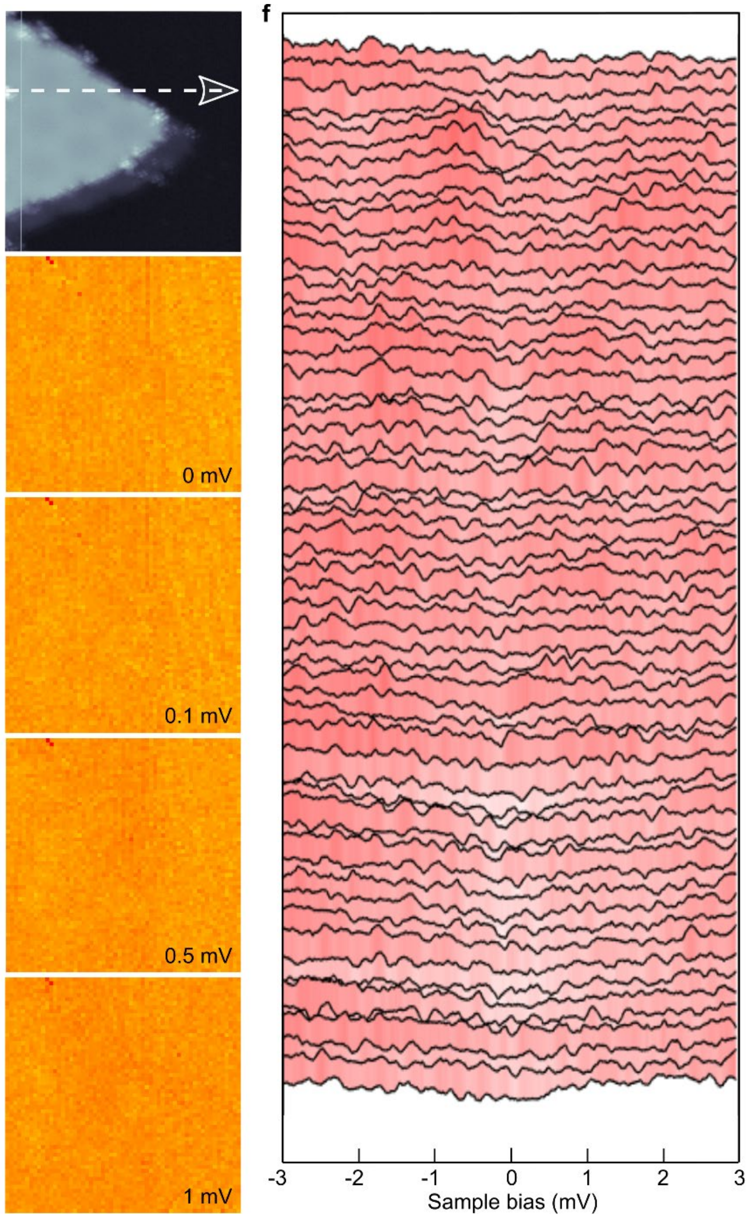
LDOS map at E_F:



S. Kezilebieke et al. "Topological superconductivity in a van der Waals heterostructure", Nature 588, 424 (2020)

Is it topological superconductivity?

- Appears on all island edges
- Removing superconductivity (quenching with an external magnetic field) also removes the edge state completely
- Not Kondo, not standard edge state

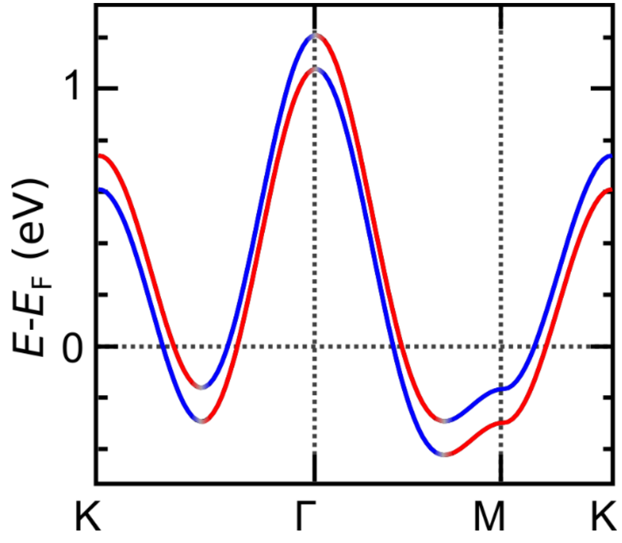


External field 4T

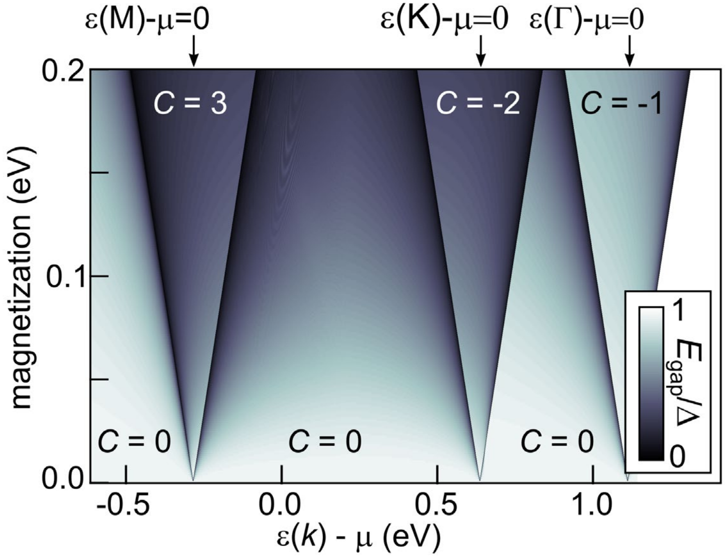
Discussion

- Many things match between theory and experiment.
- What does not?
- Chemical potential does not match – what is going on?
- Modulation of the edge mode intensity along straight edges

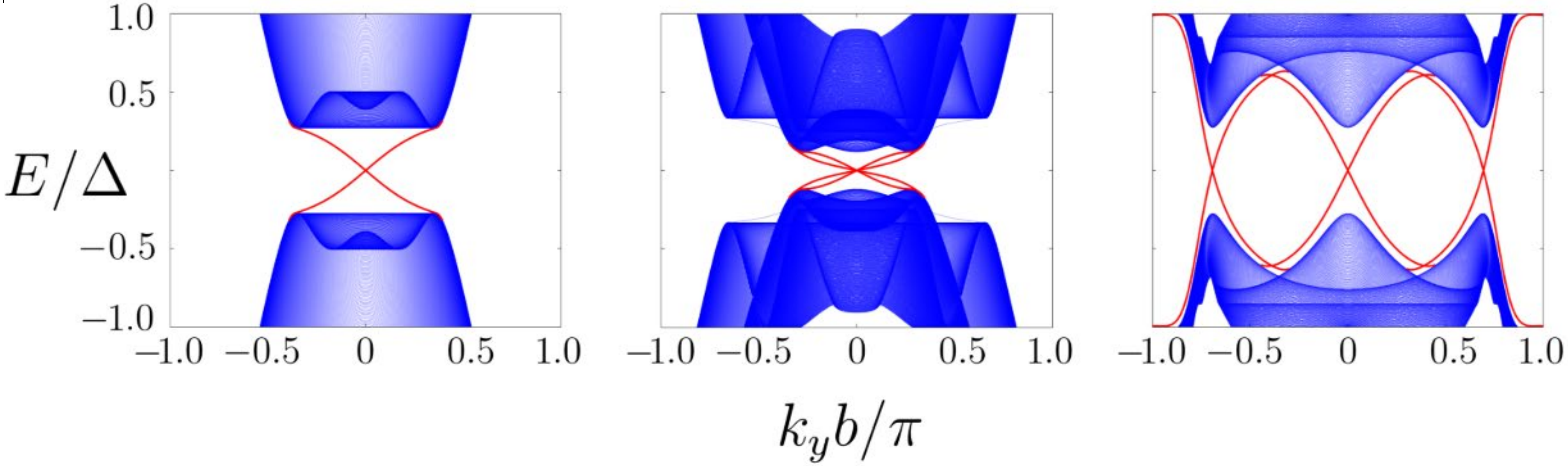
NbSe$_2$ with Rashba + Zeeman



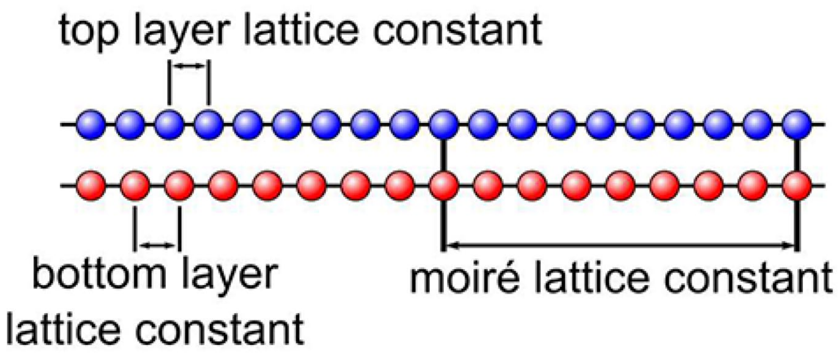
Resulting phase diagram:



