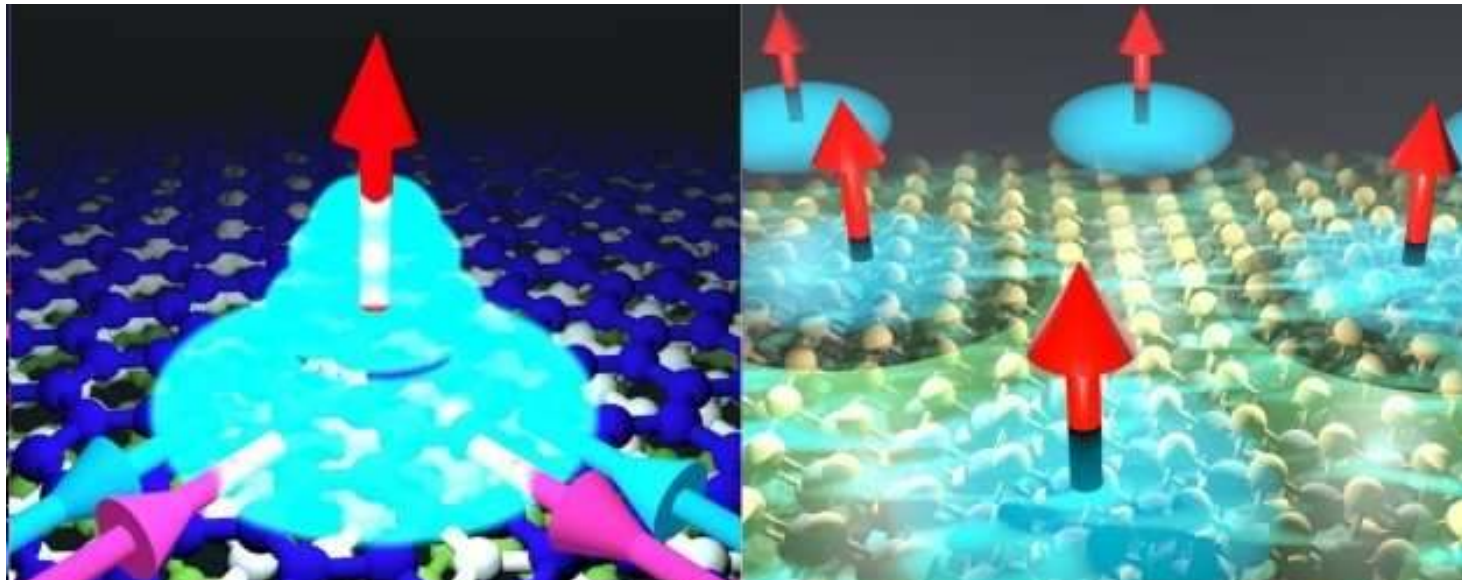


Engineering artificial heavy-fermion quantum matter in twisted van der Waals materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



Phys. Rev. Lett. 127, 026401 (2021)

Nature 599, 582–586 (2021)

Correlations in Topological Quantum Matter, Lammi, Finland
September 7th 2022

Complexity, universality and emergence

Psychology

Medicine

Biology

Chemistry

Physics

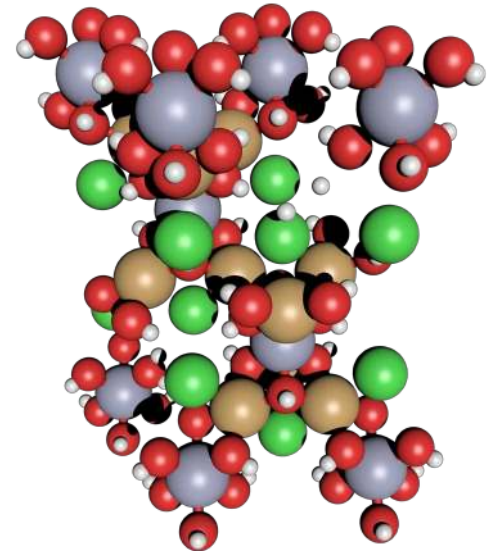
Complexity

Fish schools



Phys. Rev. Lett. 120, 198101 (2018)

Complex compounds



Complex systems allow to have new phenomena that did not exist before

Exotic physics in quantum materials

Quantum spin liquids



Unconventional superconductivity



Topological matter



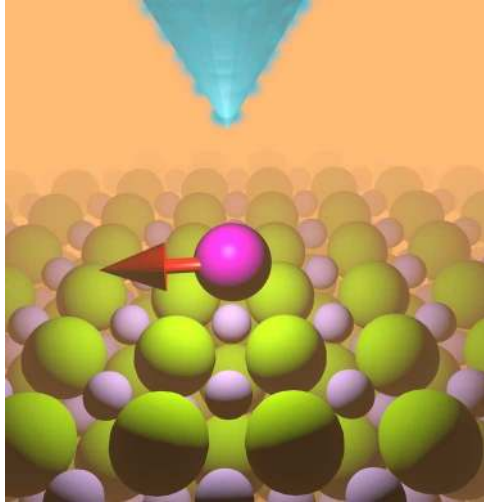
Topological quantum computing

Low consumption electronics

New physics not yet realized in nature (fractional quantum particles)

Building quantum matter with artificial lattices

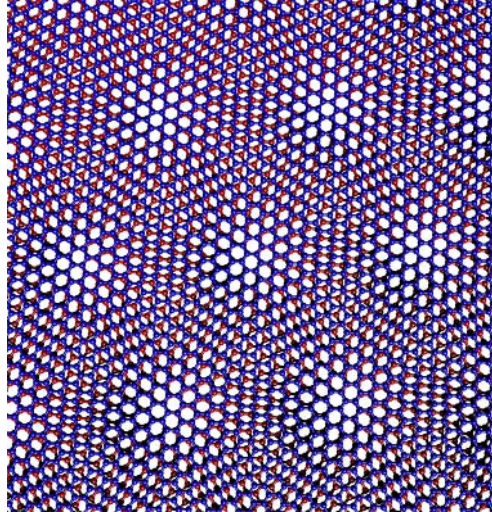
Atomic lattices



$a = 0.5 \text{ nm}$

Nature Physics 12 (7), 656 (2016)
Nature Physics 13 (7) 668 (2017)
Science 335 (6065), 196-199 (2012)

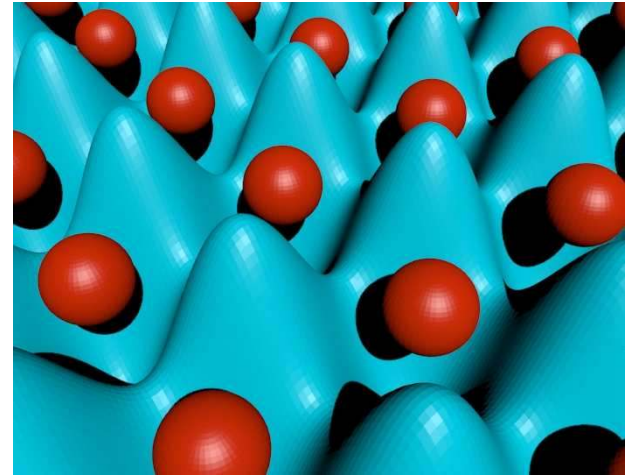
Twisted 2D moire materials



$a = 30 \text{ nm}$

Phys. Rev. Lett. 99, 256802 (2007)
Nature Physics 6, 109–113 (2010)
PNAS 108 (30) 12233-12237 (2011)
Nature 556, 80–84 (2018)

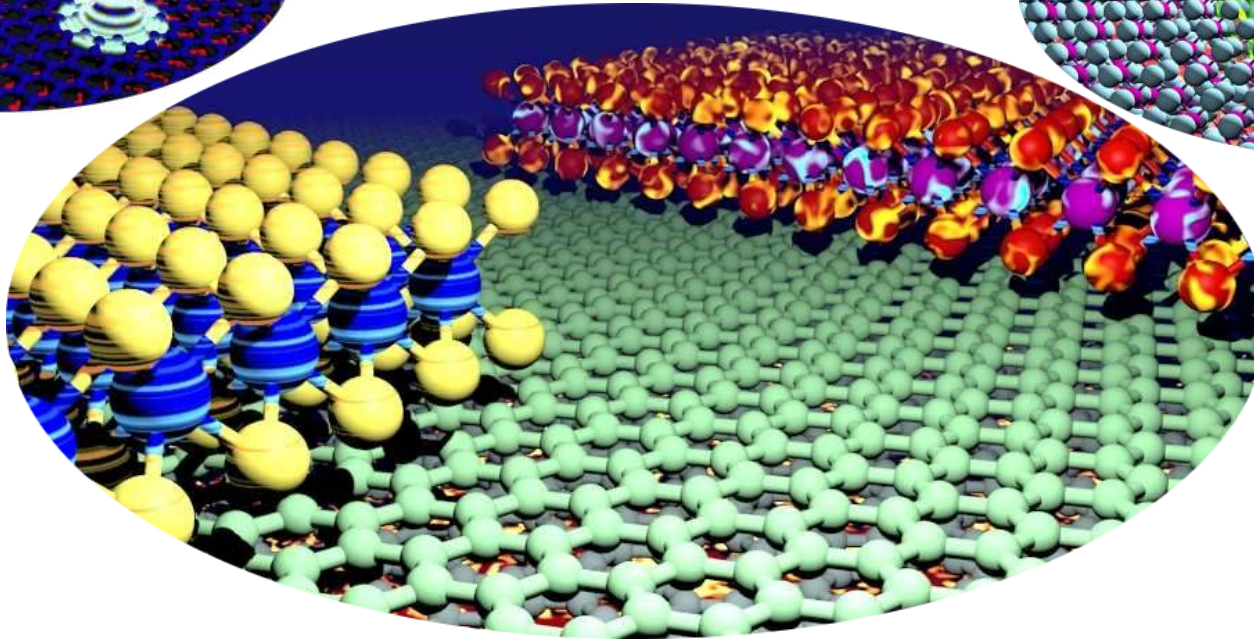
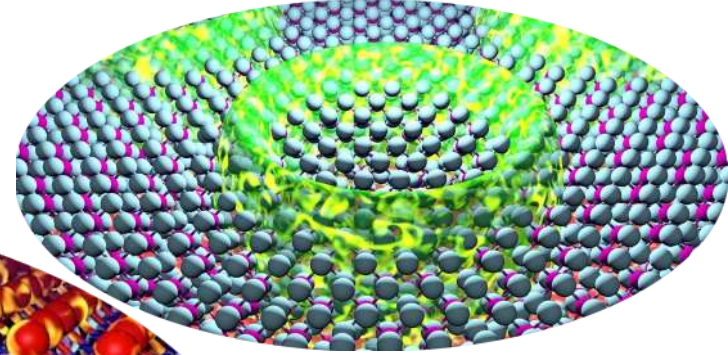
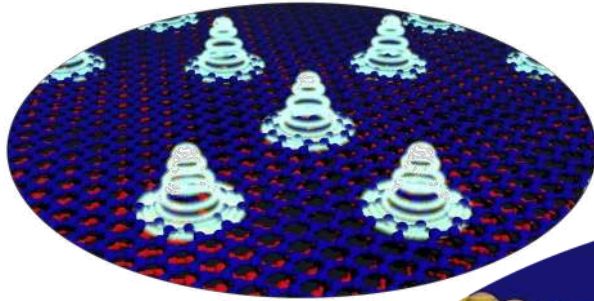
Cold atoms



$a = 1000 \text{ nm}$

Nature 545, 462–466 (2017)
Nature 455, 204–207 (2008)
Phys. Rev. Lett. 111, 185307 (2013)

A new universe in each van der Waals heterostructure

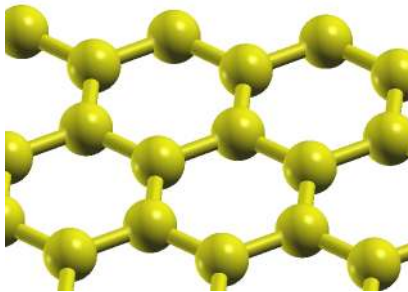


Each van der Waals heterostructure allows creating a whole new universe for electrons

The two-dimensional materials world

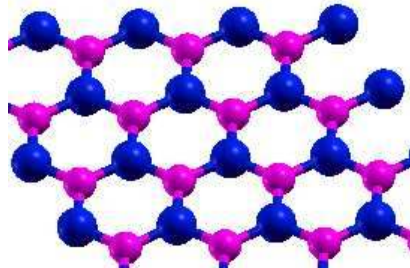
Semimetal

Graphene



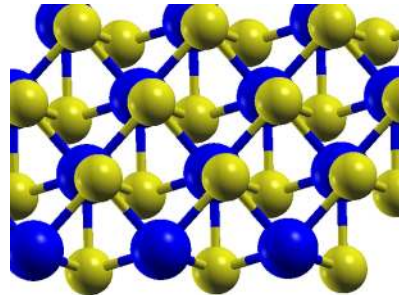
Insulator

BN



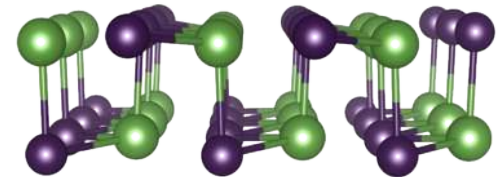
Superconductor

NbSe₂



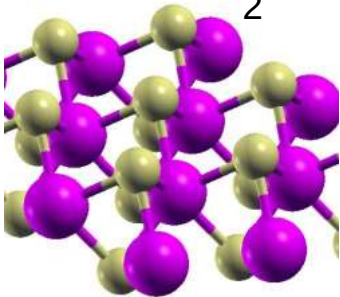
Ferroelectric

SnTe



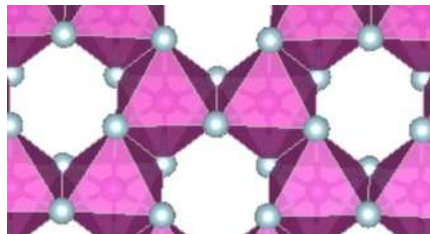
Semiconductor

WSe₂



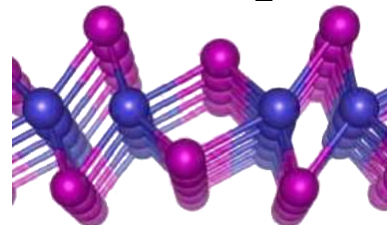
Ferromagnet

CrI₃



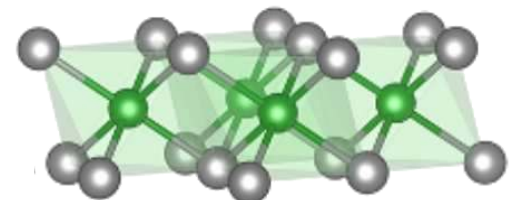
**Quantum spin
Hall insulator**

WTe₂



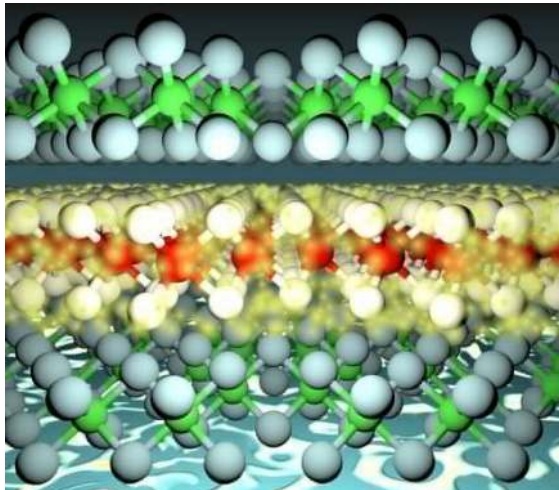
Multiferroic

NiI₂



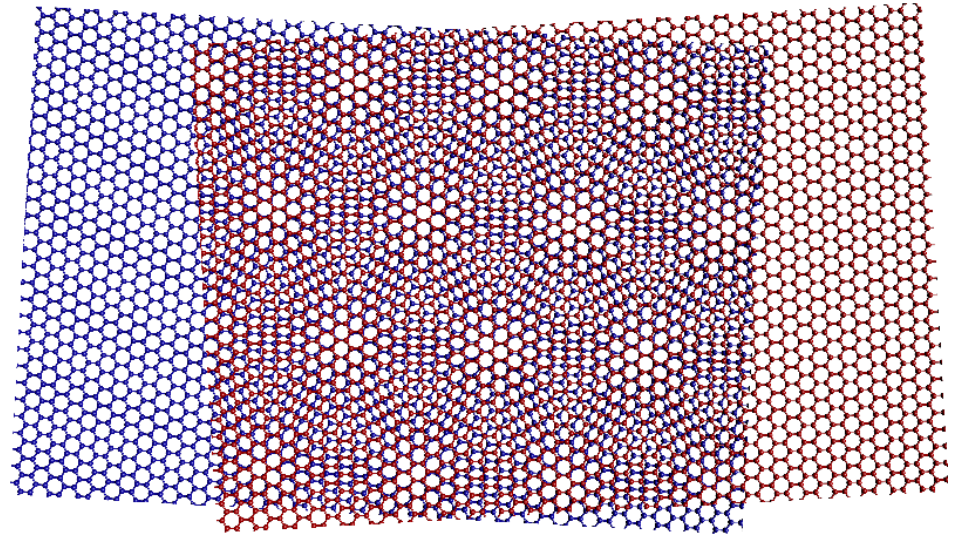
The flexibility of two-dimensional materials

They can be stacked



Nature 499.7459 419. (2013)

They can be rotated

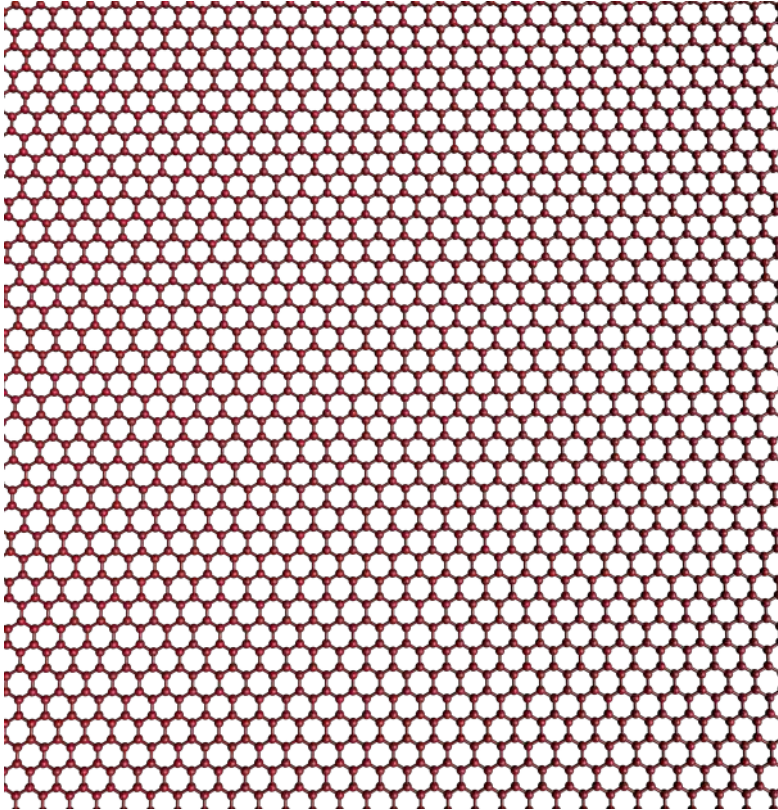


Science 361.6403 690-693. (2018)

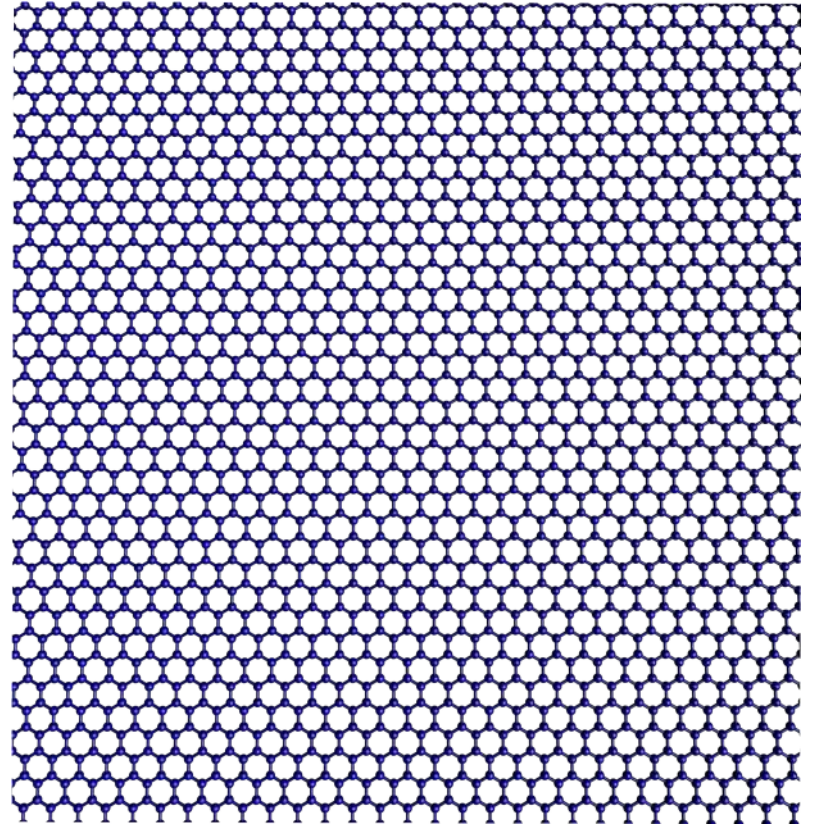
These are unique features of two-dimensional materials

