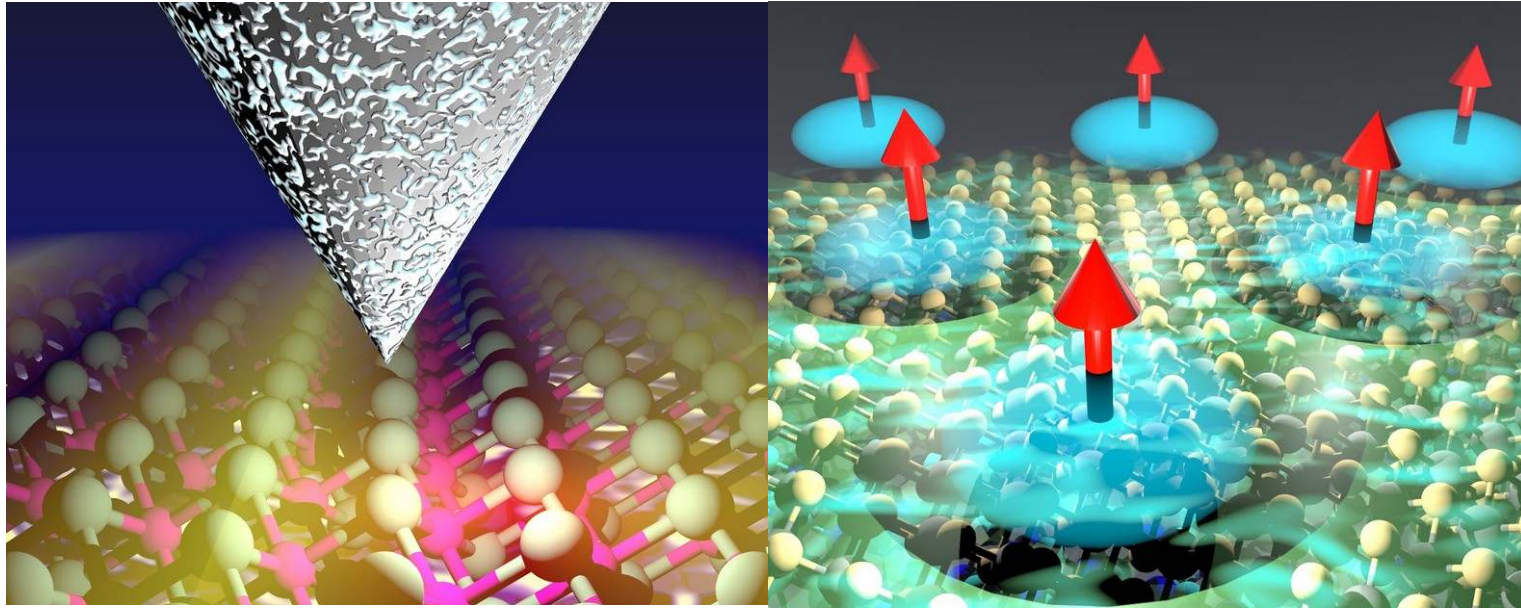


Engineering artificial quantum matter with two-dimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



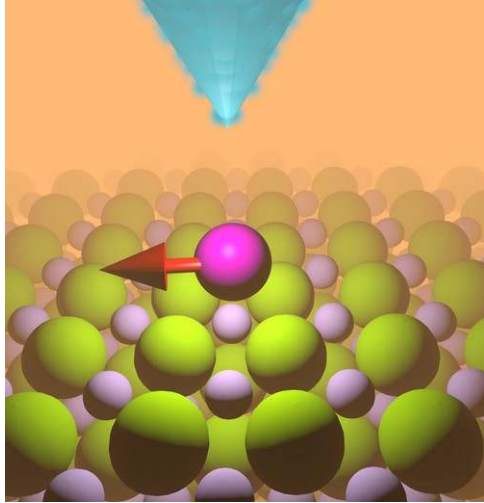
2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)
Advanced Materials, 2311342 (2024)

Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)
Nano Letters 24 (14), 4272–4278 (2024)

17th European School on Molecular Nanoscience (ESMolNa2024), Spain, May 23rd 2024

Building quantum matter with artificial lattices

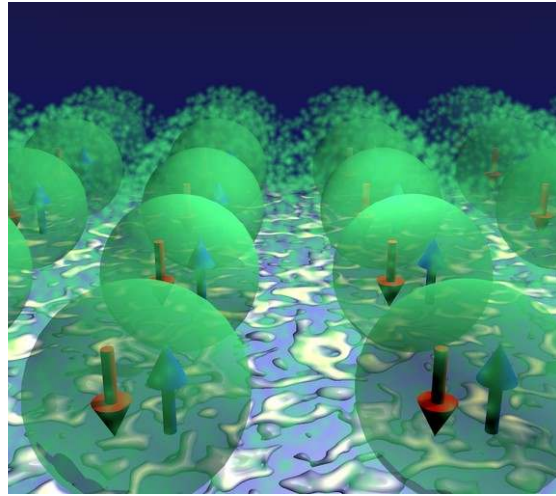
Atomic lattices



$a = 0.5 \text{ nm}$

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

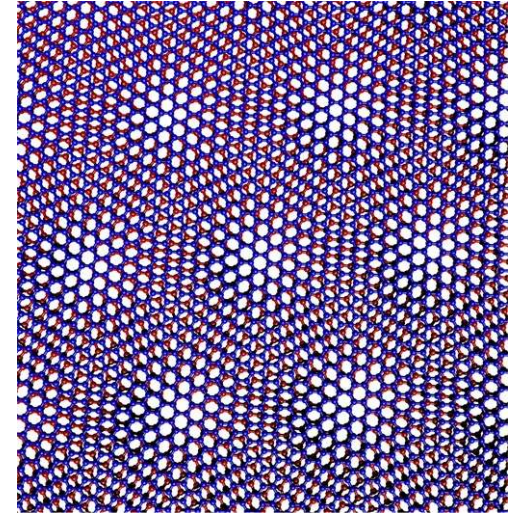
Molecular lattices



$a = 3 \text{ nm}$

Nature 408, 447–449 (2000)
Nature Rev. Mat. 5 (2), 87-104 (2020)
Nature 598, 287–292 (2021)
Phys. Rev. Lett. 130, 100401 (2023)

Twisted 2D moire materials



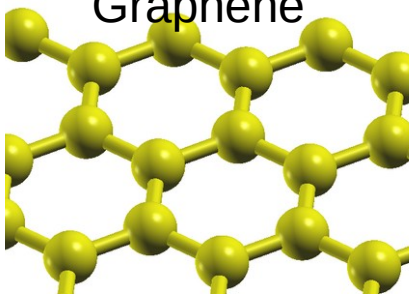
$a = 30 \text{ nm}$

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

The world of two-dimensional materials

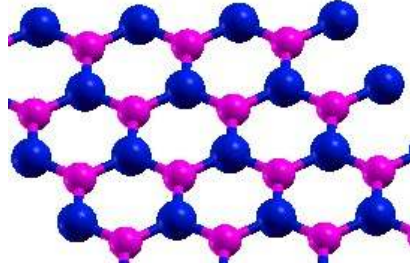
Semimetal

Graphene



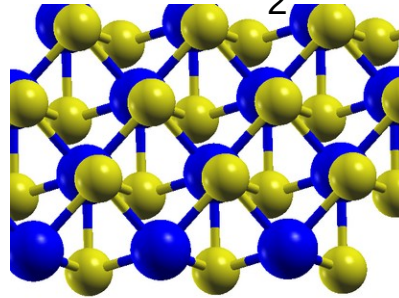
Insulator

BN



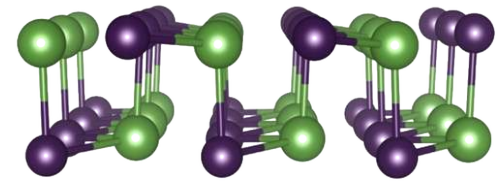
Superconductor

NbSe₂



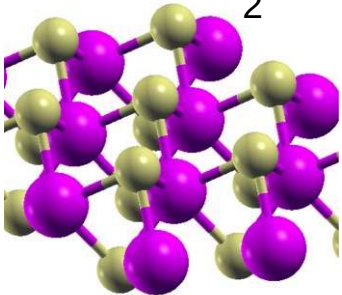
Ferroelectric

SnTe



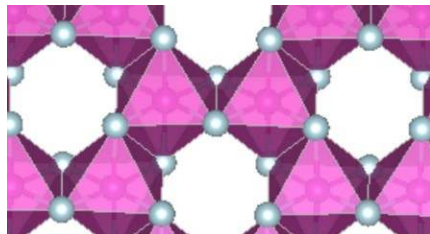
Semiconductor

WSe₂



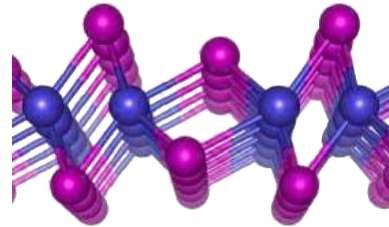
Ferromagnet

CrI₃



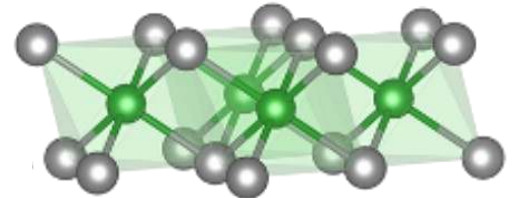
**Quantum spin
Hall insulator**

WTe₂



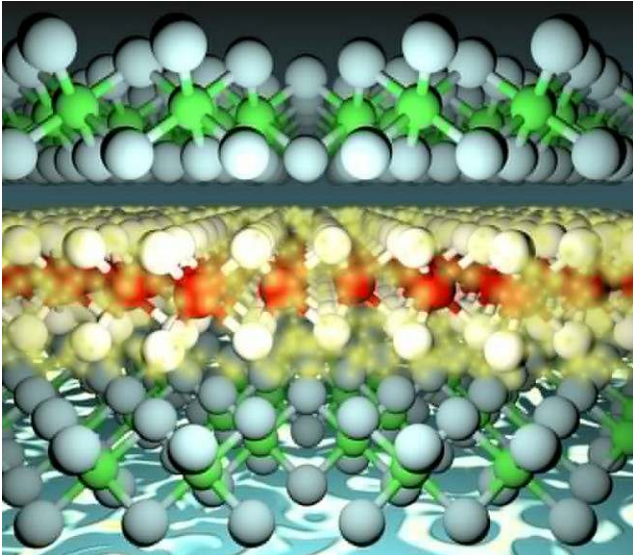
Multiferroic

NiI₂

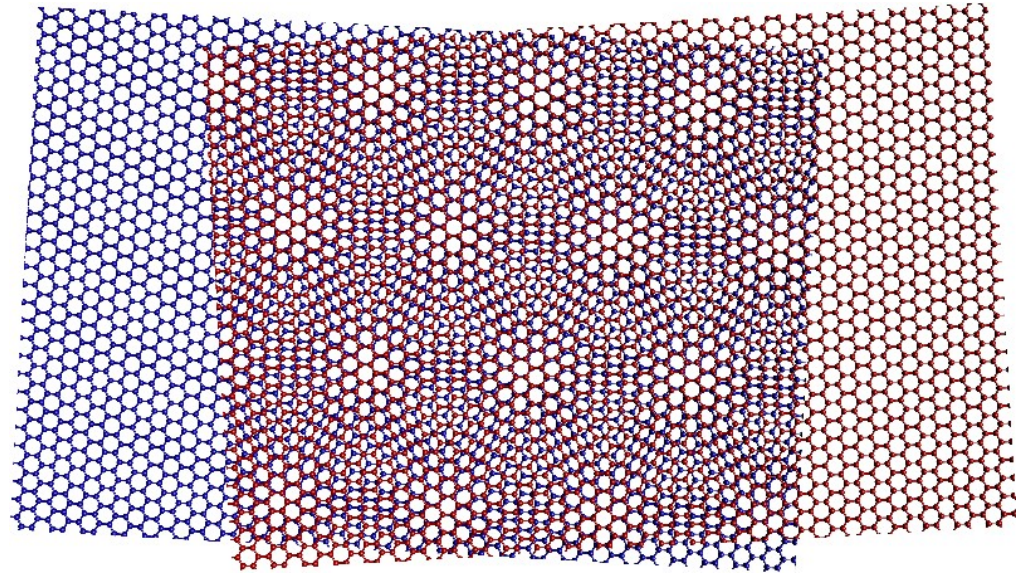


The flexibility of two-dimensional materials

They can be stacked



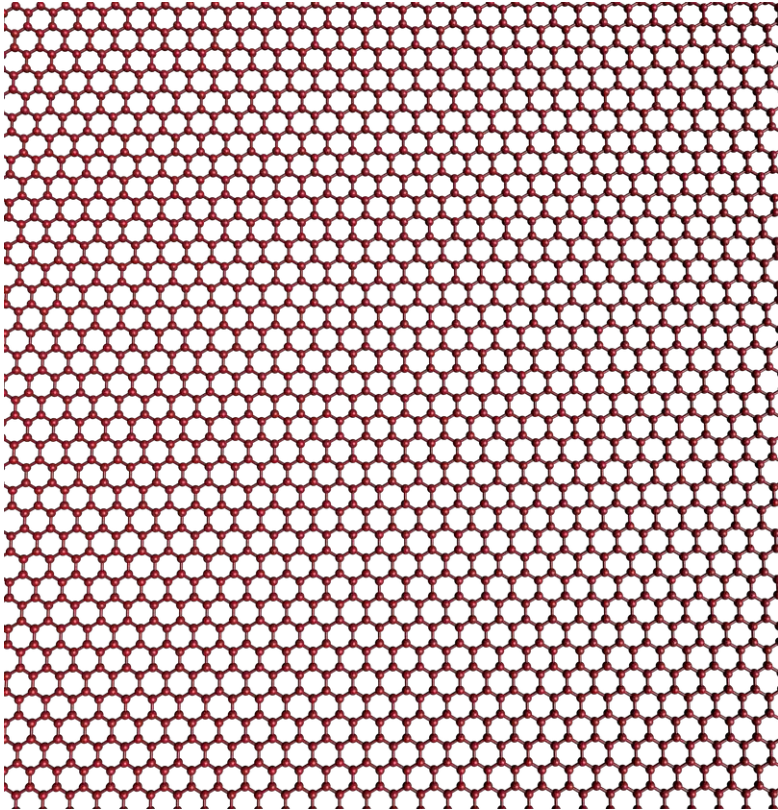
They can be rotated



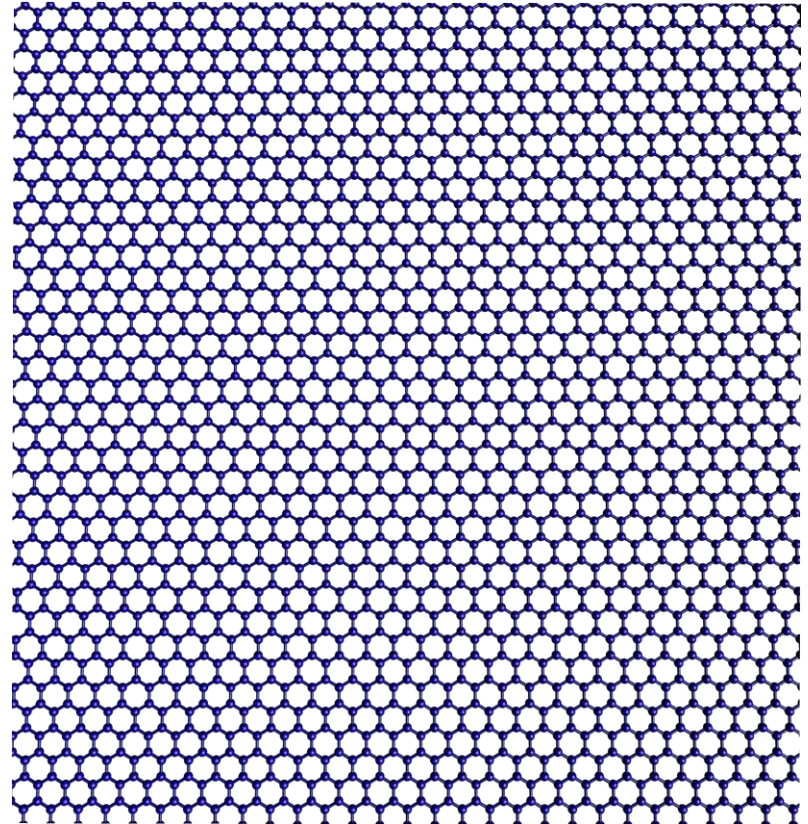
These are unique features of two-dimensional materials