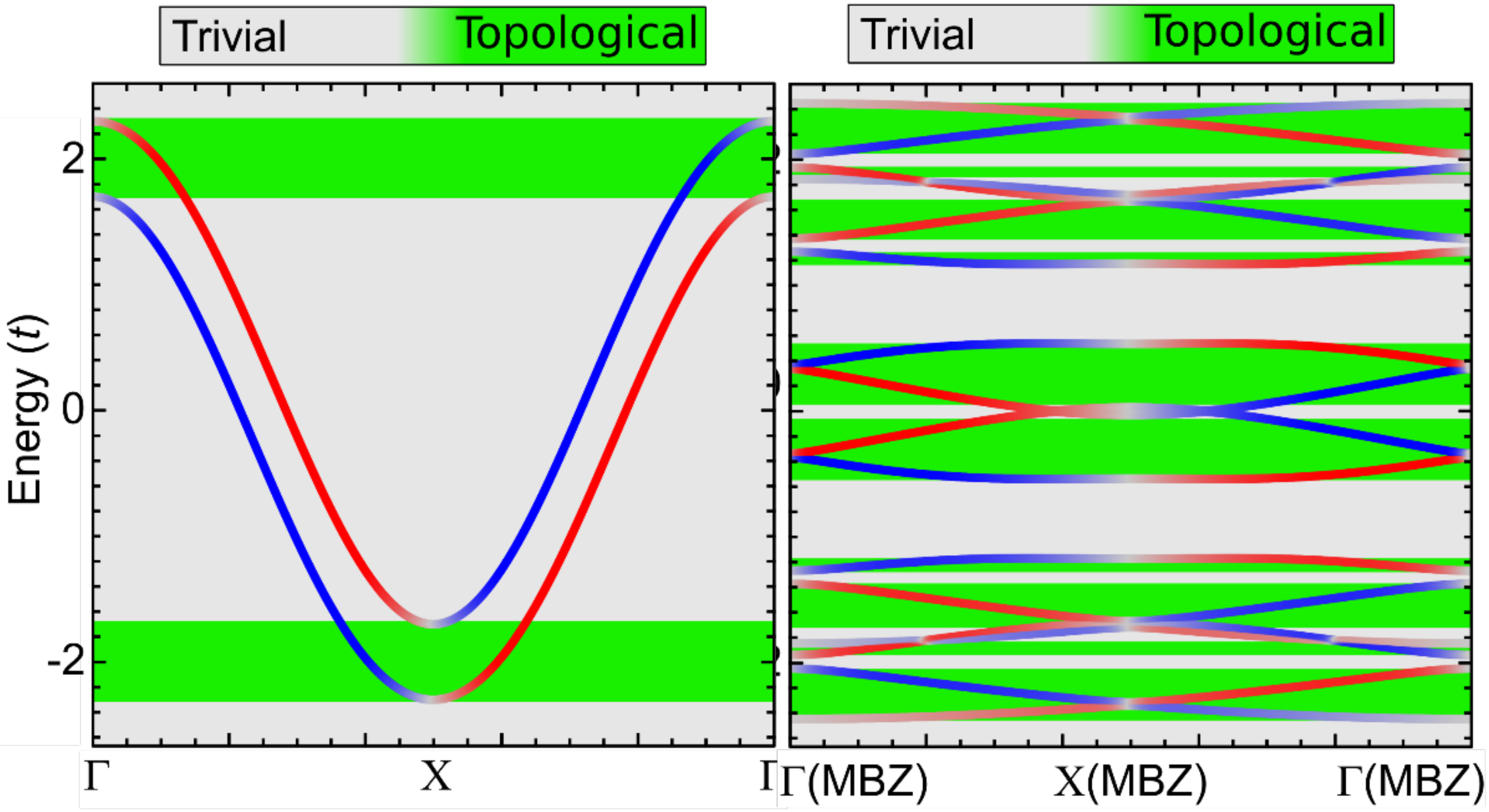
⇒ **The effect of the moiré pattern**



Details: effect of the moiré pattern



- Moiré modulates the electrostatic potential, exchange coupling, superconducting pairing,...
- Folding the band structure
- Creates more regions with pseudo-helical states
  ⇒ topological superconductivity



⇒ Also happens in a realistic model: topological phase emerges at charge neutrality

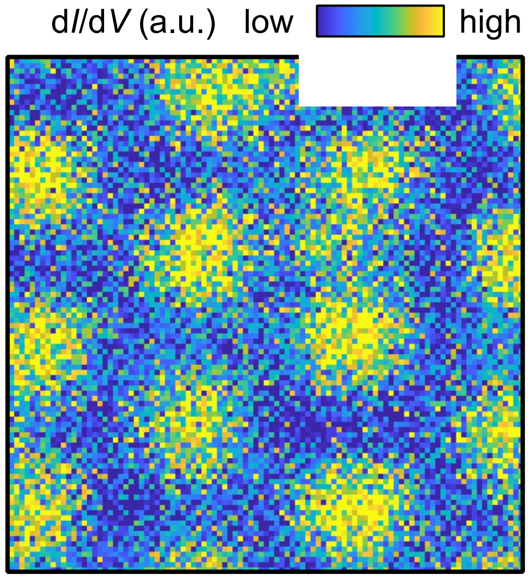
Kezilebieke et al. Moiré-enabled topological superconductivity in a vdW heterostructure, Nano Lett. 22, 328 (2022)

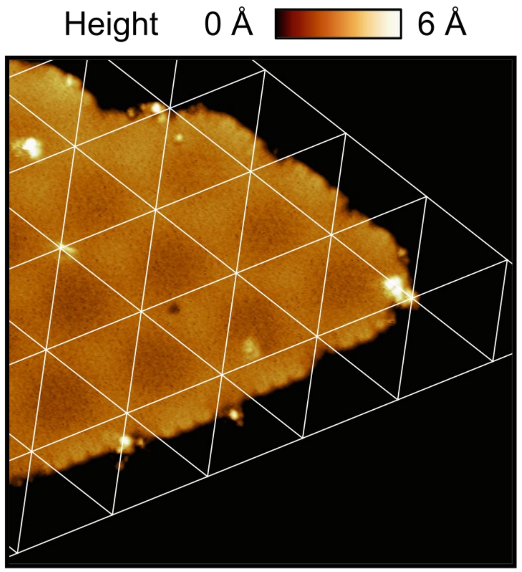
See the effect of the moiré pattern

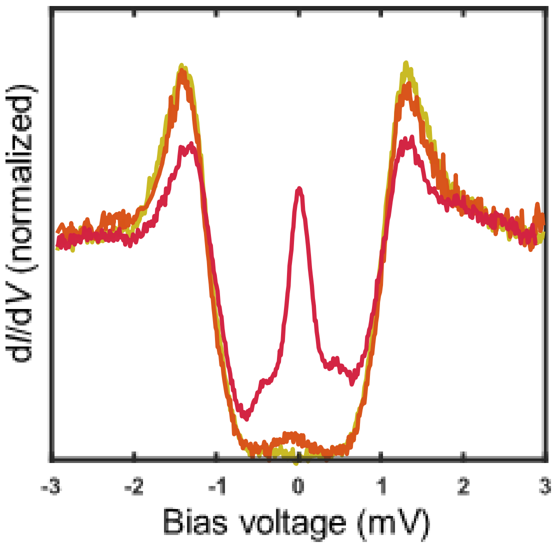
Conclusions / outlook:

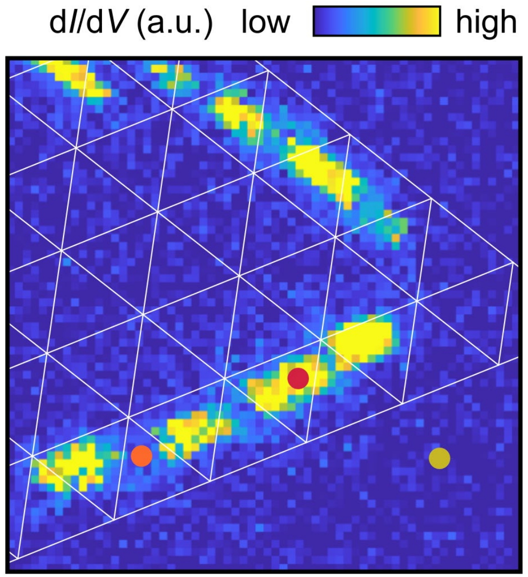
- Moiré-enabled topological superconductivity
- Other experimental probes / channels for the detection / confirmation of the Majorana modes
- Moiré engineering, other magnetic materials etc.
- Definite proof of Majorana modes still lacking – quantum statistics