Moire in twisted bilayer graphene

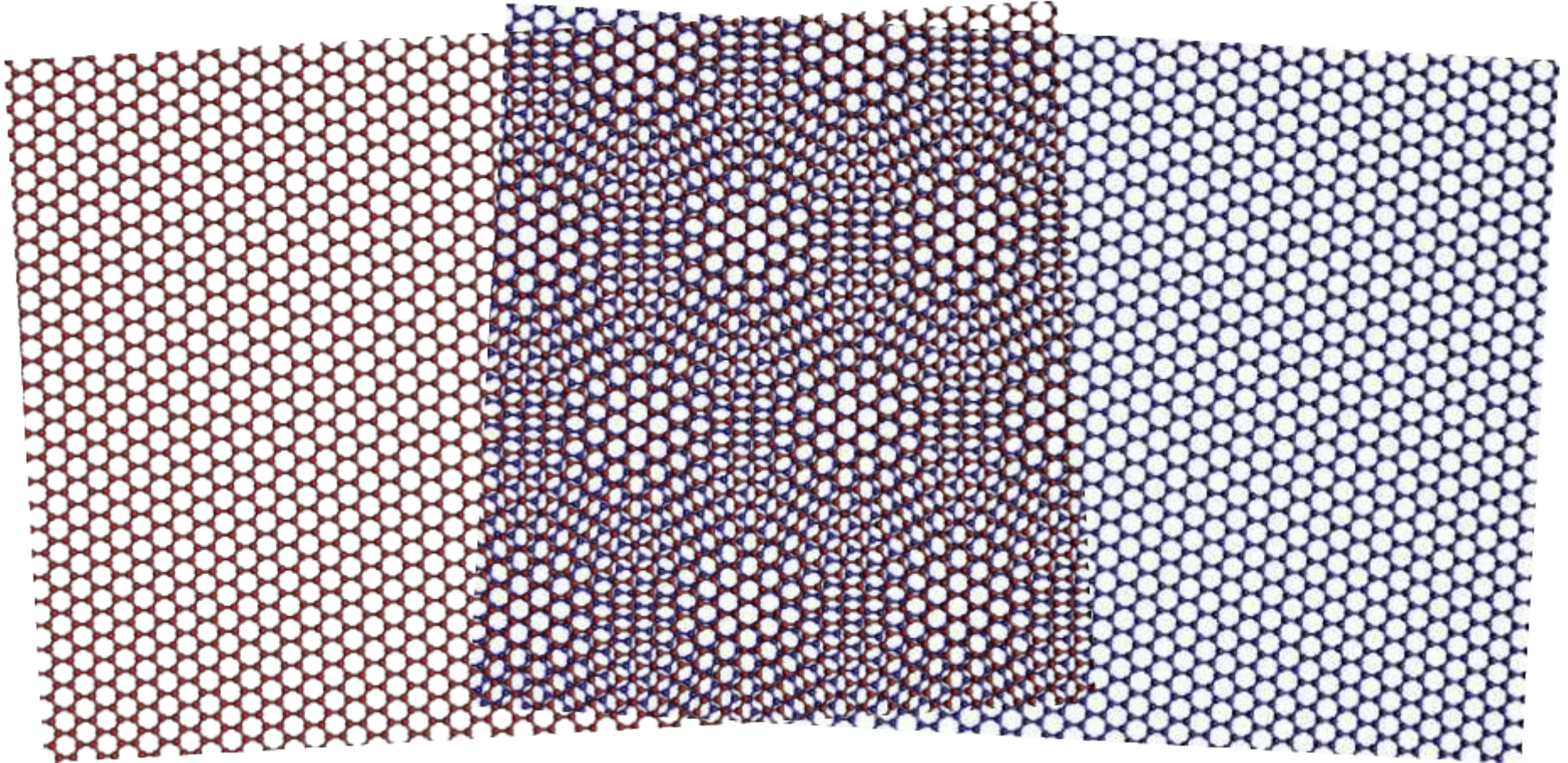
Upper graphene layer



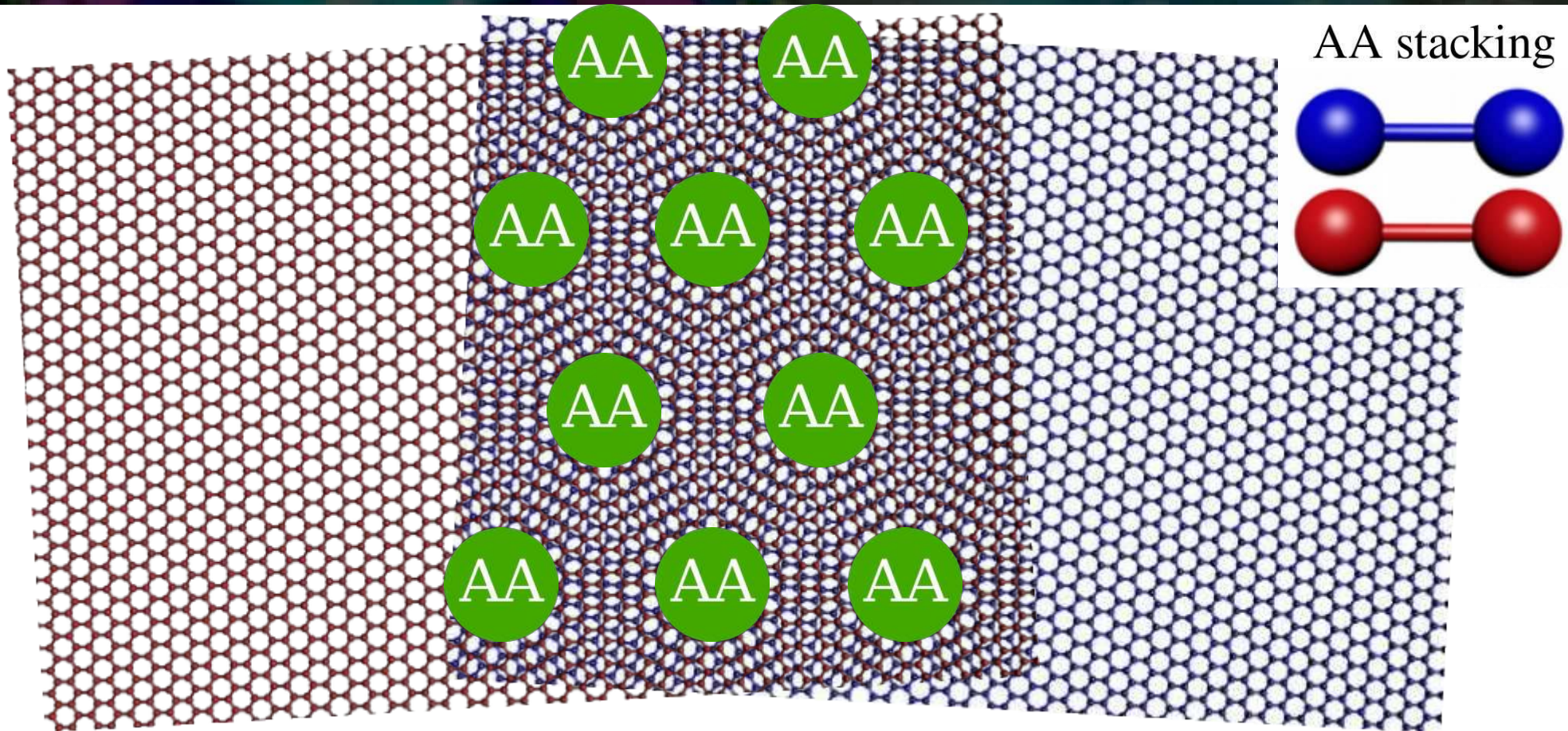
Lower graphene layer



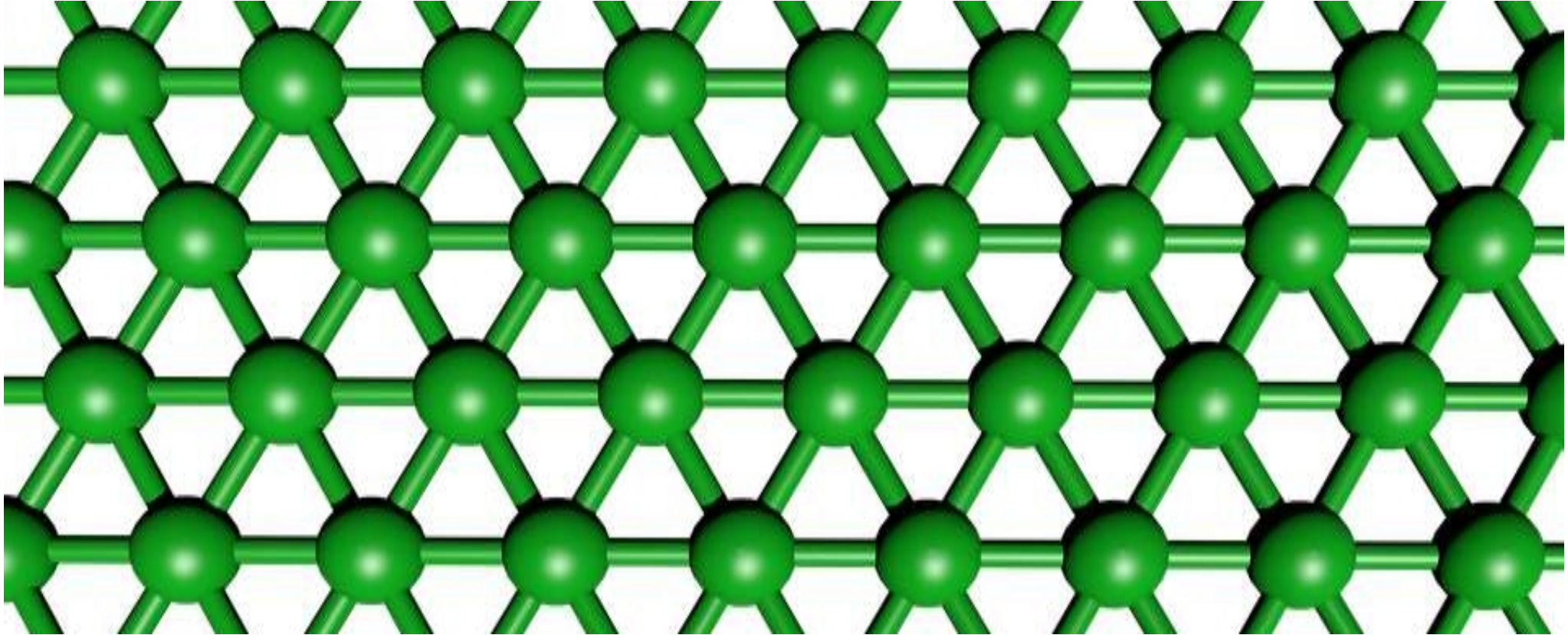
Moire in twisted bilayer graphene



Moire in twisted bilayer graphene

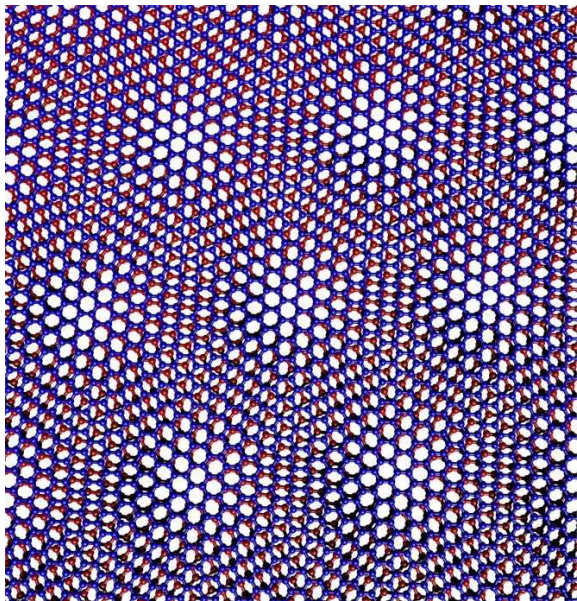


Moire in twisted bilayer graphene

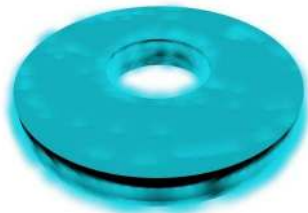


One material, a zoo of electronic phases

Twisted bilayer graphene

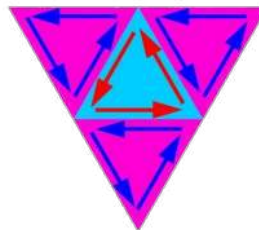


Superconductivity



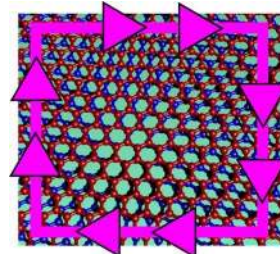
Nature 556, 43–50 (2018)

Topological networks



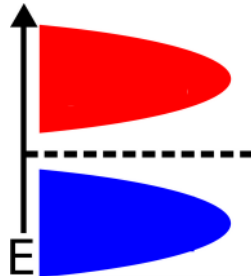
Nano Lett. 18, 11, 6725-6730 (2018)

Chern insulators



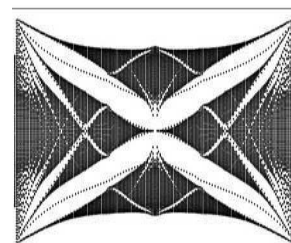
Science 365, 605-608 (2019)

Correlated insulators



Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782-786 (2018)

Fractional Chern insulators



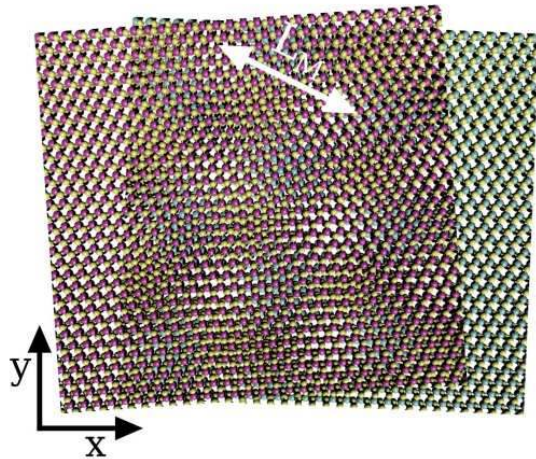
Nature 600, 439–443 (2021)

A bilayer of a van der Waals material realizes a variety of widely different electronic states

The generality of correlated physics in twisted 2D materials

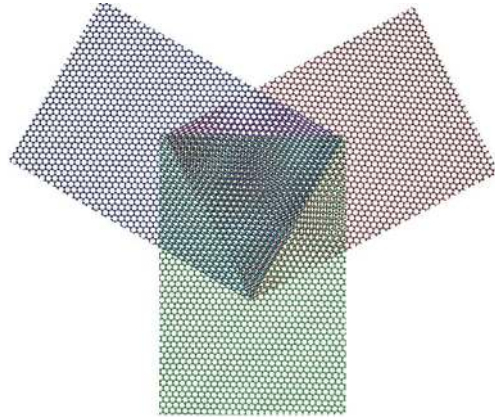
Superconductivity & correlated behavior found also in

Twisted dichalcogenides



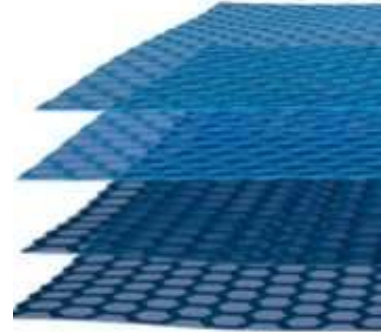
Nature 579, 359–363 (2020)

Twisted graphene trilayers



Nature 590, 249–255 (2021)

Twisted graphene tetralayers



Nature 583, 221–225 (2020)

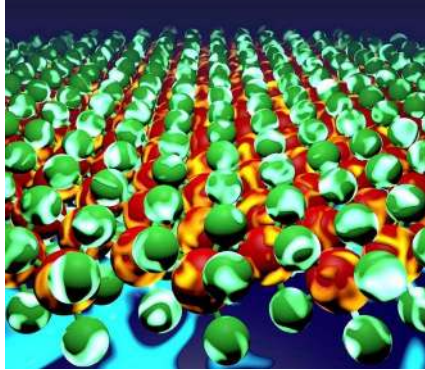
Some of new physics observed were only realized before in rare, heavy or even toxic compounds

Controlling electronic states in van der Waals materials

*Of course, we can use the typical knobs of bulk compounds
(pressure, chemical doping, etc)*

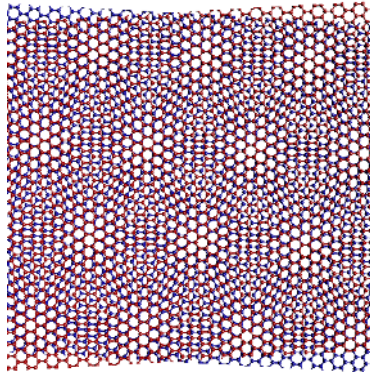
Most importantly, we can exploit the full tunability of van der Waals materials

Gating



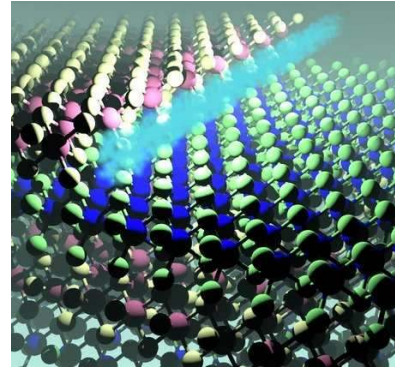
Science 306, 5696, 666-669 (2004)

Twist engineering



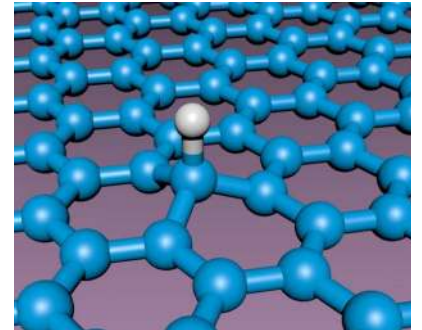
Nat, Rev, Mat, 6, 201–206 (2021)

Materials engineering



Nature 499, 419–425 (2013)

Atomic engineering

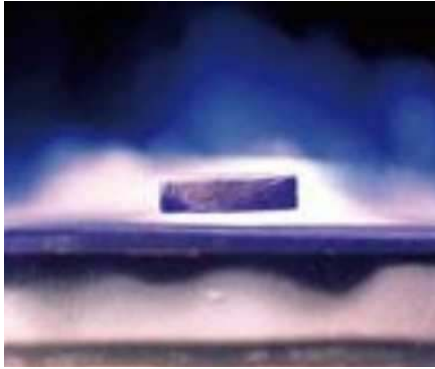


Science, 352(6284), 437-441 (2016)

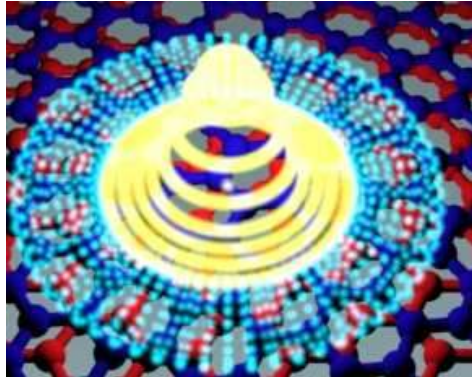
Allowing to independently control different features of the electronic structure

The opportunities of twisted van der Waals materials

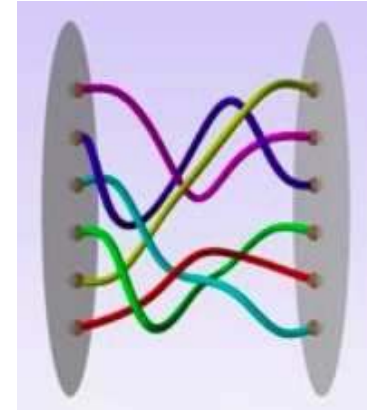
“Unveil” the nature of high T_c superconductivity (?)



Controllable exotic quantum matter (?)



A platform for topological quantum computing (?)