Moire in twisted bilayer graphene

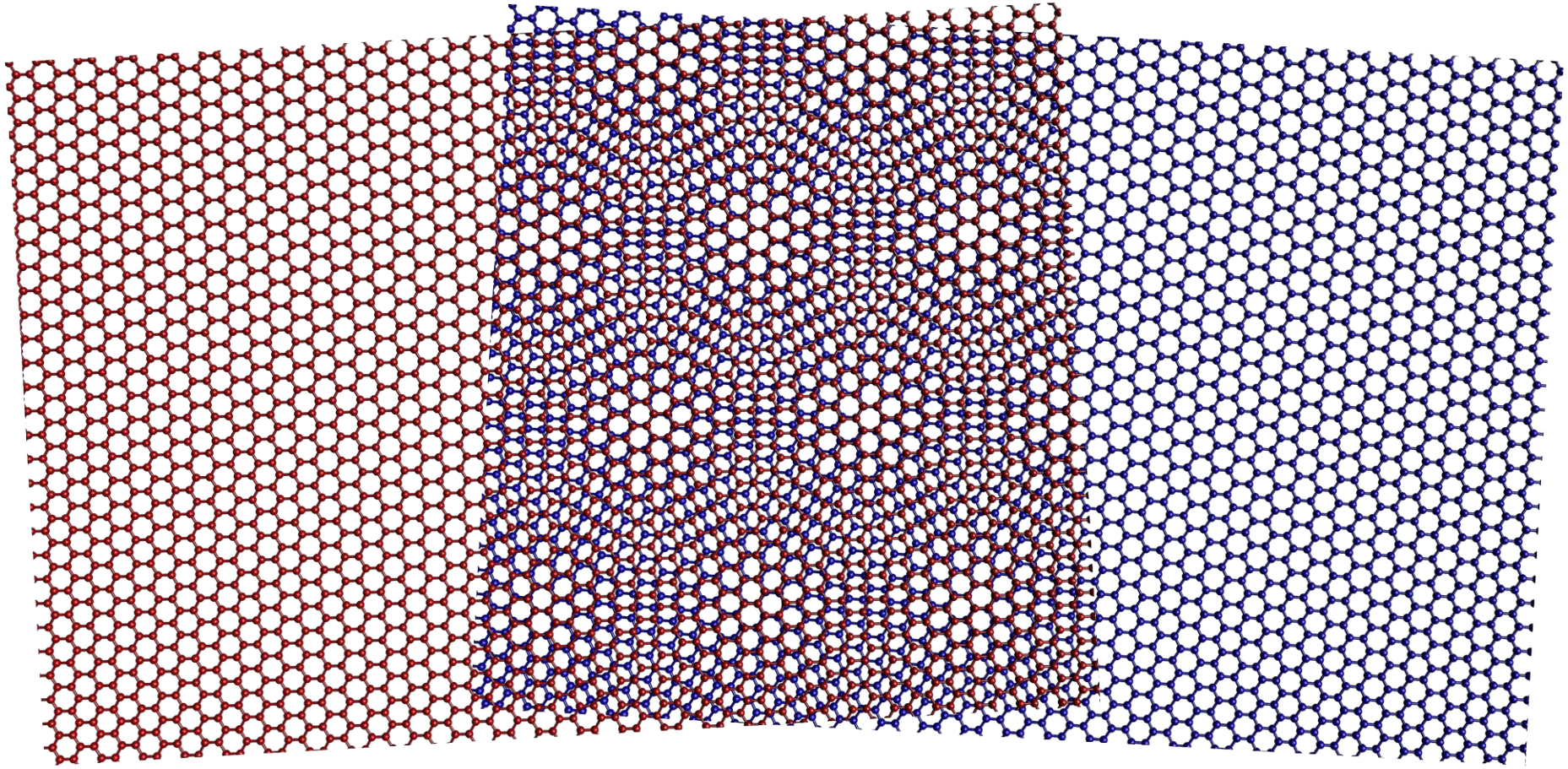
Upper graphene layer



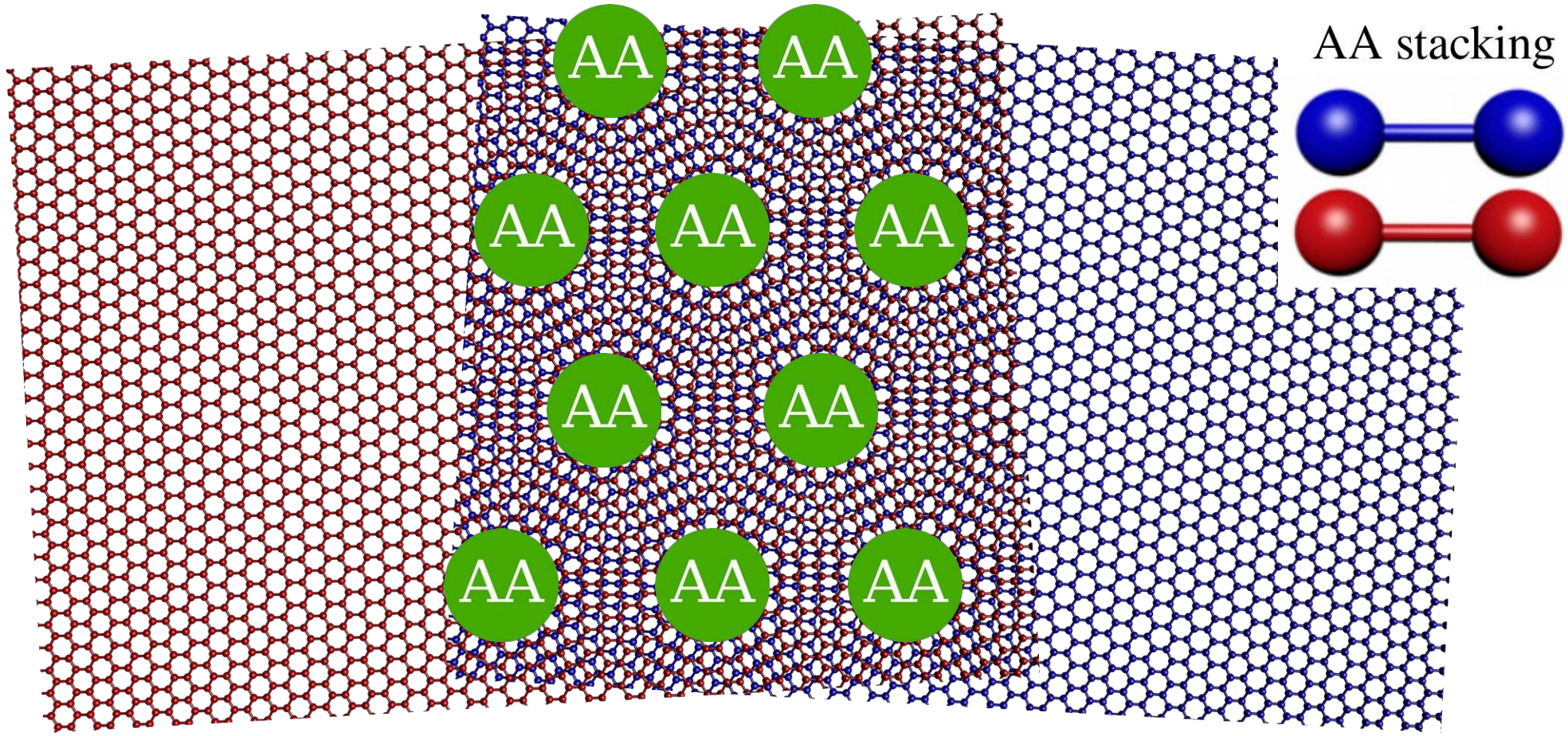
Lower graphene layer



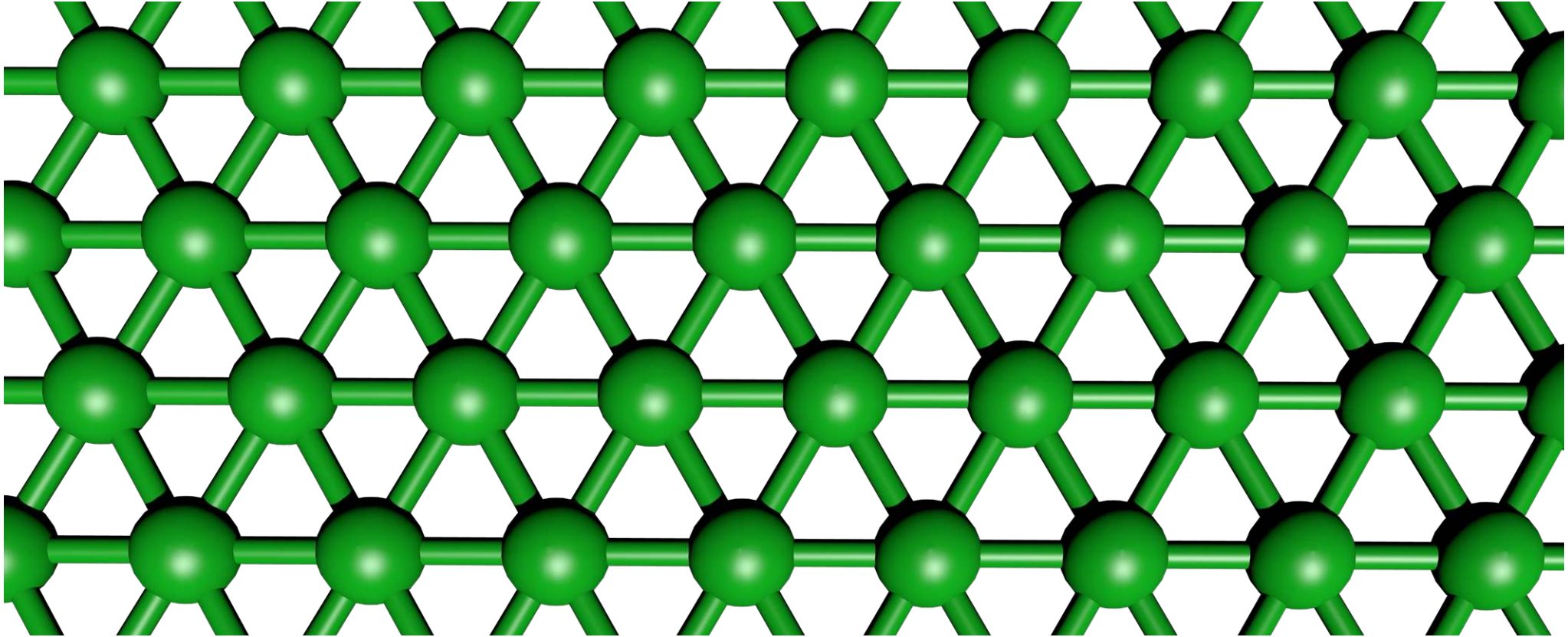
Moire in twisted bilayer graphene



Moire in twisted bilayer graphene

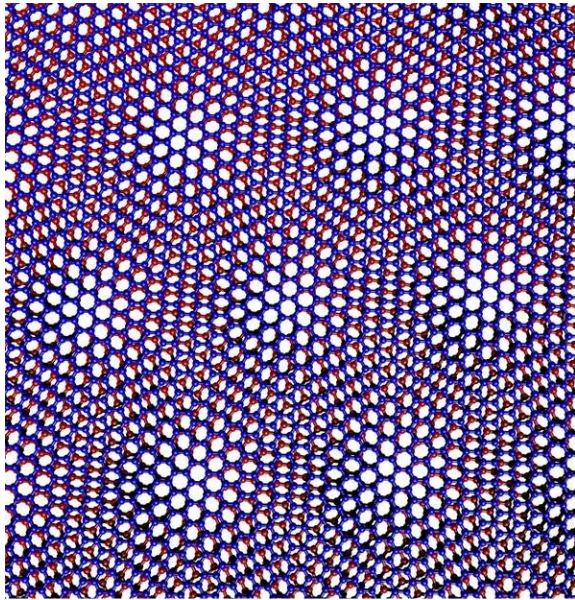


Moire in twisted bilayer graphene

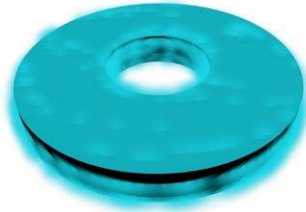


One material, a zoo of electronic phases

Twisted bilayer graphene

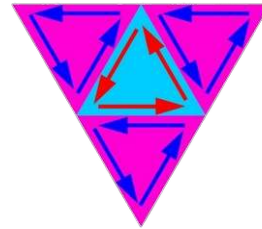


Superconductivity



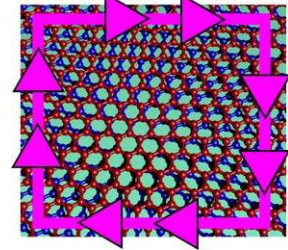
Nature 556, 43–50 (2018)
Nature 600, 240–245 (2021)

Topological networks



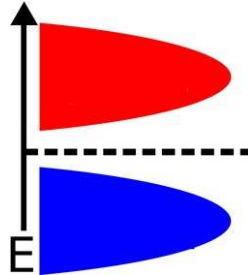
Nano Lett. 18, 11,
6725–6730 (2018)

Chern insulators



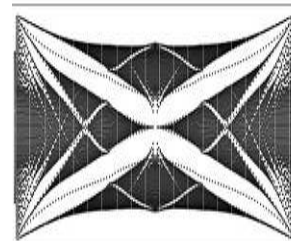
Science 365, 605–608 (2019)

Correlated insulators



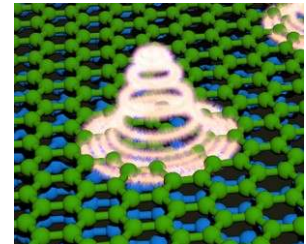
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782–786 (2018)

Fractional Chern insulators

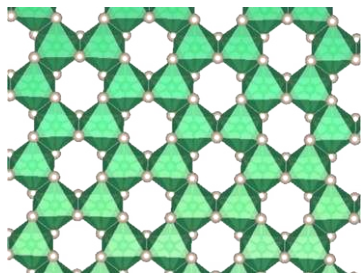


Nature 600, 439–443 (2021)

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

Van der Waals magnetic materials

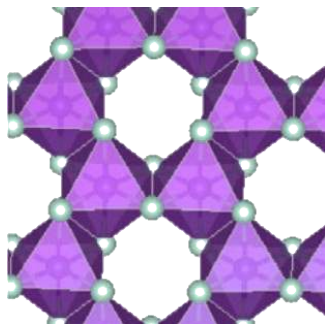
**Ferromagnet,
antiferromagnets**



CrI_3 , CrCl_3 , CrBr_3

Nature 546, 270–273 (2017)

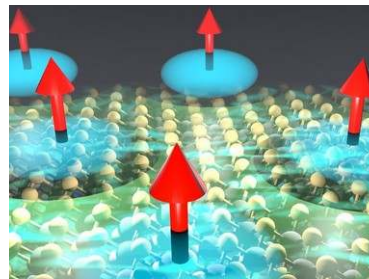
**(proximal)
Quantum
spin-liquids**



RuCl_3 , 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

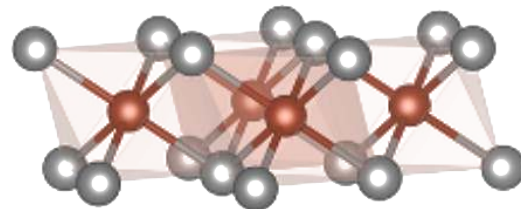
**Heavy-fermion
Kondo insulators**



$1\text{T-TaS}_2/1\text{H-TaS}_2$

Nature 599, 582–586 (2021)

Multiferroics



NiI_2

Nature 602, 601–605 (2022)

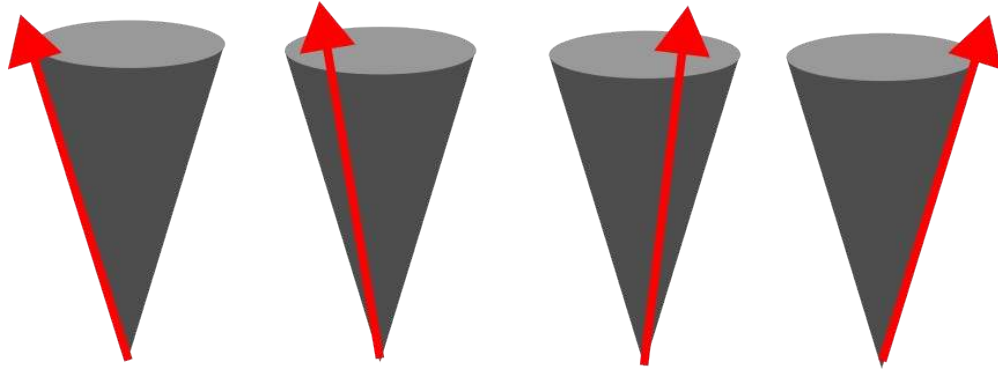
Break time-reversal

Do not break time-reversal

Break time-reversal
and inversion symmetry

Emergent excitations in van der Waals magnets

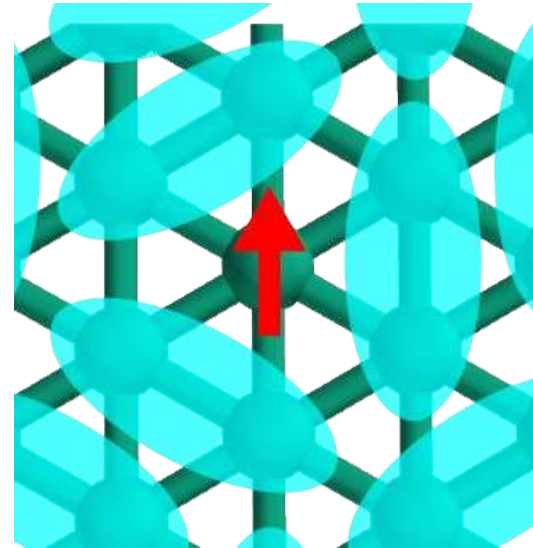
Magnons



$S=1$
No charge

Instrumental for magnonics

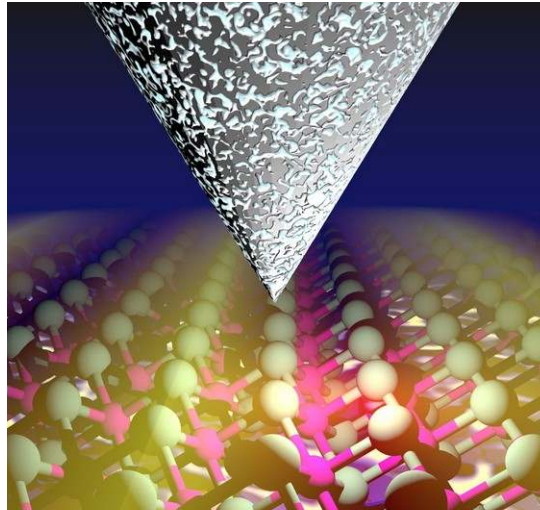
Spinons



$S=1/2$
No charge

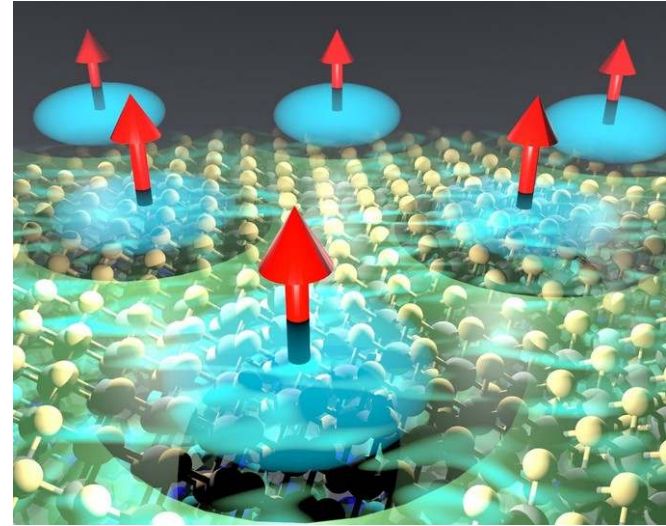
Plan for today

Engineering and probing van der Waals multiferroics



2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)
Advanced Materials, 2311342 (2024)

Engineering and probing van der Waals heavy fermion Kondo magnets



Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)
Nano Letters 24 (14), 4272–4278 (2024)

A reminder about
magnetism

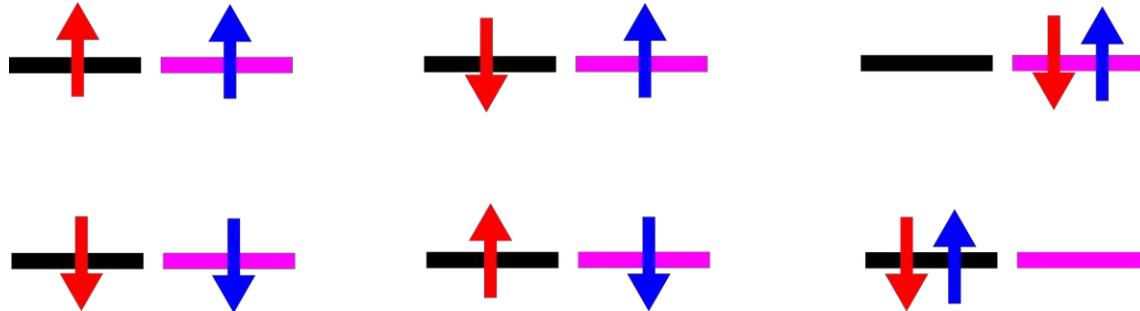
A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$



The full Hilbert space at half filling is

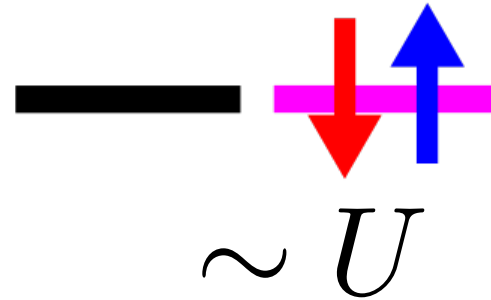
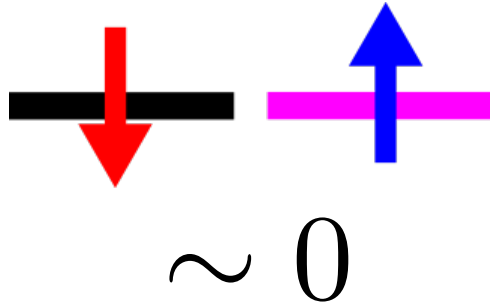


A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

The energies in the strongly interacting limit are $U \gg t$



A reminder about magnetism, the origin of local moments

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$



The low energy manifold is



Just one electron in each site for $U \gg t$

Local $S=1/2$ at each site

A reminder about magnetism, the origin of the Heisenberg model

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

In the strongly correlated (half-filled) limit we obtain a Heisenberg model

$$\mathcal{H} = J \vec{S}_0 \cdot \vec{S}_1$$

$$J \sim \frac{|t|^2}{U}$$

The lattice Heisenberg model

For a generic lattice and a generic interacting model, the Hamiltonian is

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

In those generic cases, the exchange couplings can be positive or negative

$$J_{ij} > 0$$

Antiferromagnetic coupling

$$J_{ij} < 0$$

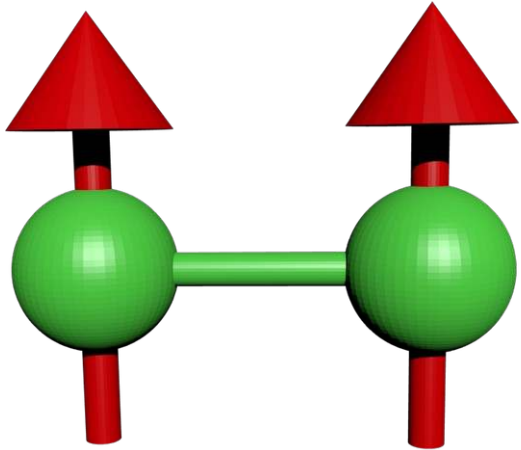
Ferromagnetic coupling

Spin-orbit coupling introduces anisotropic couplings

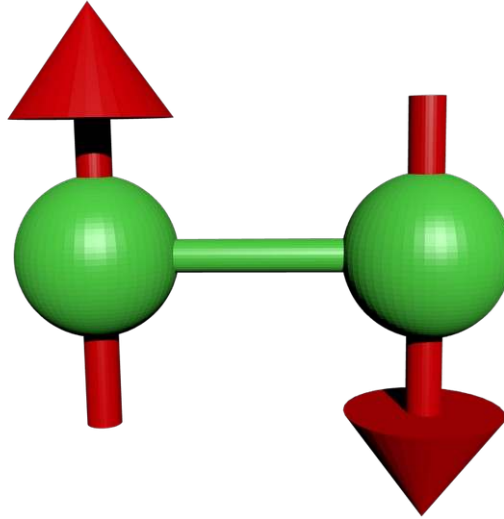
$$\mathcal{H} = \sum_{ij} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta$$

The classical Heisenberg model

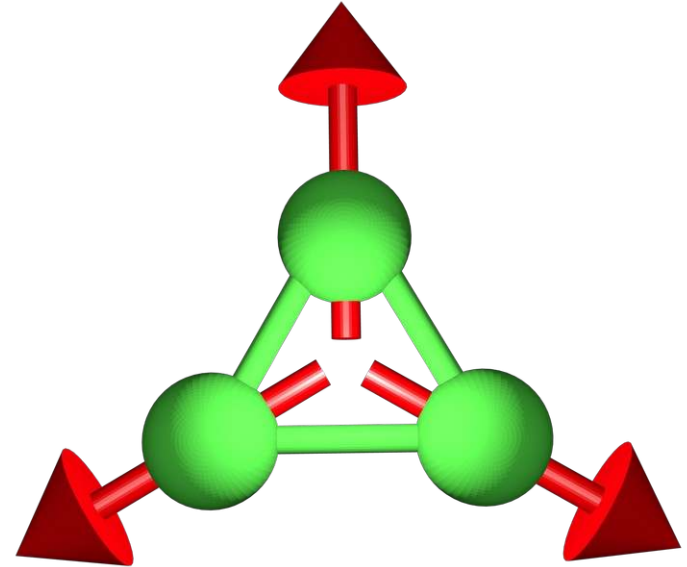
Ferromagnetism



Antiferromagnetism



Frustrated magnetism



The quantum Heisenberg model

What is the ground state of the quantum Heisenberg Hamiltonian?

$$\mathcal{H} = J \vec{S}_0 \cdot \vec{S}_1 \quad [S_j^\alpha, S_j^\beta] = i\epsilon_{\alpha\beta\gamma} S_j^\gamma$$

The ground state is unique, and does not break time-reversal

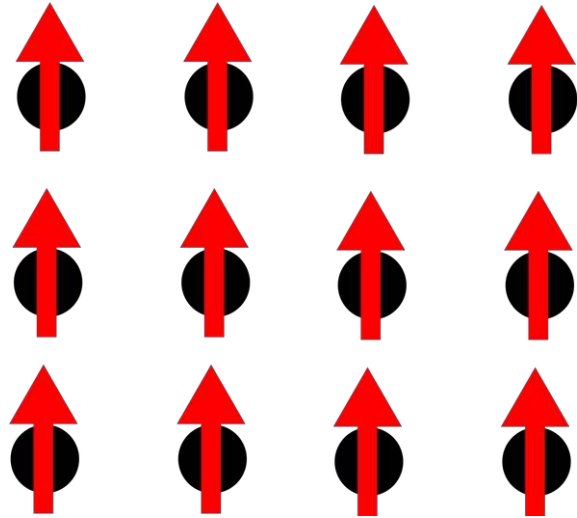
$$|GS\rangle = \frac{1}{\sqrt{2}} [|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle] \quad \langle \vec{S}_i \rangle = 0$$

The state is maximally entangled

A quantum magnet is the macroscopic version of the previous state $\langle \vec{S}_i \rangle = 0$

Classical and quantum magnets

Ferromagnet, antiferromagnets & helimagnets

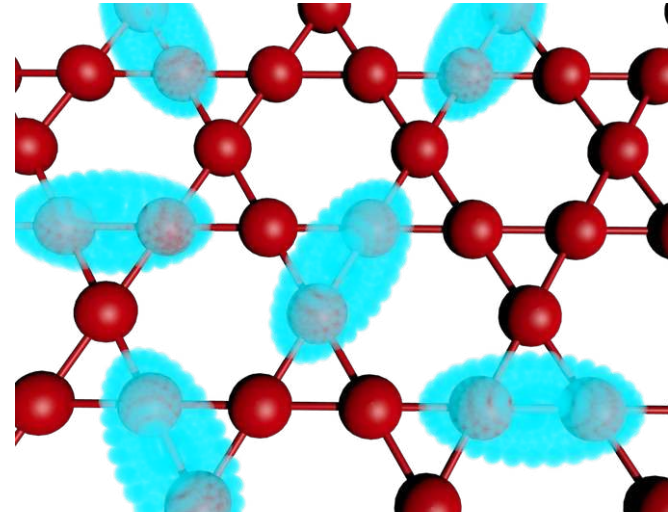


$$\langle \vec{S}_n \rangle \neq 0$$

They break time-reversal symmetry

Classical magnets

Quantum spin liquids and heavy fermion Kondo magnets



$$\langle \vec{S}_n \rangle = 0$$

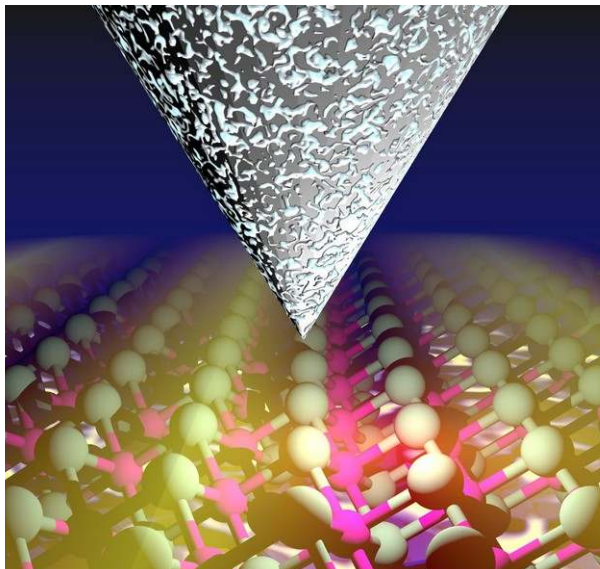
They do not break time-reversal symmetry

Quantum entangled magnets

Van der Waals multiferroic magnets

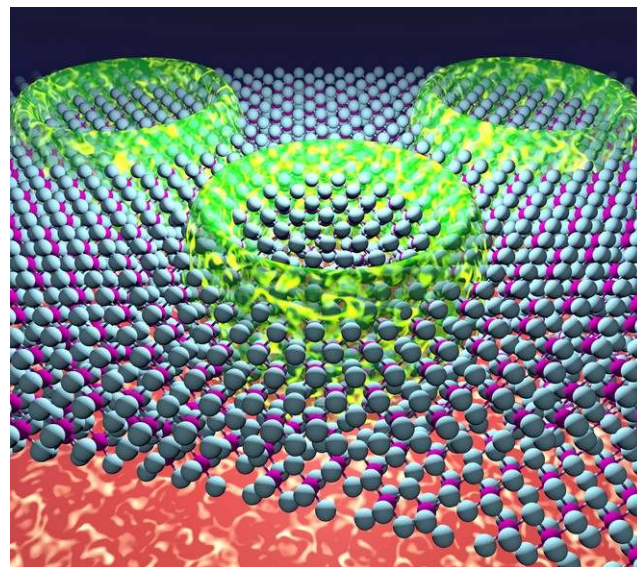
Plan for 2D multiferroics

Multiferroicity in NiI_2



2D Materials 9 (2), 025010 (2022)
Advanced Materials, 2311342 (2024)

Artificial multiferroicity by twisting 2D magnets



2D Materials 10 (2), 025026 (2023)

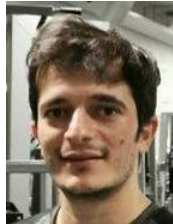
Behind the scenes

Intrinsic multiferroics, NiI_2 (theory and experiment)

V. Vaňo



M. Amini



A. Fumega



H. Gonzalez
Herrero



S. Kezilebieke



P. Liljeroth



2D Materials 9 (2), 025010 (2022)
Advanced Materials, 2311342 (2024)

Artificial multiferroics, twisted CrBr_3 bilayers

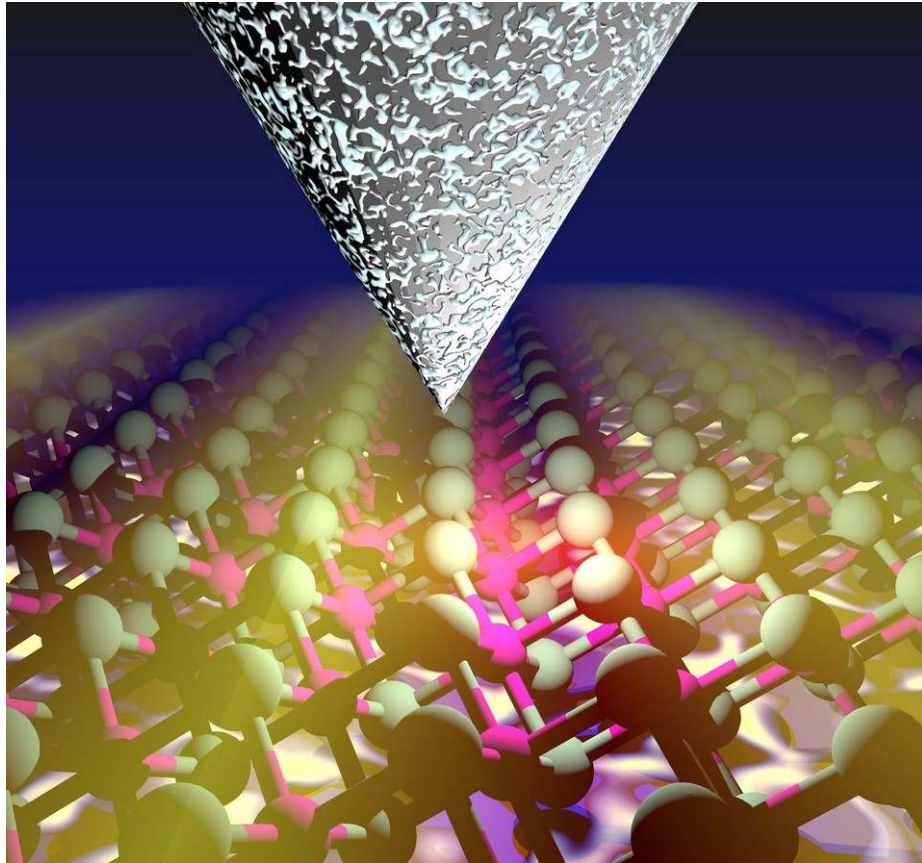
(theory)