Shiba band LDOS





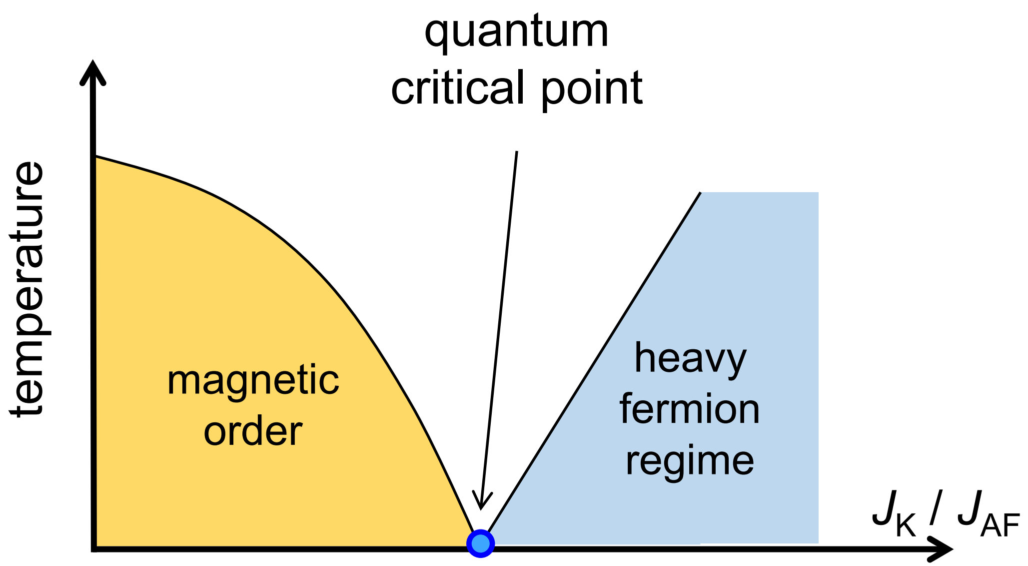




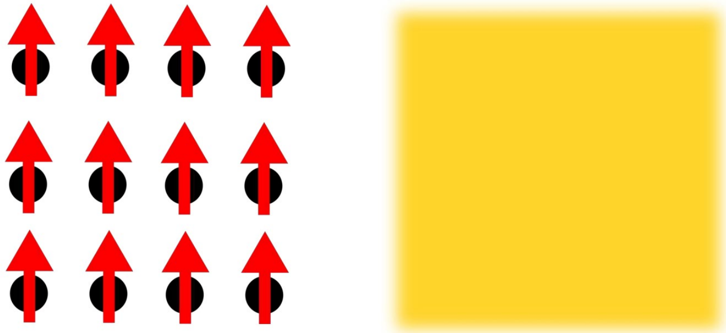
Edge mode LDOS

What else can you do? Artificial heavy fermions

- Heavy fermions:
  - Strongly correlated matter (Kondo lattice physics)
  - Quantum criticality and unconventional superconductivity
  - Found in rare-earth compounds
- Relevant parameters:
  - Antiferromagnetic coupling between the localized moments: $\boldsymbol{J_{AF}}$
  - Exchange coupling between the localized moments and itinerant electrons: $\boldsymbol{J_K}$
  - Ratio J_K / J_{AF} controls the phase diagram



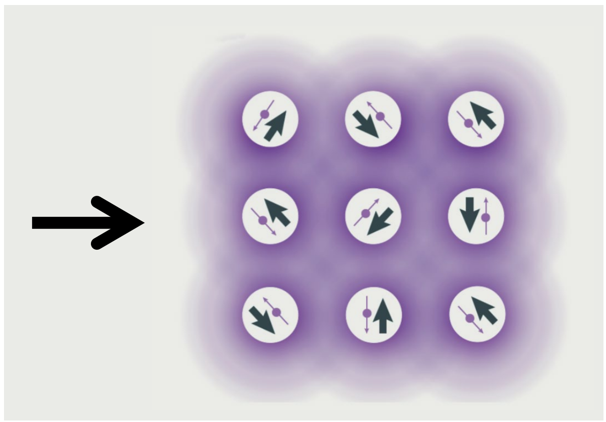
Lattice of Kondo impurities
(f-electrons in rare-earth compounds)



Dispersive electron gas

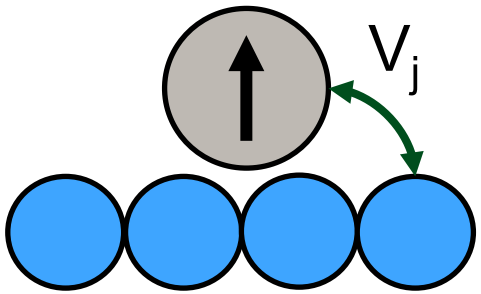
s/p/d-electrons in rare-earth compounds

Formation of a Kondo lattice

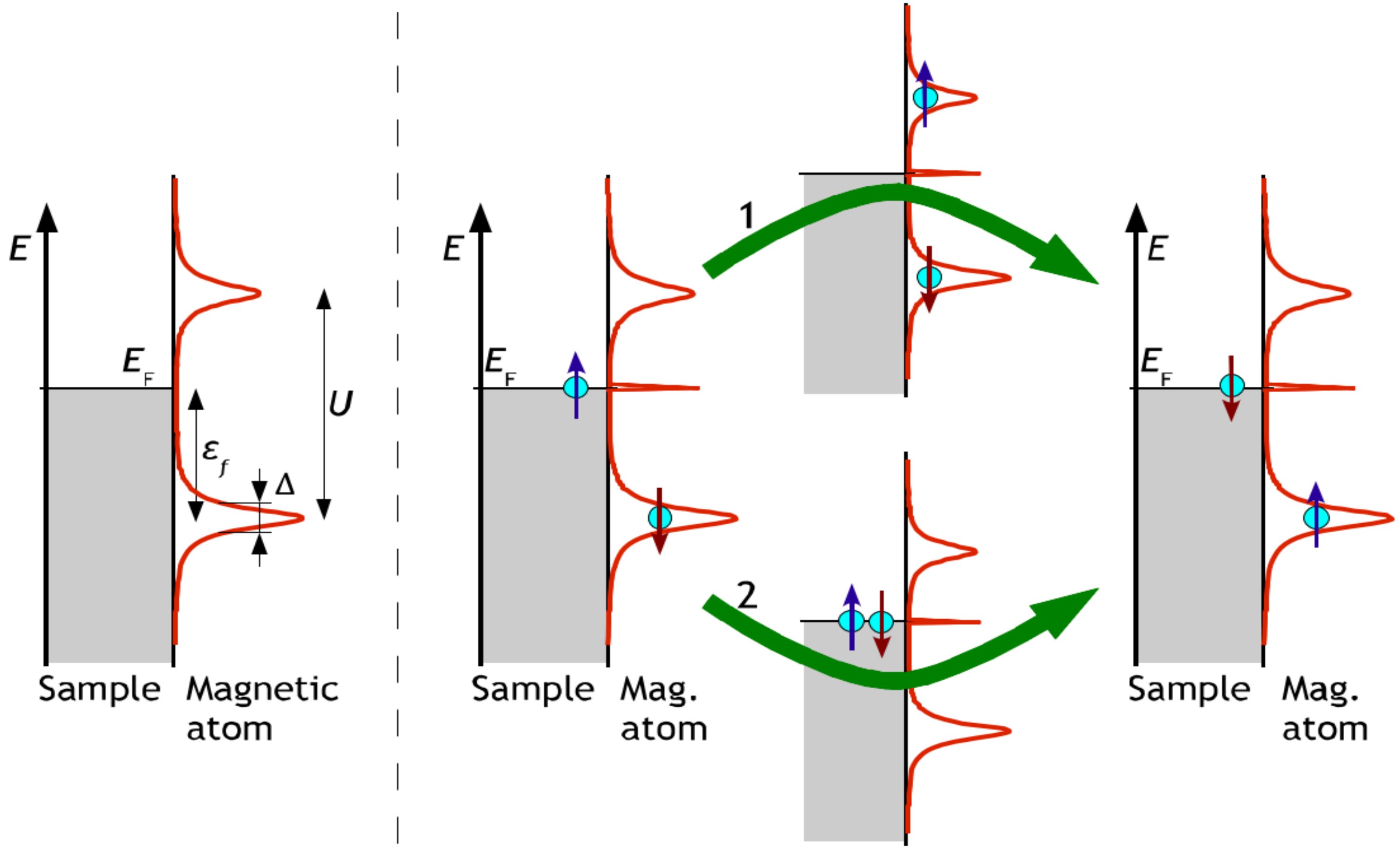


S. Wirth, F. Steglich, Exploring heavy fermions from macroscopic to microscopic length scales. *Nat. Rev. Mater.* **1**, 16051 (2016)