Explore exotic physics with a degree of control unprecedented in any other material

- *Control of single particle and many-body interactions*
- *Explore physics that only appear in rare or toxic compounds*

Designing quantum matter in twisted materials

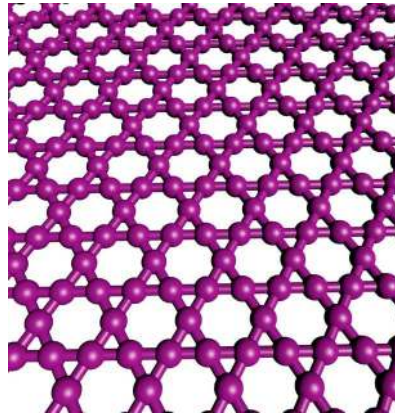
*The strength of
electronic correlations*

$$V_{ijkl}c_i^\dagger c_j c_k^\dagger c_l$$



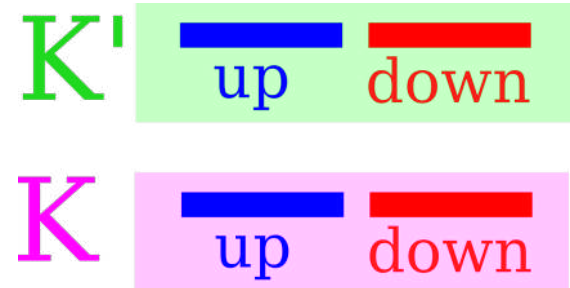
Twist angle control

*The details of the
electronic dispersion*



Interlayer bias

*The local quantum
degrees of freedom*



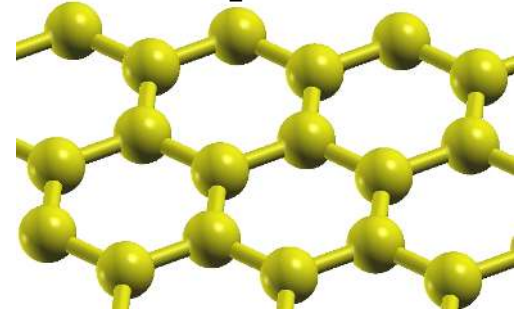
Magnetic 2D materials

Goal: engineer local quantum degrees of freedom via 2d magnetism

Designing heavy-fermion quantum matter with twisted magnetic van der Waals materials

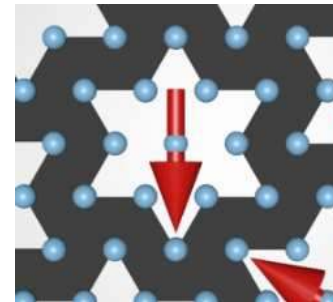


Graphene



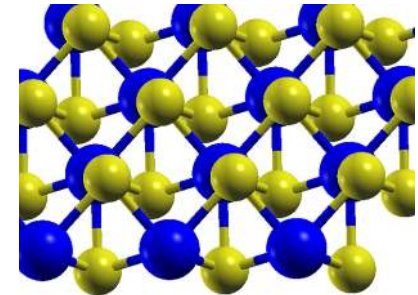
Magnet

1T-TaS₂



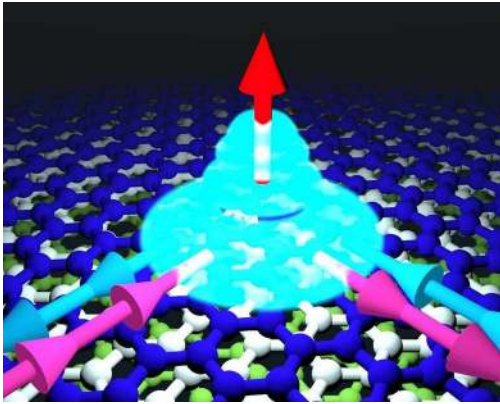
Metal

1H-TaS₂



Behind the scenes

Heavy fermions in twisted graphene trilayers

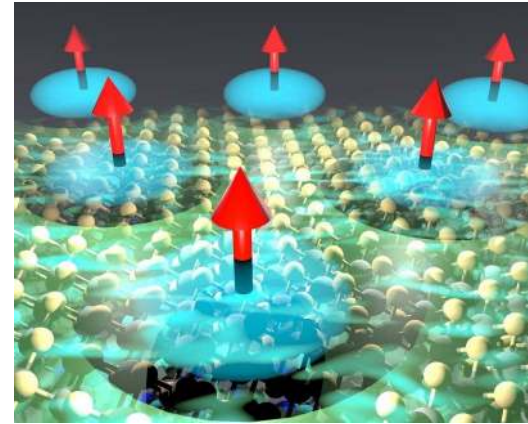


Aline Ramires



Phys. Rev. Lett. 127, 026401 (2021)

Heavy fermions in twisted dichalcogenide bilayers



V. Vaňo



S. Kezilebieke



P. Liljeroth

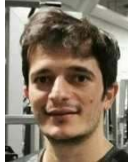


Nature 599, 582–586 (2021)

S. Ganguli



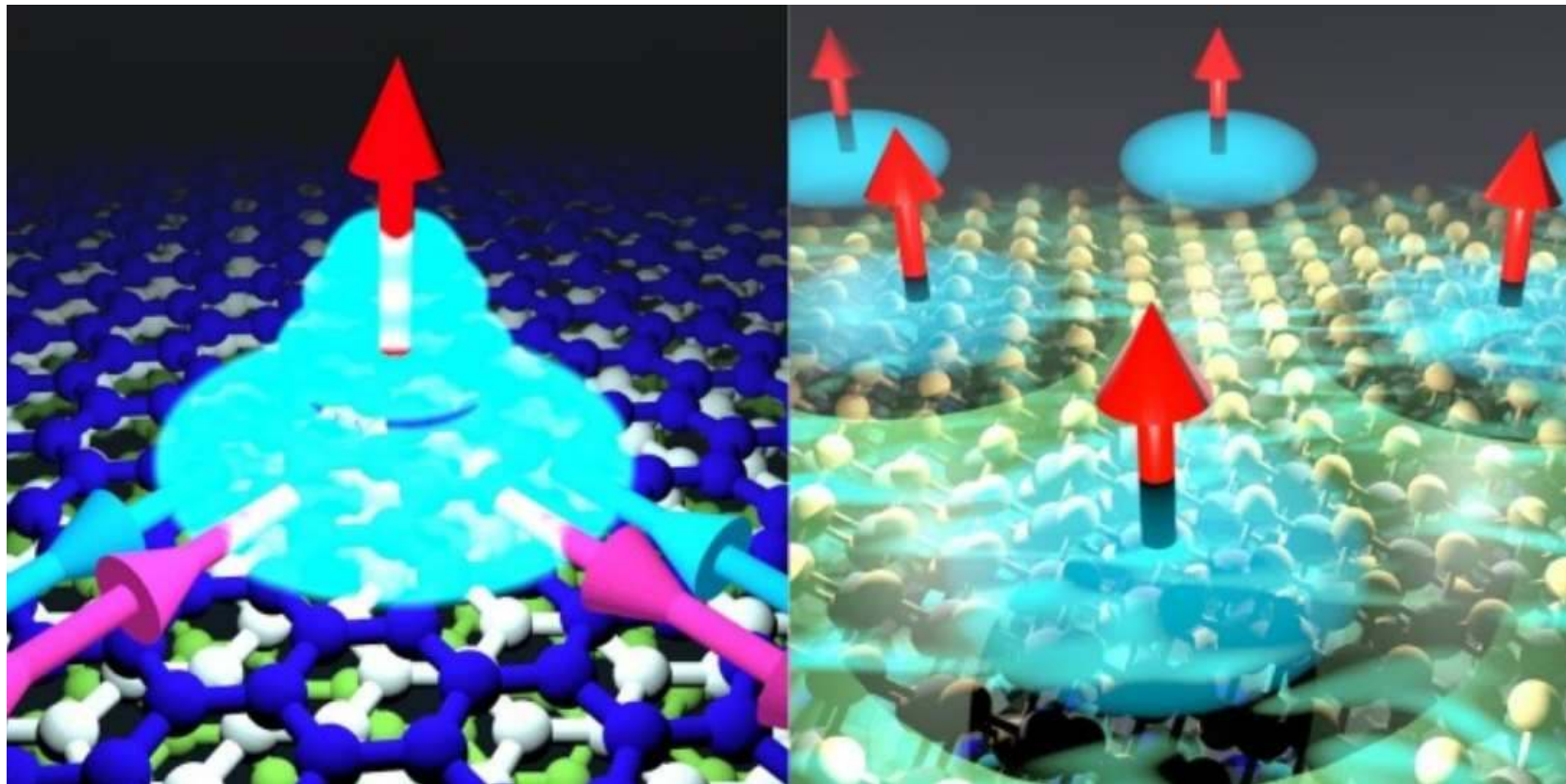
M. Amini



G. Chen



Heavy fermions in van der Waals materials

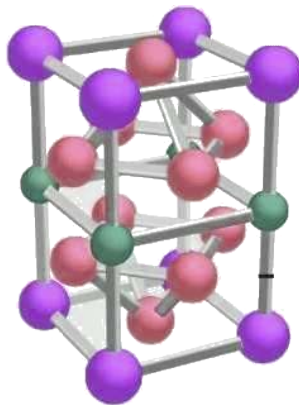


Searching for new heavy fermion compounds

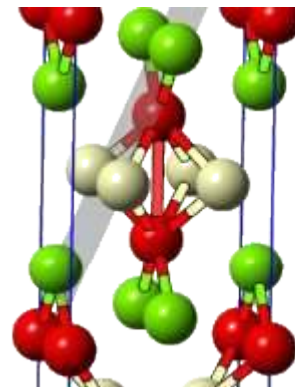
We have a rich family of bulk heavy-fermion compounds



Science 375, 6576, 76-81 (2021)



Nature 579, 523–527 (2020)



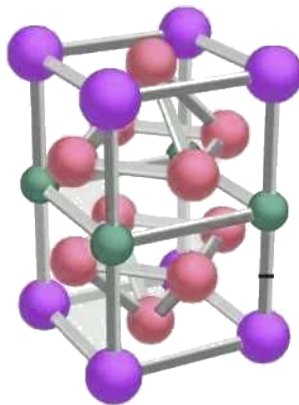
- Strongly correlated matter, dominated by Kondo lattice physics
- Unique system to realize quantum criticality and unconventional superconductivity
- Found in rare-earth compounds, such as UTe_2 or UPt_3

Searching for new heavy fermion compounds

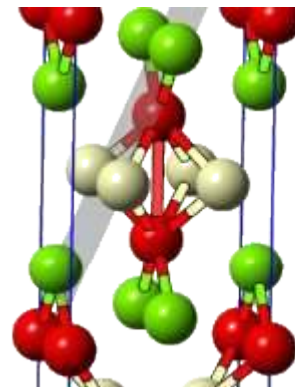
We have a rich family of bulk heavy-fermion compounds



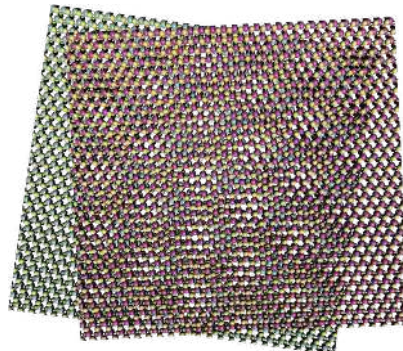
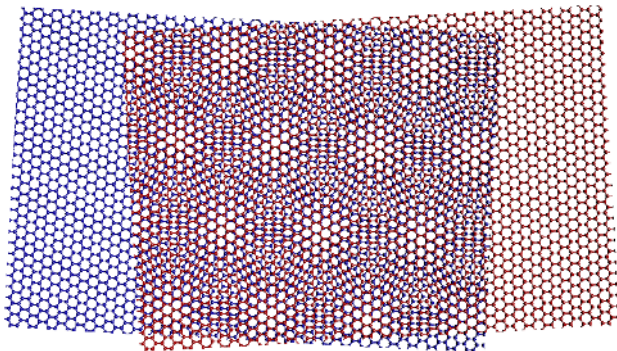
Science 375, 6576, 76-81 (2021)



Nature 579, 523–527 (2020)



Can we realize heavy-fermion physics with two-dimensional materials?

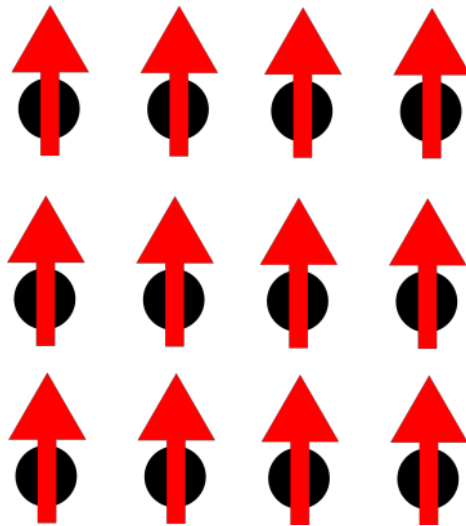


Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)
Phys. Rev. Research 3, 043173 (2021)
arXiv:2110.11962
arXiv:2207.00096

Building an artificial heavy fermion state



Lattice of Kondo impurities



f-electrons in
rare-earth compounds

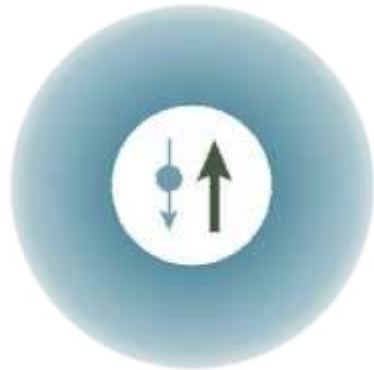
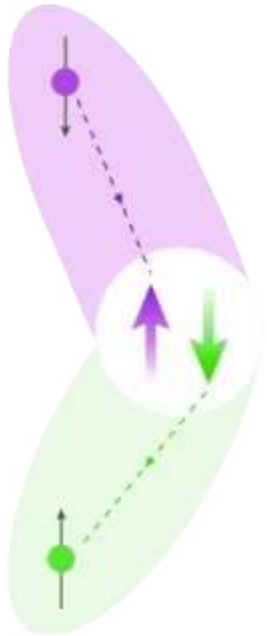
Dispersive electron gas



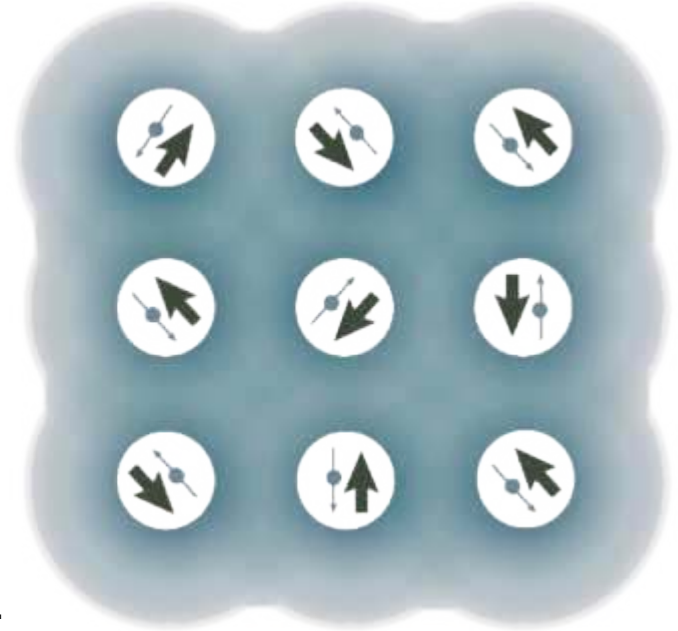
s/p/d-electrons in
rare-earth compounds

Building an artificial heavy fermion state

Conduction electrons form
Kondo singlets with the impurities



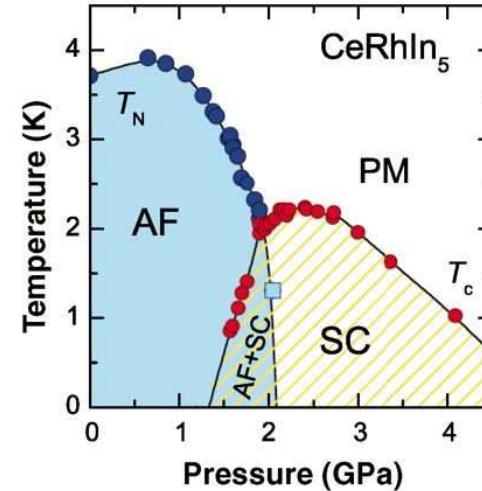
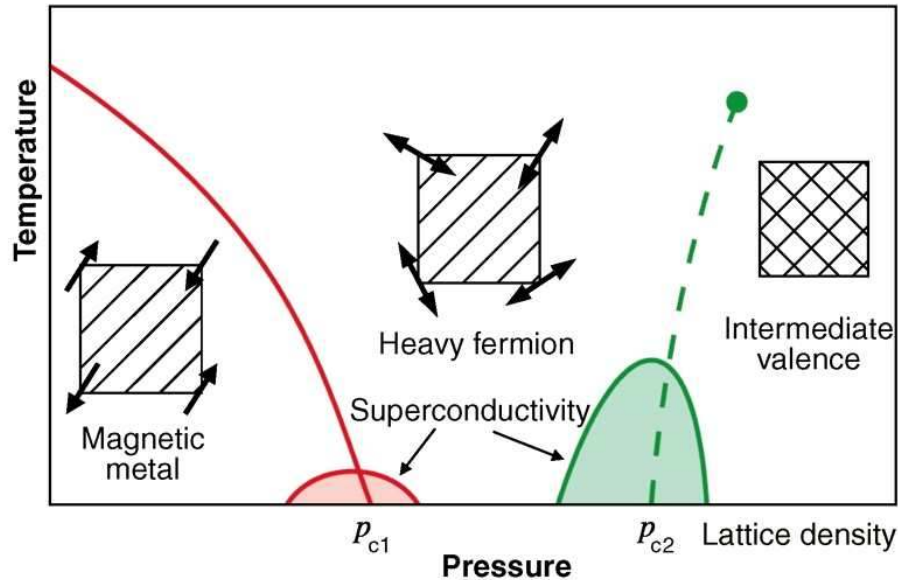
Kondo-lattice model



Associated with Kondo lattice physics:

- Colossal mass enhancement of electrons
- Quantum criticality
- Unconventional (topological) superconductivity

Basics of heavy fermion physics



Science 332.6026, 196-200 (2011)

Associated with Kondo lattice physics:

- Colossal mass enhancement of electrons
- Quantum criticality
- Unconventional (topological) superconductivity

The Kondo lattice problem

The Kondo problem

Conduction electrons

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right)$$

Kondo coupling

$$H_K = \sum_{\alpha\beta} \left(c_{0\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{0\alpha} \right) \cdot \vec{S}$$

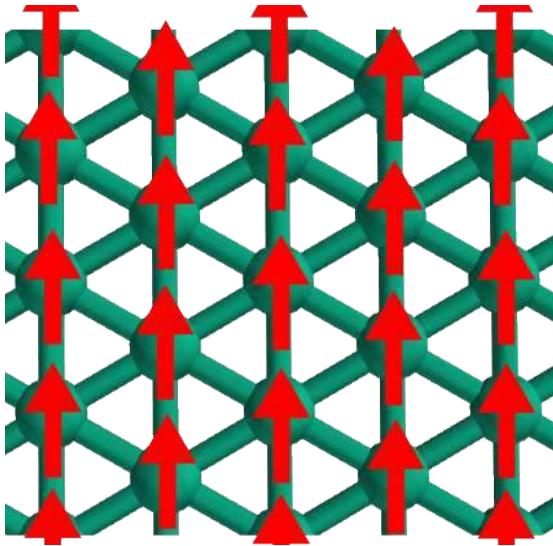
We now take a quantum spin $S=1/2$

$$|GS\rangle \sim \frac{1}{\sqrt{2}} [|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle]$$



The Kondo lattice problem

Lattice of Kondo impurities



Dispersive electron gas



$$J_K$$

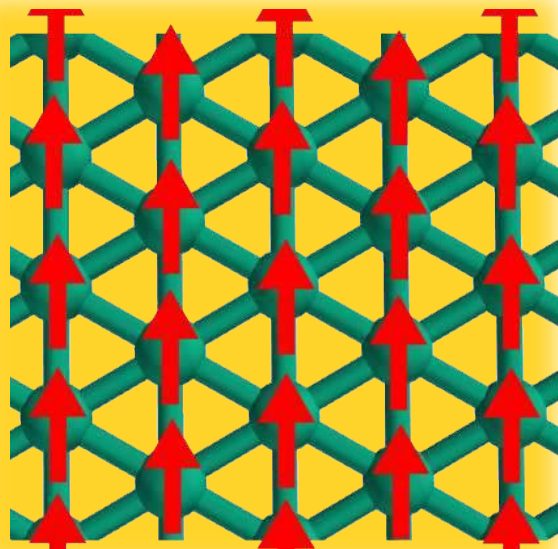

Both ingredients coupled through Kondo coupling

The Kondo lattice problem

The Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Conduction electrons



Kondo coupling

Kondo sites

Solving the Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Replace the spin sites by auxiliary fermions

$$S_{\alpha\beta}(j) \sim f_{j\alpha}^\dagger f_{j\beta} - \frac{n_f(j)}{N} \delta_{\alpha\beta}$$

This makes the effective Hamiltonian an “interacting” fermionic Hamiltonian

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger f_{j\beta} \right) \left(f_{j\alpha}^\dagger c_{j\alpha} \right)$$

Solving the Kondo lattice problem

Now we decouple the fermions with a mean-field approximation

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} \left(c_{j\beta}^{\dagger} f_{j\beta} \right) \left(f_{j\alpha}^{\dagger} c_{j\alpha} \right)$$