A. Fumega



2D Materials 10 (2), 025026 (2023)

Origin and visualization of multiferroicity in NiI_2



V. Vaňo



M. Amini



A. Fumega



H. Gonzalez
Herrero



S. Kezilebieke



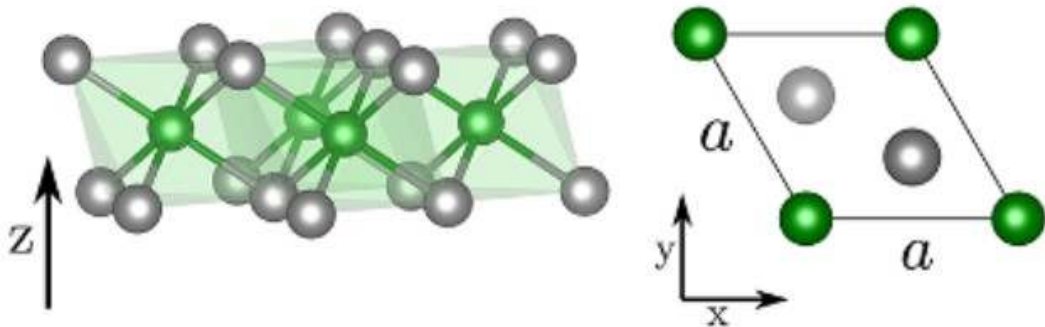
P. Liljeroth



2D Materials 9 (2), 025010 (2022)
Advanced Materials, 2311342 (2024)

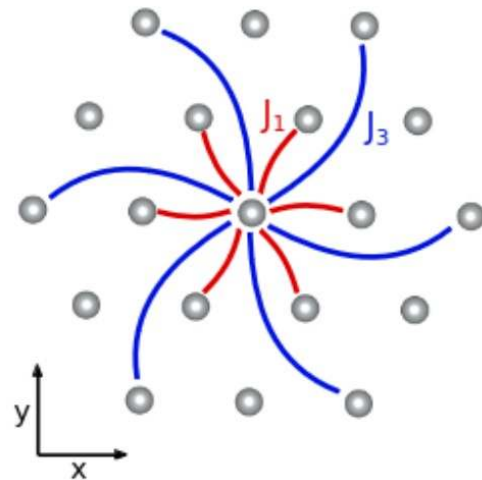
Monolayer NiI_2

Structure

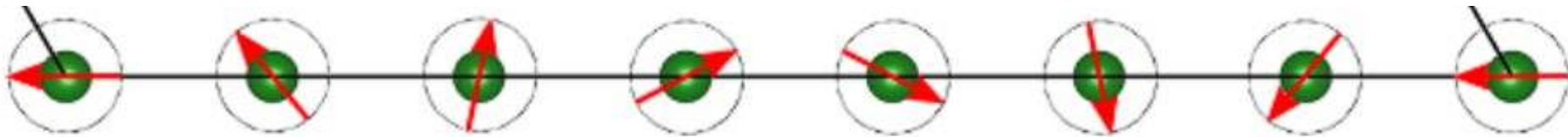


Nature 602, 601–605 (2022)

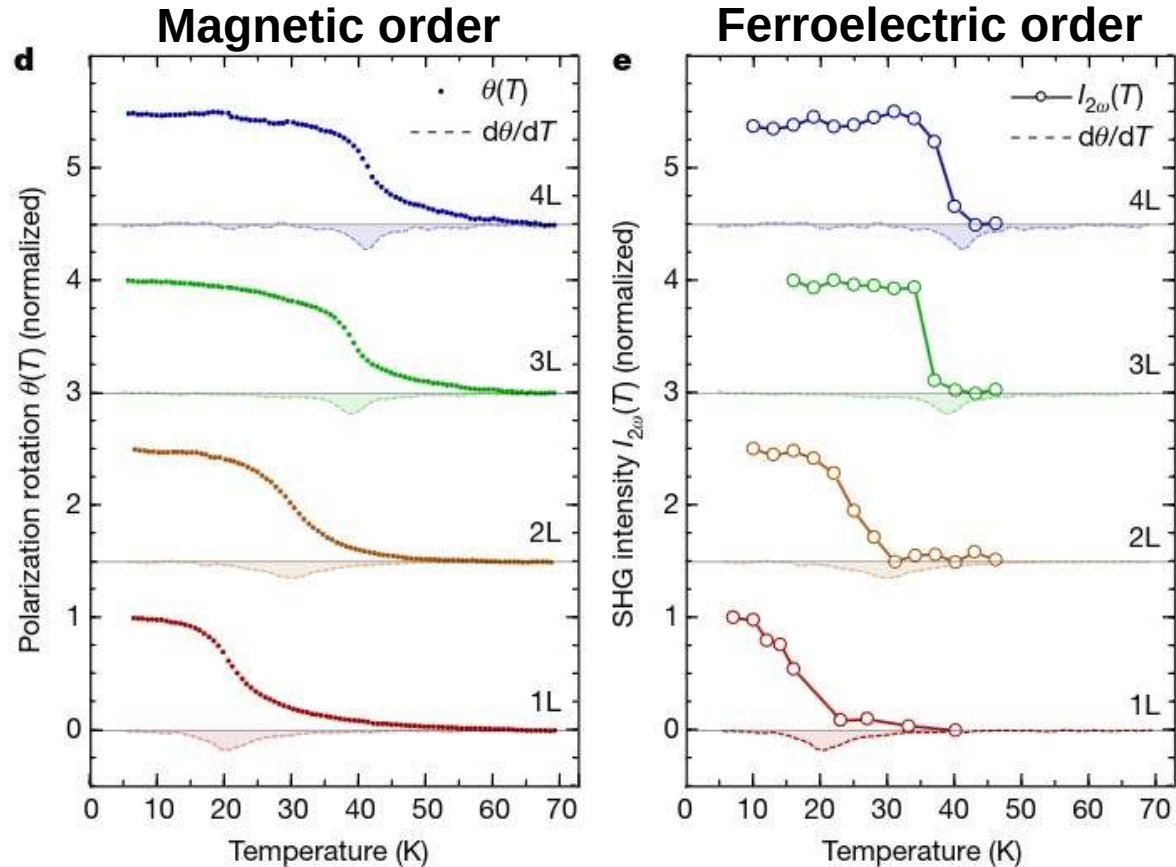
Frustrated magnetic
Heisenberg model



Magnetic structure



Multiferroicity in monolayer NiI_2



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

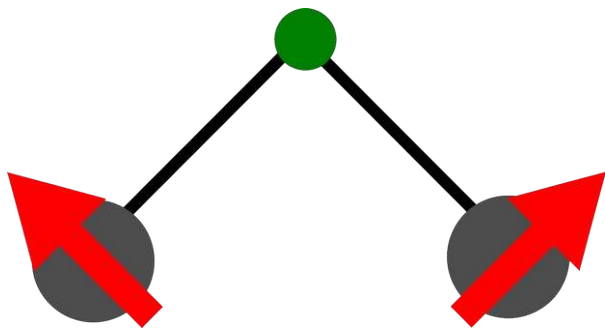
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

In the presence of spin-orbit coupling, new terms can appear in the Hamiltonian

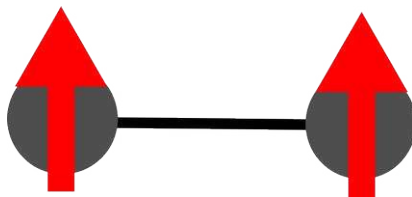
Antisymmetric exchange

$$(\mathbf{r}_{ik} \times \mathbf{r}_{kj}) \cdot \vec{S}_i \times \vec{S}_j$$



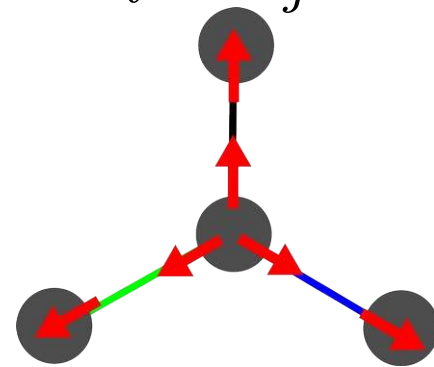
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

$$S_i^{\alpha(i)} S_j^{\alpha(j)}$$



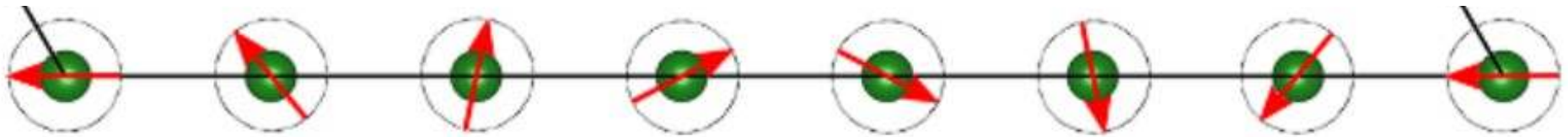
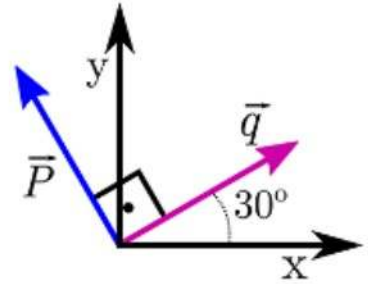
In NiI_2 , the non-collinear order is promoted by isotropic exchange

In NiI_2 , the ferroelectricity is created by inverse antisymmetric exchange

The origin of ferroelectricity in NiI_2

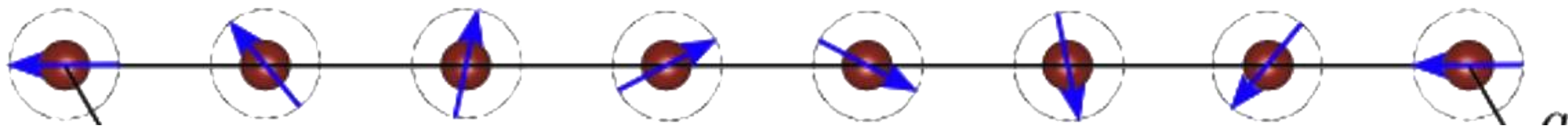
Magnetically driven ferroelectric order $\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$

$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e} \cdot \mathbf{S})(\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}_0))} \mathbf{M}(\mathbf{r}_0)$$



The emergence of non-collinear order is drives ferroelectricity

The origin of ferroelectricity in NiI_2



$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e}$$

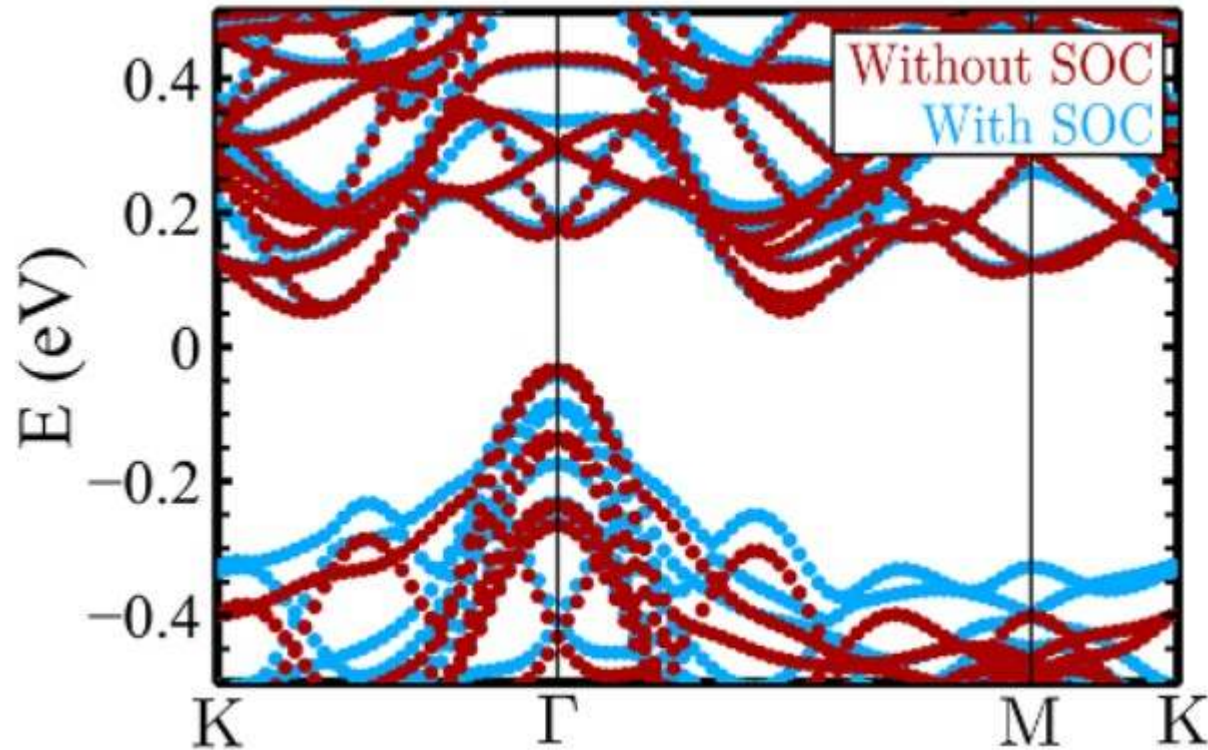
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

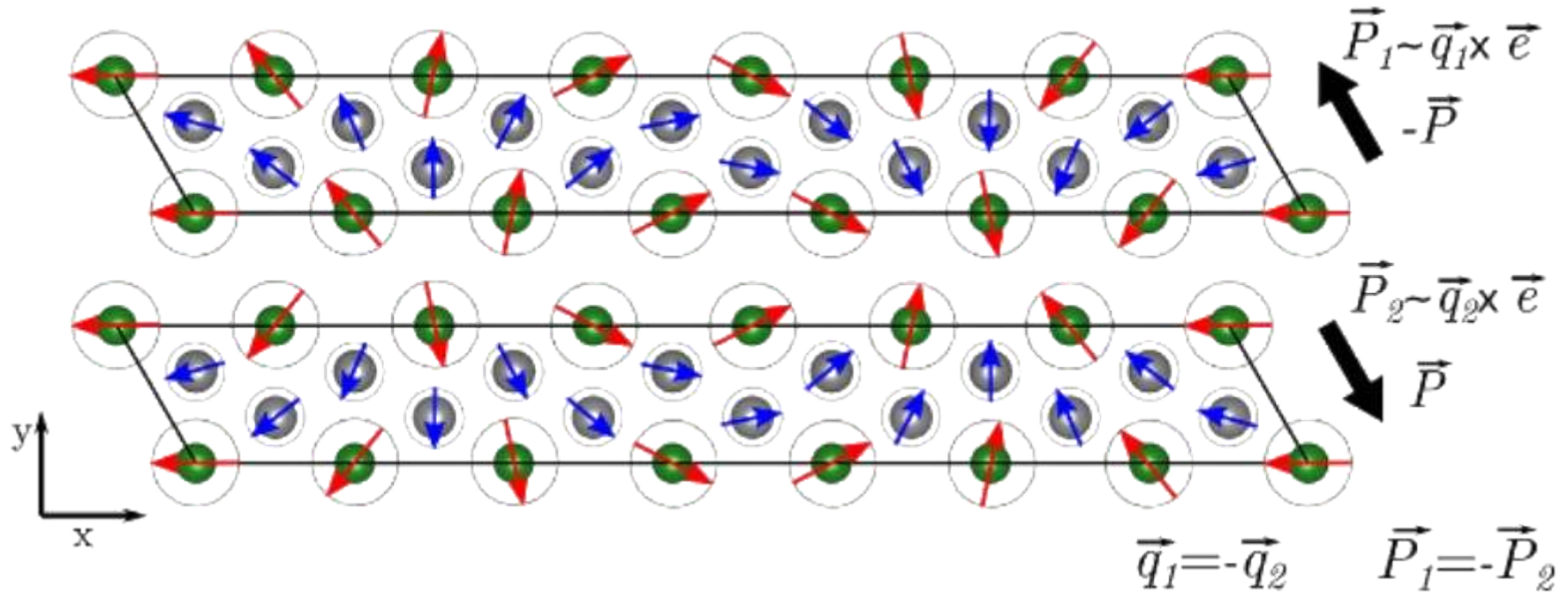
The impact of spin-orbit coupling in the electronic structure



Spin-orbit coupling introduces corrections of 100 meV close to the Fermi level

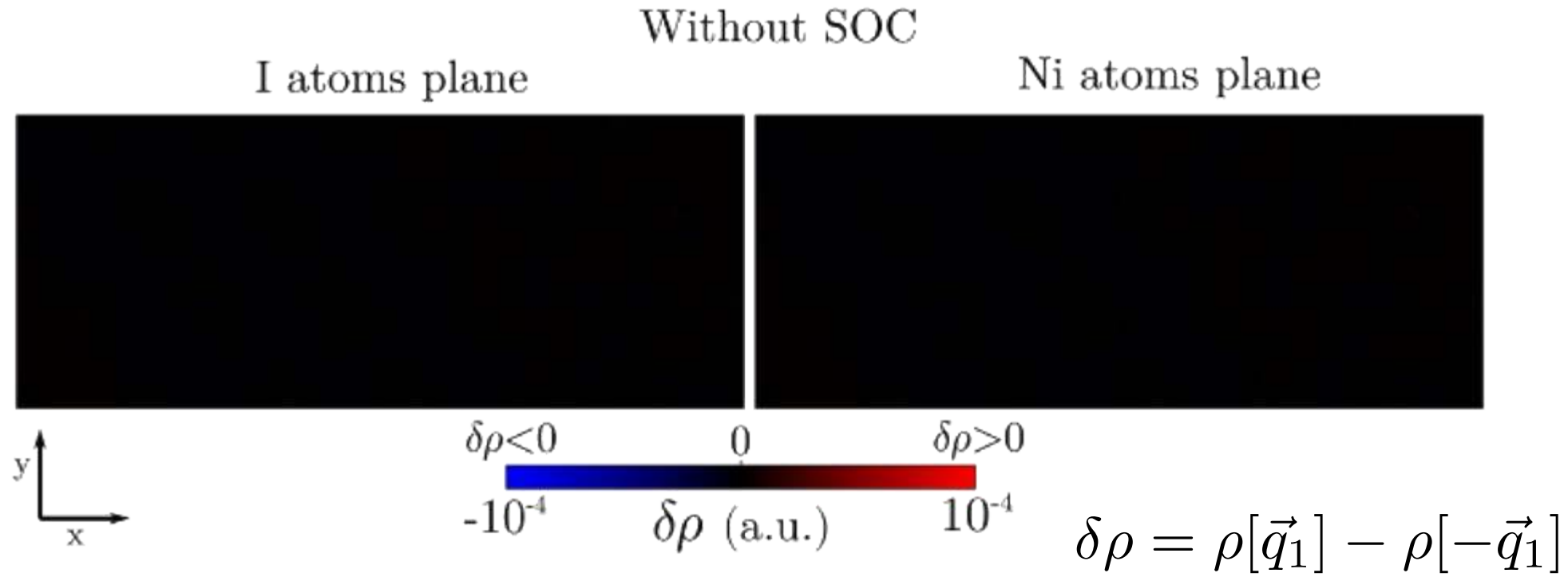
There is a major role of the halide close to the Fermi level

Ferroelectric correction to the electronic density



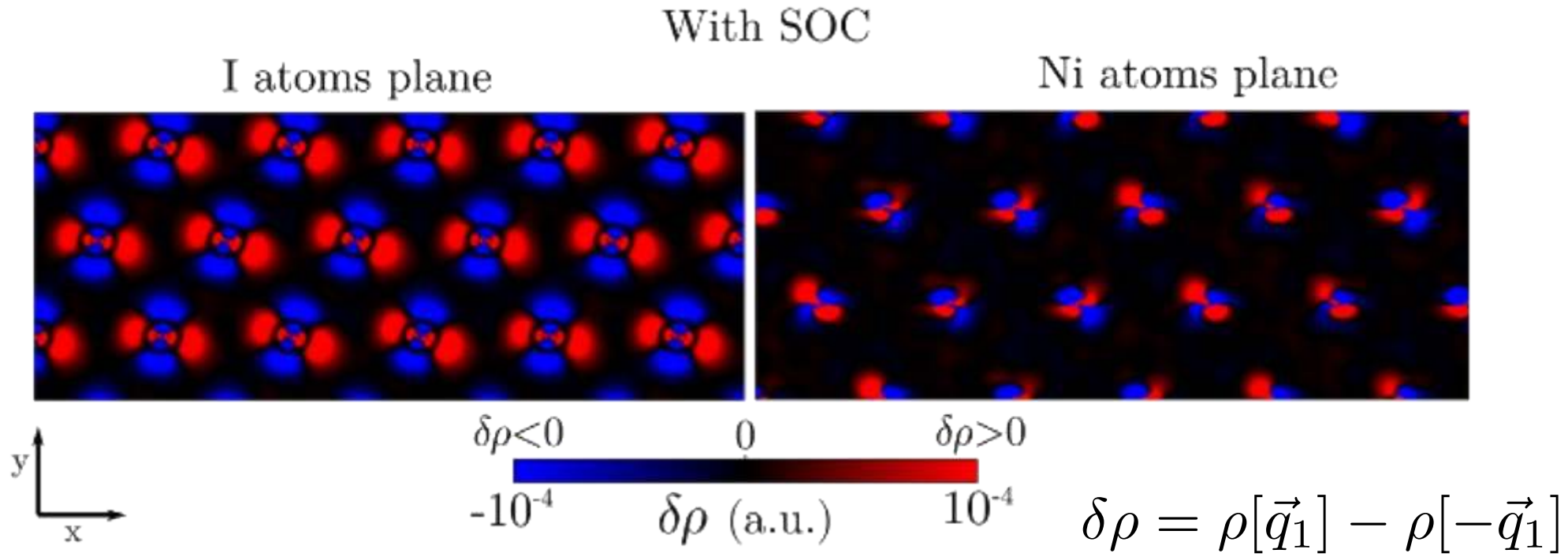
We will examine the ferroelectric density correction $\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$

Ferroelectric correction to the electronic density



In the absence of SOC, no ferroelectric density appears

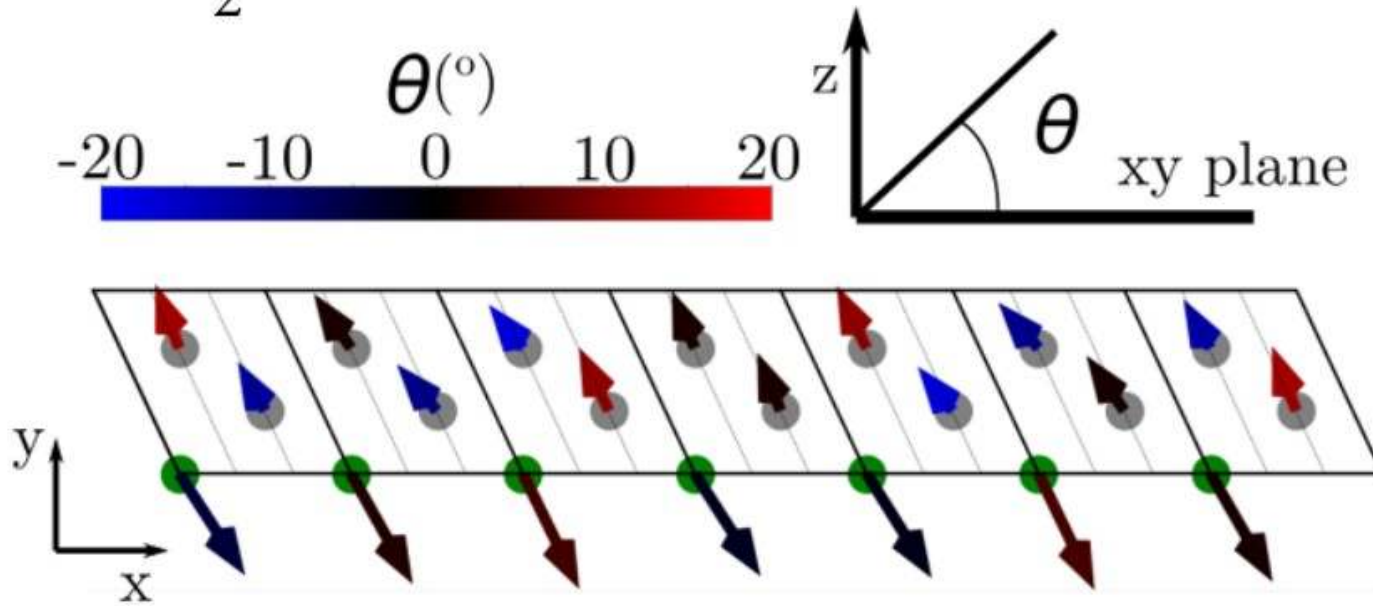
Ferroelectric correction to the electronic density