Kondo effect

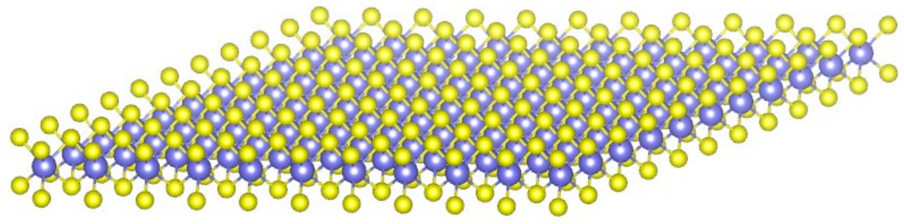


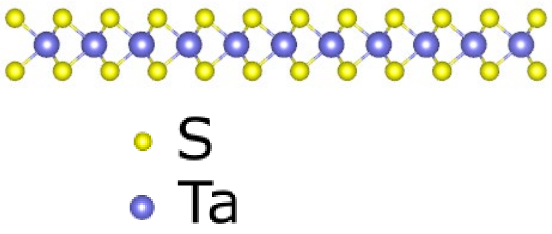
Magnetic impurity on a metal



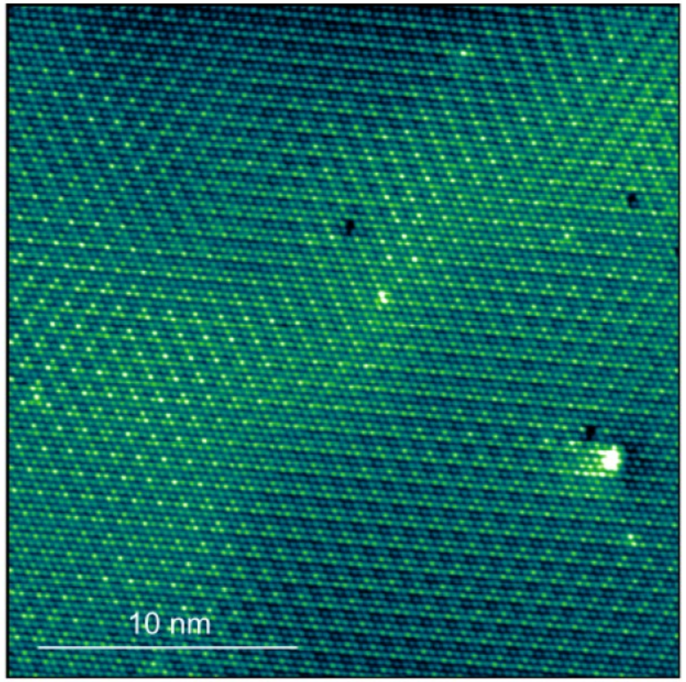
- Kondo effect can be understood within the Anderson impurity model
- The Kondo effect is a higher order scattering process that happens through virtual states (Ternes *et al J. Phys.: Condens. Matter* **21** 053001 (2008))

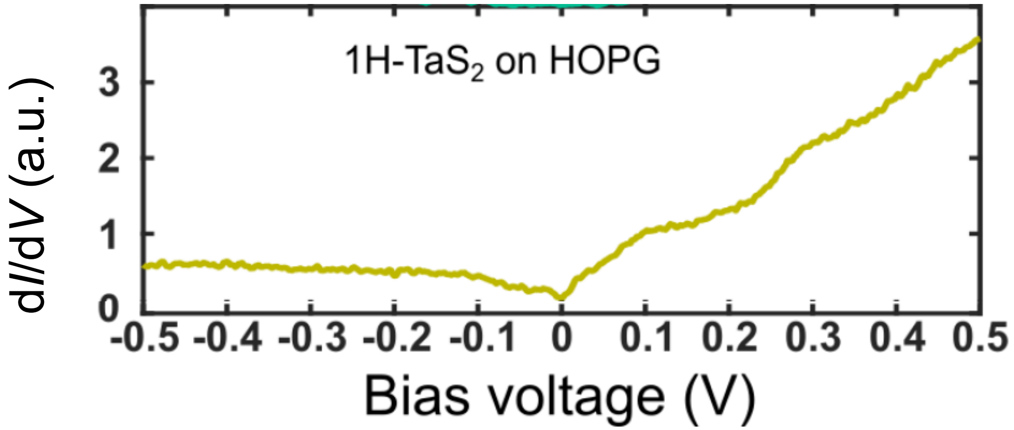
Itinerant electrons: 1H-TaS_2





- Growth via MBE
  - Ta using e-beam evaporation
  - S from FeS_2 from a Knudsen cell
- Metallic behaviour
  - Similar to 1H-$NbSe_2$, see e.g. de la Barrera *et al. Nat Commun* **9,** 1427 (2018).
  - Charge density wave reconstruction
  - Superconductivity at low temperatures

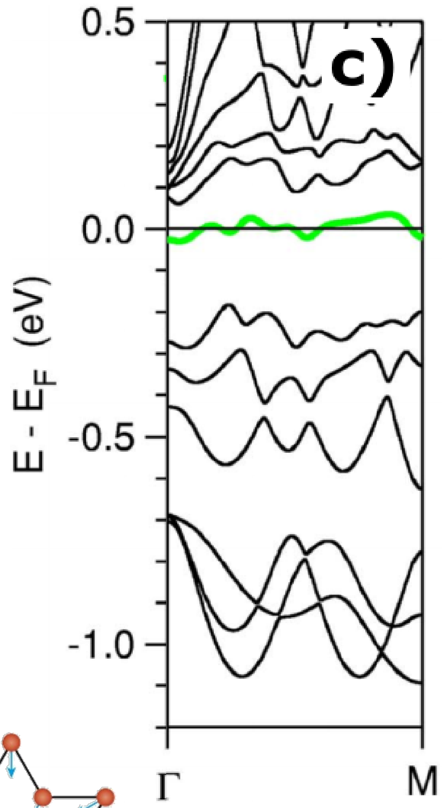


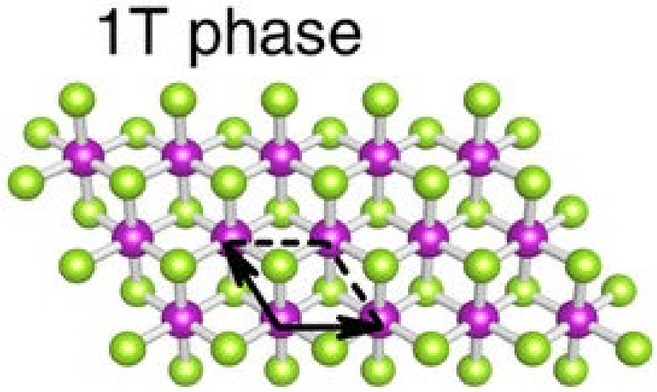


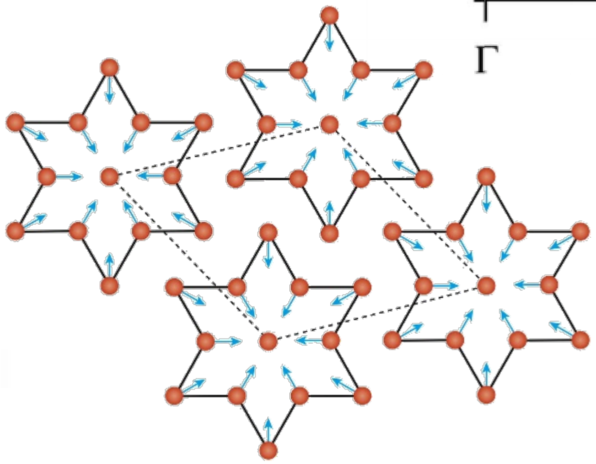
V. Vaňo, S.C. Ganguli et al. Evidence of nodal superconductivity in monolayer 1H-TaS_2 with hidden order fluctuations, Adv. Mater. 35, 2305409 (2023).