Obtaining a quadratic Hamiltonian

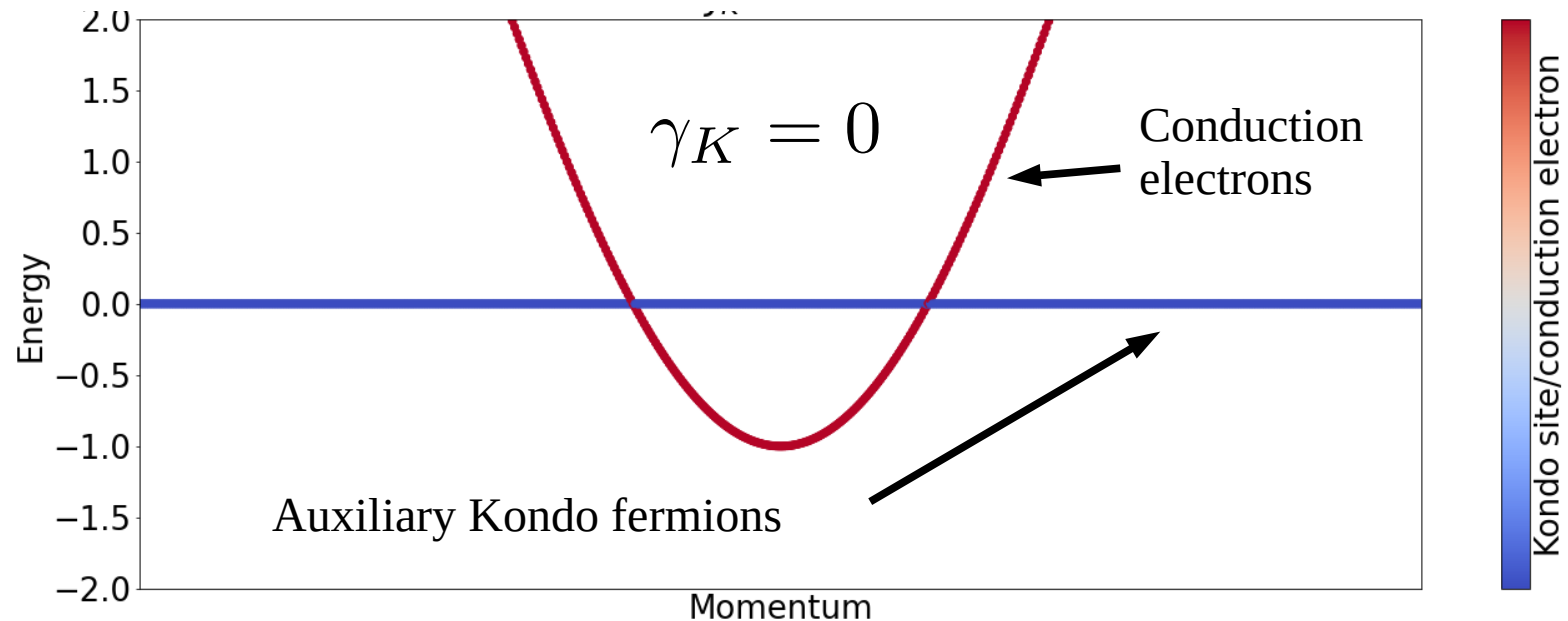
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

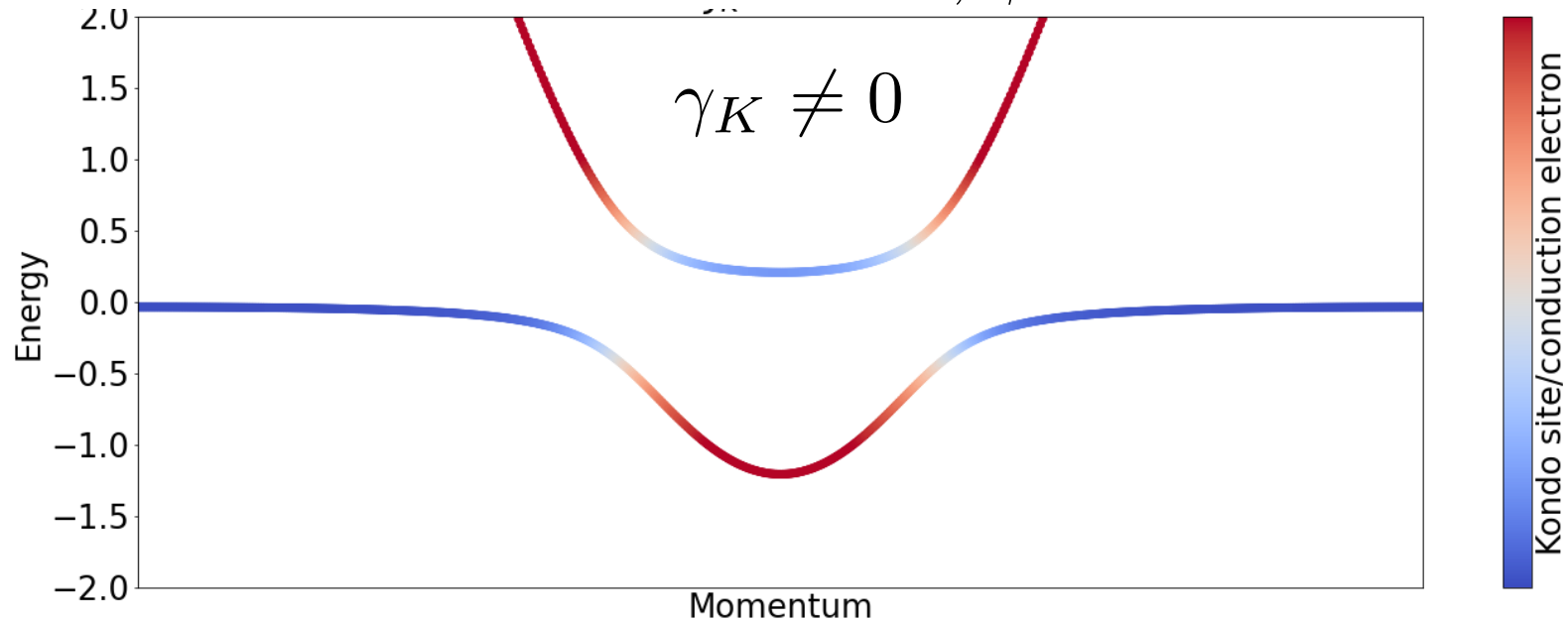
Electronic structure of the Kondo lattice problem

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k}, \alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$



Electronic structure of the Kondo lattice problem

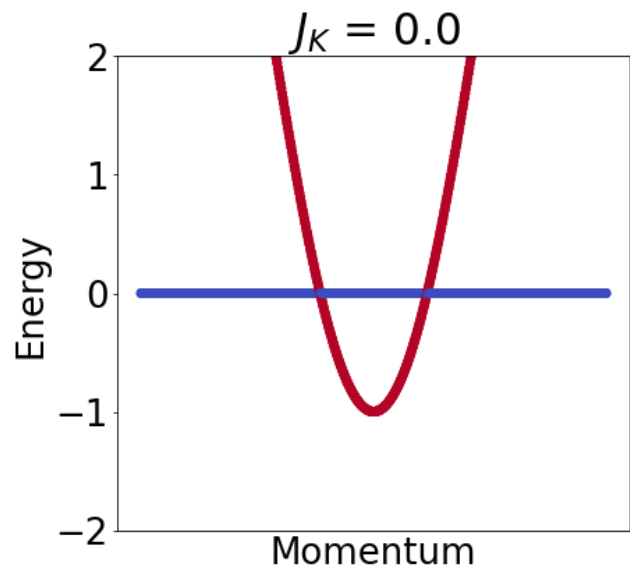
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k}, \alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$



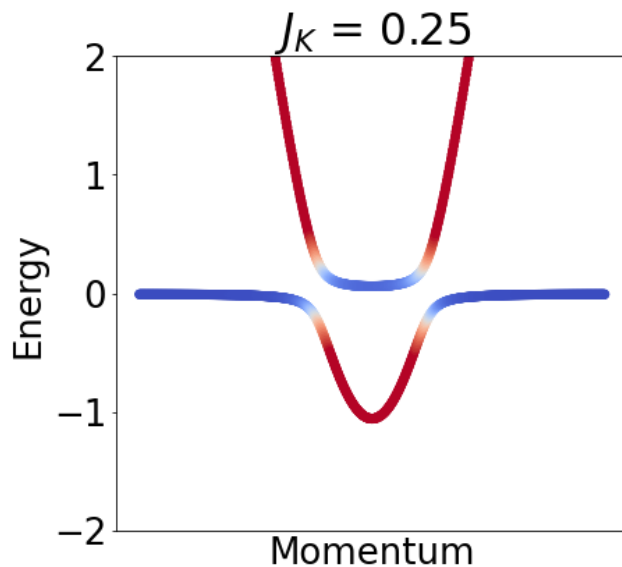
The Kondo coupling opens up a gap in the electronic structure

Dependence on the Kondo coupling

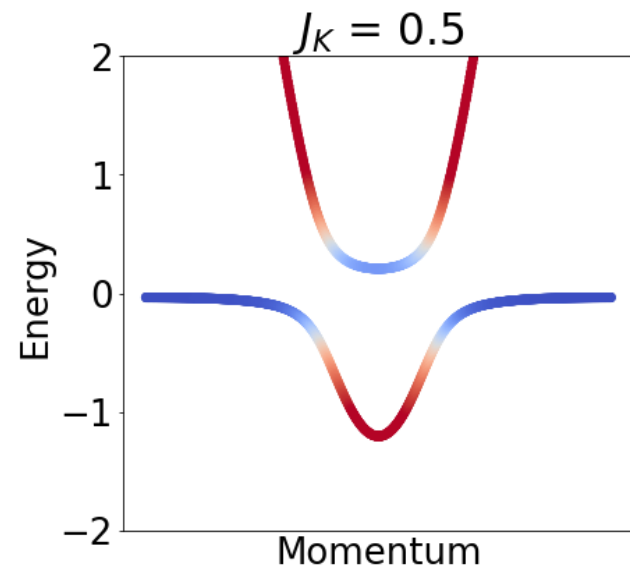
The heavy-fermion gap becomes bigger as the Kondo coupling increases



Kondo site/conduction electron

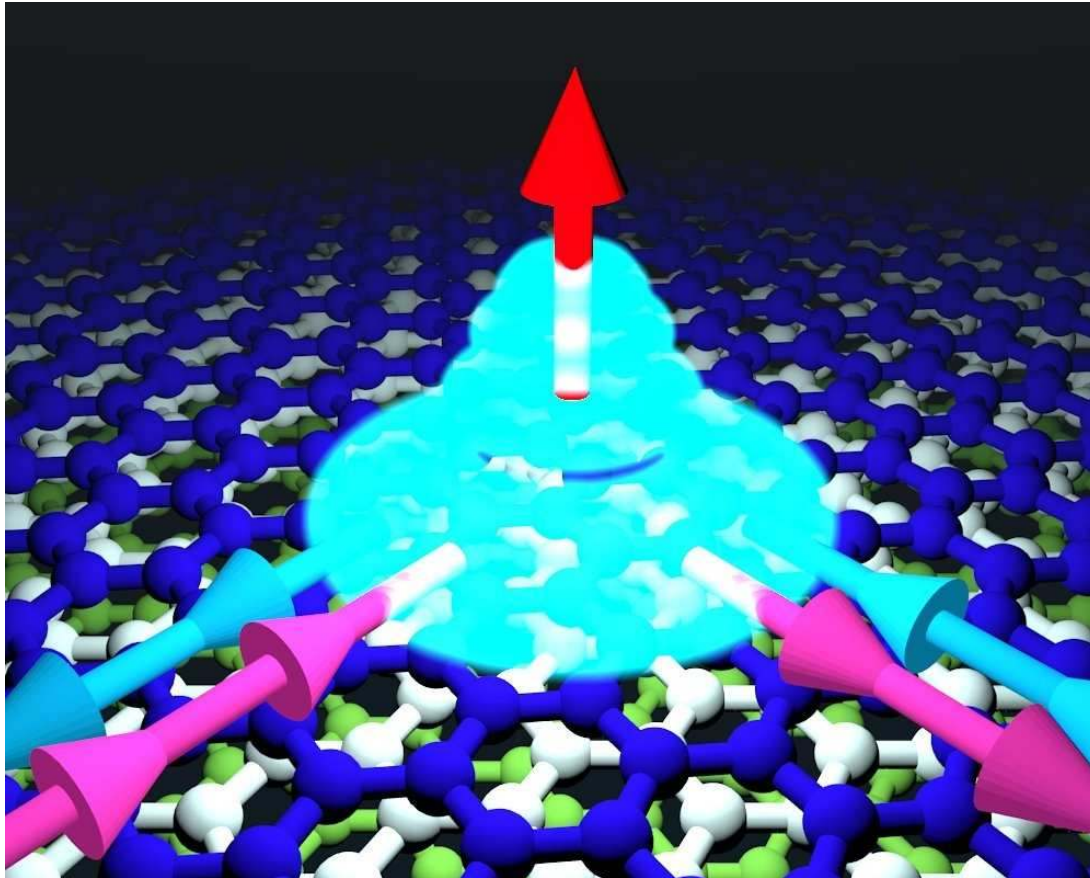


Kondo site/conduction electron



Kondo site/conduction electron

Heavy-fermions in twisted graphene trilayers



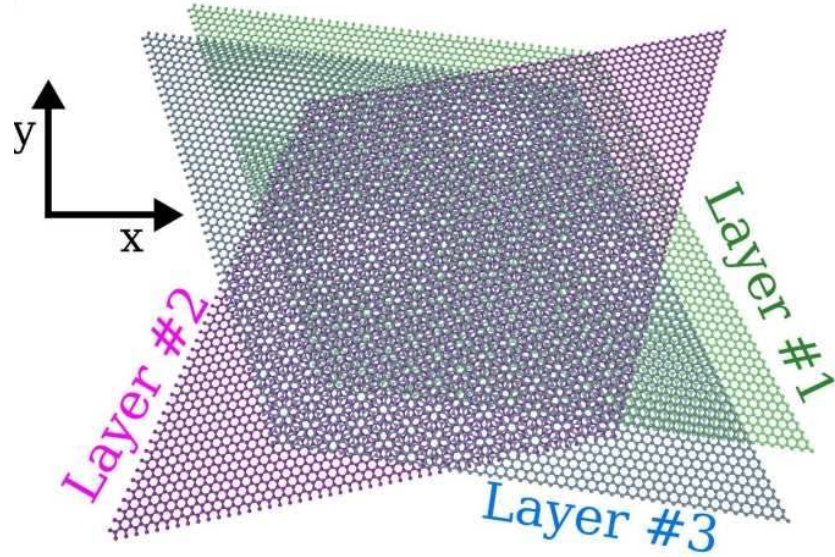
Aline Ramires



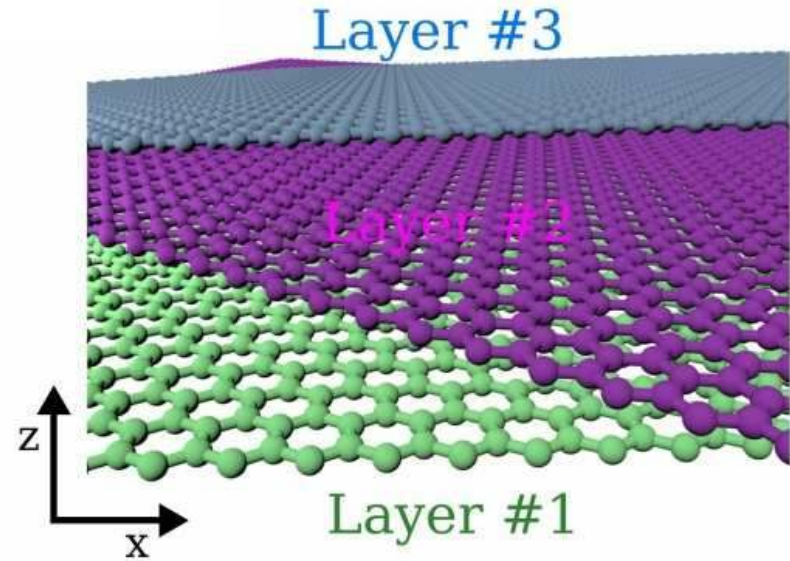
Phys. Rev. Lett. 127, 026401 (2021)