In the presence of SOC, a ferroelectric density appears both in Ni and I

The ferroelectric force

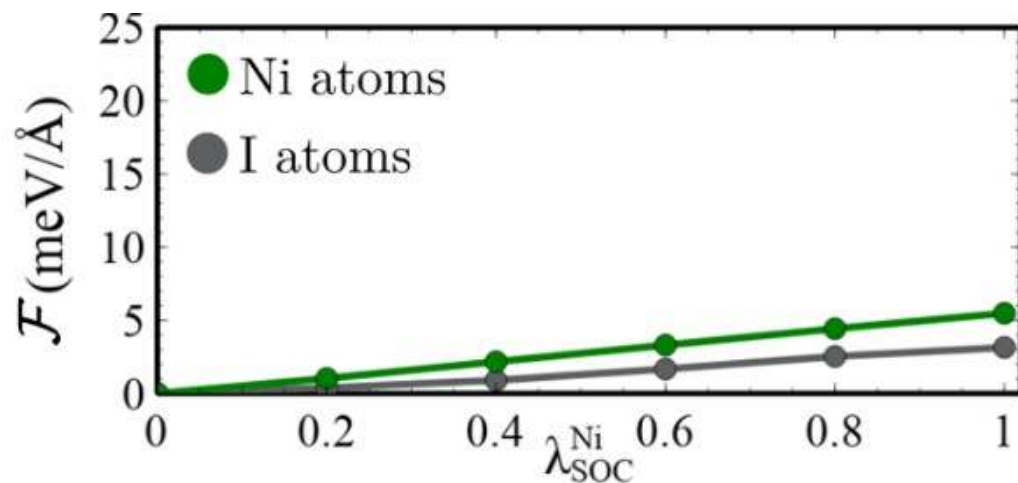
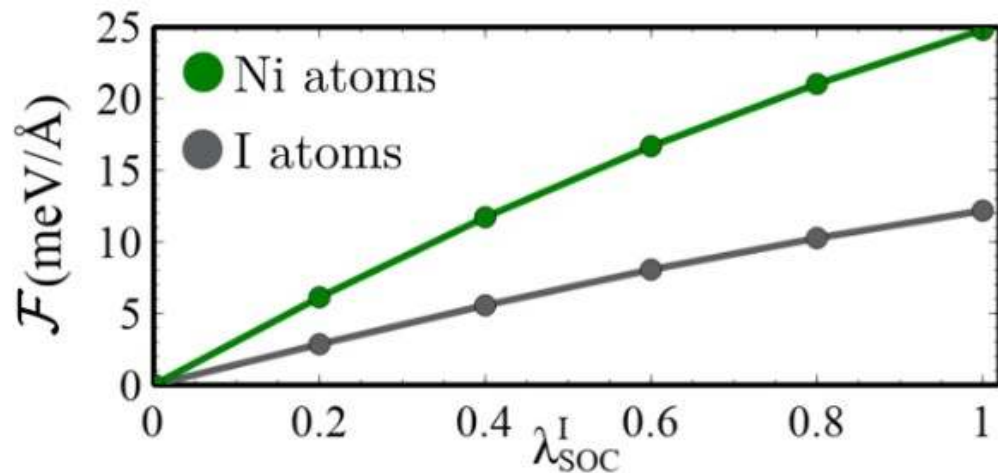
$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$



The forces between the two magnetic configuration account for the ferroelectric displacement

The ferroelectric force

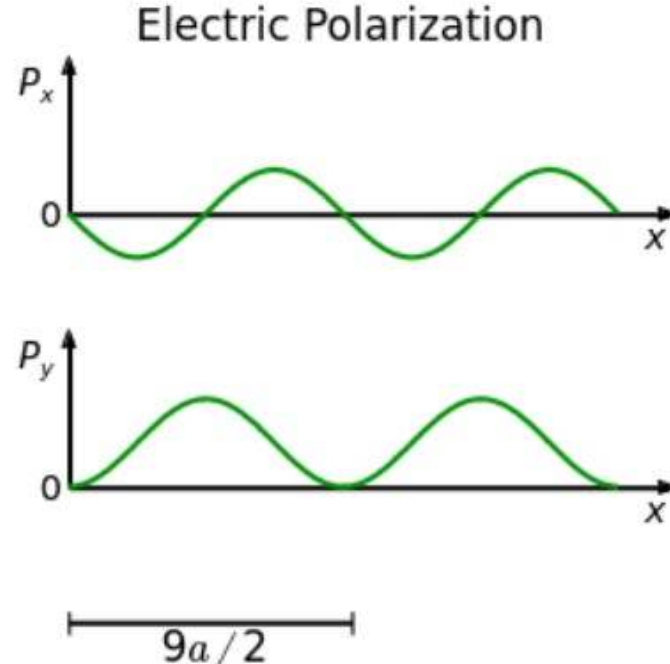
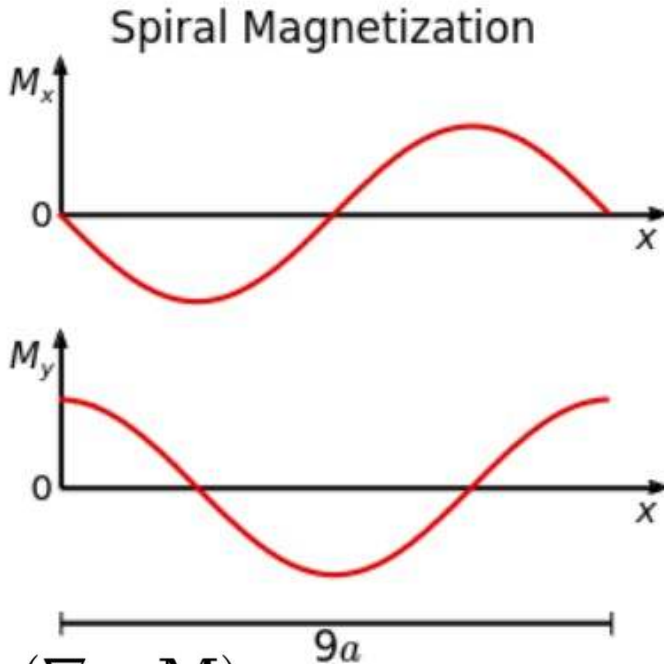
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\text{SOC}}^{\text{Ni}} \mathcal{H}_{\text{SOC}}^{\text{Ni}} + \lambda_{\text{SOC}}^{\text{I}} \mathcal{H}_{\text{SOC}}^{\text{I}},$$



The dominant contribution to the ferroelectric force comes from iodine

Modulated multiferroicity in monolayer NiI_2

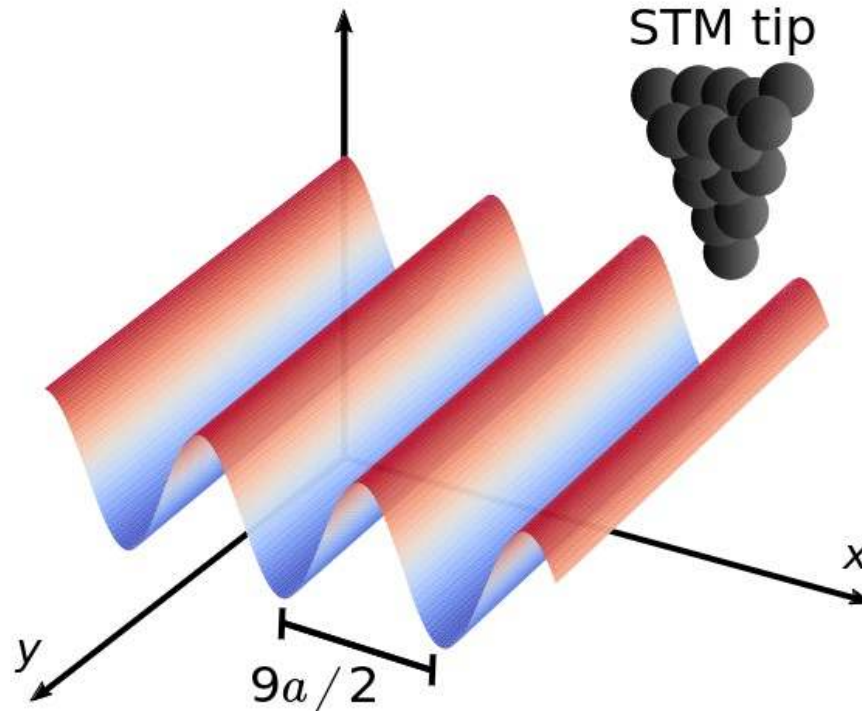
The ferroelectric order has actually a modulation in real space



$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{M^2} = \Lambda \mathbf{q} (-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

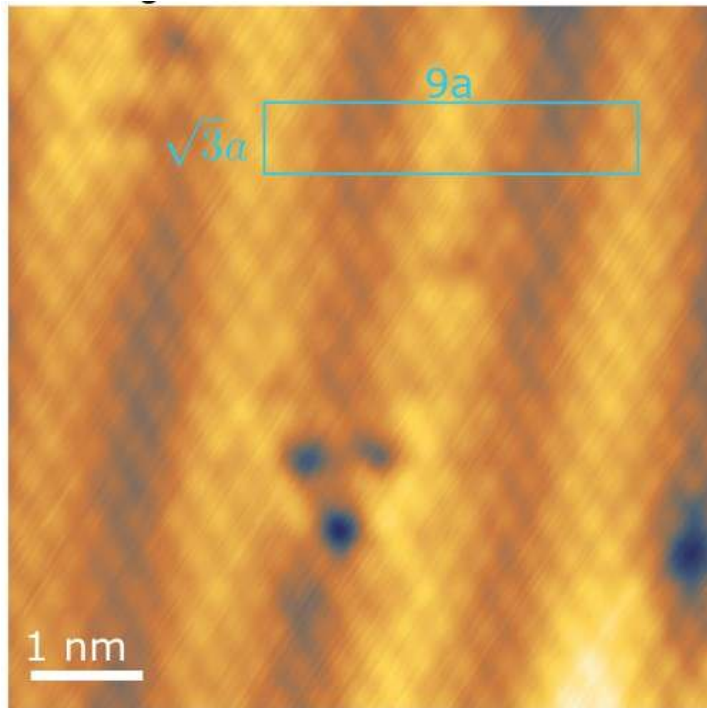
Visualizing multiferroicity in monolayer NiI_2

Modulated electrostatic potential

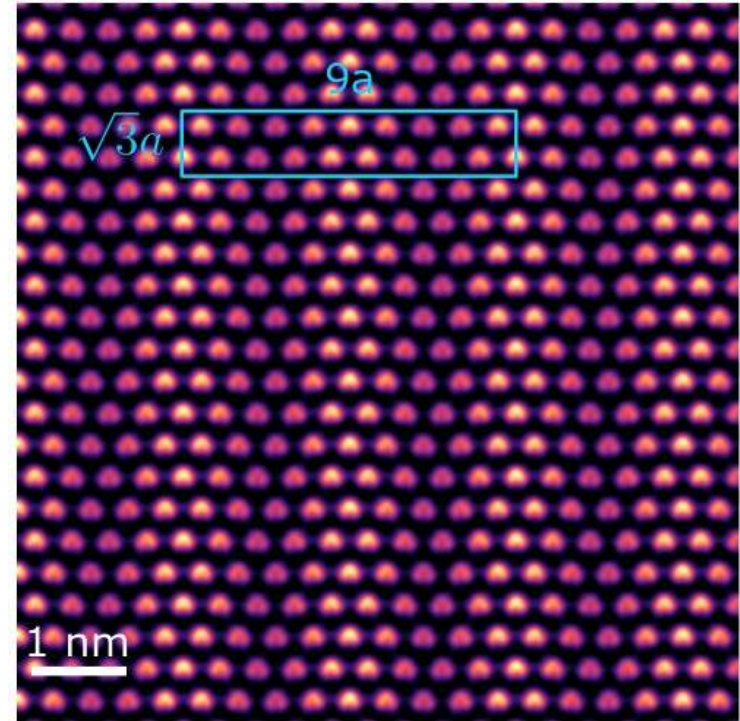


Visualizing multiferroicity in monolayer NiI_2

Experiment



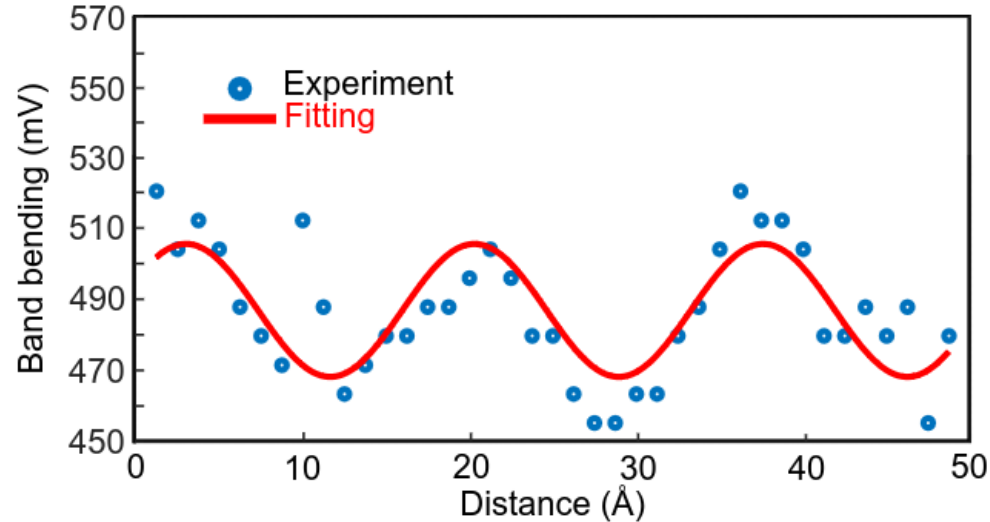
Theory



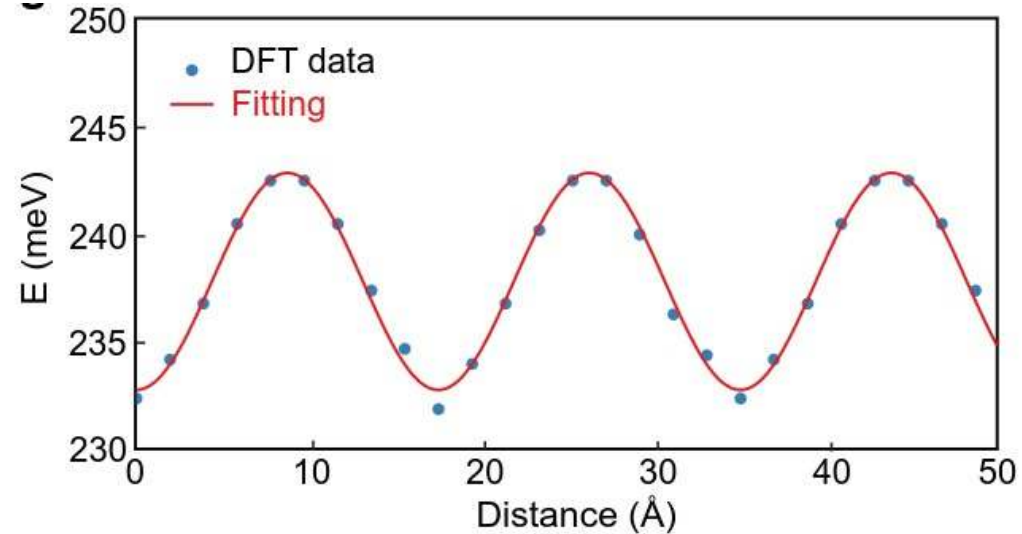
$$\Lambda_{\mathbf{q}}(-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

Ferroelectricity from modulated band offset

Band offset (experiment)

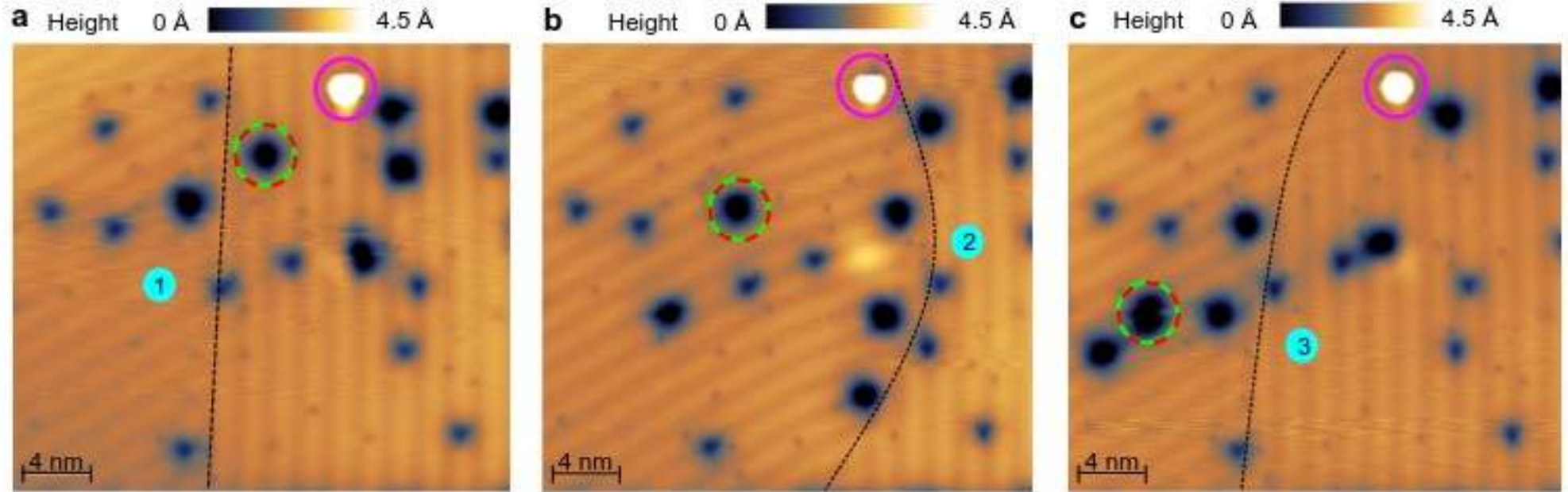


Band offset (theory)



$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{M^2} = \Lambda q (-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

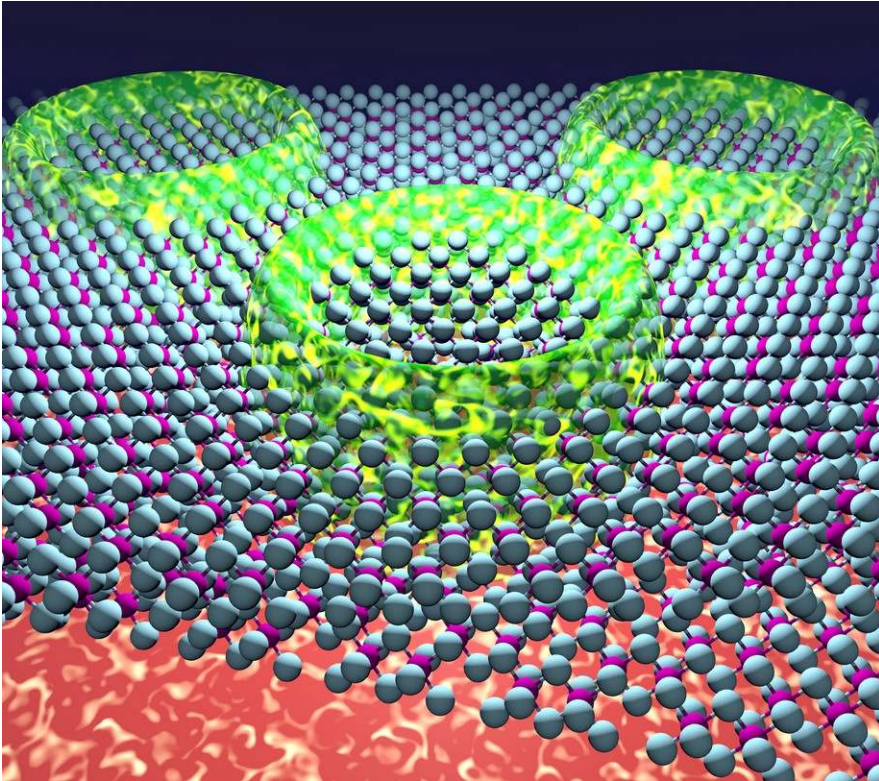
Controlling multiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

Advanced Materials,
2311342 (2024)

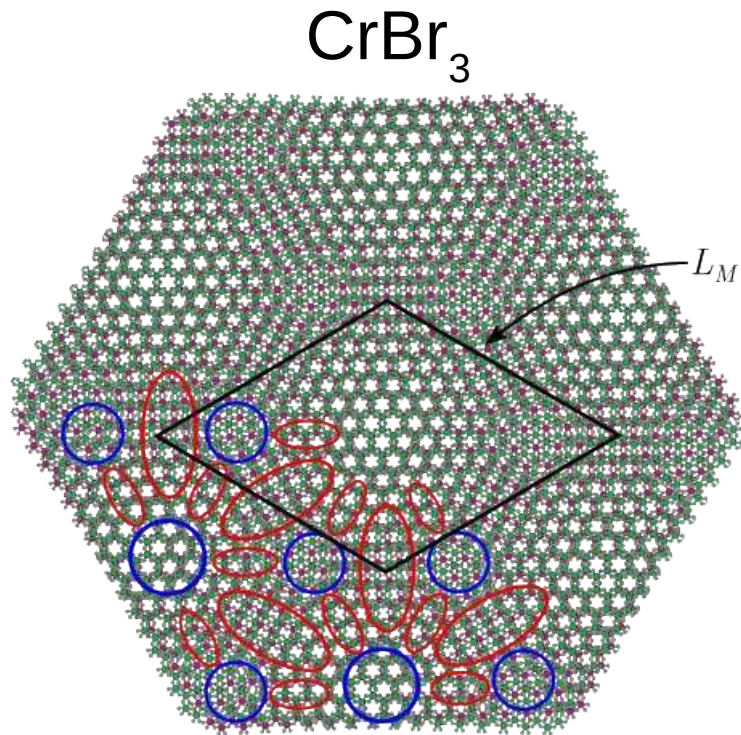
Engineering artificial multiferroics with twisted van der Waals materials