Localized moments: 1T-TaS_2

- CDW state in 1T-TaS_2 has one localized unpaired electron per CDW unit cell
  - Mott insulator
- Growth via MBE





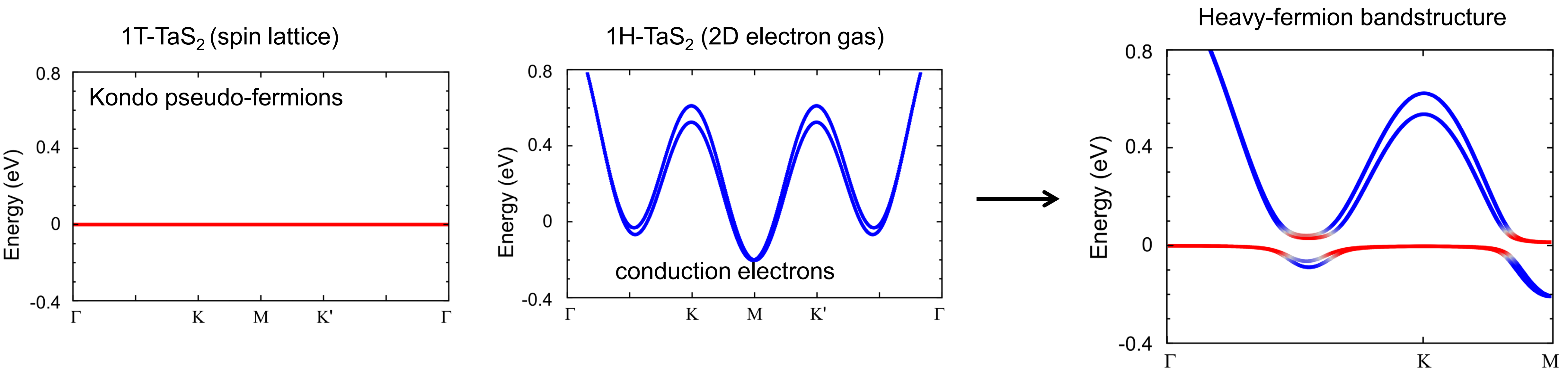


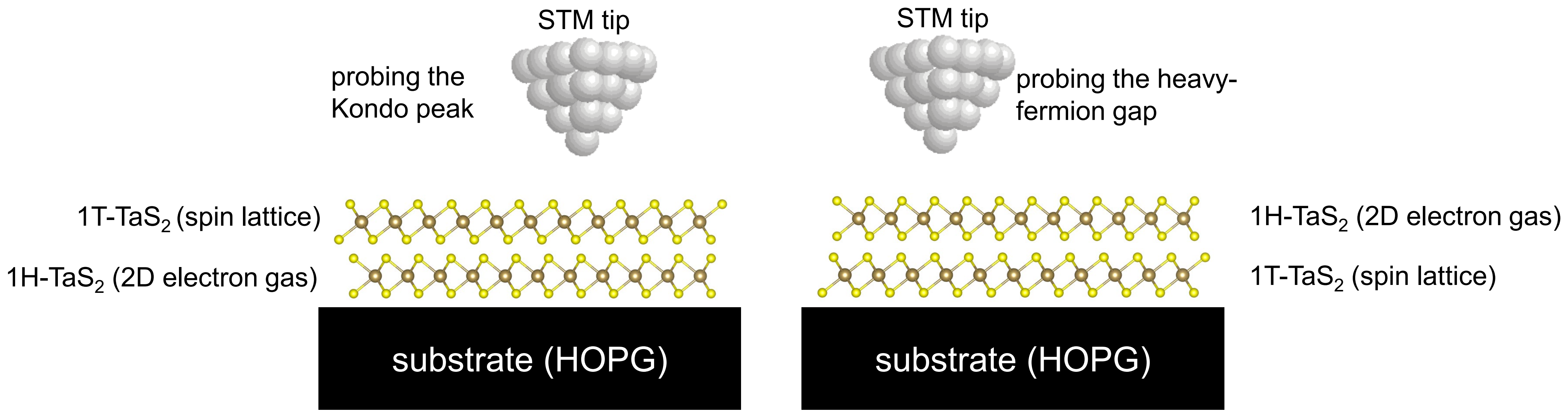
1T-TaS_2: K. Rossnagel, N. V. Smith, Phys. Rev. B 73, 073106 (2006);
K.T. Law, P.A. Lee, PNAS 114. 6996 (2017)

1T-$TaSe_2$: Y. Chen et al. Nat. Phys. 16, 218 (2020).
W. Ruan et al. Nat. Phys. 17, 1154 (2021)

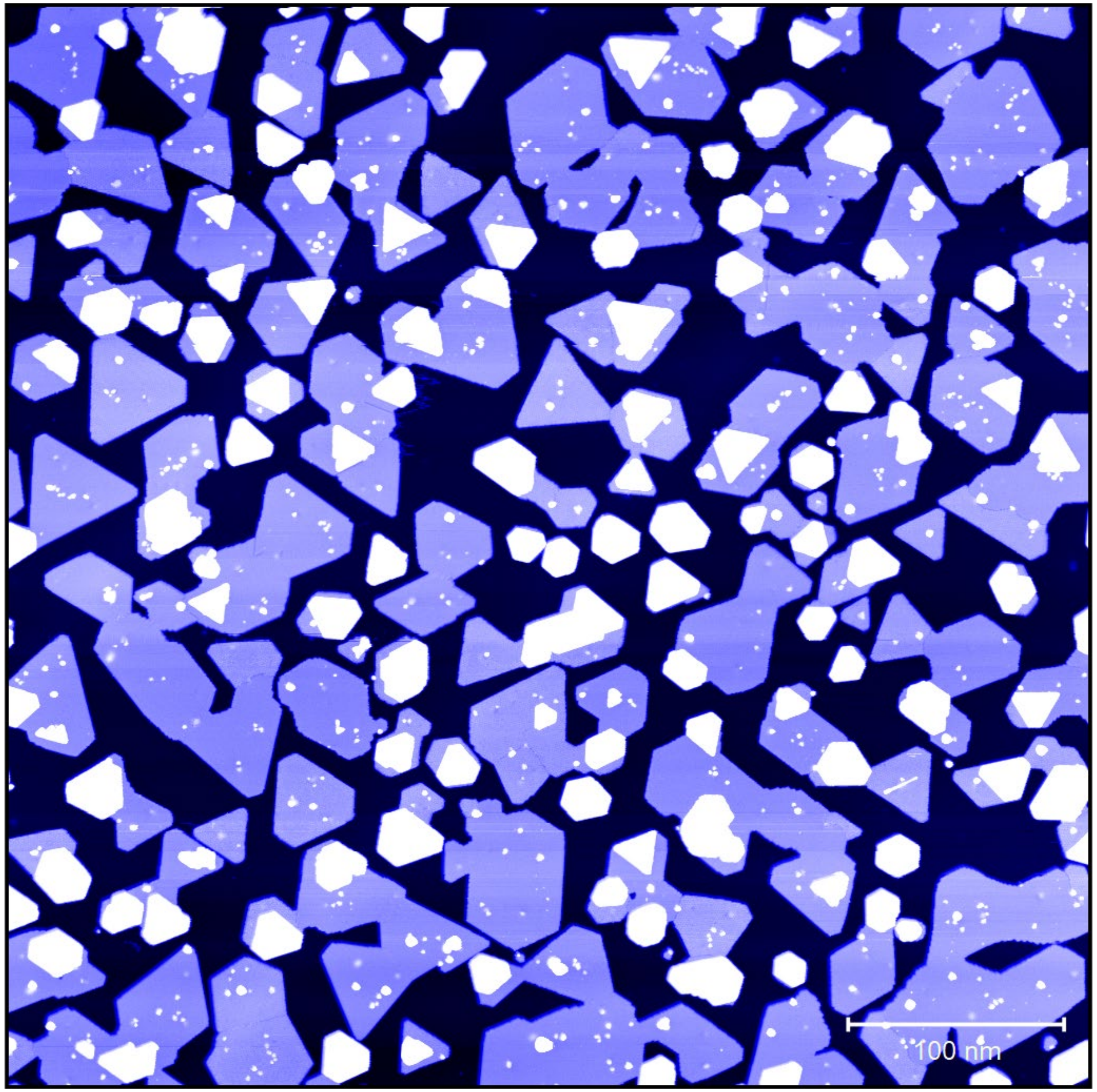
Growth: J. Hall, 2D Mater. 5, 025005 (2018).
H. Lin et al. Nano Res. 11, 4722–4727 (2018).