Heavy-fermions in twisted graphene trilayers

Top view

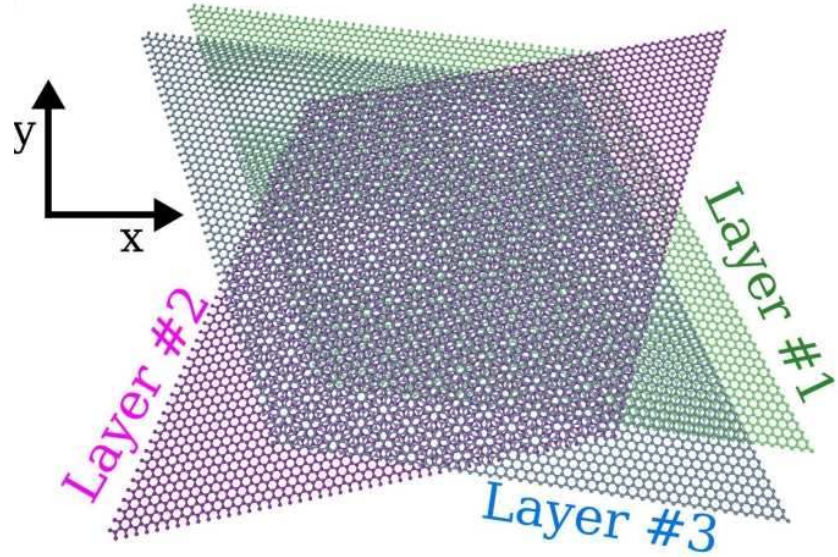


Lateral view

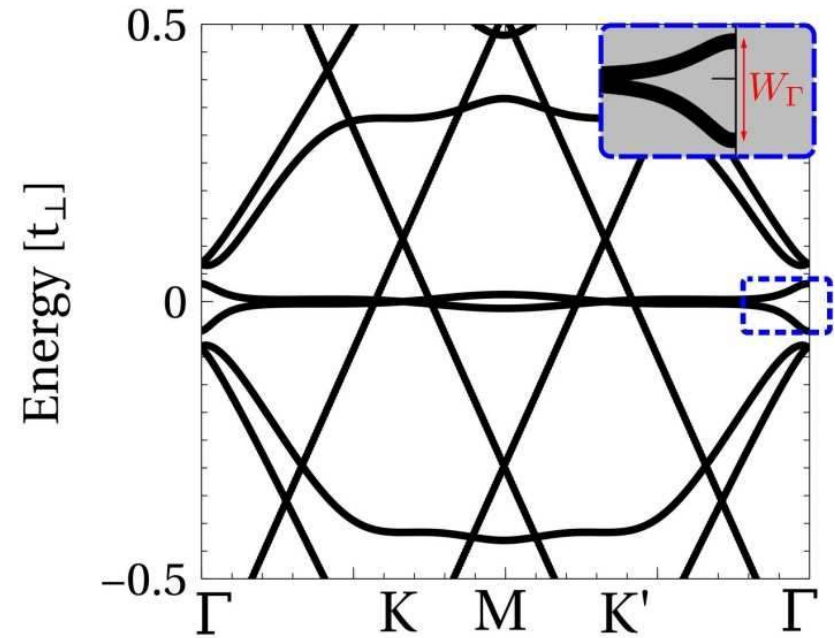


Heavy-fermions in twisted graphene trilayers

Top view

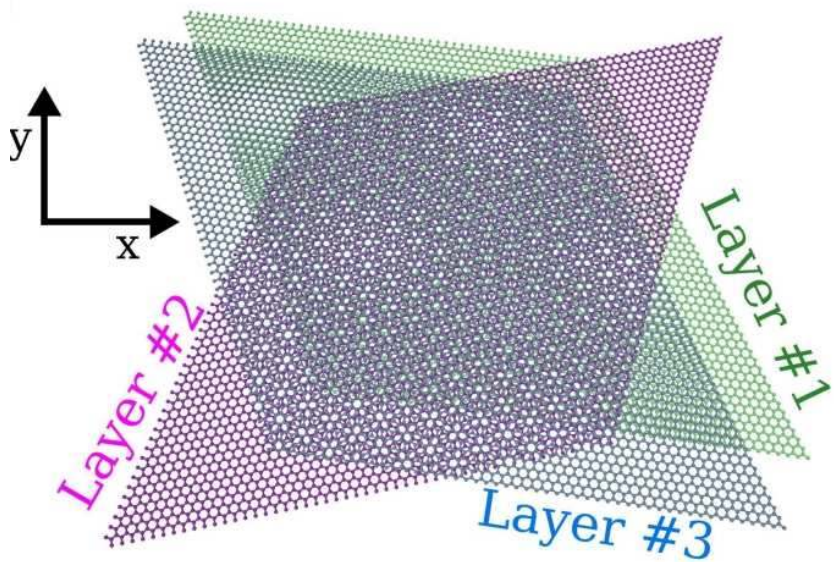


Band structure

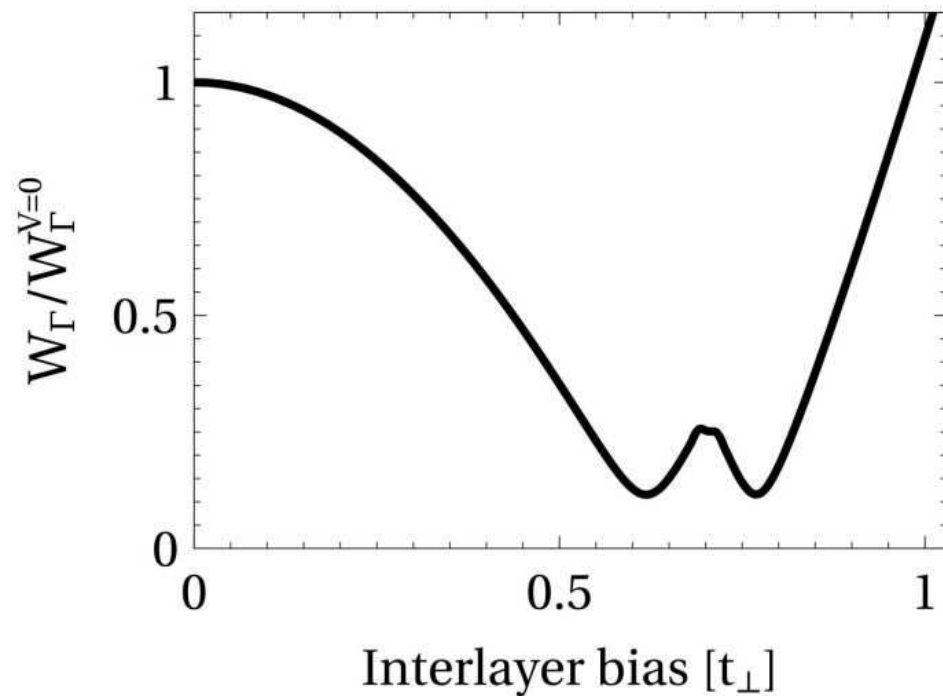


Heavy-fermions in twisted graphene trilayers

Top view



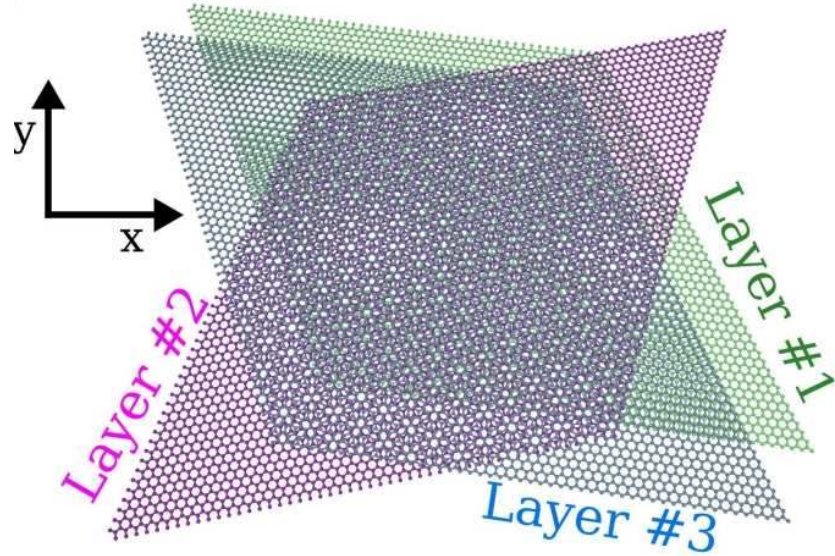
Bandwidth of nearly flat bands



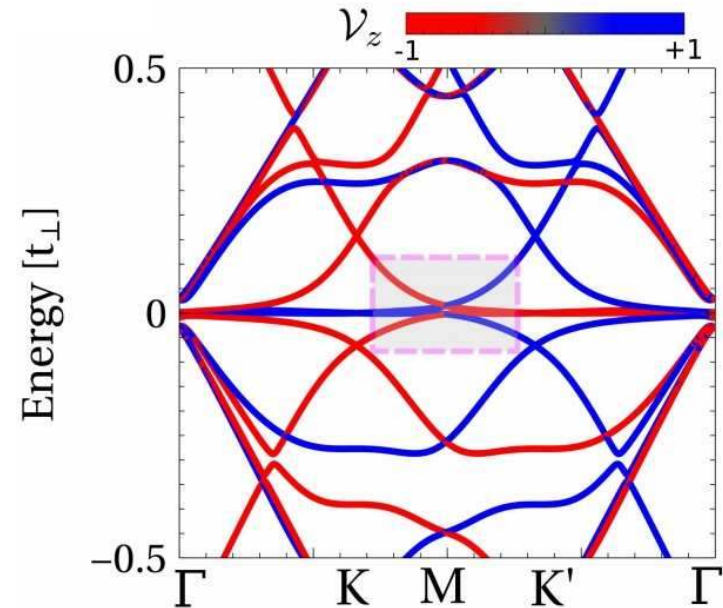
Band flattening as an interlayer bias is applied

Heavy-fermions in twisted graphene trilayers

Top view

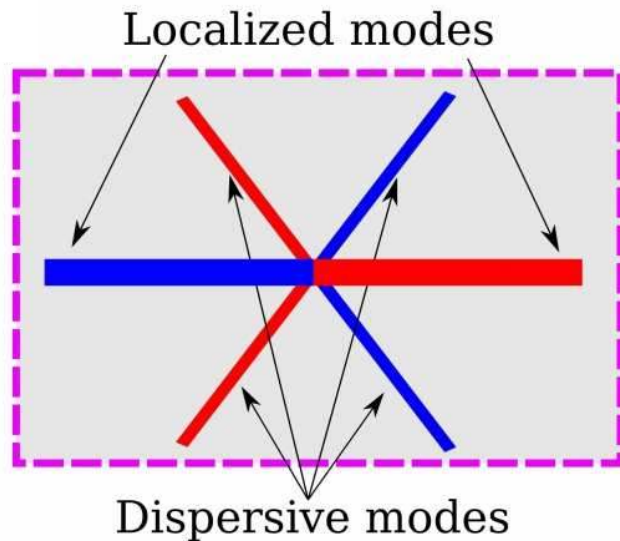


Electronic structure in the presence of an electric bias

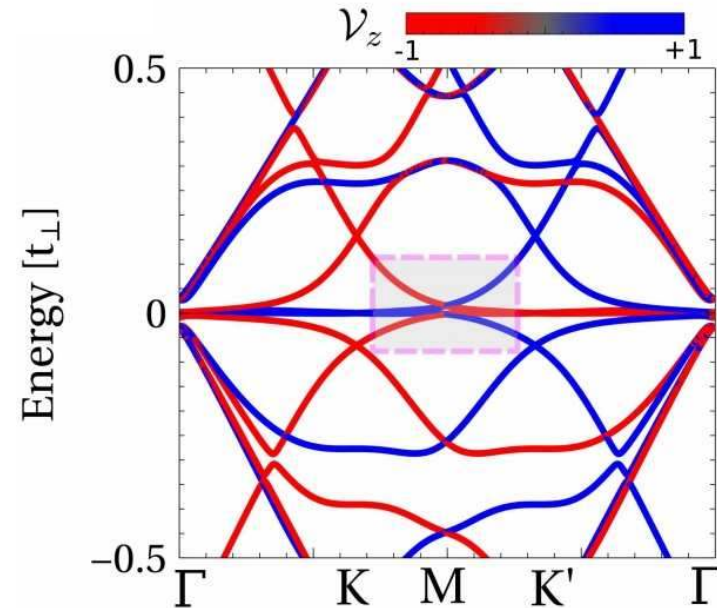


Heavy-fermions in twisted graphene trilayers

Low energy model

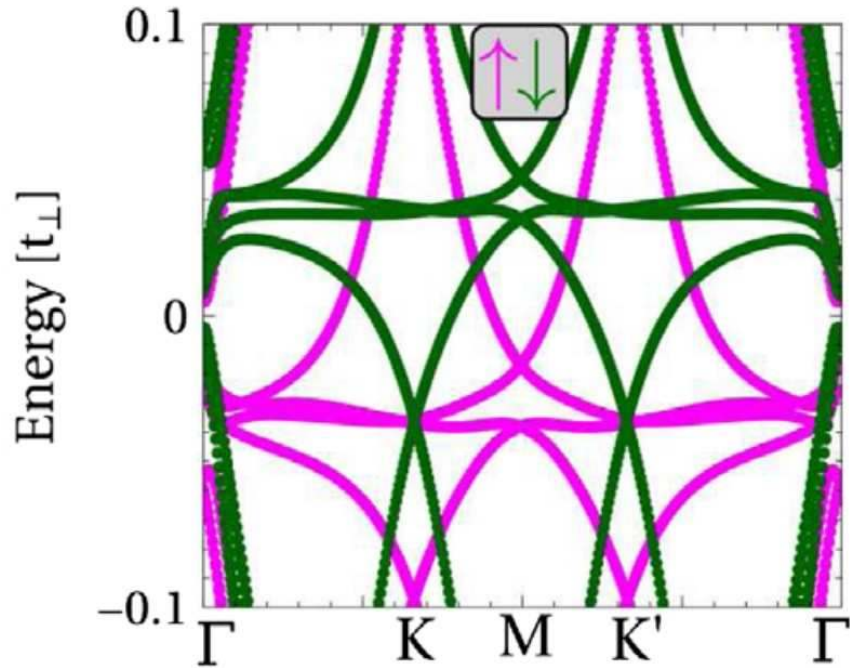


Electronic structure in the presence of an electric bias

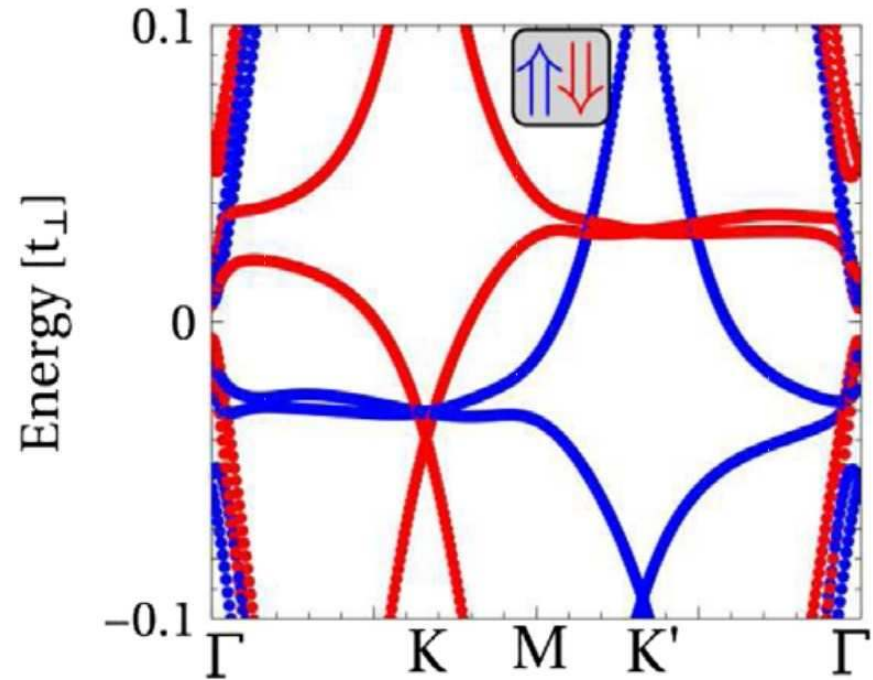


Mean-field electronic structure at half filling

Promoting spin moment formation



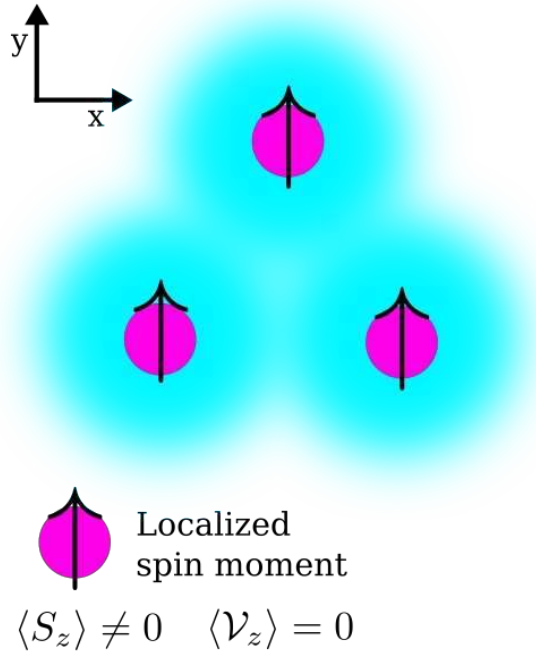
Promoting valley moment formation



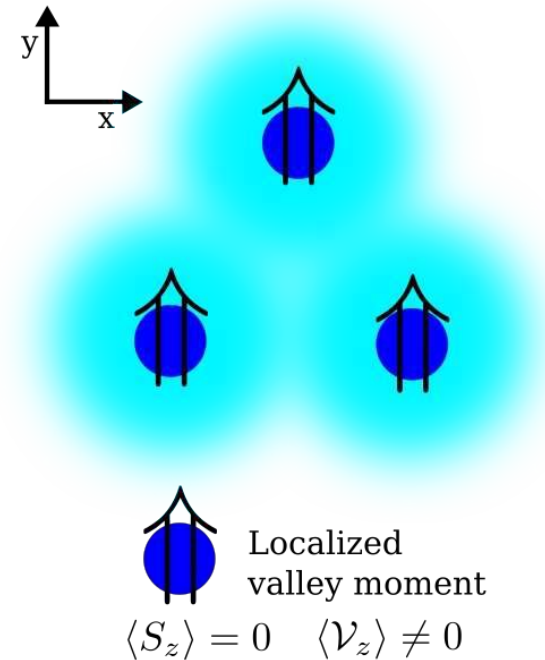
Local spin and valley moments appear when including electronic interactions at the mean-field level

Mean-field electronic structure at half filling

Promoting spin moment formation



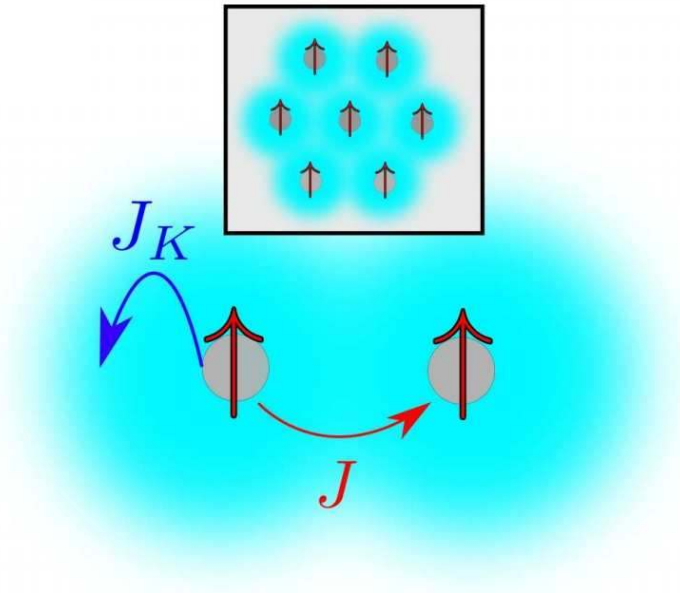
Promoting valley moment formation



Local spin and valley moments appear when including electronic interactions at the mean-field level

Kondo lattice model in the presence of interactions

Interactions of the Kondo lattice model



Kondo coupling Exchange coupling

$$H = H_{\text{dispersive}} + J_K \sum_{\langle i\alpha \rangle} \mathbf{S}_i \cdot \mathbf{s}_\alpha + J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

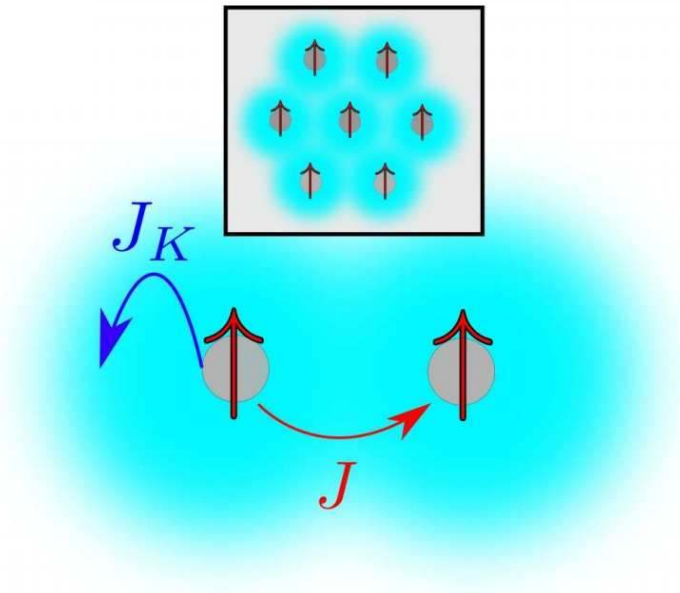
Promotes Kondo screening Promotes magnetic order

A heavy fermion regime appears when the Kondo coupling dominates over the exchange coupling

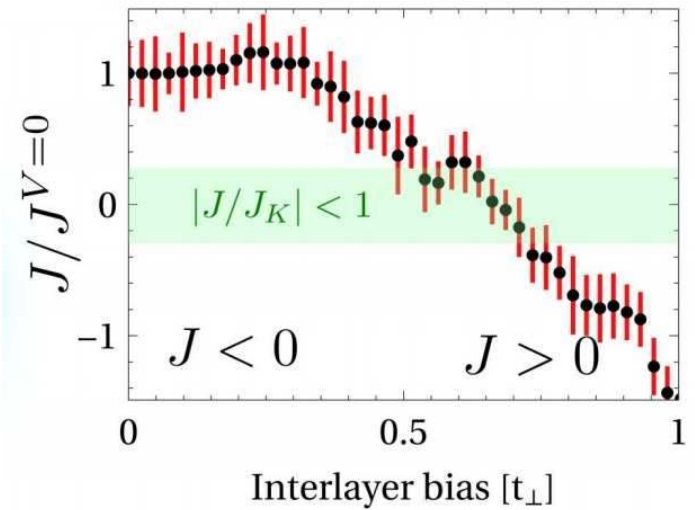
$$|J/J_K| < 1$$

Kondo lattice model in the presence of interactions

Interactions of the Kondo lattice model



Electric control of the Kondo regime

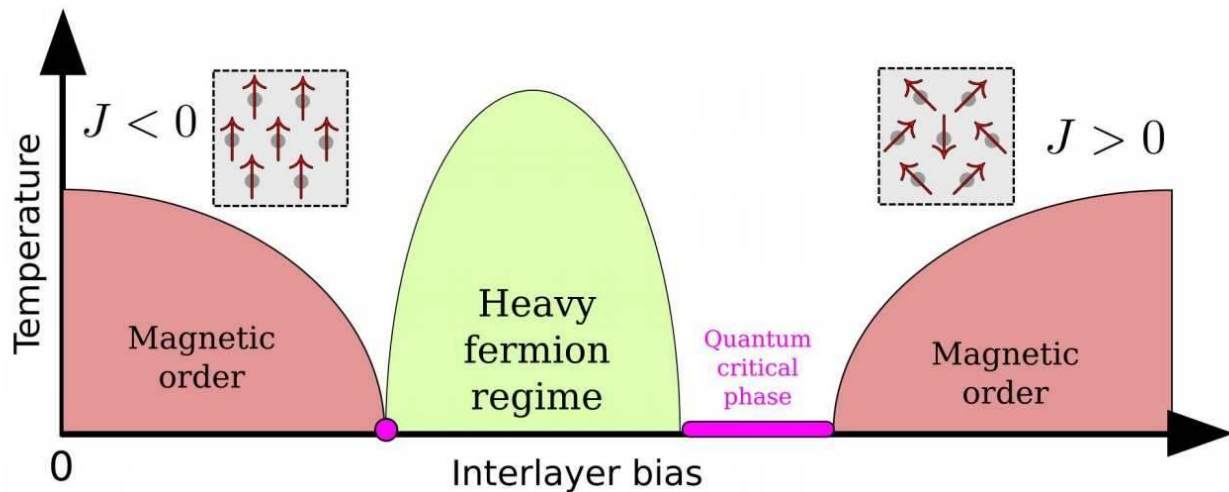


A heavy fermion regime appears when the Kondo coupling dominates over the exchange coupling