A. Fumega

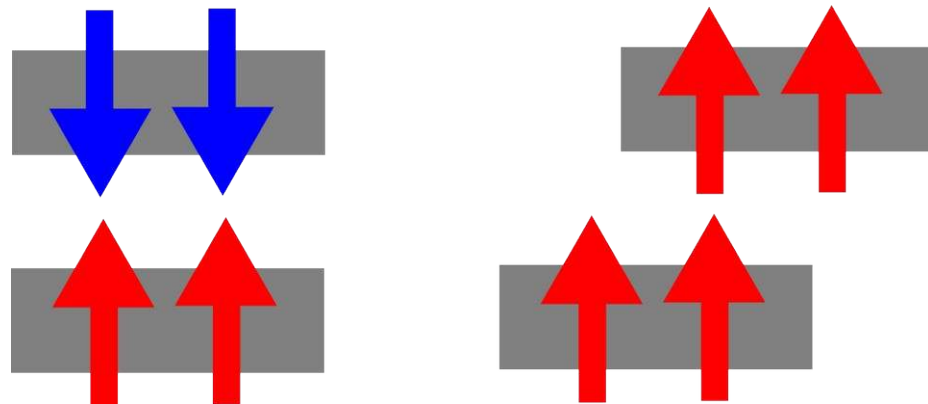


Twisted 2D trihalides



Bilayer Stacking

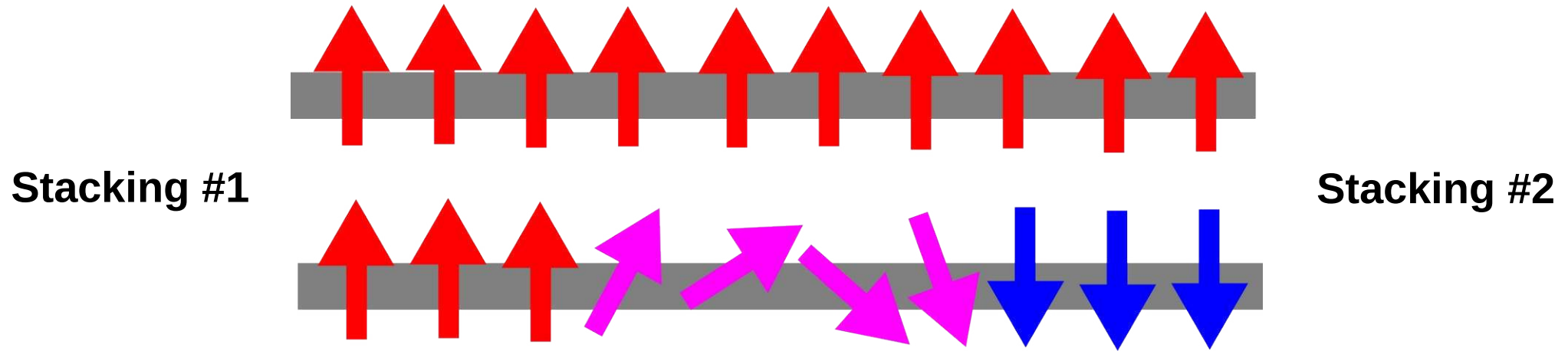
(M)	Monoclinic
(R)	Rhombohedral



The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Mixed FE/AF domains

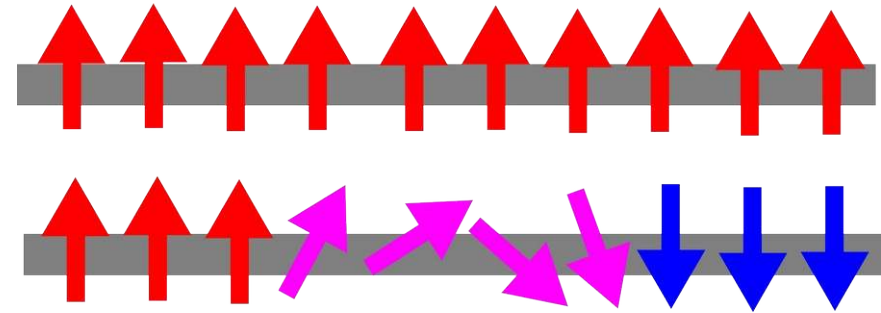
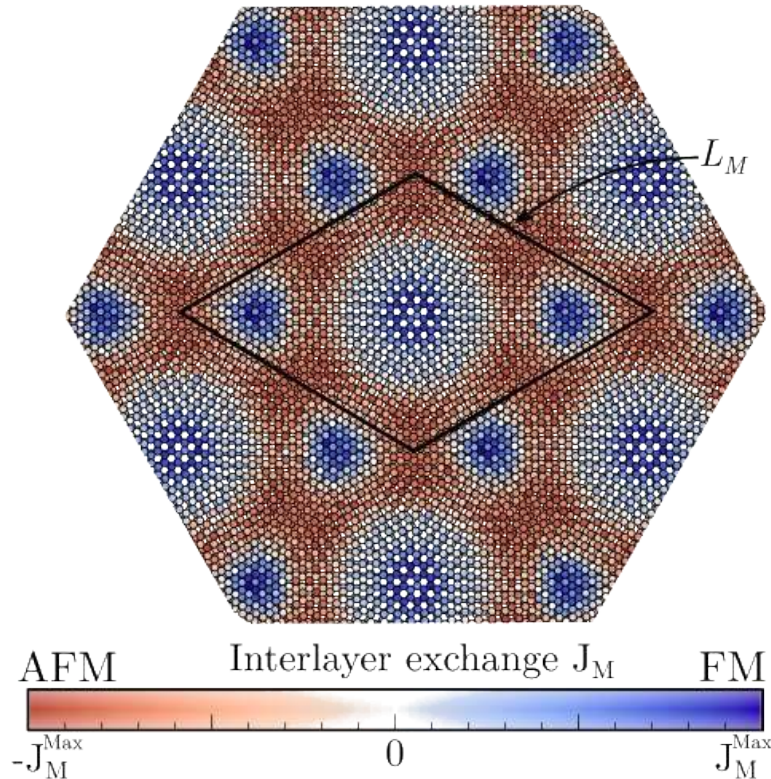
Non-collinear magnetism

Nature Nanotechnology 17, 143–147 (2022)

arXiv:2204.01636

Twisted 2D magnetic materials

Non-collinear magnetism and multiferroic order appear due to the moire



$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e}$$

Electric
polarization

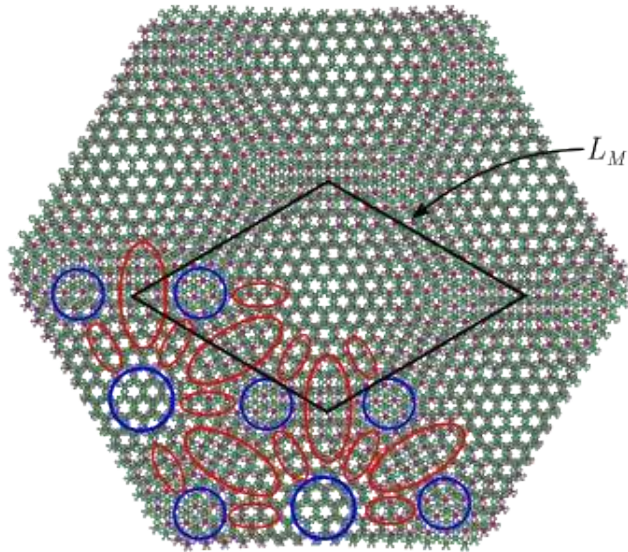
SOC driven

Spin rotation axis

Wavevector of the spin spiral

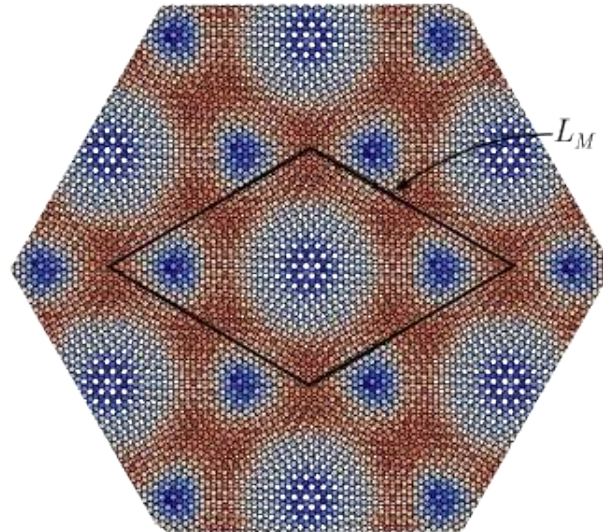
Stacking, magnetic order and electric polarization

Structure



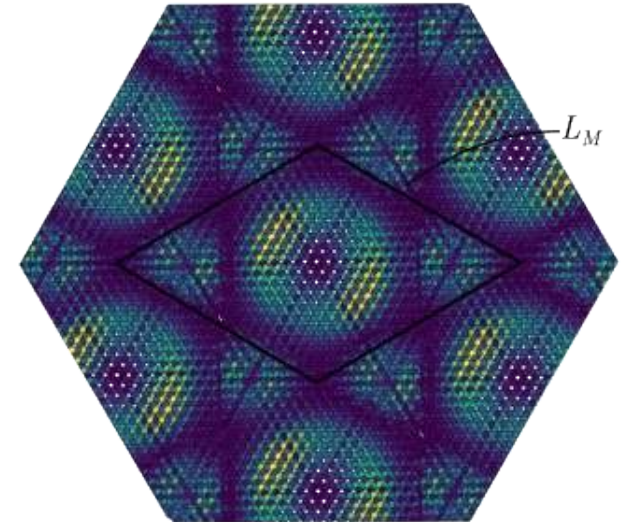
Bilayer Stacking $\left\{ \begin{array}{l} \text{(M)} \text{ Monoclinic} \\ \text{(R)} \text{ Rhombohedral} \end{array} \right.$

Magnetic order



AFM Interlayer exchange J_M FM
 $-J_M^{\text{Max}}$ 0 J_M^{Max}

Ferroelectric order

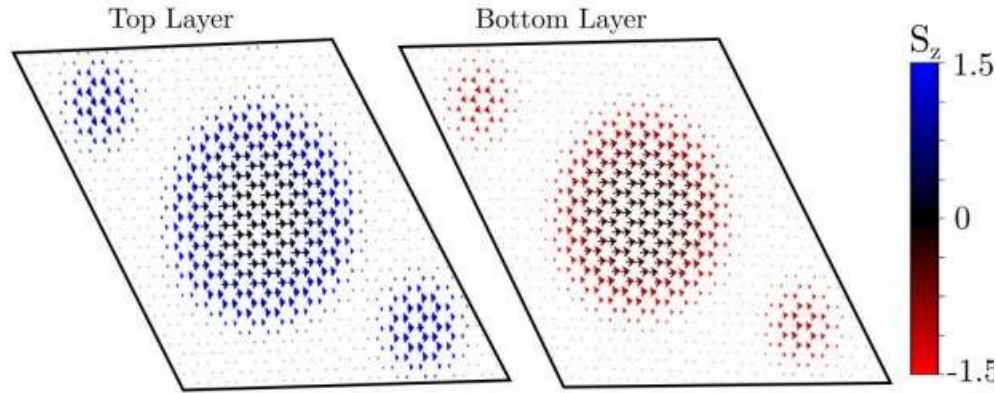


Electric Polarization
0 P^{Max}

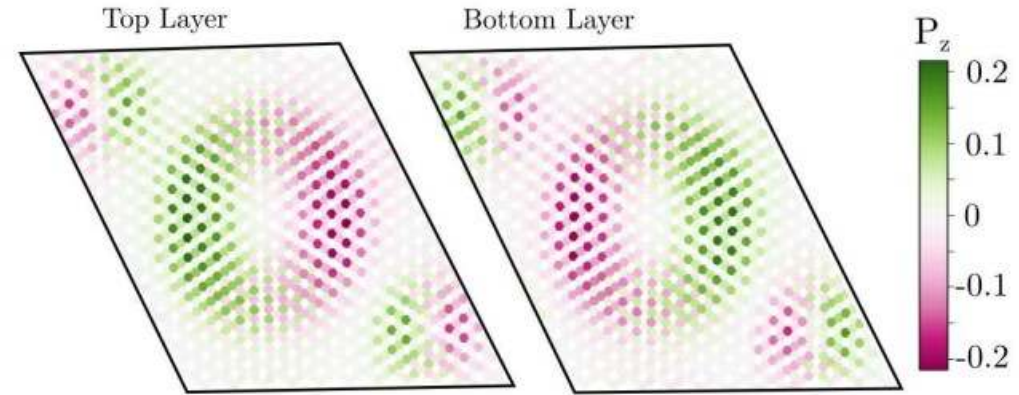
Stacking, magnetic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

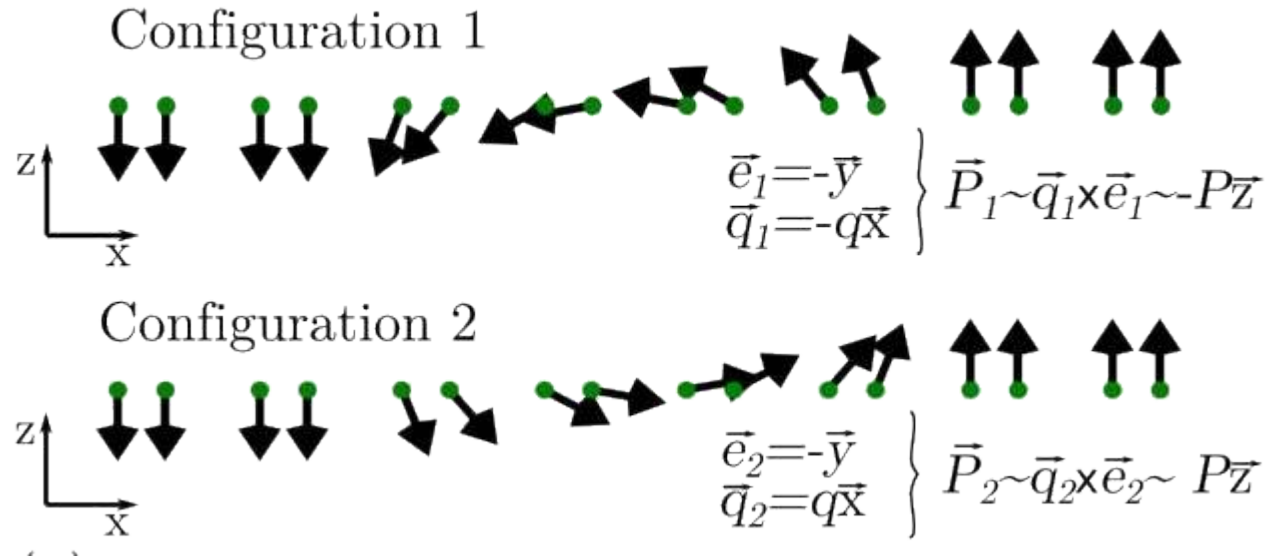


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



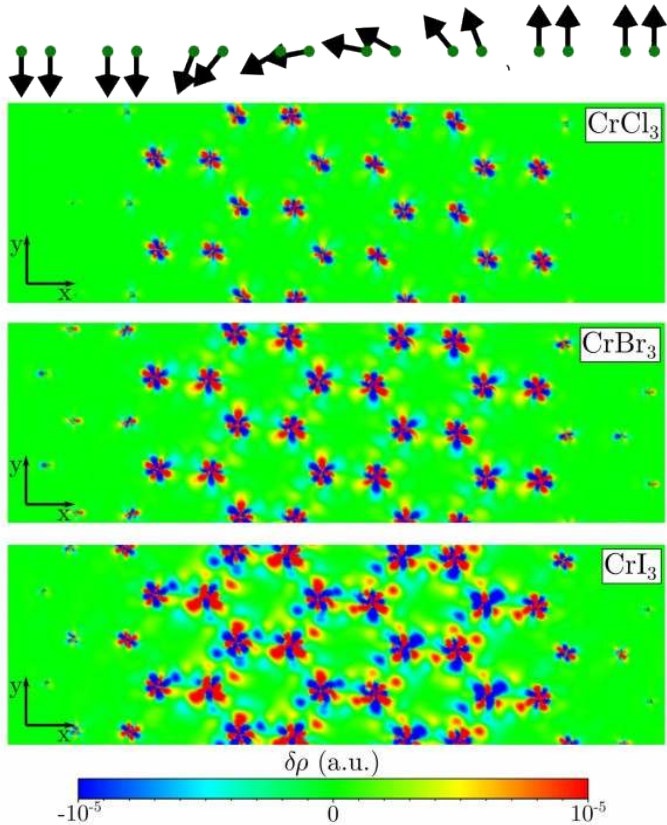
Ferroelectric electronic density

$$\delta\rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

Ferroelectric force

$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$

The ferroelectric electronic correction



Ferroelectric electronic density

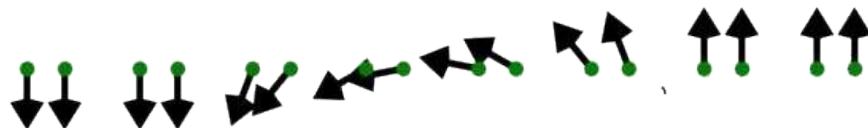
$$\delta\rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

The ferroelectric electronic correction emerges in the region with non-collinear magnetization

It becomes stronger the heavier the halide

The ferroelectric force

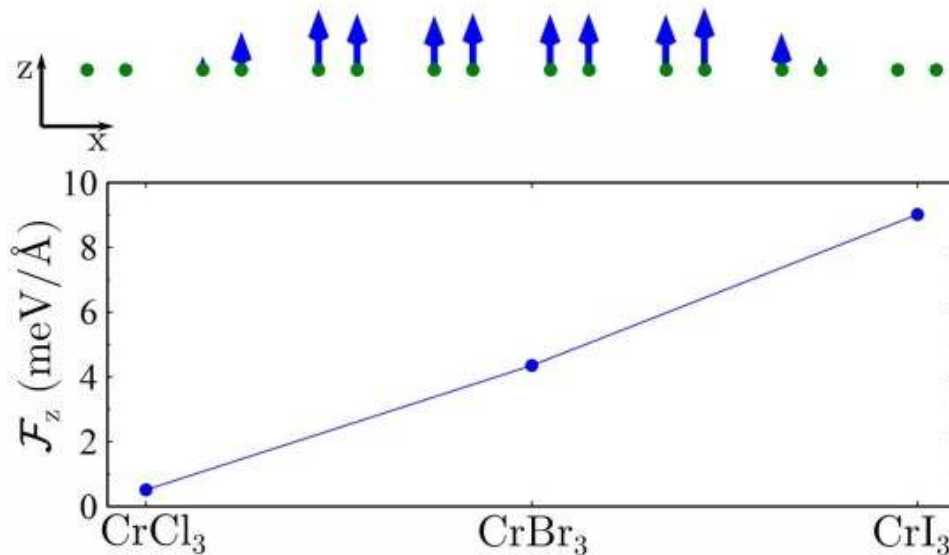
Magnetic configuration



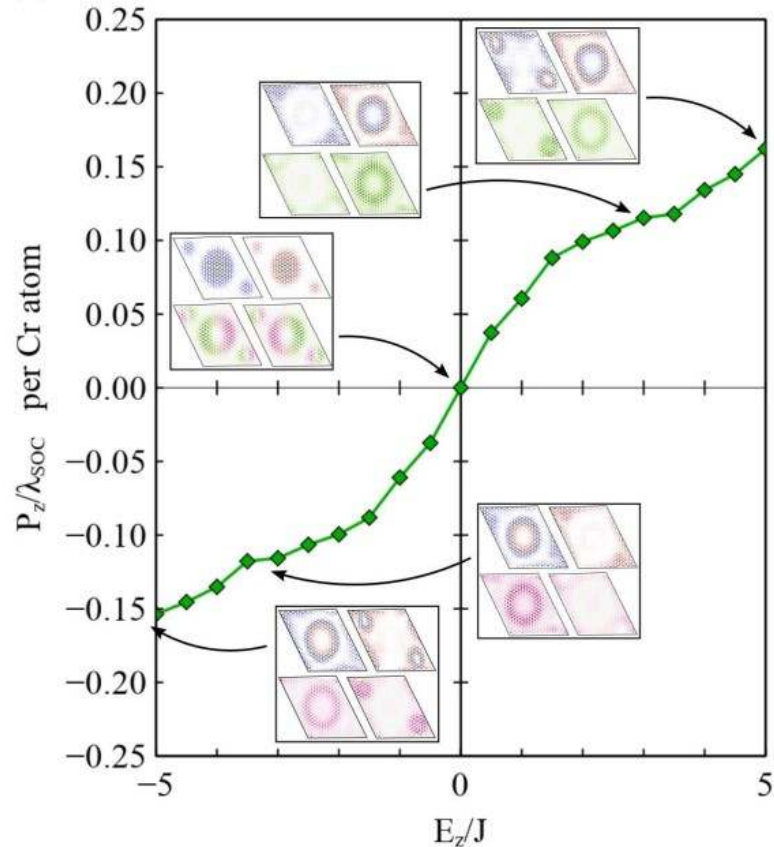
Ferroelectric force

$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$

The ferroelectric force becomes
Stronger for heavier halides



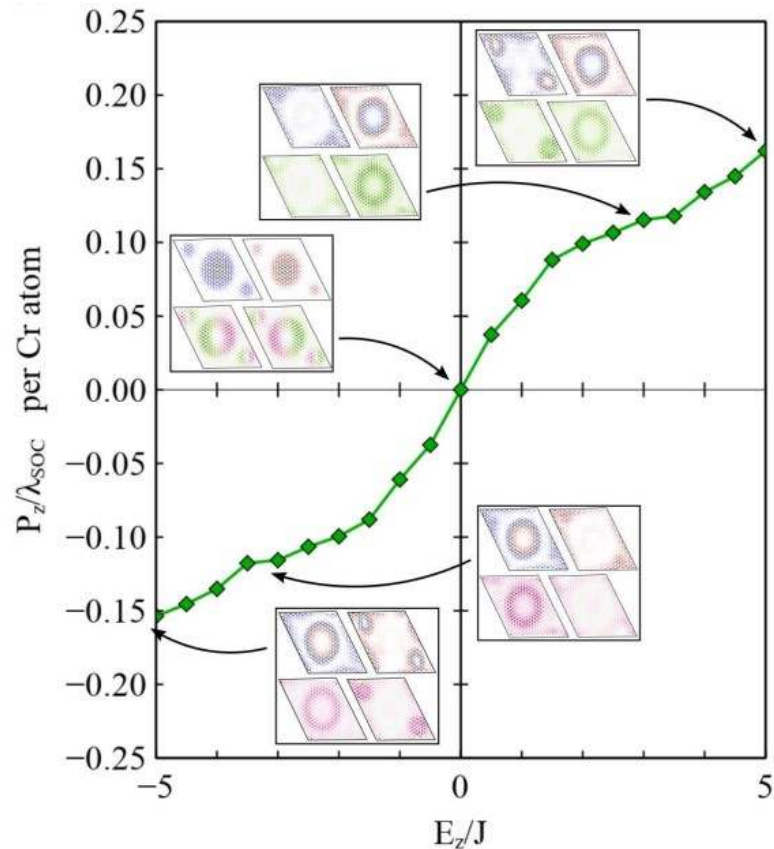
Electrically controllable skyrmions in twisted magnets



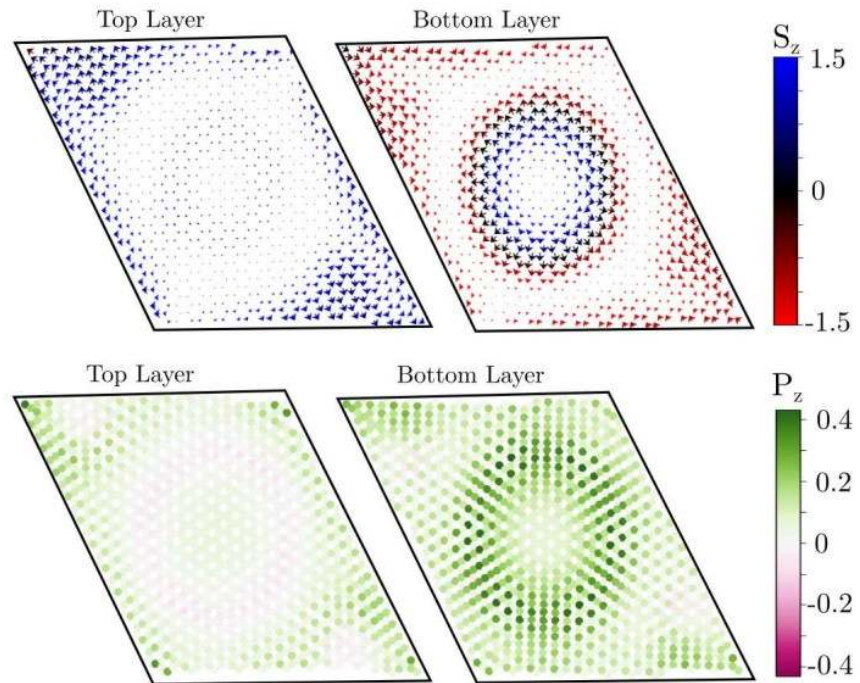
The electric polarization directly couples to an out-of plane electric field

$$\mathcal{H}_E = \frac{1}{2} \sum_{ij} \mathbf{E} \cdot \mathbf{P}_{ij}.$$

Electrically controllable skyrmions in twisted magnets



Skyrmions at finite interlayer bias

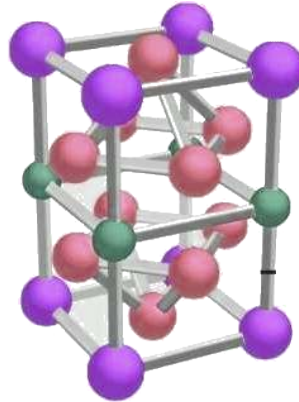


Van der Waals heavy-
fermion Kondo magnets

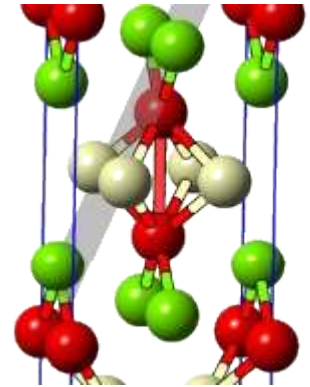
The world of heavy fermion compounds

We have a rich family of bulk heavy-fermion compounds

CeCoIn_5
Science 375, 6576, 76-81 (2021)



UTe_2
Nature 579, 523–527 (2020)



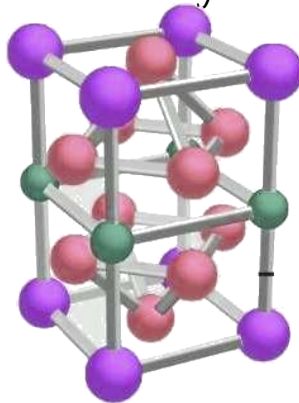
- Strongly correlated matter, dominated by Kondo lattice physics
- Unique system to realize quantum criticality and unconventional superconductivity
- Found in rare-earth compounds, such as UTe_2 or UPt_3

Searching for new heavy fermion compounds

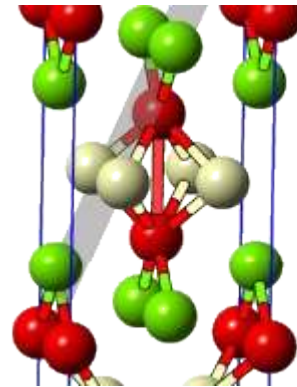
We have a rich family of bulk heavy-fermion compounds



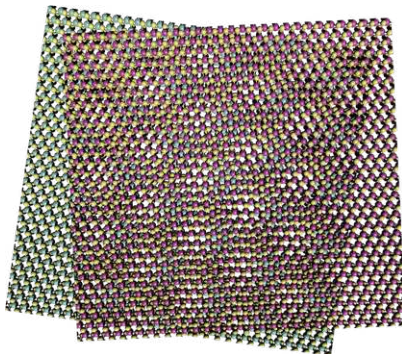
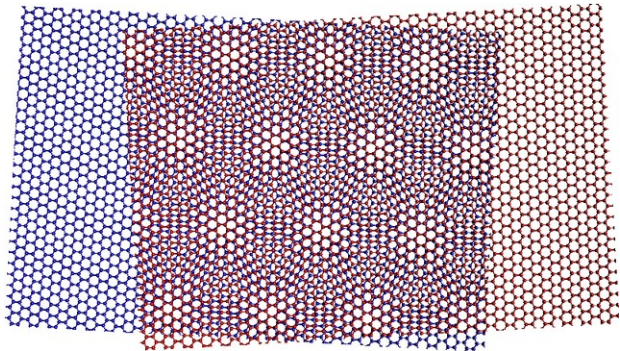
Science 375, 6576, 76-81 (2021)



Nature 579, 523–527 (2020)



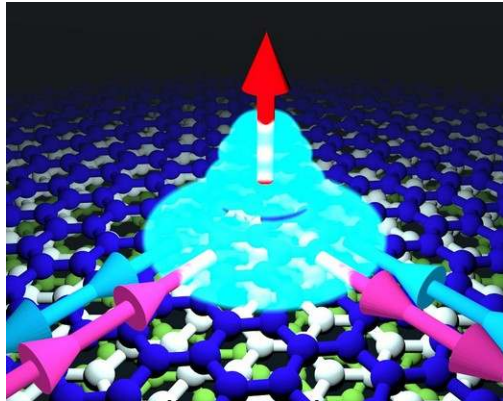
Can we realize heavy-fermion physics with two-dimensional materials?



Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)
Phys. Rev. Research 3, 043173 (2021)
Phys. Rev. B 106, L041116 (2022)
Phys. Rev. Lett. 129, 047601 (2022)
Nature 616, 61–65 (2023)
Nature 625, 483–488 (2024)
Nano Letters 24 (14), 4272–4278 (2024)

Behind the scenes

Heavy fermions in twisted graphene trilayers

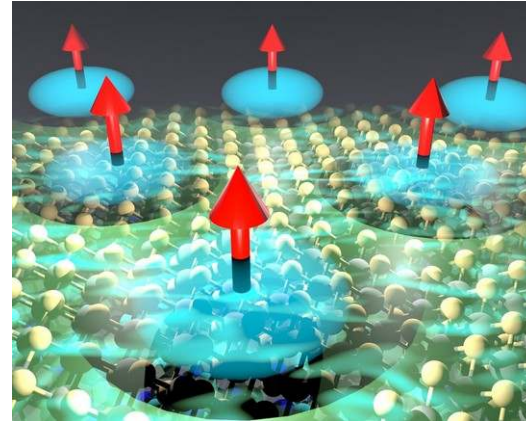


Aline Ramires



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



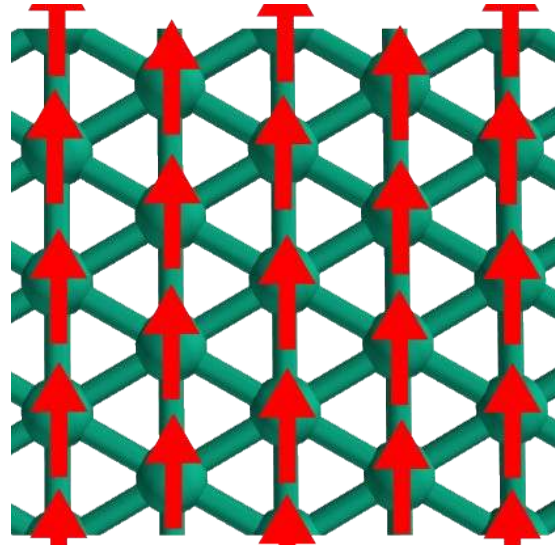
G. Chen



Building an artificial heavy fermion state



Lattice of Kondo impurities



f-electrons in
rare-earth compounds

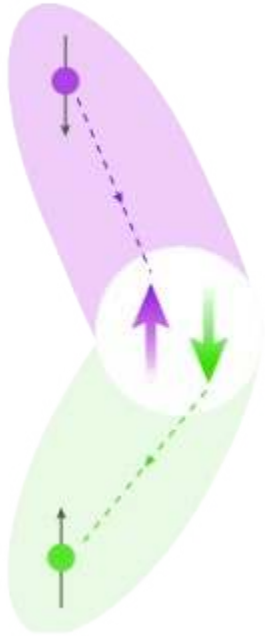
Dispersive electron gas



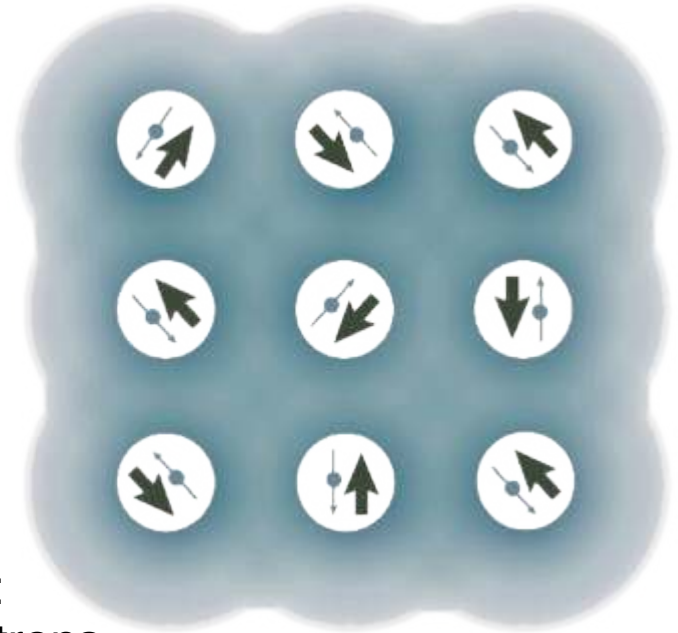
s/p/d-electrons in
rare-earth compounds

Building an artificial heavy fermion state

**Conduction electrons form
Kondo singlets with the impurities**



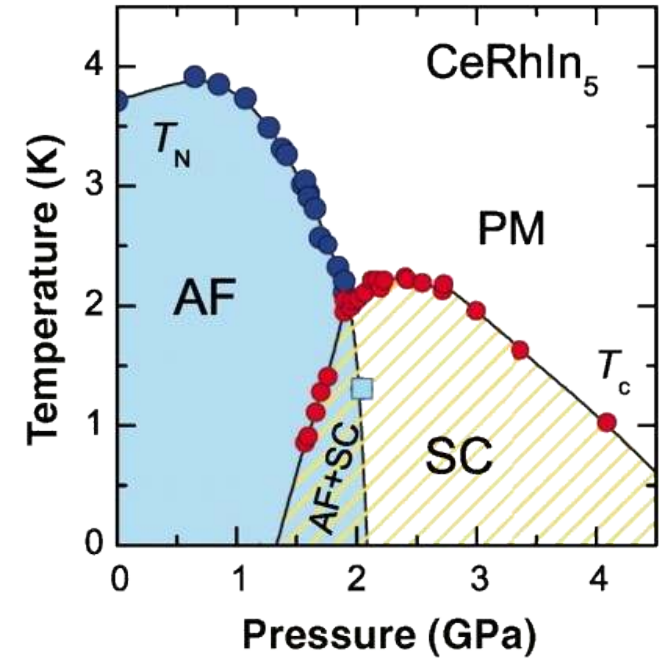
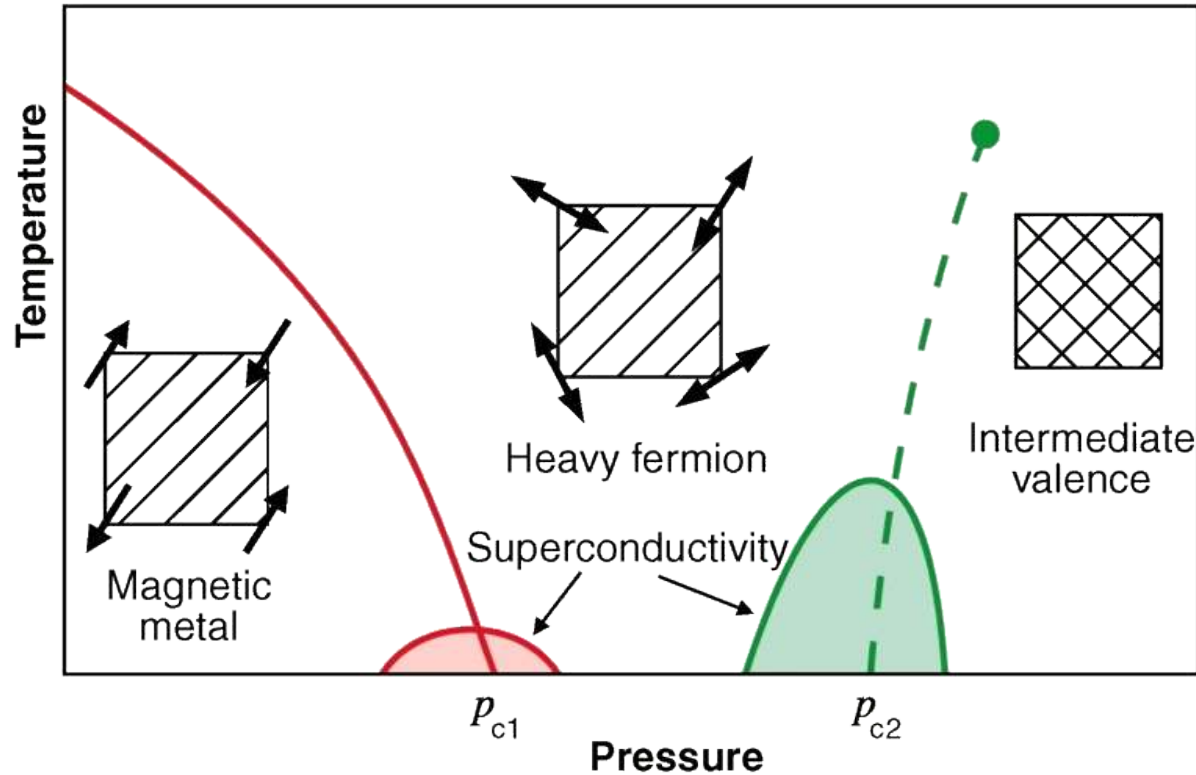
Kondo-lattice model



Associated with Kondo lattice physics:

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

Basics of heavy fermion physics



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

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

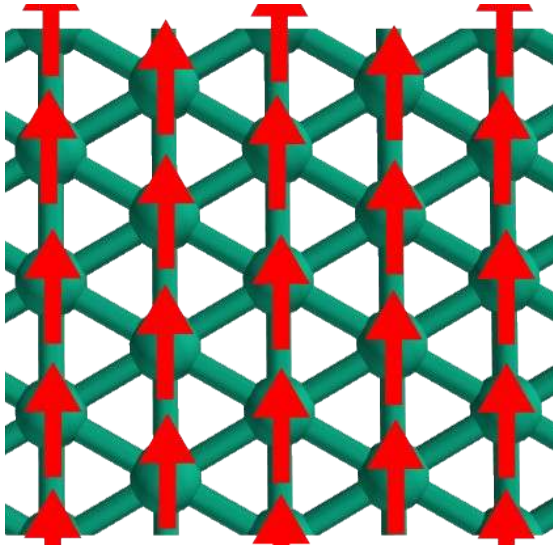
We now take a quantum spin $S=1/2$

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



The Kondo lattice problem

Lattice of Kondo impurities



Dispersive electron gas



$$J_K$$

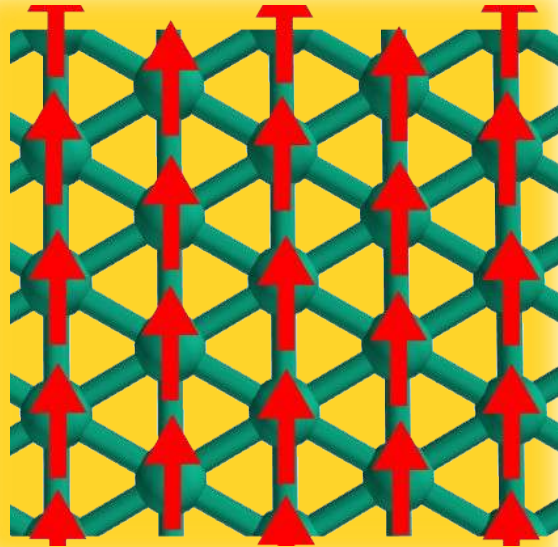

Both ingredients coupled through Kondo coupling

The Kondo lattice problem

The Kondo lattice problem

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

Conduction electrons



Kondo coupling

Kondo sites

Solving the Kondo lattice problem

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

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

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

Solving the Kondo lattice problem

Now we decouple the fermions with a mean-field approximation

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

Obtaining a quadratic Hamiltonian

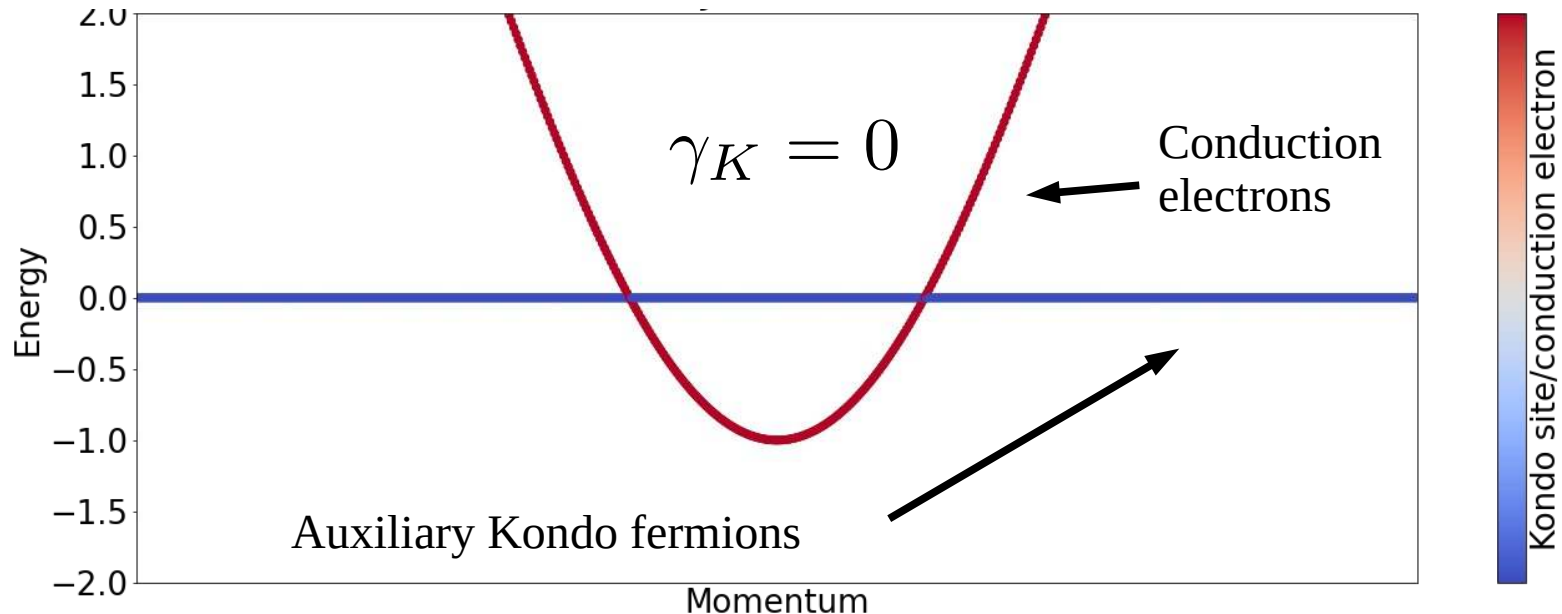
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

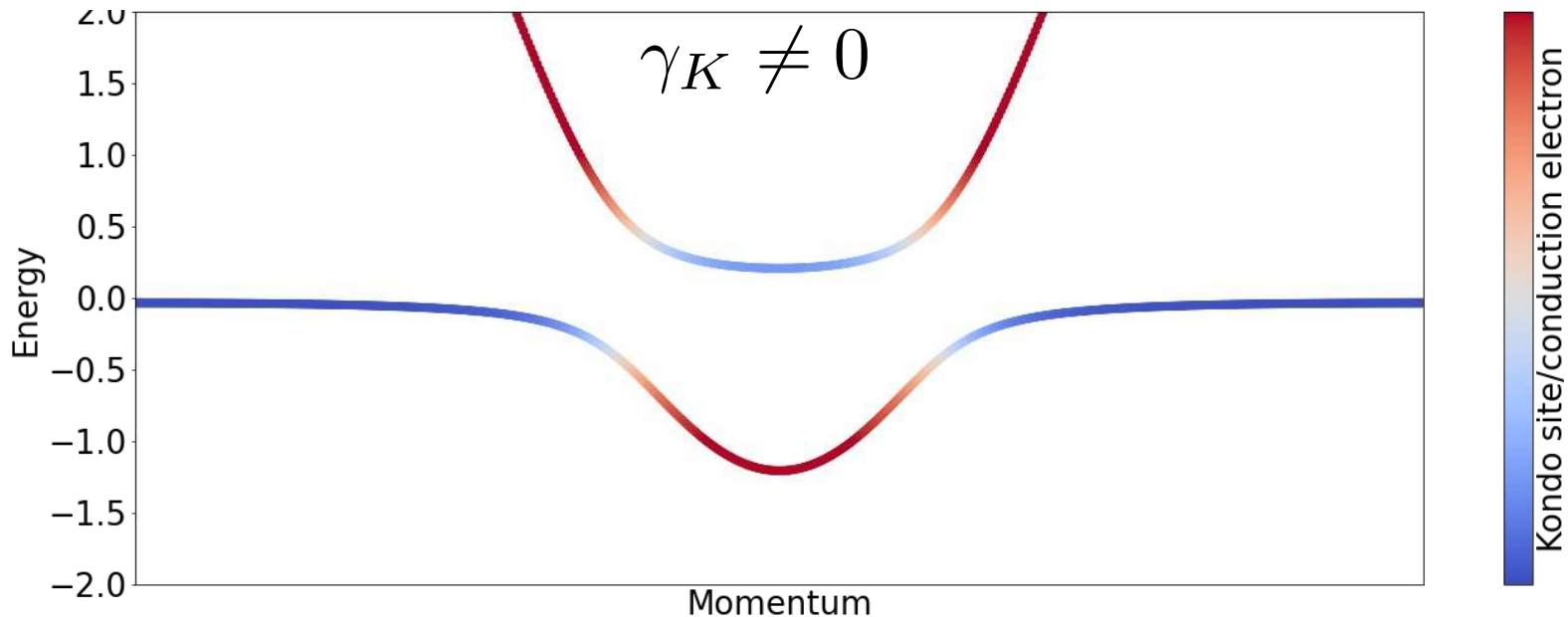
Electronic structure of the Kondo lattice problem

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



Electronic structure of the Kondo lattice problem

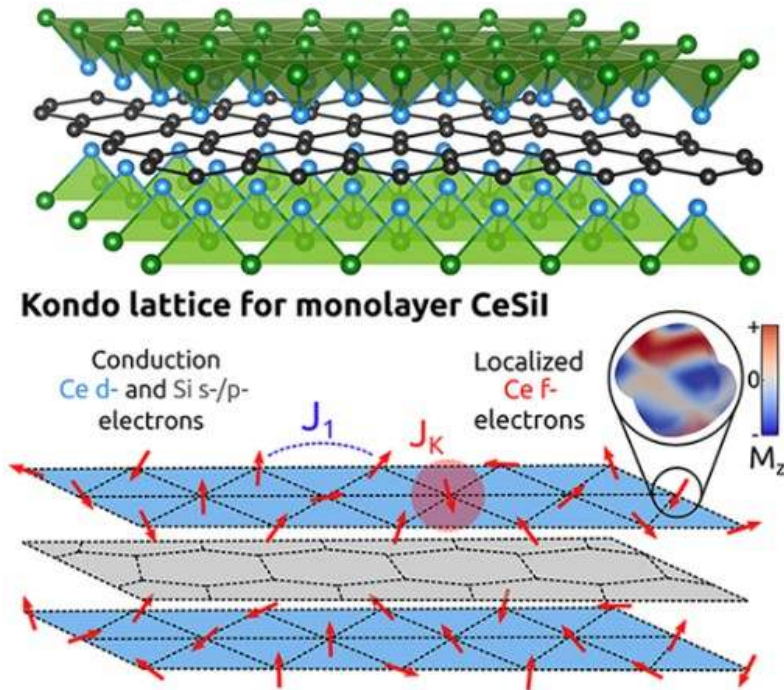
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k}, \alpha\beta} f_{\mathbf{k}\alpha}^{\dagger} c_{\mathbf{k}\alpha} + h.c.$$



The Kondo coupling opens up a gap in the electronic structure

Electronic structure of a rare-earth van der Waals heavy-fermion material CeSiI

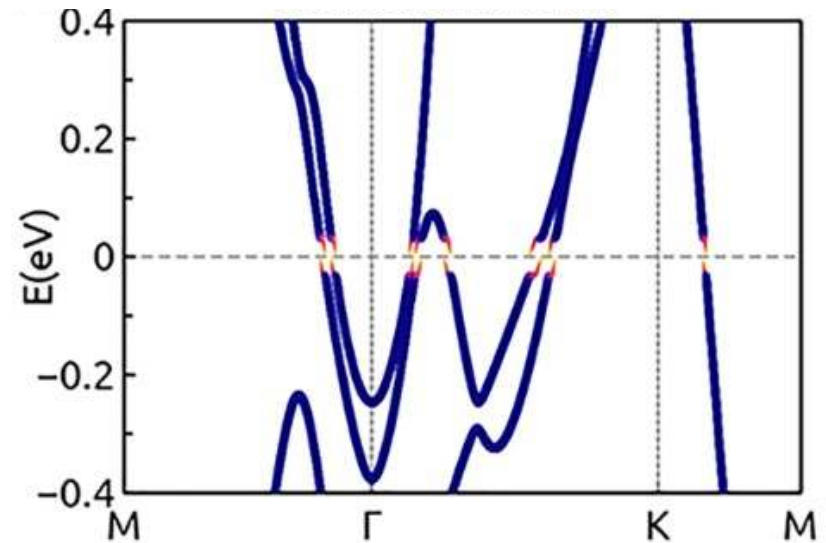
Monolayer CeSiI



Ultrathin heavy-fermion isolated in

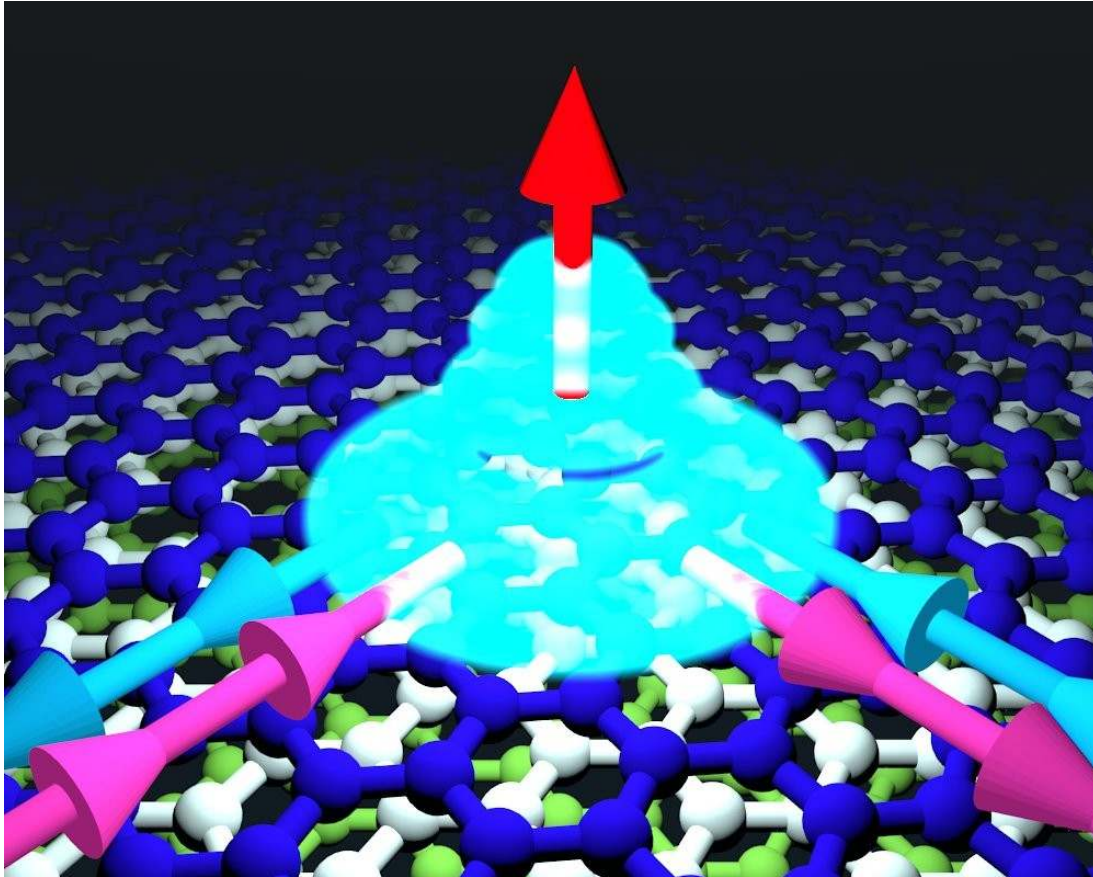
Nature 625, 483–488 (2024)