Putting it together



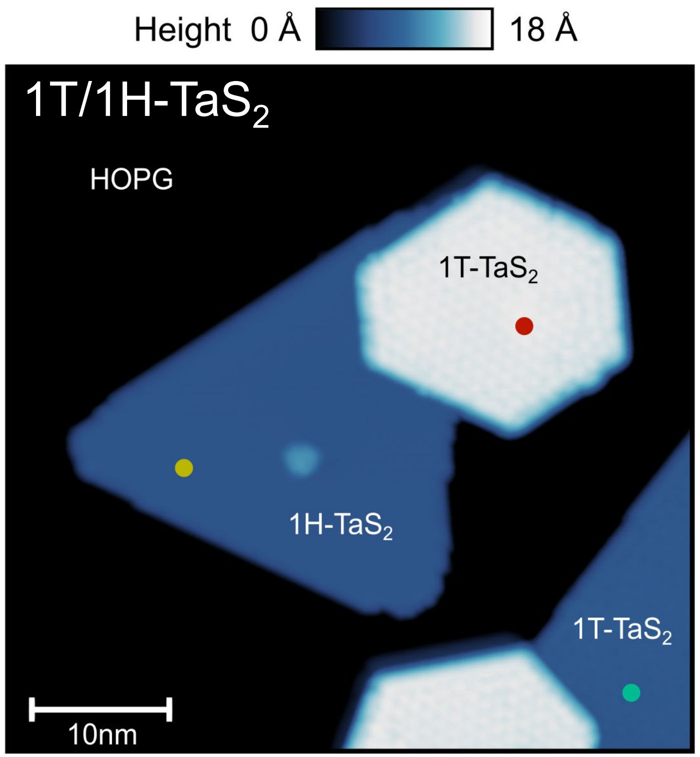


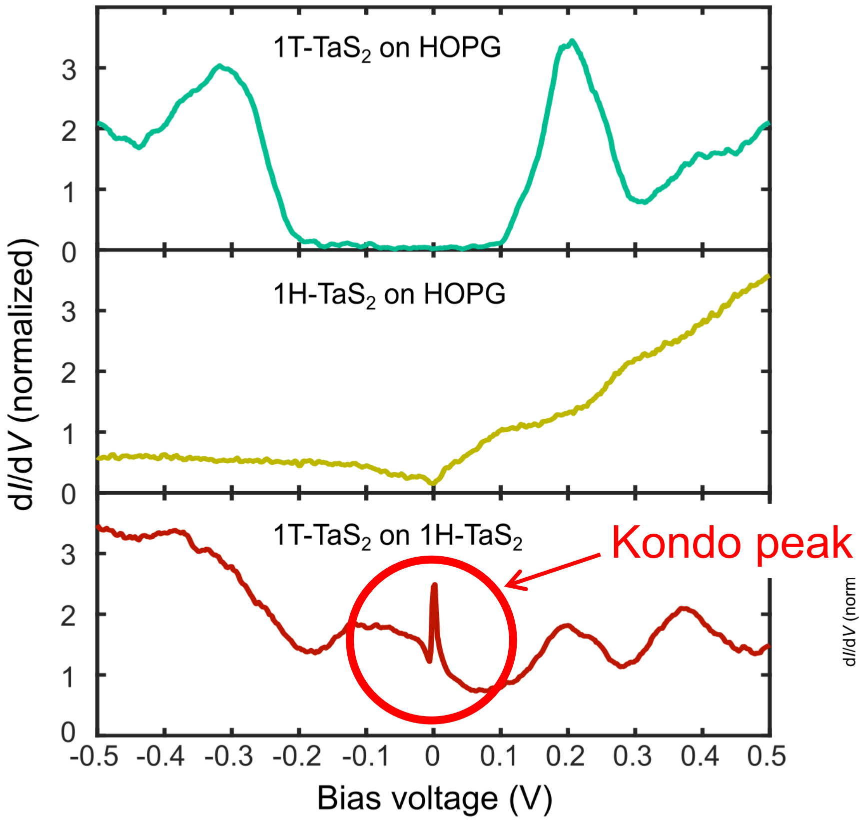
Preparing the heterostructures



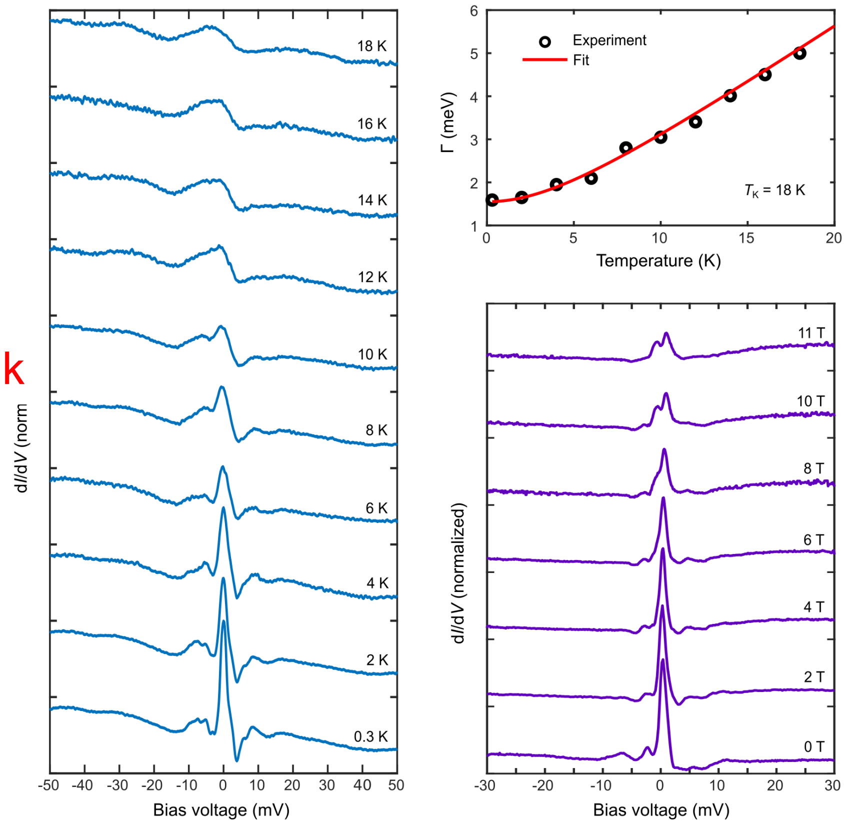
- Ta using e-beam evaporation, S from FeS_2 from a Knudsen cell
  - See e.g. J. Hall, …, T. Michely, 2D Mater. 5, 025005 (2018) for the use of FeS_2
- Growth at ~ 680°C on HOPG
- Sample temperature and coverage control 1T / 1H ratio
  - Lower growth temperature ⇒ 1H
  - Higher temperature ⇒ 1T
  - Higher coverage ⇒ 1H (perhaps during sample cooling)
  - Ref: H. Lin, W. Huang, K. Zhao, C. Lian, W. Duan, X. Chen, S.-H. Ji, Nano Res. 11, 4722 (2018)