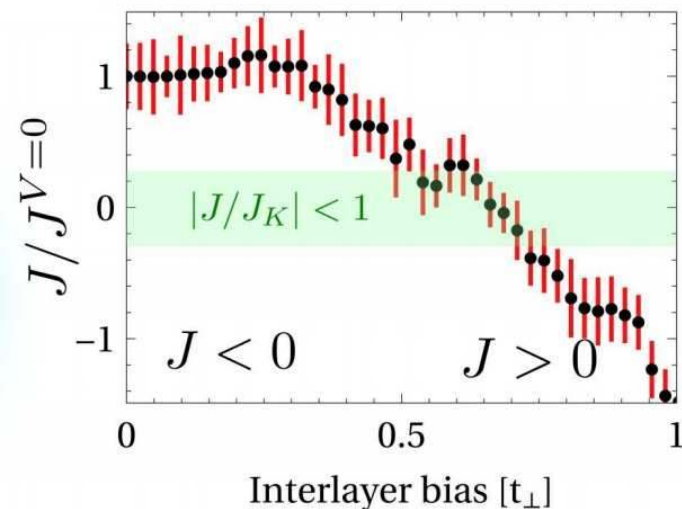
$$|J/J_K| < 1$$

Heavy-fermions in twisted graphene trilayers

Heavy-fermion (Doniach) phase diagram



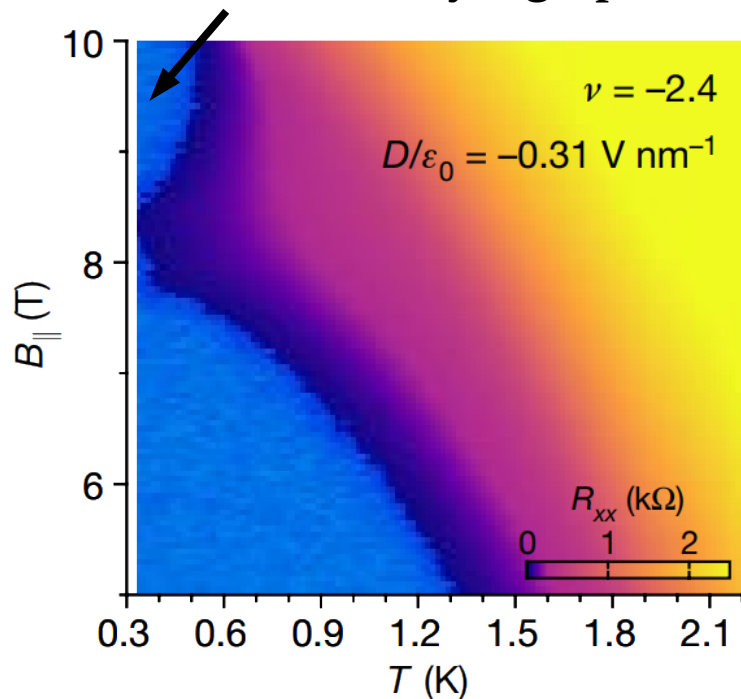
Electric control of the Kondo regime



The full phase diagram of the heavy fermion system can be explored in a single twisted graphene, by tuning the system with an electric bias

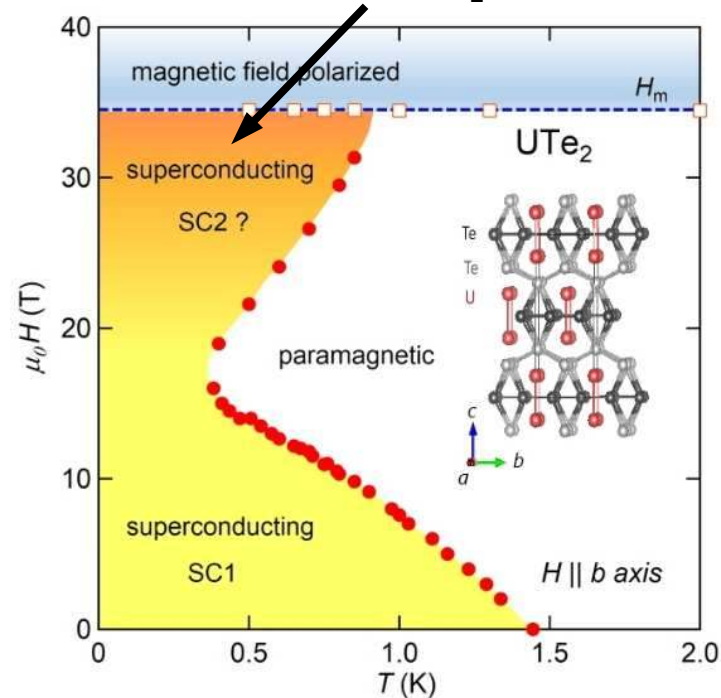
An experimental signature of heavy-fermion physics: spin-triplet superconductivity

Reentrant superconductivity
in twisted trilayer graphene



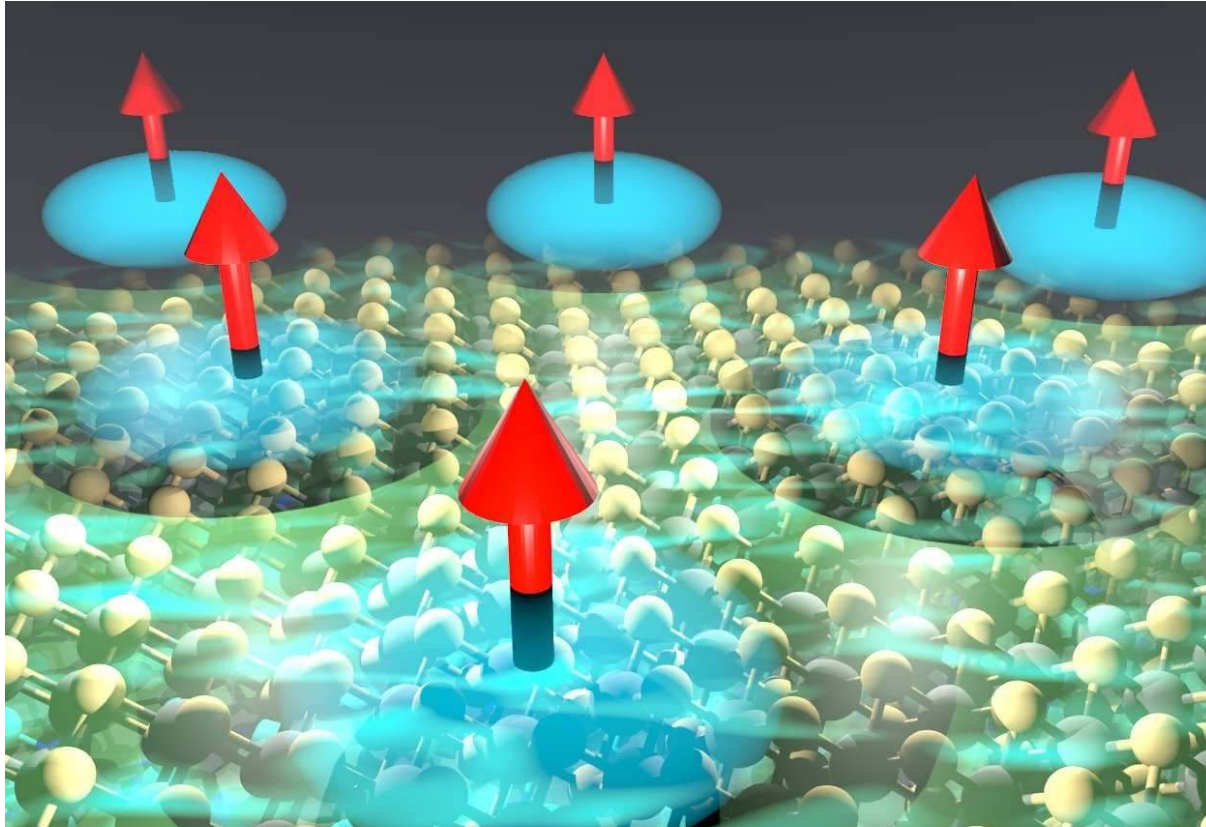
Nature 595, 526–531 (2021)

Reentrant superconductivity
in UTe_2



Nat. Phys. 15, 1250 (2019)
Science 365, 684 (2019)

Heavy-fermions in a van der Waals dichalcogenide heterostructure



S. Ganguli



M. Amini



G. Chen



V. Vaňo



S. Kezilebieke



P. Liljeroth



Nature 599, 582–586 (2021)

Heavy-fermions in dichalcogenide bilayers

**Triangular lattice of
local magnetic moments**



1T-TaS₂

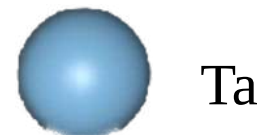
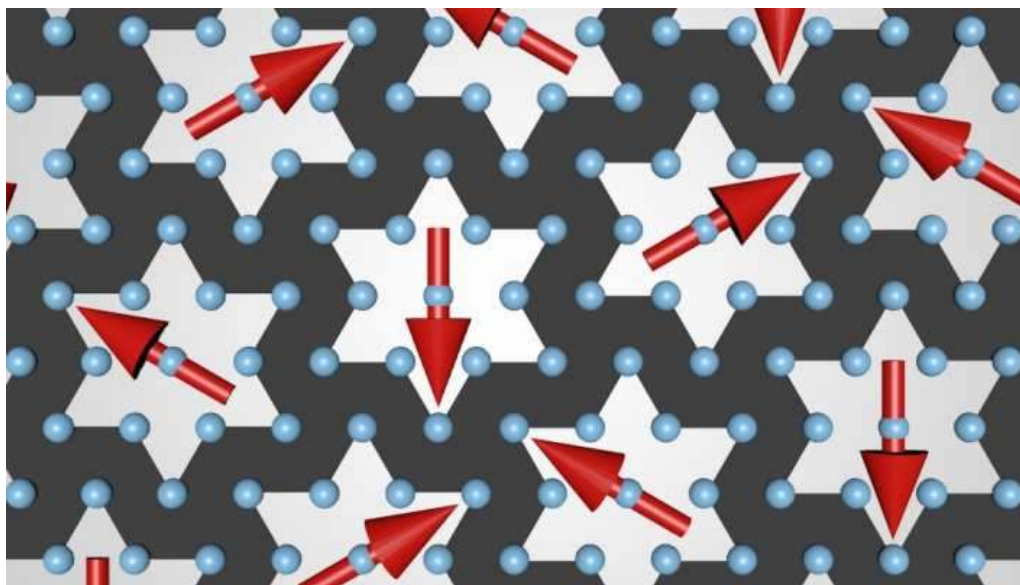
Two-dimensional electron gas



1H-TaS₂

Moment formation in 1T-TaS₂

Charge-density wave reconstruction, leading to a localized orbital in a $\sqrt{13} \times \sqrt{13}$ unit cell



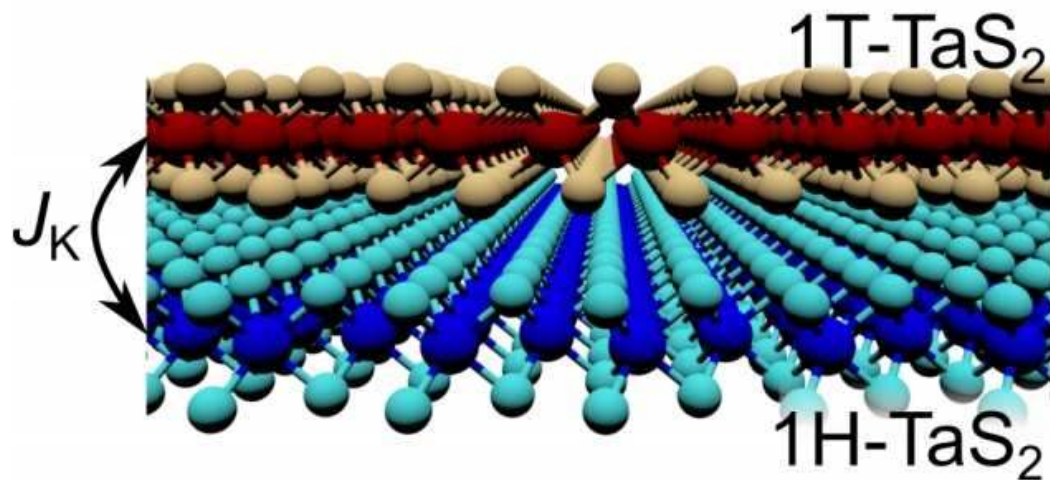
Strong interactions give rise to local moment formation

Effectively described by an S=1/2 Heisenberg model in a triangular lattice

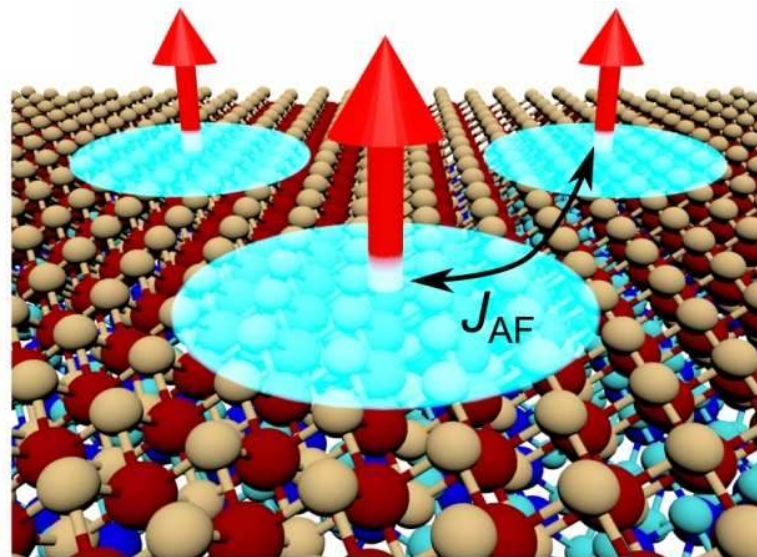
PNAS 114.27 (2017)
Phys. Rev. X 7, 041054 (2017)
Phys. Rev. B 96, 195131 (2017)

Heavy-fermions in dichalcogenide bilayers

Interlayer coupling creates
the Kondo coupling



Intralayer exchange between
local moments

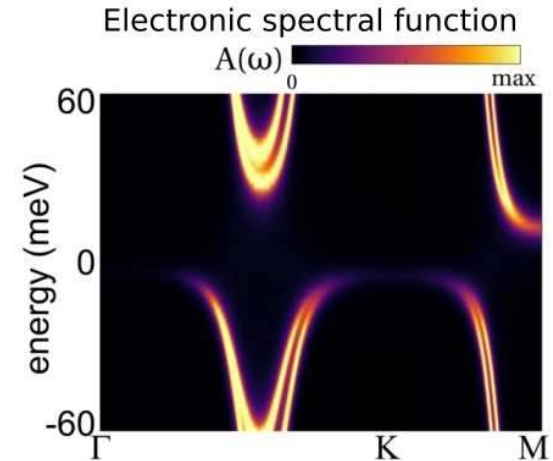
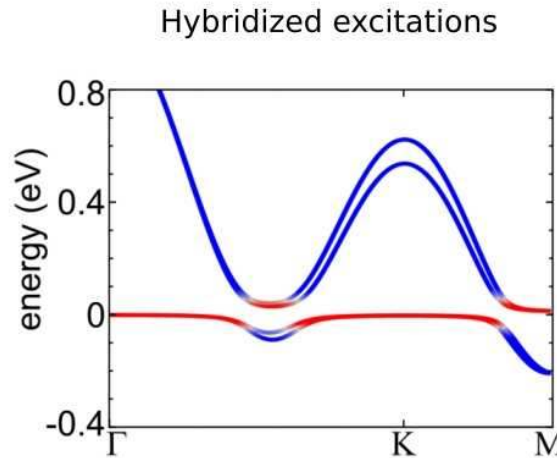
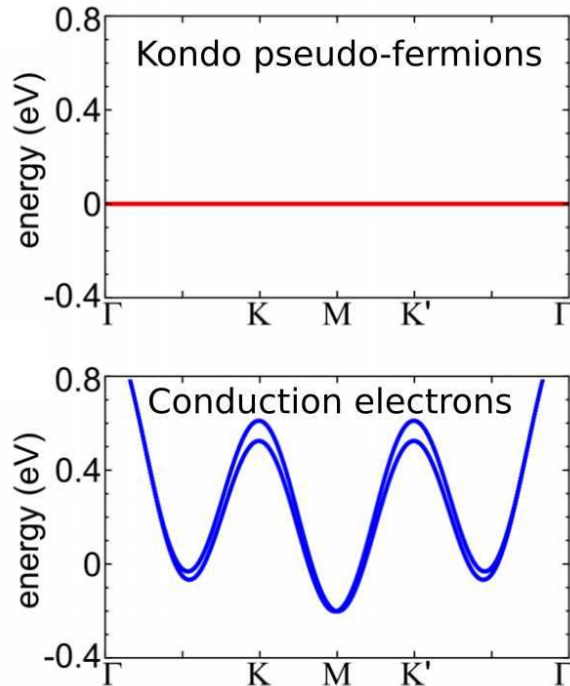


The bilayer realizes all the ingredients for heavy-fermion physics

Brief theory of heavy-fermions

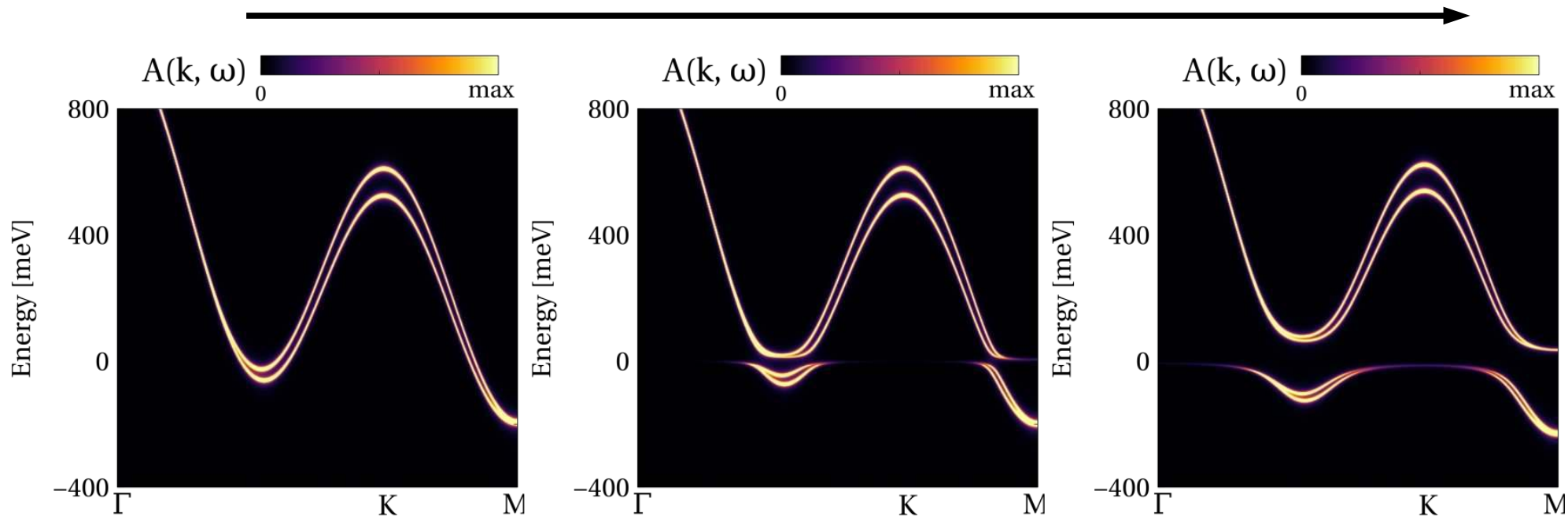
Kondo physics introduces resonant pseudo-fermions at the chemical potential

Leading to the opening of a heavy-fermion gap



Heavy-fermion hybridization gap

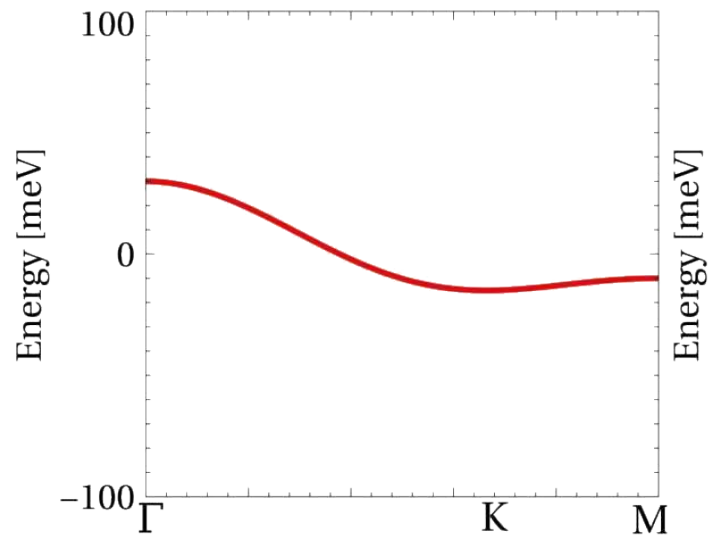
Increasing Kondo coupling



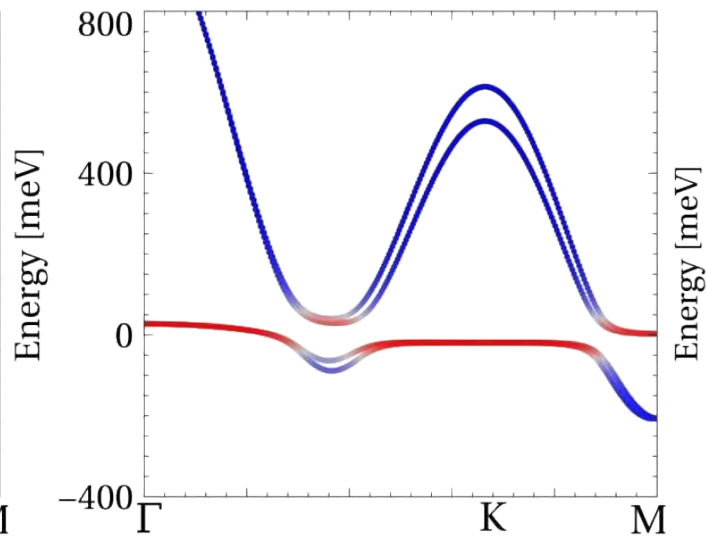
The Kondo coupling leads to a heavy-fermion gap opening in the metal