Heavy-fermion (gapped) electronic structure



Nano Letters 24 (14), 4272–4278 (2024)

Heavy-fermions in twisted graphene trilayers



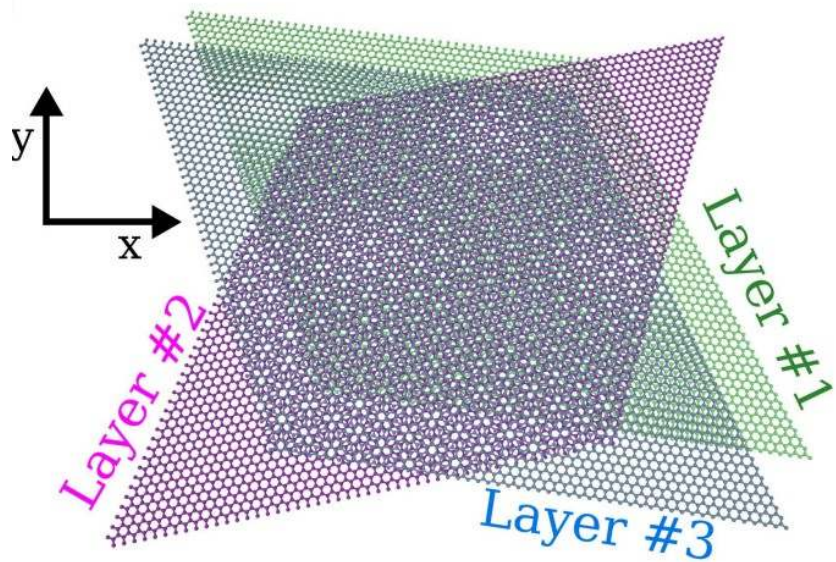
Aline Ramires



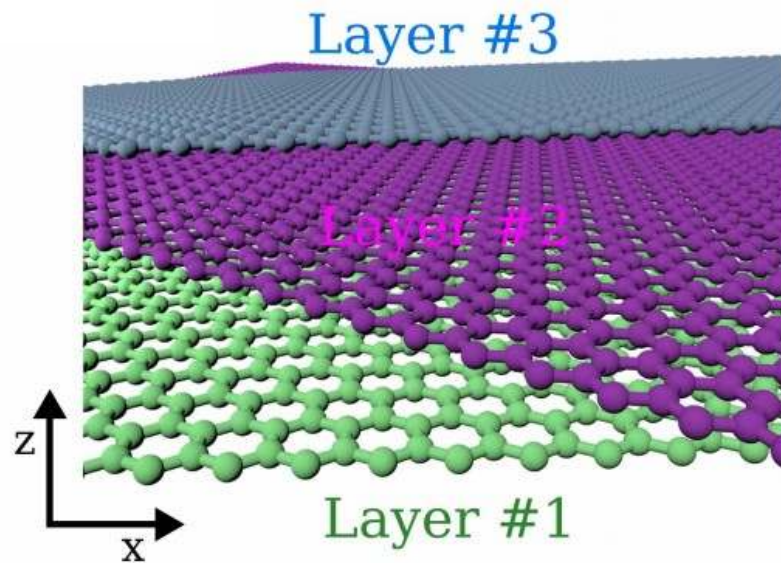
Phys. Rev. Lett. 127, 026401 (2021)

Heavy-fermions in twisted graphene trilayers

Top view

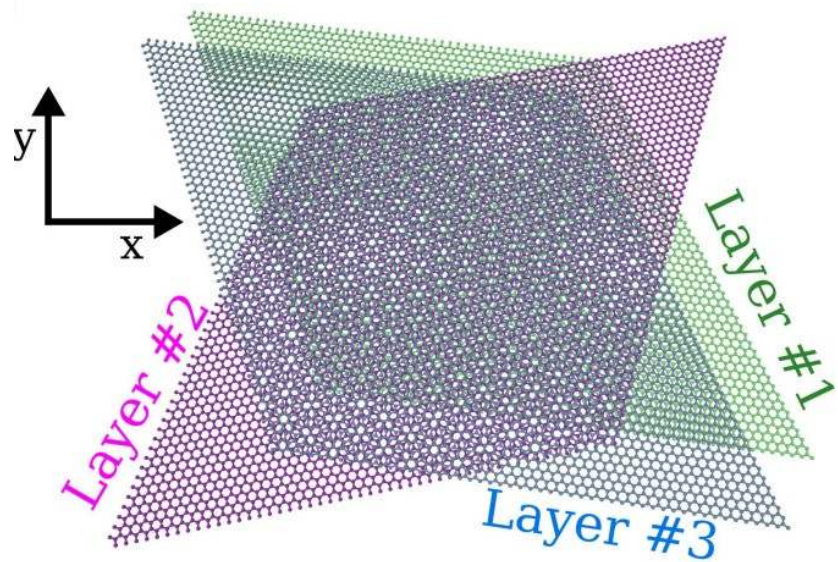


Lateral view

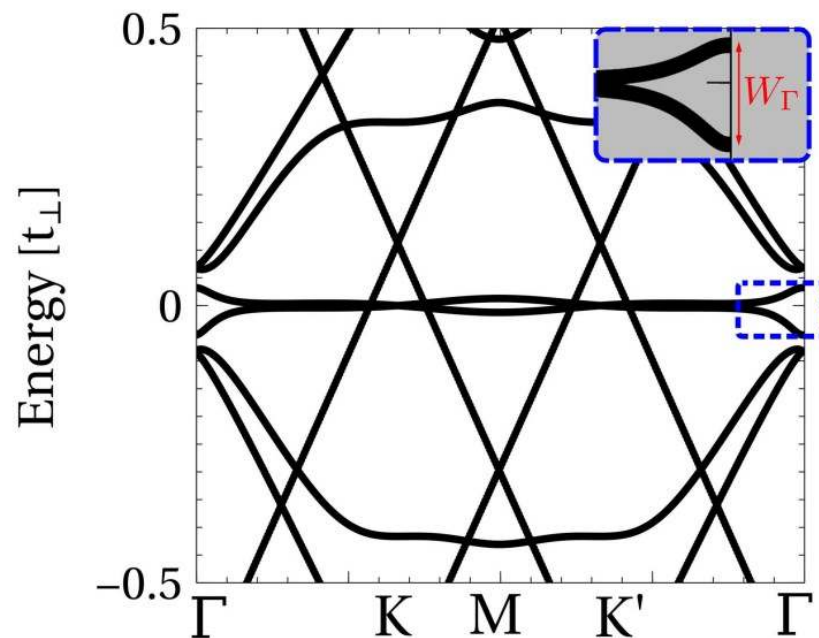


Heavy-fermions in twisted graphene trilayers

Top view

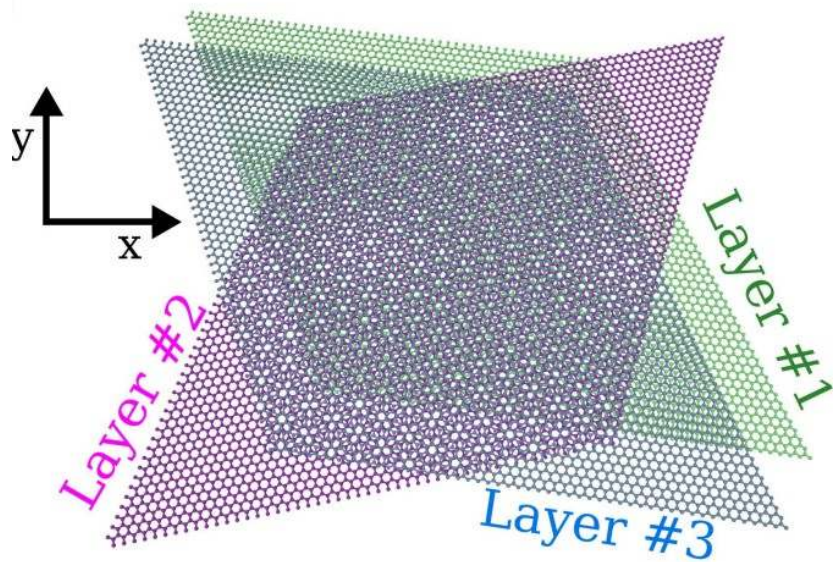


Band structure

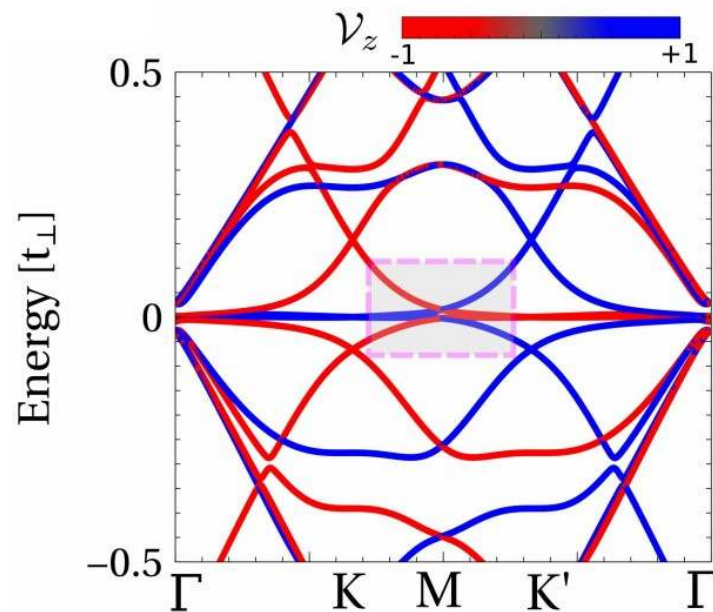


Heavy-fermions in twisted graphene trilayers

Top view

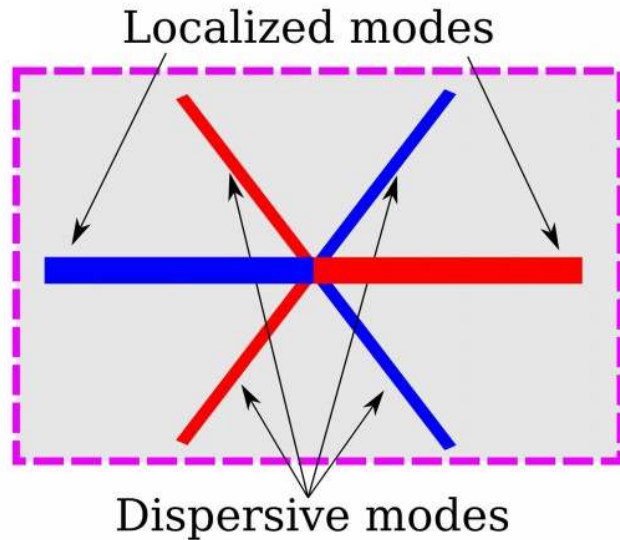


Electronic structure in the presence of an electric bias

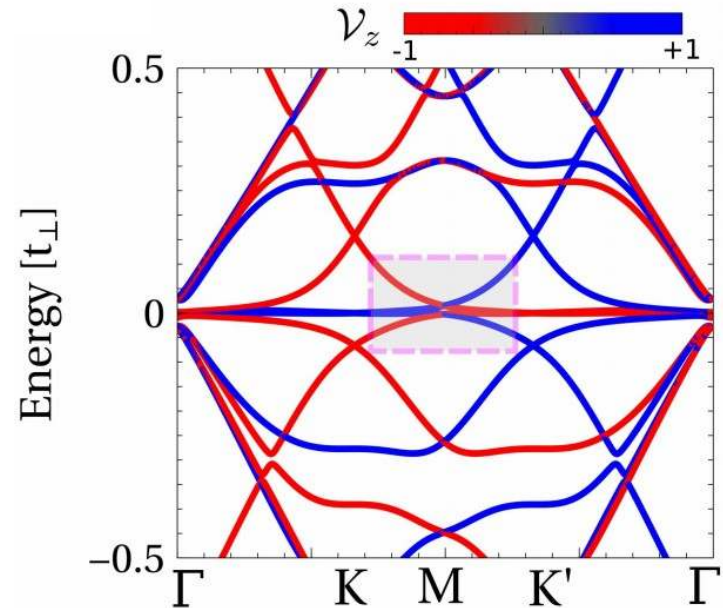


Heavy-fermions in twisted graphene trilayers

Low energy model

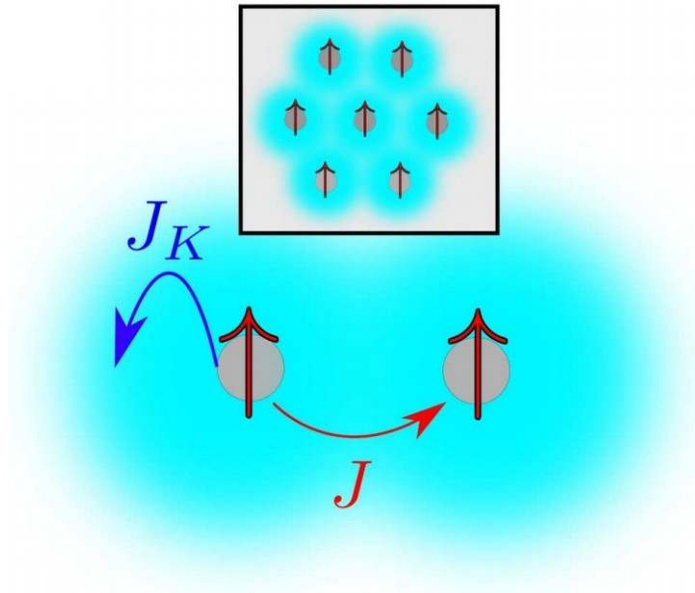


Electronic structure in the presence of an electric bias



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

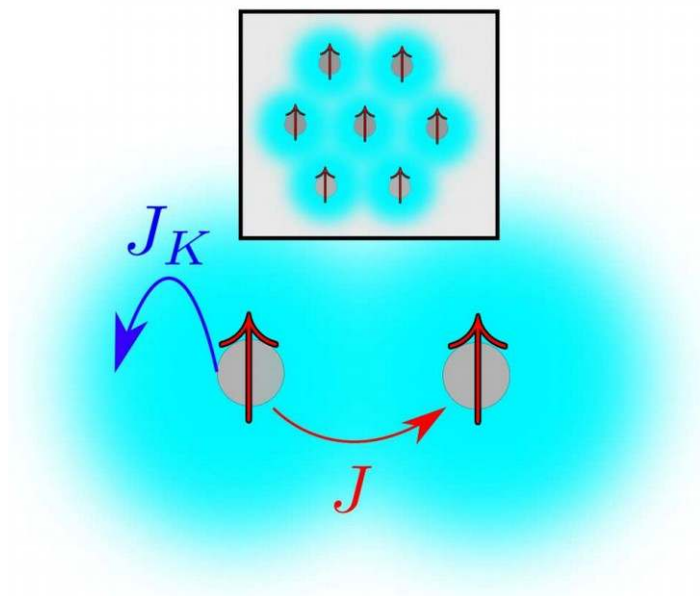
Promotes
magnetic order

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

$$|J/J_K| < 1$$

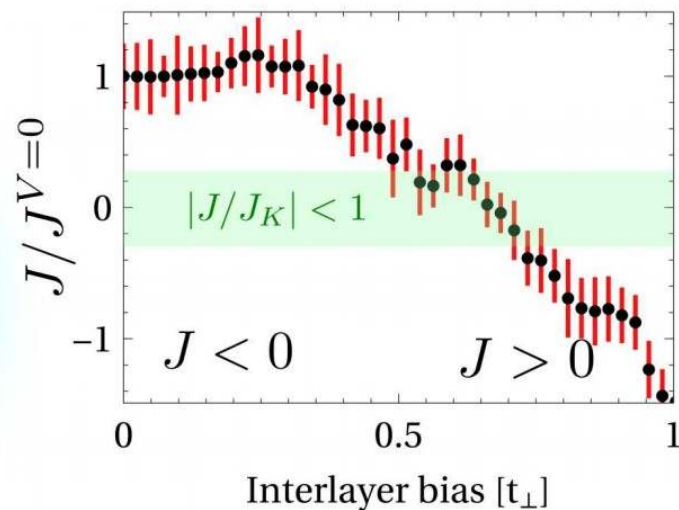
Kondo lattice model in the presence of interactions

Interactions of the Kondo lattice model



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

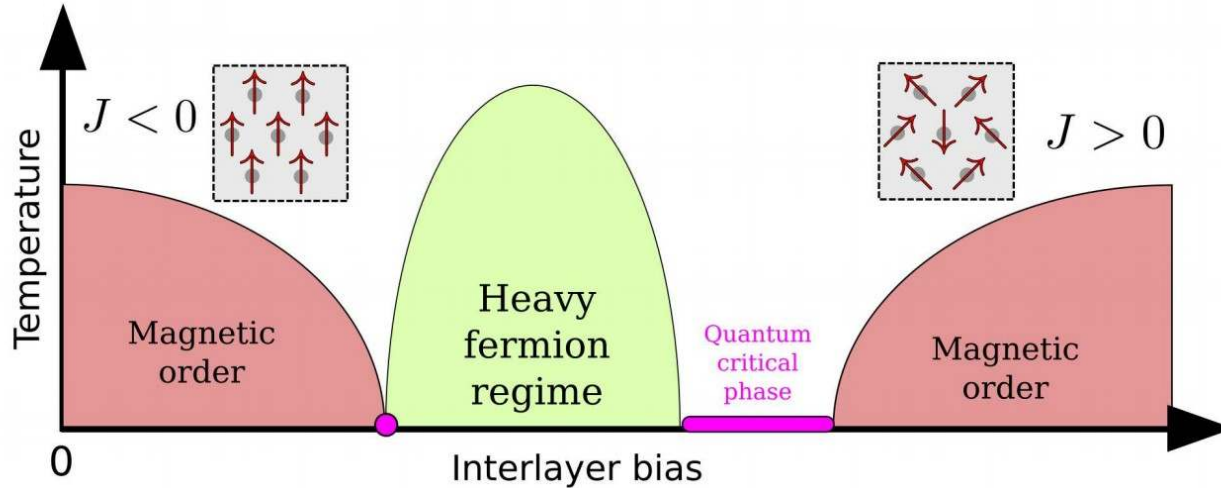
Electric control of the Kondo regime



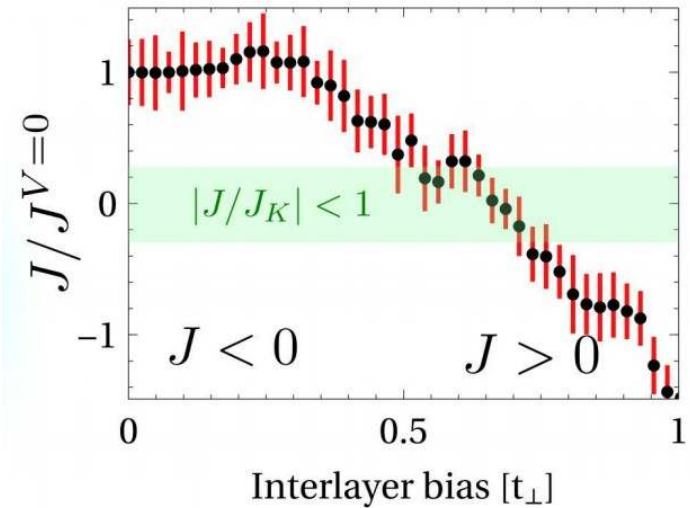
$$|J/J_K| < 1$$

Heavy-fermions in twisted graphene trilayers

Heavy-fermion (Doniach) phase diagram



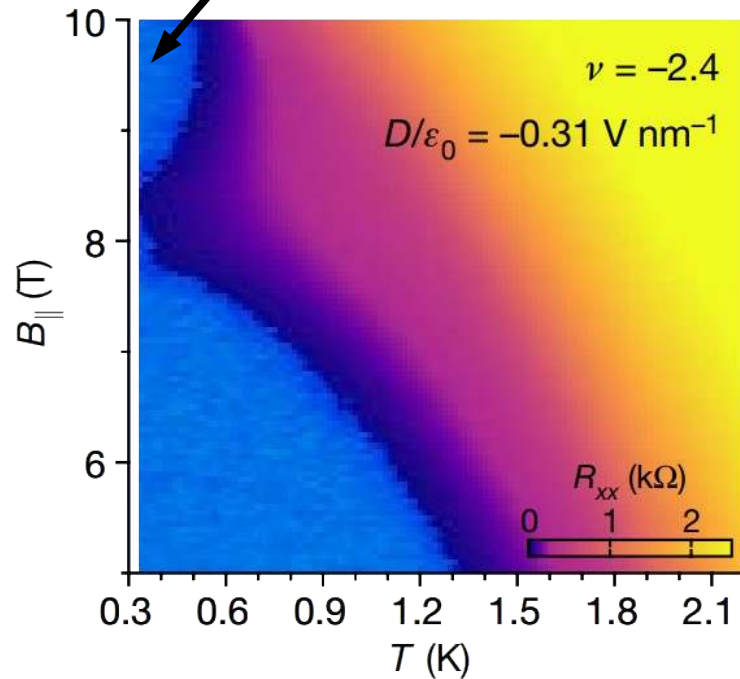
Electric control of the Kondo regime



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

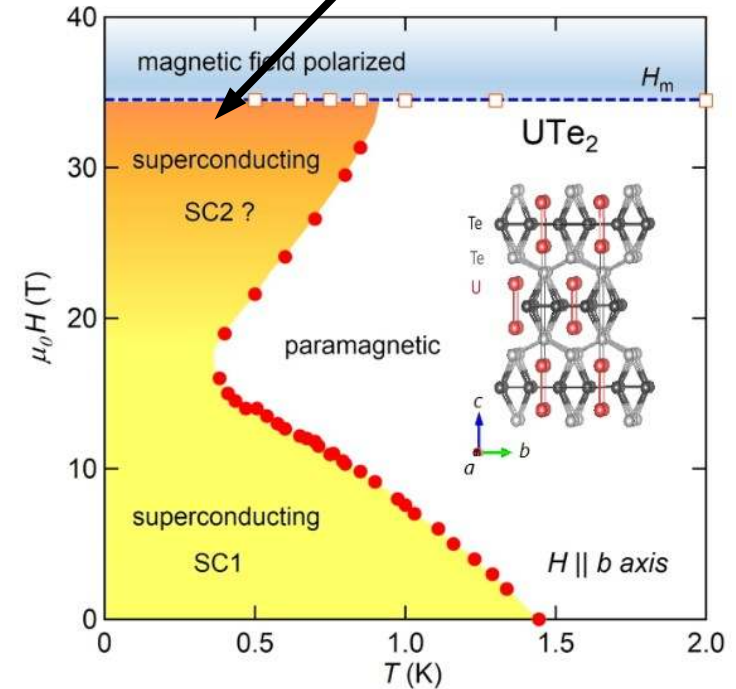
An experimental signature of heavy-fermion physics: spin-triplet superconductivity

Reentrant superconductivity
in twisted trilayer graphene



Nature 595, 526–531 (2021)

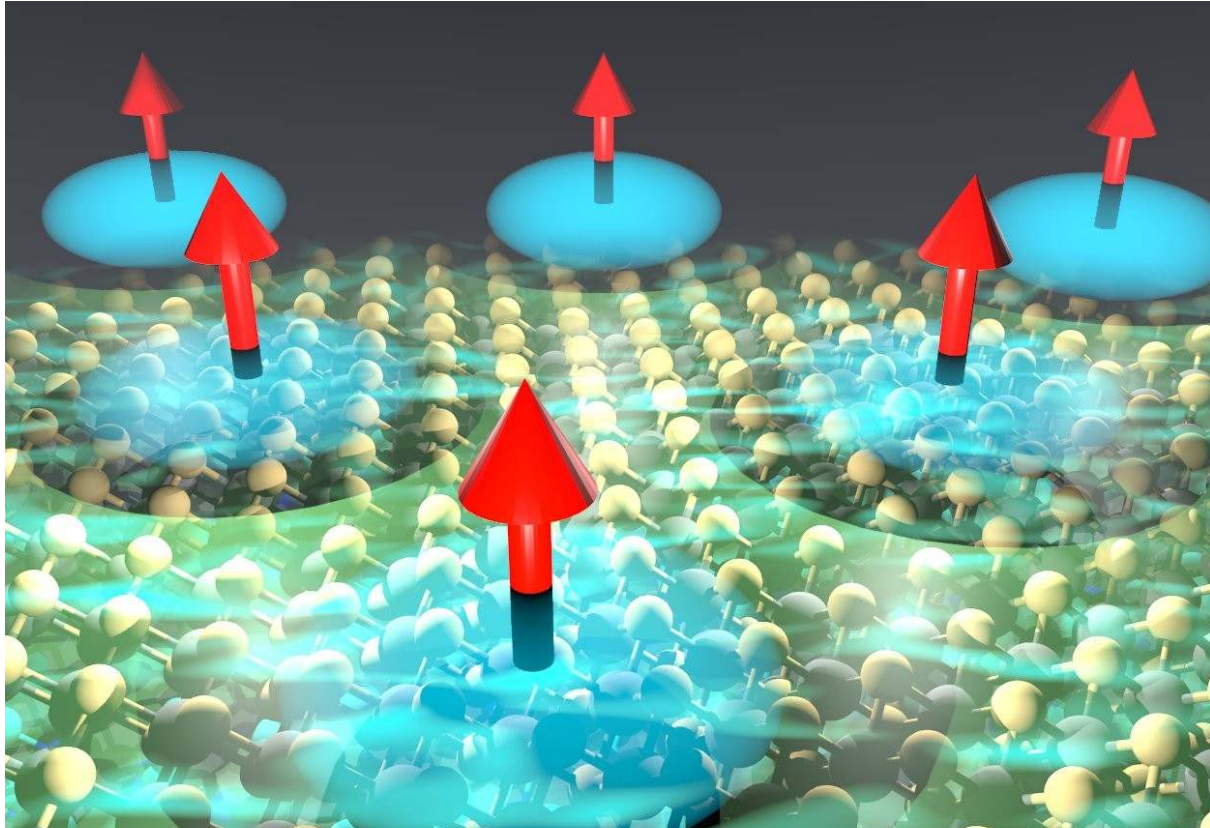
Reentrant superconductivity
in UTe_2



Nat. Phys. 15, 1250 (2019)