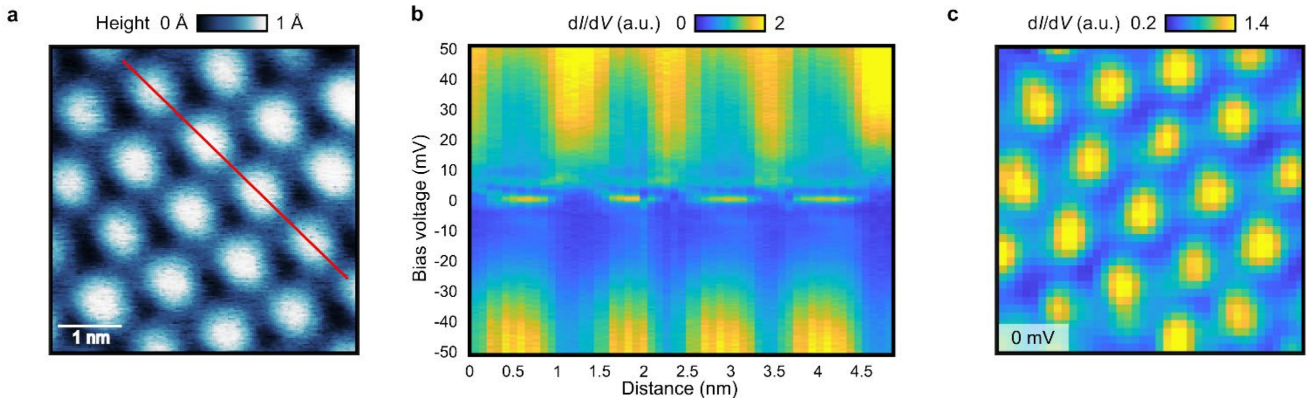
Experimental realization: Kondo



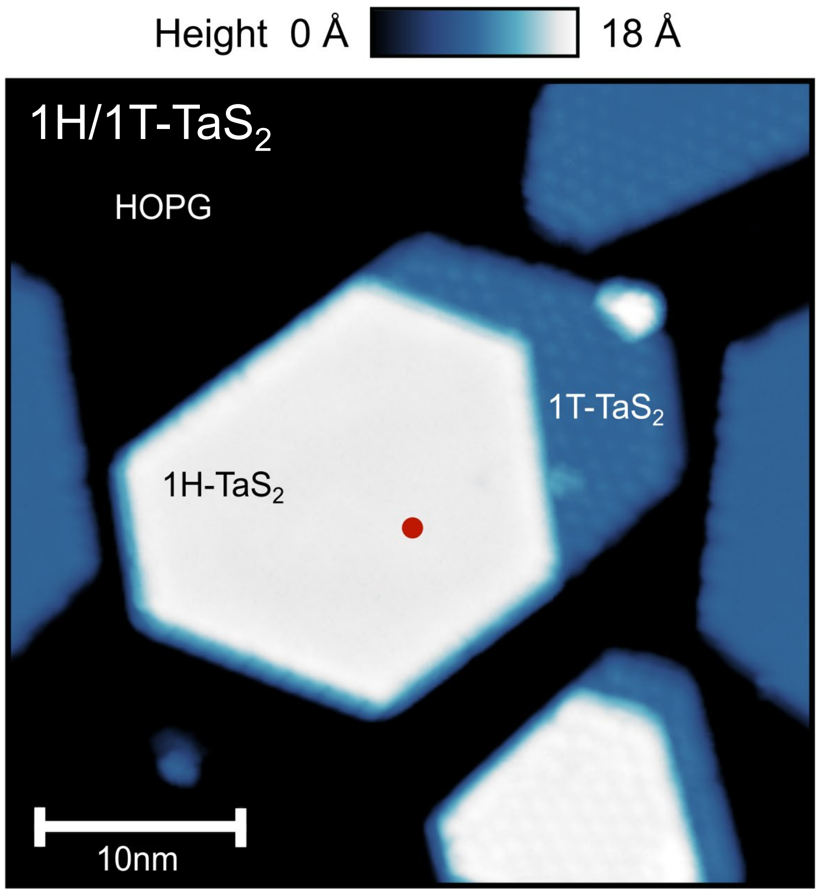


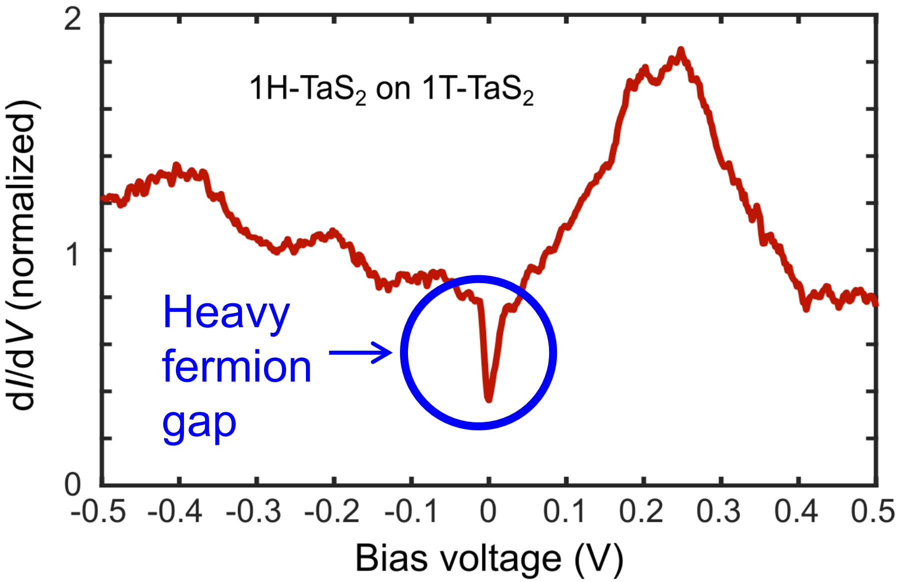
Temperature and magnetic field dependence



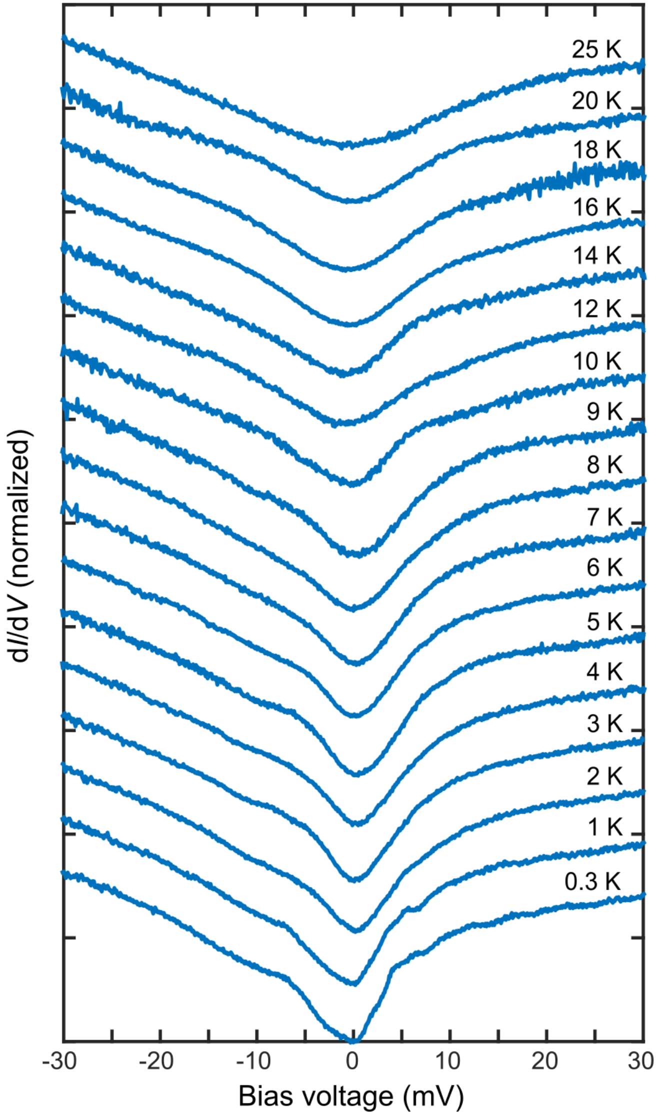


Experimental realization: heavy fermion gap





Temperature dependence

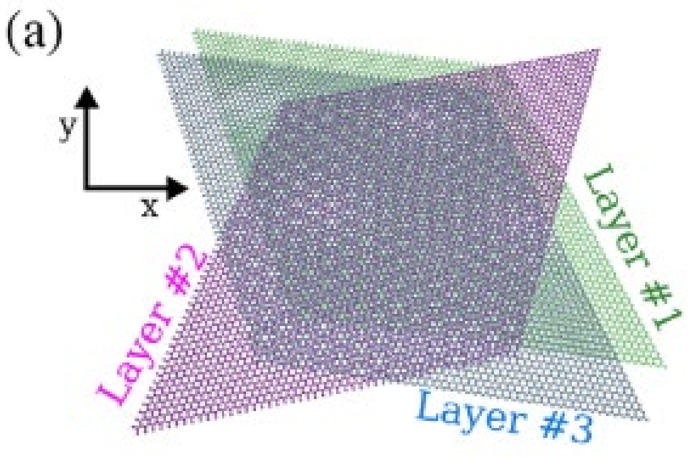


Ruled out:

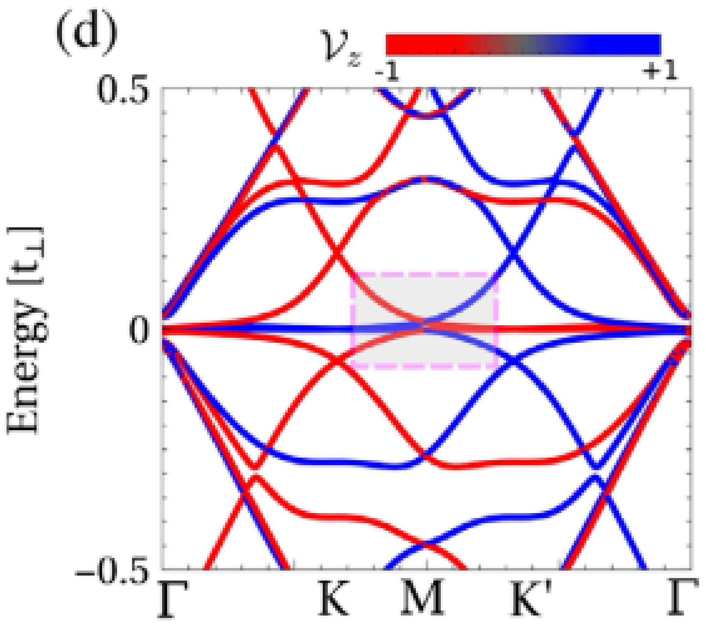
1) CDW gap, 2) SC gap, 3) Coulomb gap

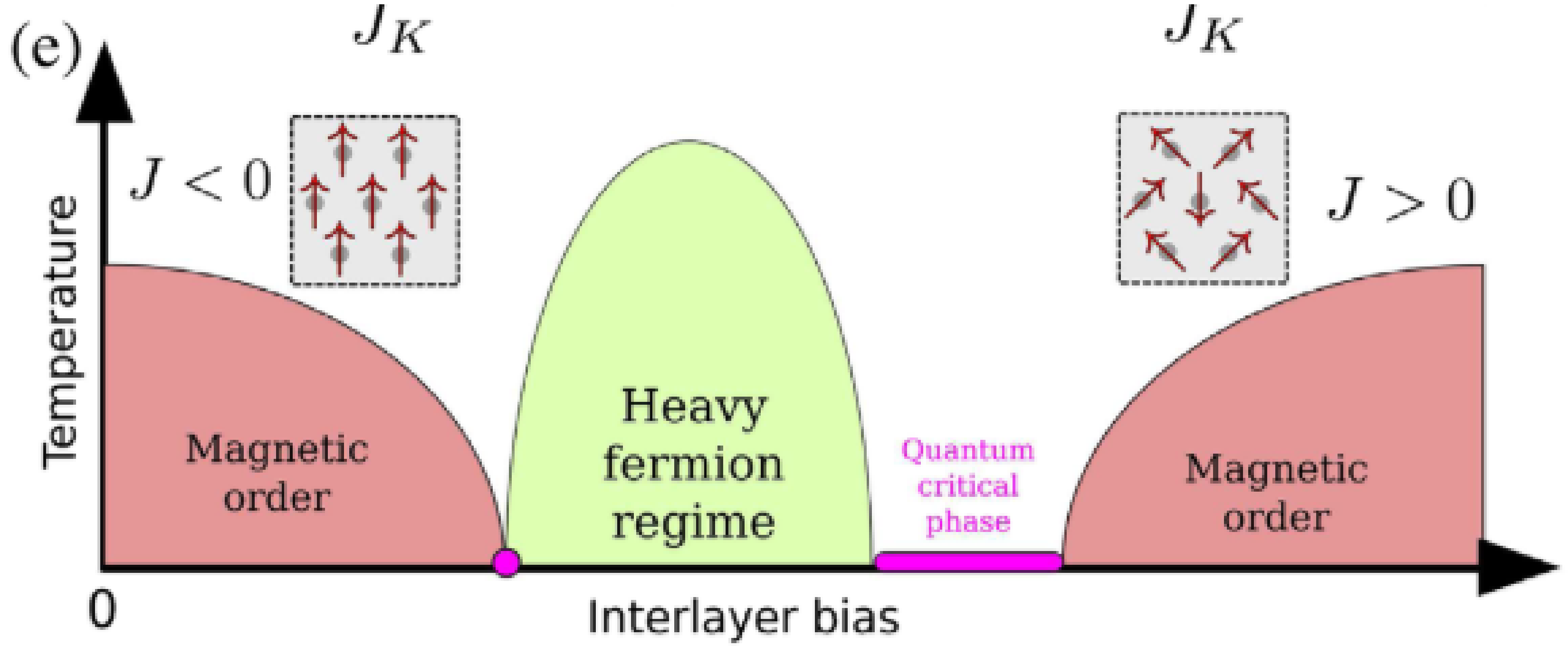
V. Vaňo et al. Artificial heavy fermions in a van der Waals heterostructure, Nature 599, 582 (2021)

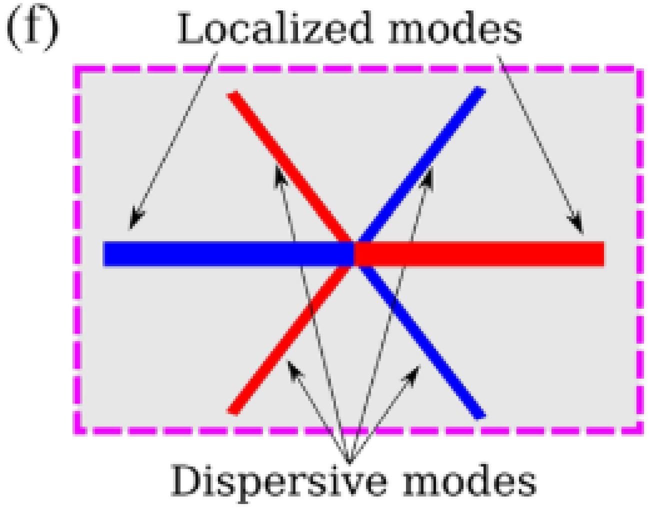
Graphene multilayers as a heavy fermion system



Aline Ramires, Jose L. Lado, Emulating heavy fermions in twisted trilayer graphene, Phys. Rev. Lett. 127, 026401 (2021)

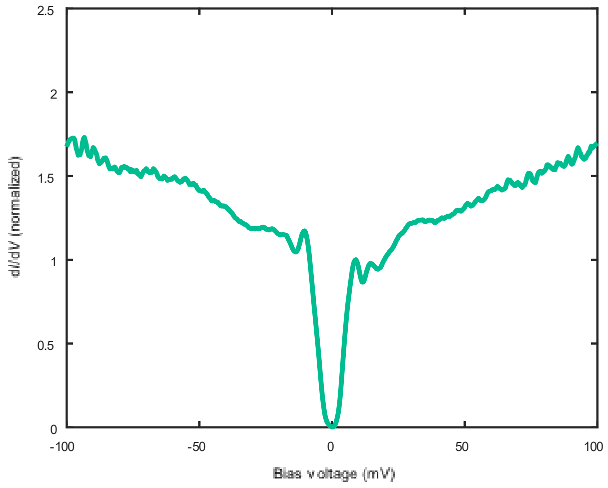


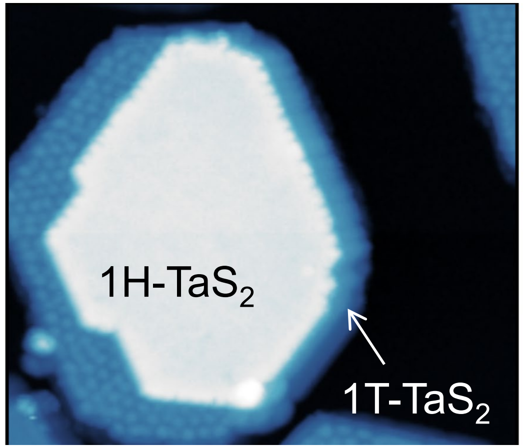


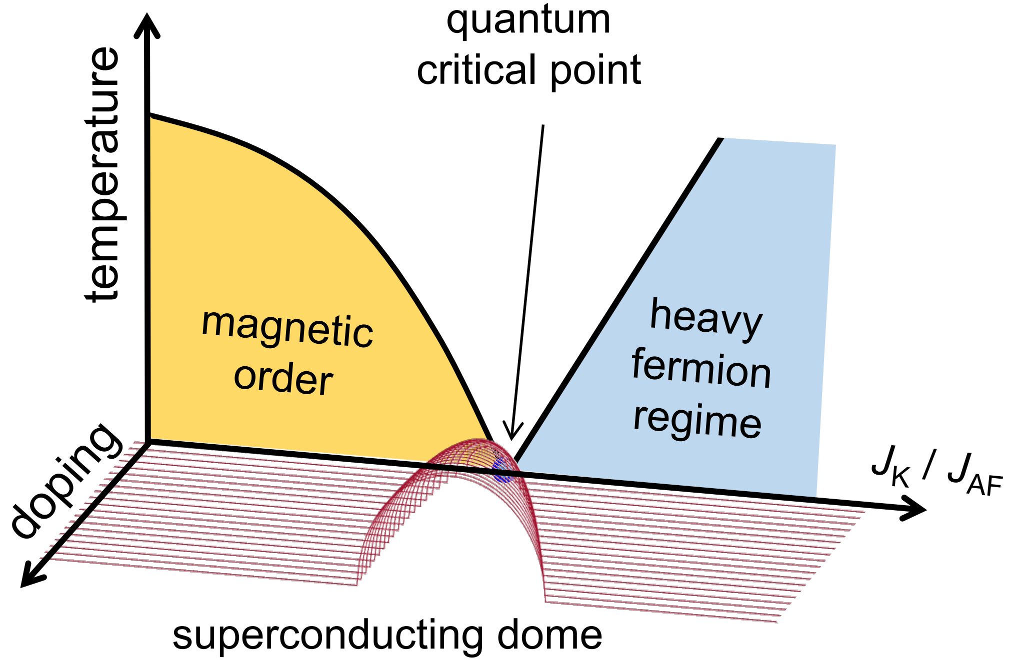


What next?

- Demostrated artificial heavy fermion behaviour in a vdW heterostructure
  - V. Vaňo et al. Artificial heavy fermions in a van der Waals heterostructure, Nature 599, 582 (2021)
- Can we play with the phase diagram?







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Evidence for ground state coherence in a two-dimensional Kondo lattice

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DFT (Aalto): Orlando Silveira, Adam Foster