Heavy-fermion hybridization gap

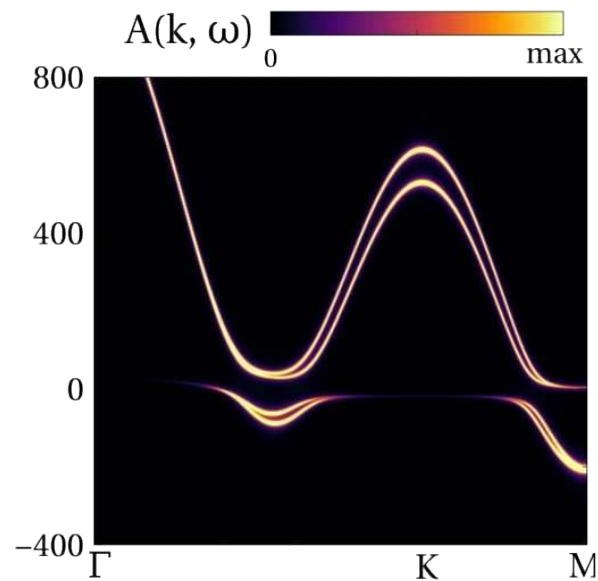
Kondo pseudofermions



Hybridized excitations



Conduction spectral function



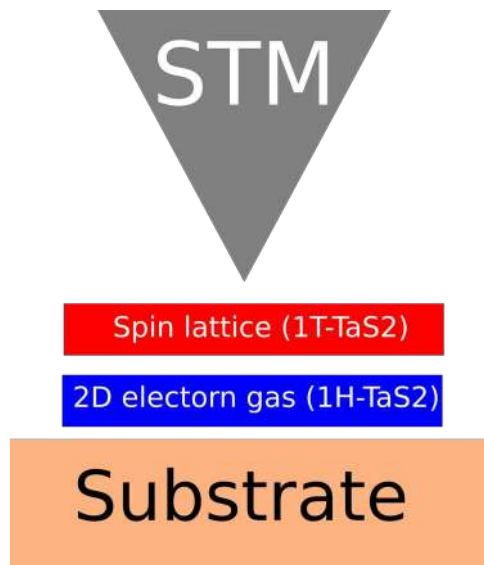
The gap remains open when including a small pseudofermion dispersion

Experimental signatures of heavy-fermion physics

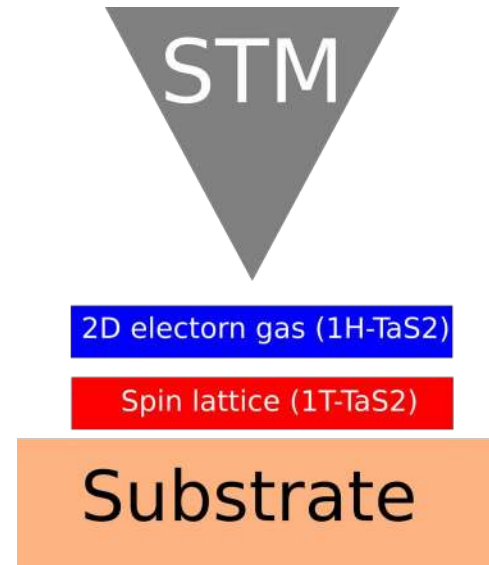
- Kondo physics in the magnetic lattice
- Gap opening in the metallic layer

Both signatures can be probed with scanning tunnel microscopy by growing two heterostructures

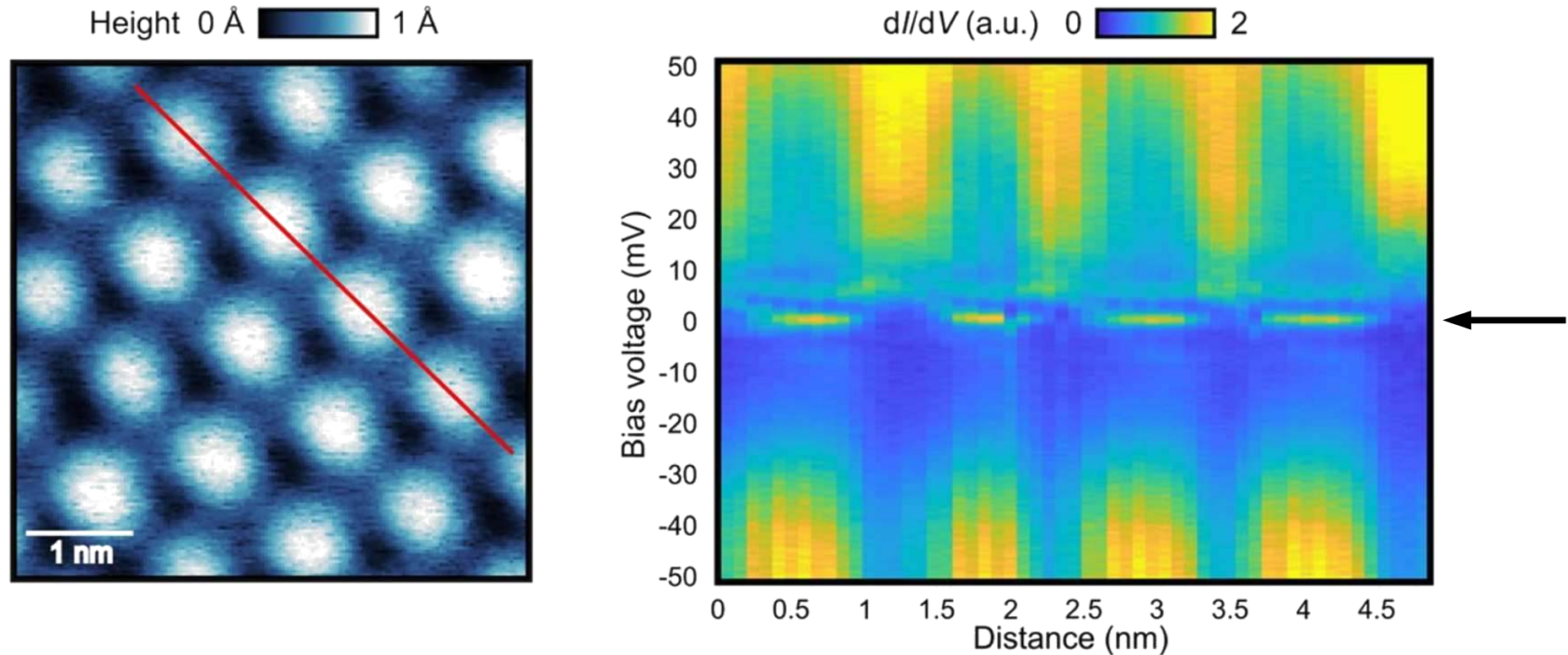
Probing the Kondo peak



Probing the heavy-fermion gap

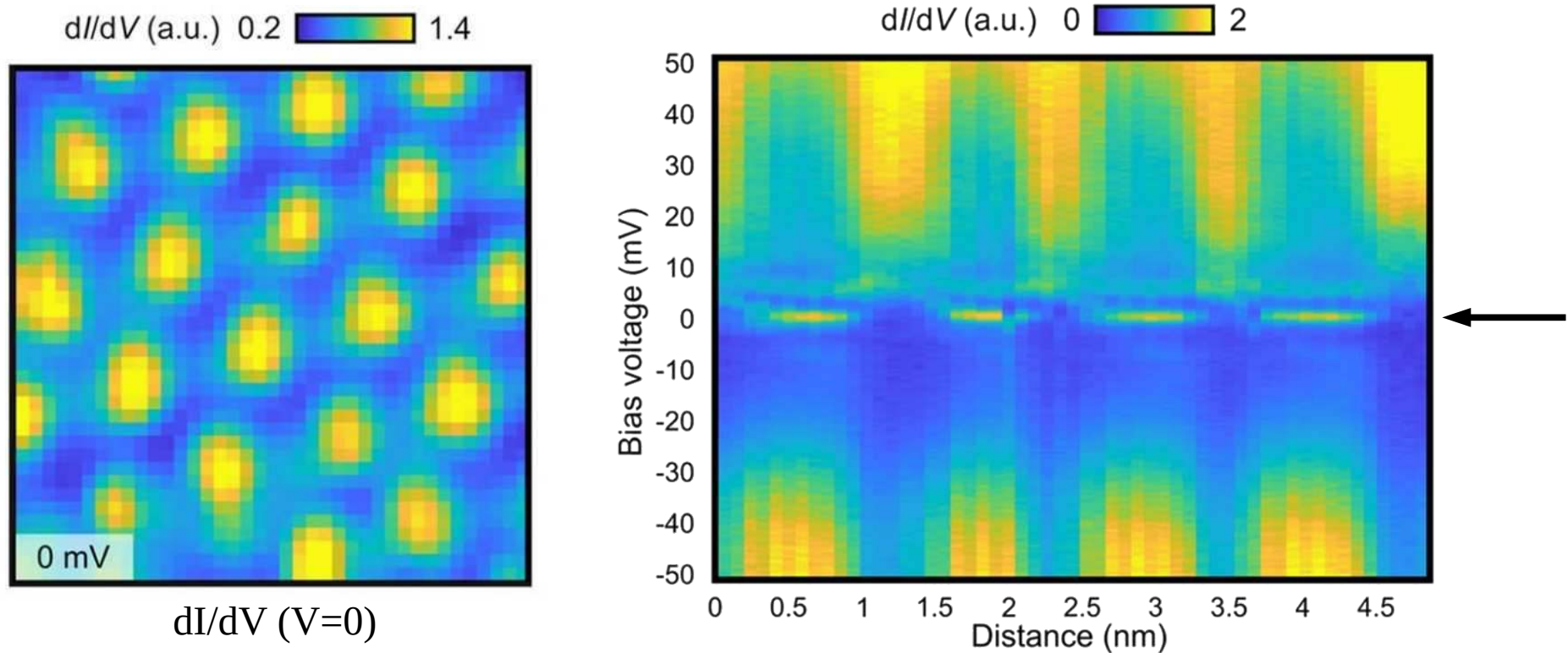


Grid spectroscopy of 1T-1H



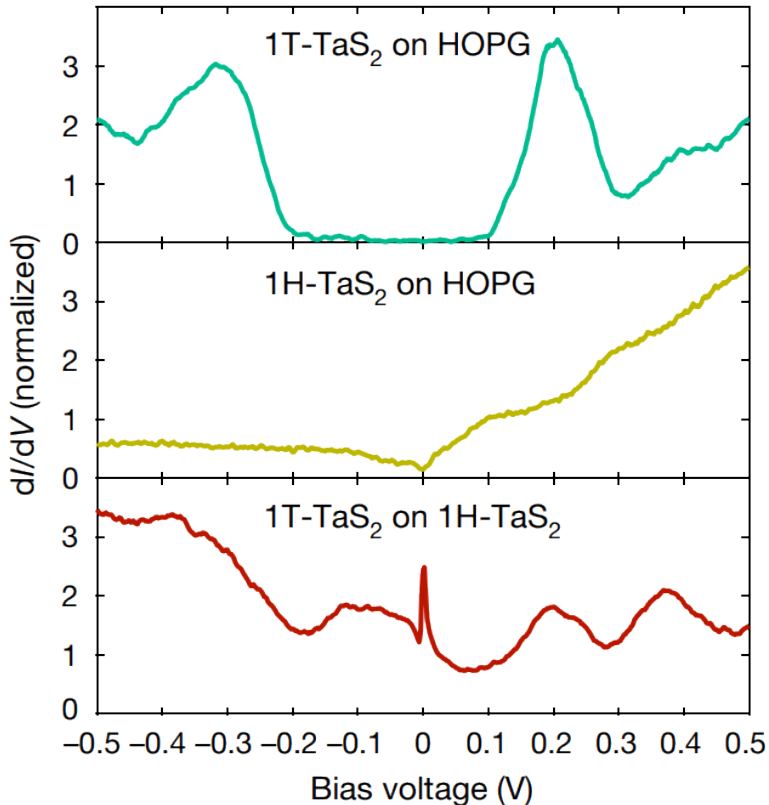
Zero bias (Kondo) peak appearing with the CDW periodicity

Grid spectroscopy of 1T-1H



Zero bias (Kondo) peak appearing with the CDW periodicity

Zero bias Kondo peak

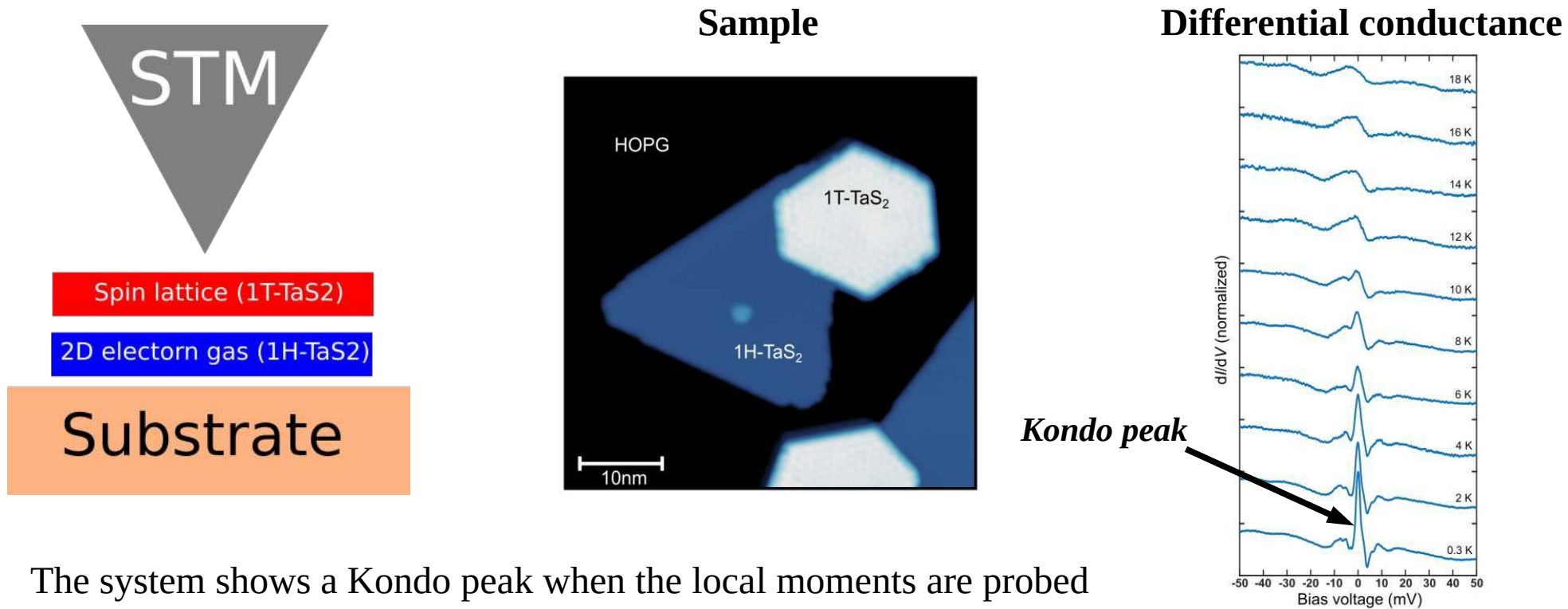


1T @ HOPG: Mott insulator

1H @ HOPG: Metal
(with a SC state at very low T)

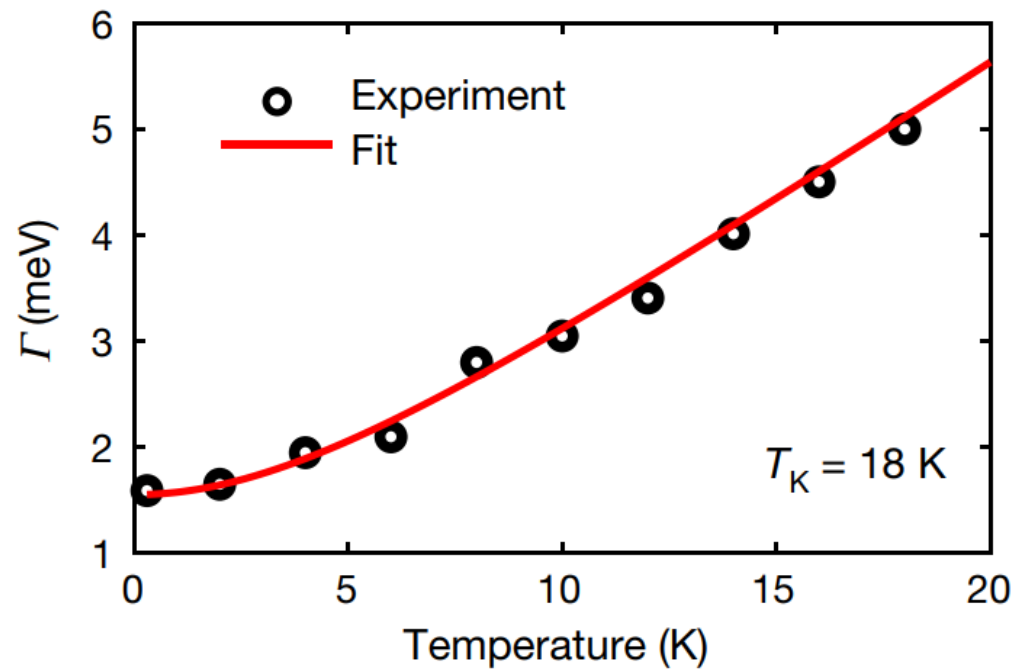
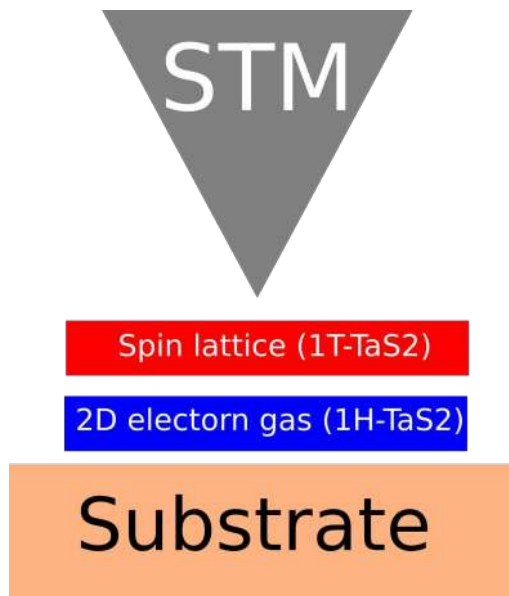
1T @ 1H: Kondo peak

Kondo physics in the magnetic lattice



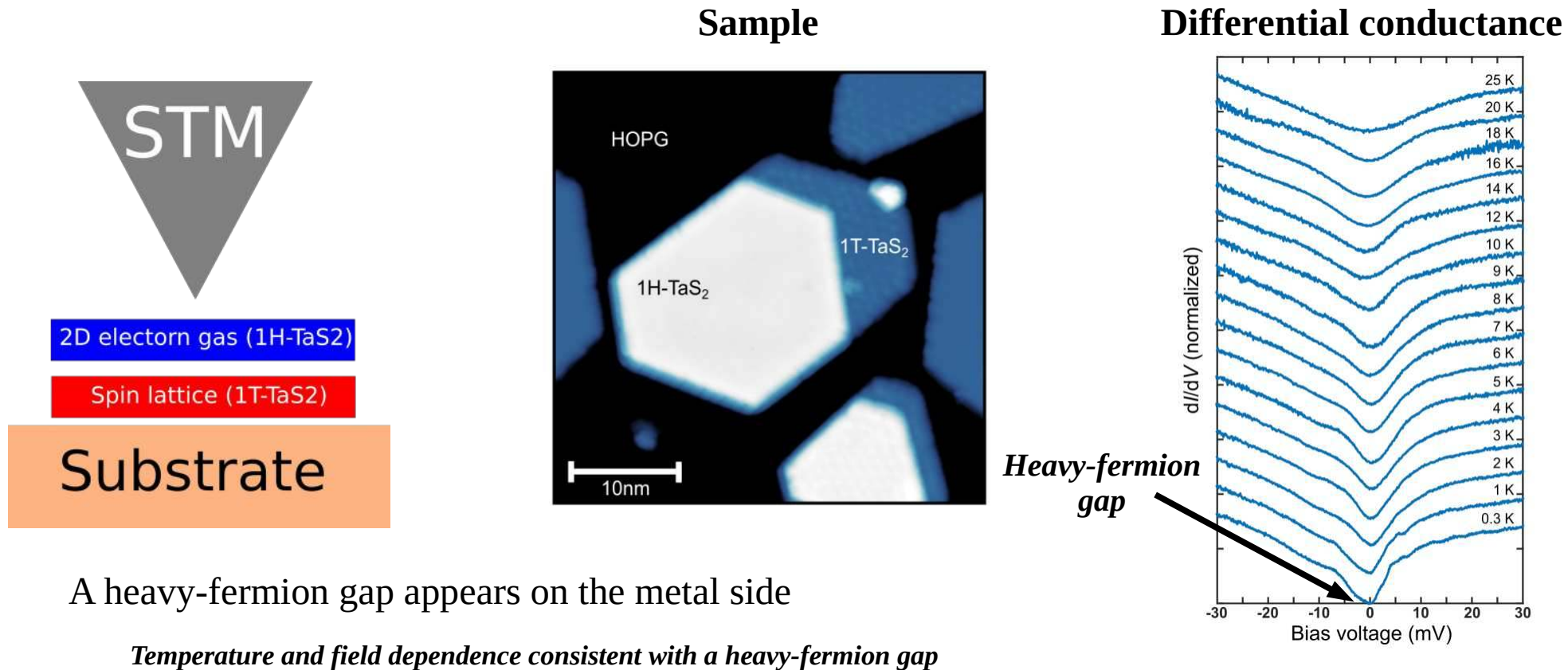
Temperature and field dependence consistent with Kondo

Kondo physics in the magnetic lattice

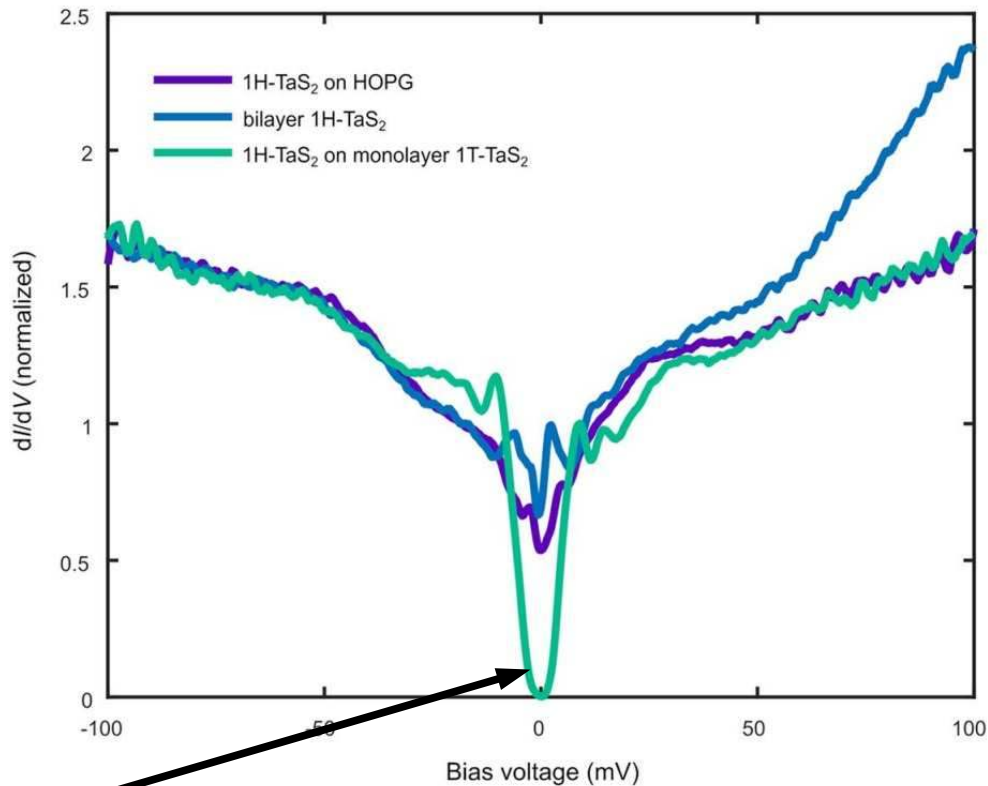
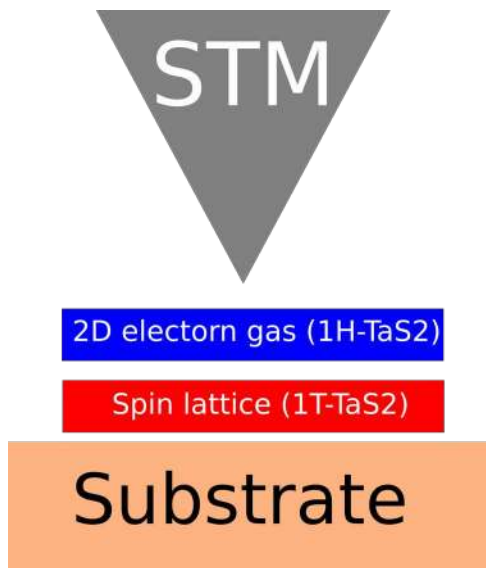


The Kondo peak follows the expected temperature dependence

Heavy-fermion gap in the 2D electron gas

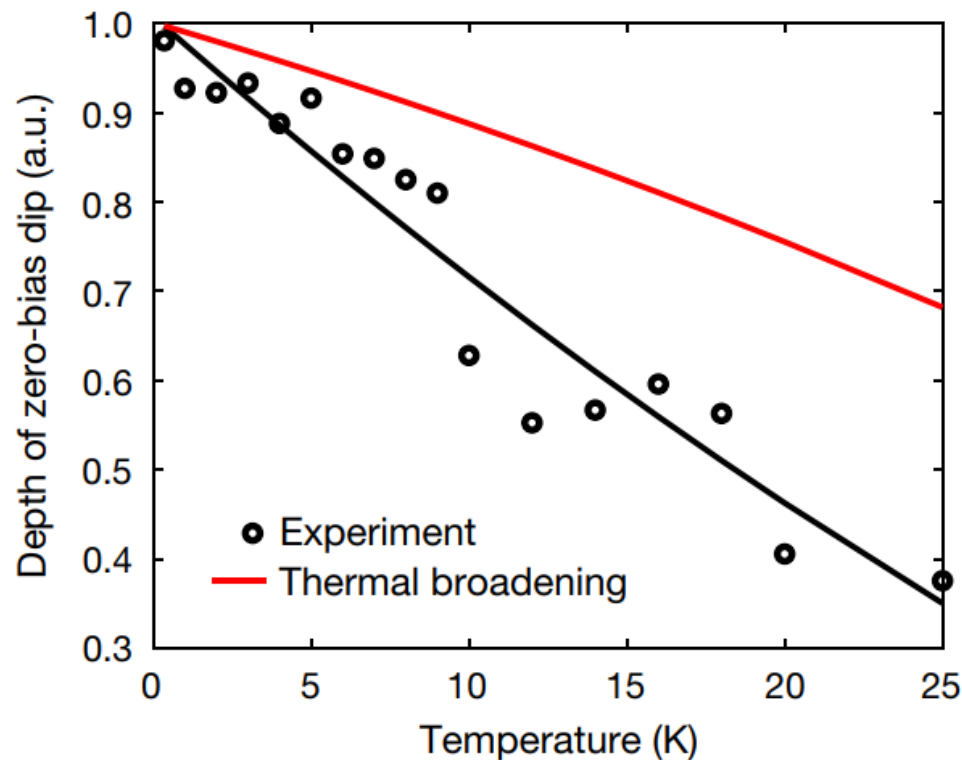
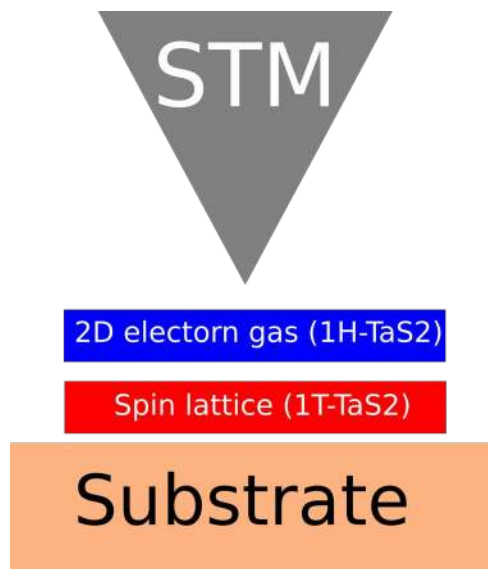


Heavy-fermion gap in the 2D electron gas



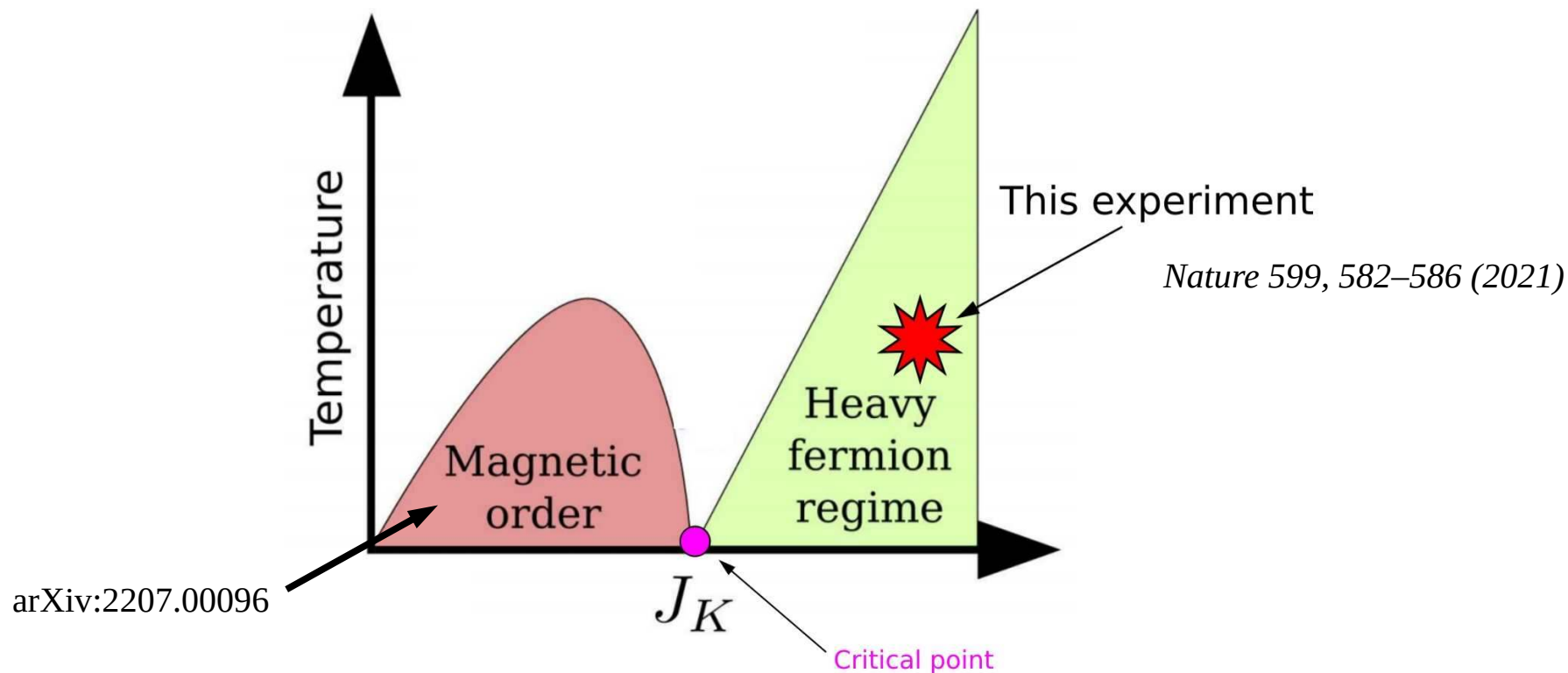
Heavy-fermion gap

Heavy-fermion gap in the 2D electron gas



The gap shows a temperature dependence beyond thermal broadening, expected from a heavy-fermion gap

Next steps in van der Waals heavy-fermions



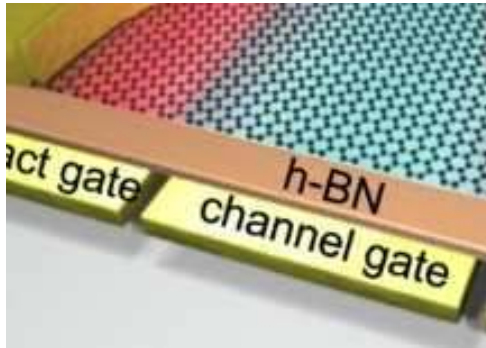
Can we push this system to critical and different unconventional superconducting regimes?

Controlling the heavy-fermion state

*Of course, we can use the typical knobs of bulk heavy fermion compounds
(pressure, chemical doping, etc)*

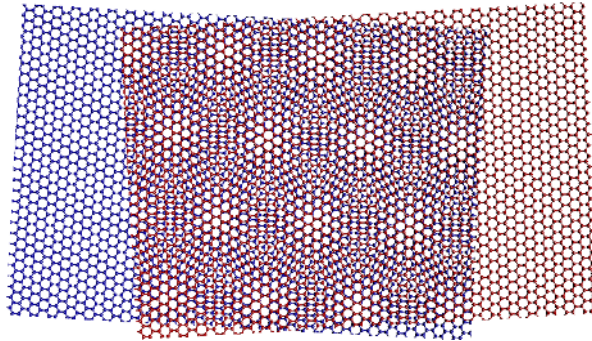
Most importantly, we can exploit the full tunability of van der Waals materials

Gating



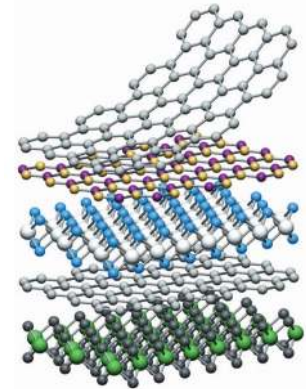
Science 306, 5696, 666-669 (2004)

Twist engineering



Nature Reviews Materials 6, 201–206 (2021)

Materials engineering

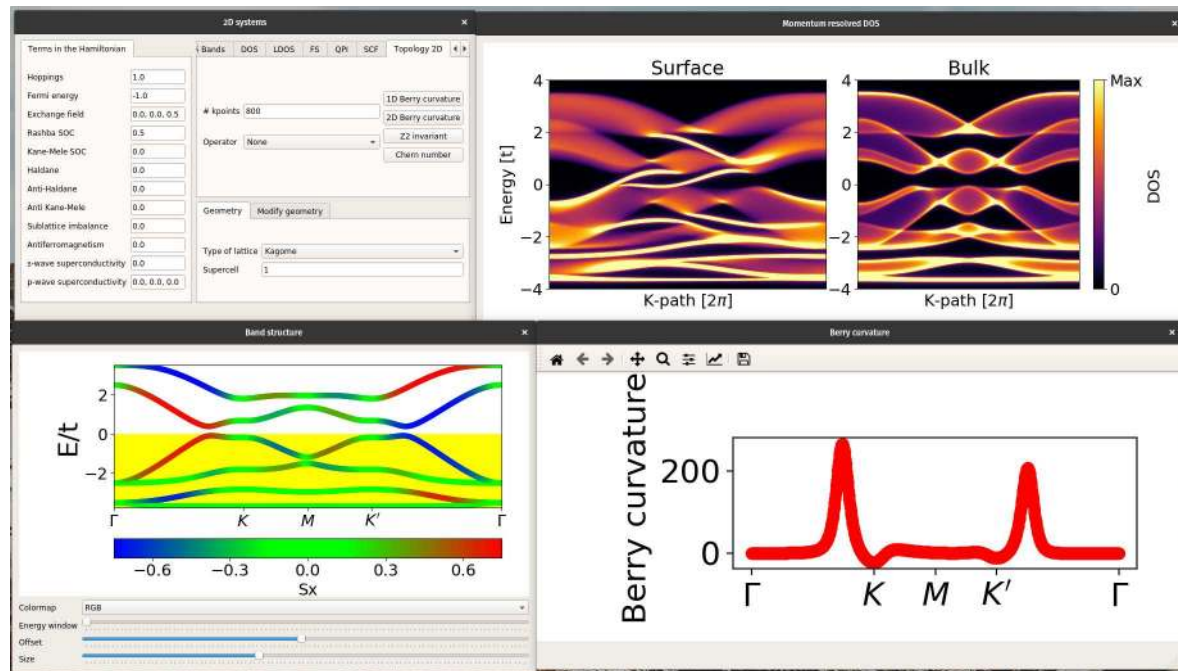
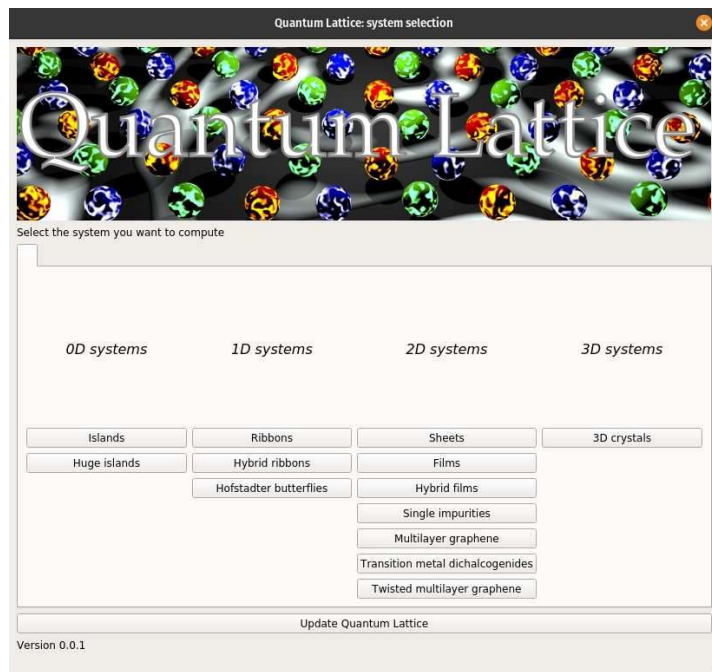


Nature 499, 419–425 (2013)

Allowing to independently control Kondo coupling, exchange coupling, doping and electronic dispersion

Computing electronic
properties

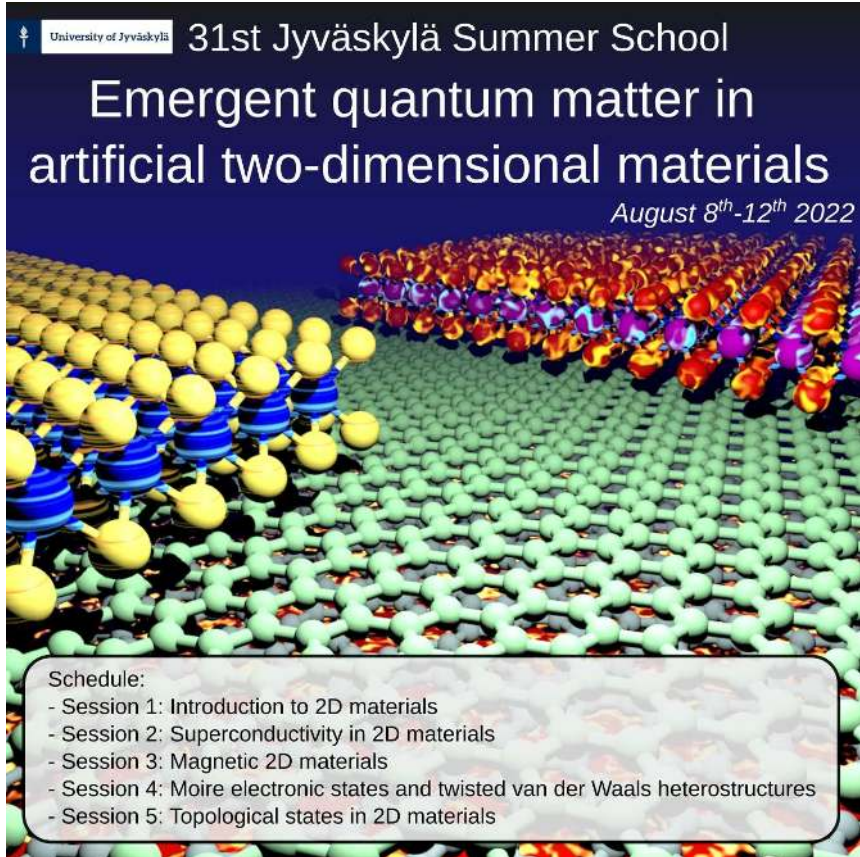
Quantum Lattice: A user interface to compute electronic properties



Quantum Lattice: open source interactive interface for tight binding modeling

<https://github.com/joselado/quantum-lattice>

Quantum matter in van der Waals materials school



University of Jyväskylä 31st Jyväskylä Summer School
Emergent quantum matter in
artificial two-dimensional materials
August 8th-12th 2022

Schedule:

- Session 1: Introduction to 2D materials
- Session 2: Superconductivity in 2D materials
- Session 3: Magnetic 2D materials
- Session 4: Moire electronic states and twisted van der Waals heterostructures
- Session 5: Topological states in 2D materials

Recordings available at

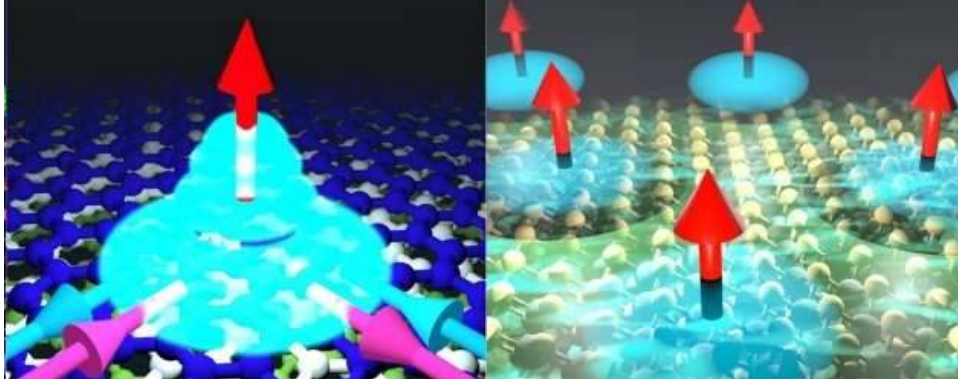


<https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ>

- Session 1: Introduction to 2D materials
- Session 2: Superconductivity in 2D materials
- Session 3: Magnetic 2D materials
- Session 4: Moire electronic states and twisted van der Waals heterostructures
- Session 5: Topological states in 2D materials

Take home

Van der Waals heterostructures provide a platform to engineer heavy-fermion physics, opening a pathway to a whole new family of emergent quantum states



Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)

Thank you!

Funding from