Science 365, 684 (2019)

Heavy-fermions in a van der Waals dichalcogenide heterostructure



V. Vaňo



M. Amini



G. Chen



S. Ganguli



S. Kezilebieke



P. Liljeroth



Nature 599, 582–586 (2021)

Heavy-fermions in dichalcogenide bilayers

Triangular lattice of
local magnetic moments



1T-TaS₂

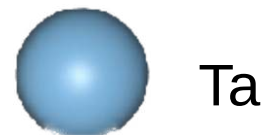
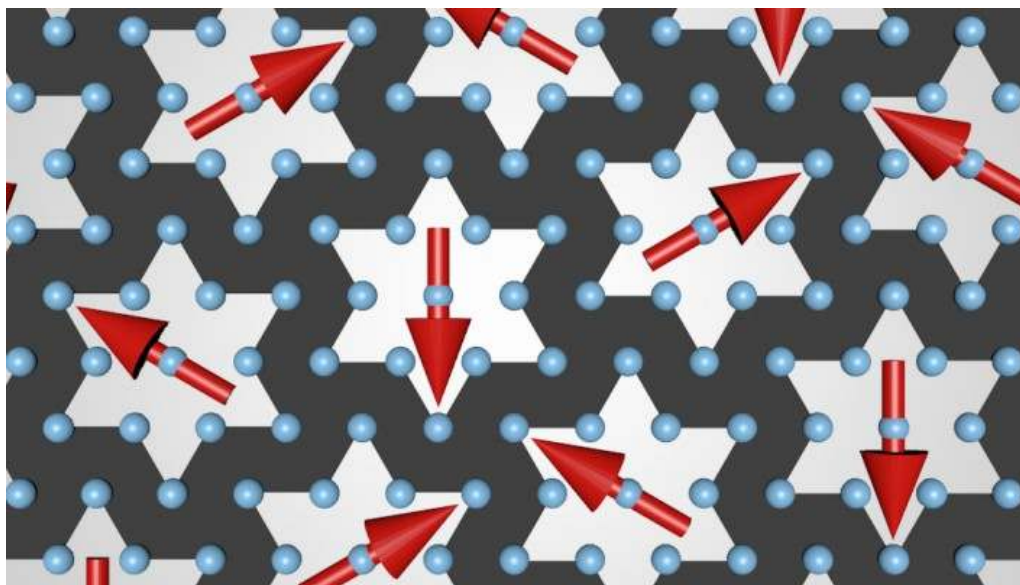
Two-dimensional electron gas



1H-TaS₂

Moment formation in 1T-TaS₂

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



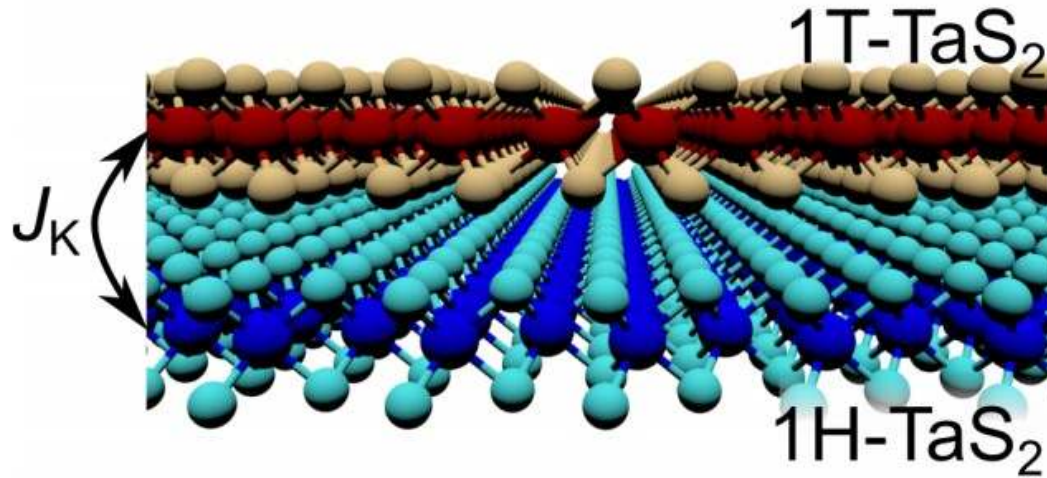
Strong interactions give rise to local moment formation

Effectively a $S=1/2$ Heisenberg model in a triangular lattice

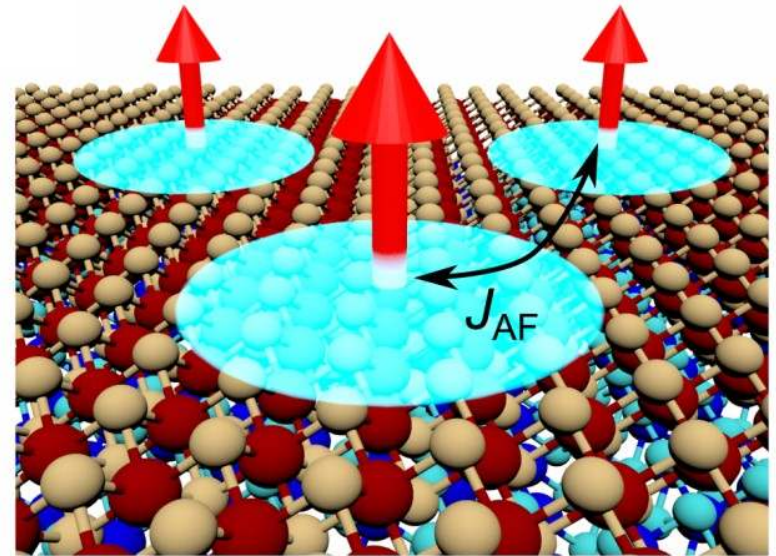
PNAS 114.27 (2017)
Phys. Rev. X 7, 041054 (2017)
Phys. Rev. B 96, 195131 (2017)

Heavy-fermions in dichalcogenide bilayers

Interlayer coupling creates
the Kondo coupling



Intralayer exchange between
local moments



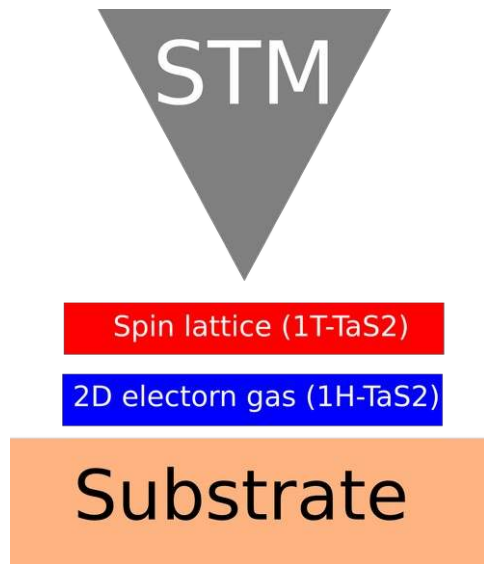
The bilayer realizes all the ingredients for heavy-fermion physics

Experimental signatures of heavy-fermion physics

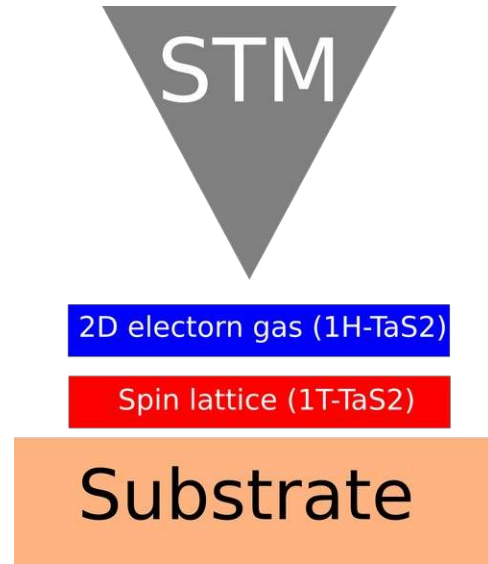
- Kondo physics in the magnetic lattice
- Gap opening in the metallic layer

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

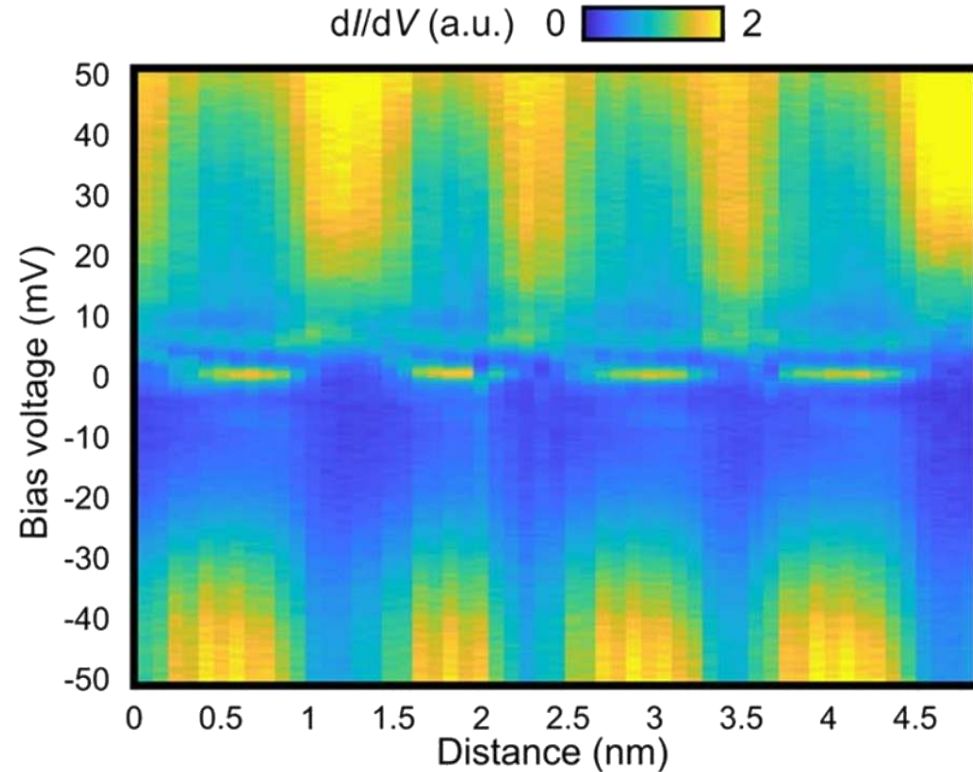
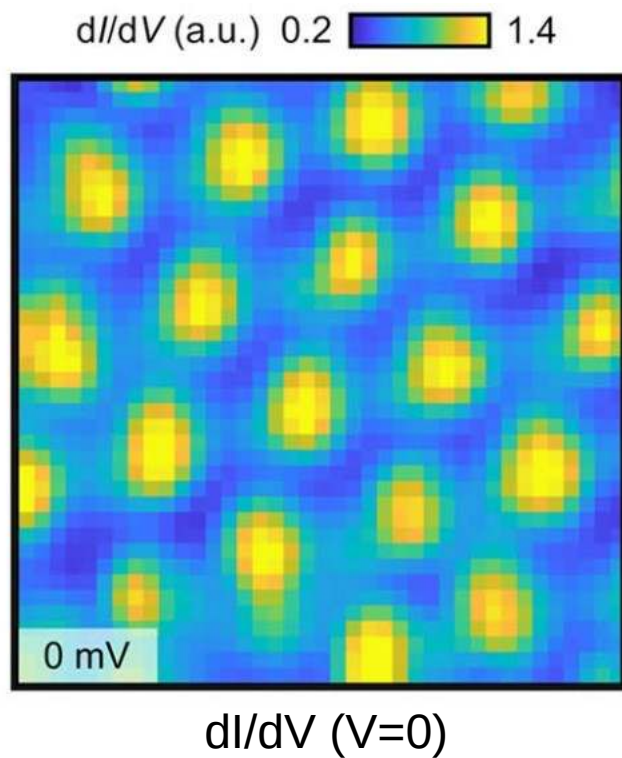
Probing the Kondo peak



Probing the heavy-fermion gap

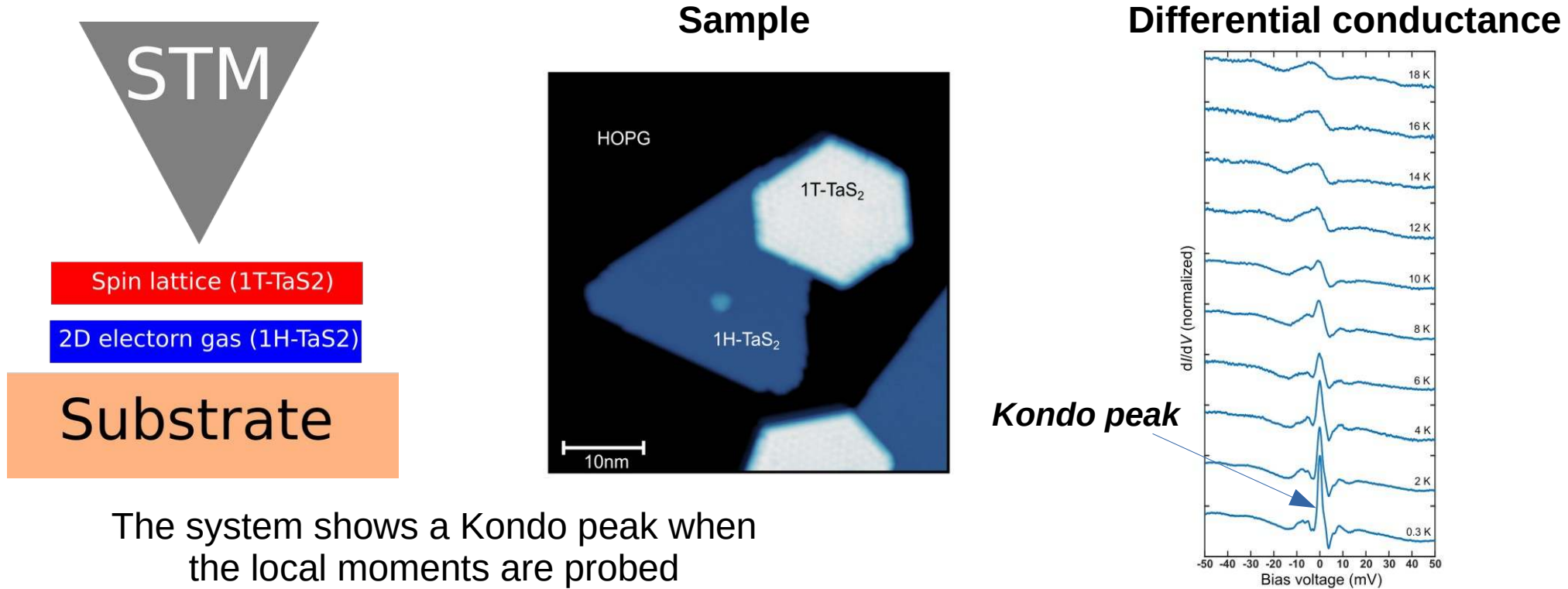


Grid spectroscopy of 1T-1H



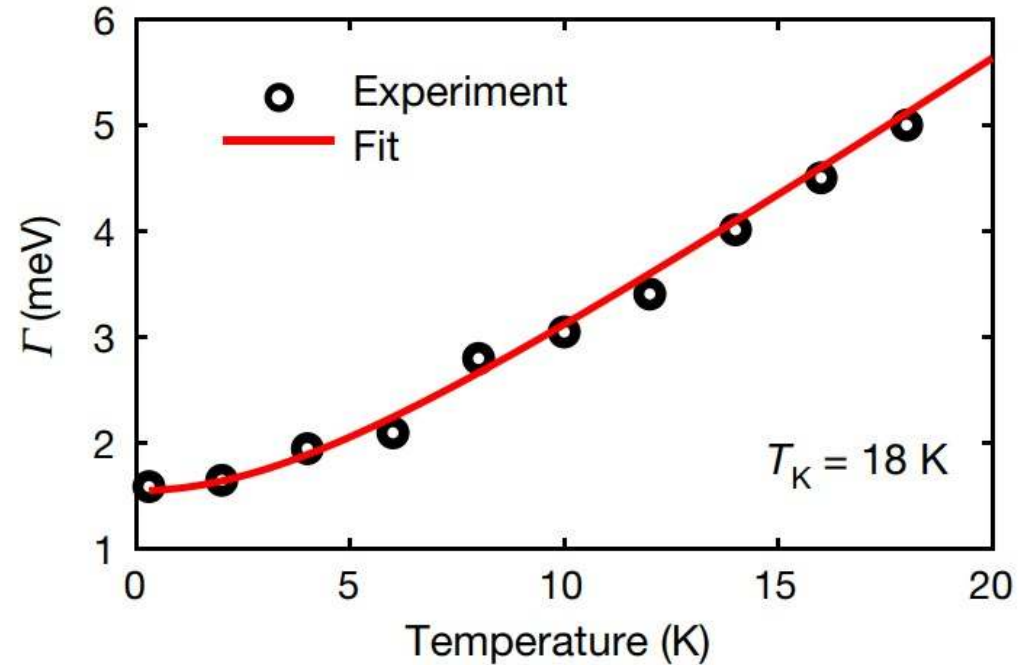
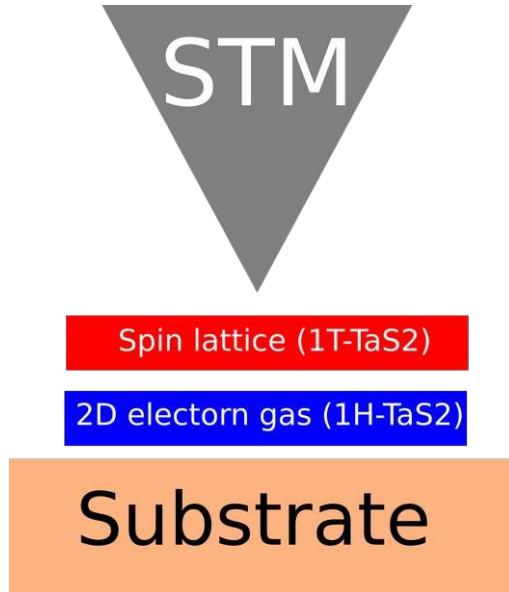
Zero bias (Kondo) peak appearing with the CDW periodicity

Kondo physics in the magnetic lattice



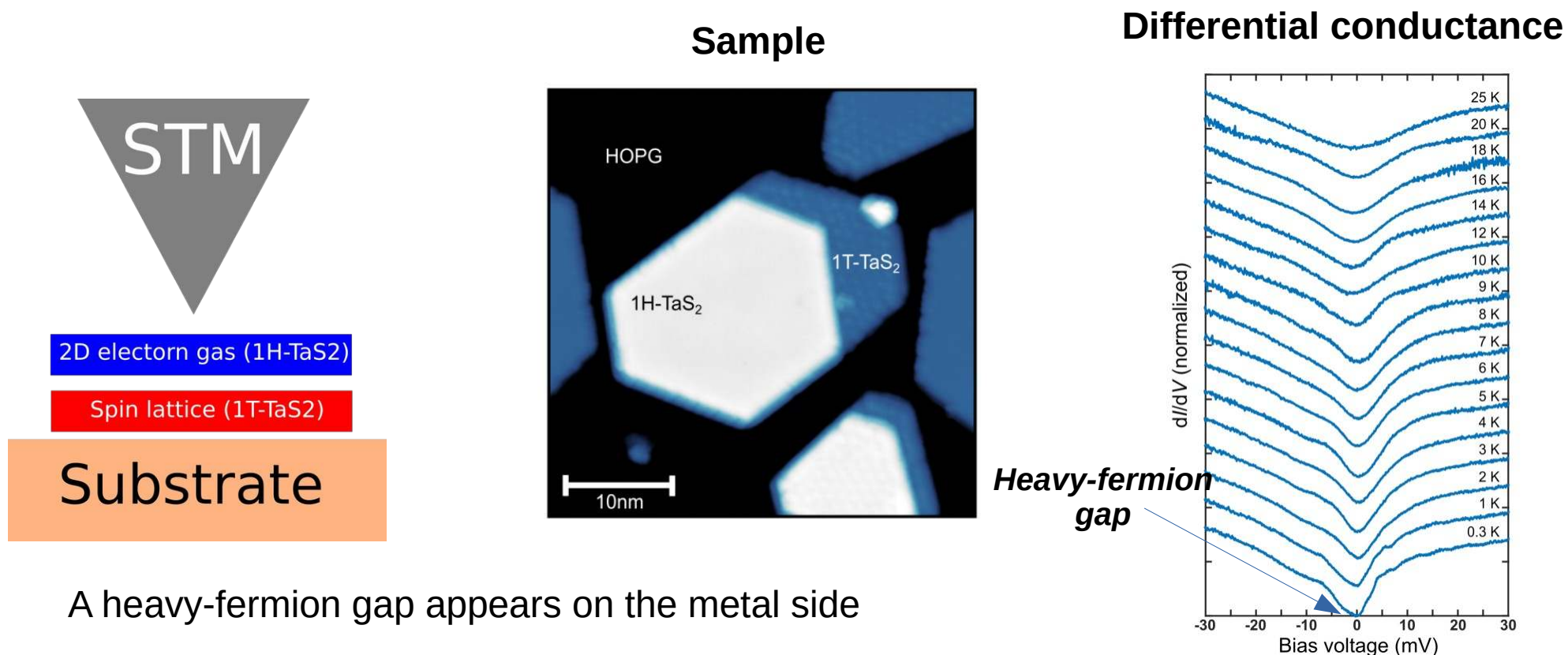
Temperature and field dependence consistent with Kondo

Kondo physics in the magnetic lattice



The Kondo peak follows the expected temperature dependence

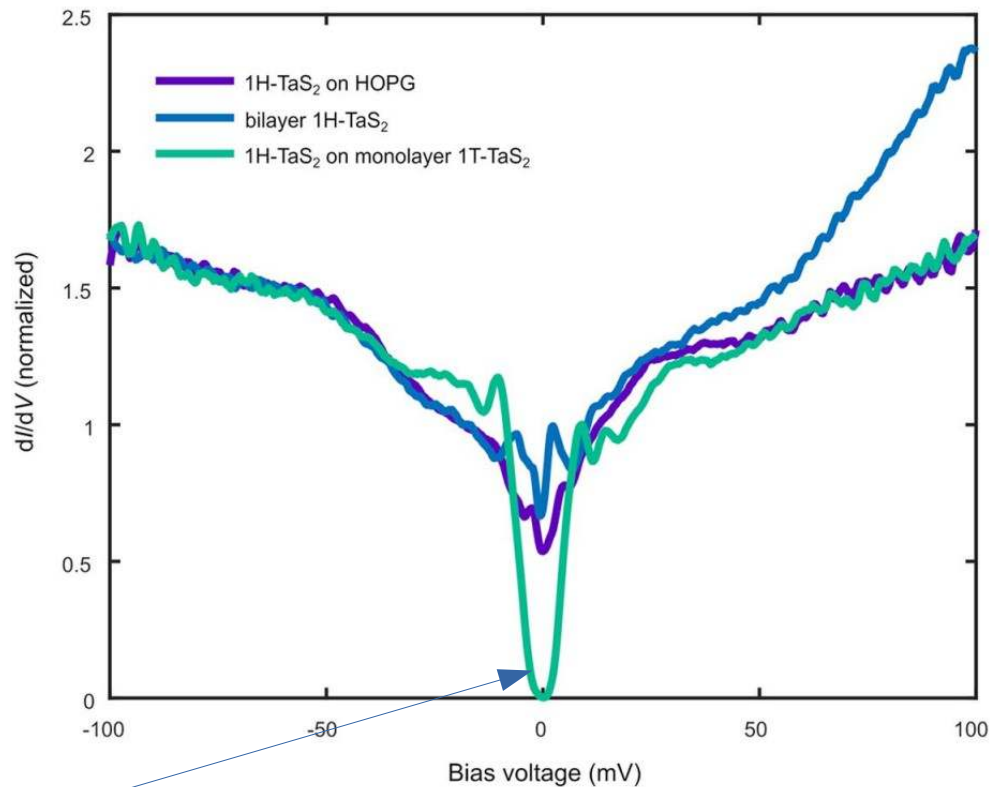
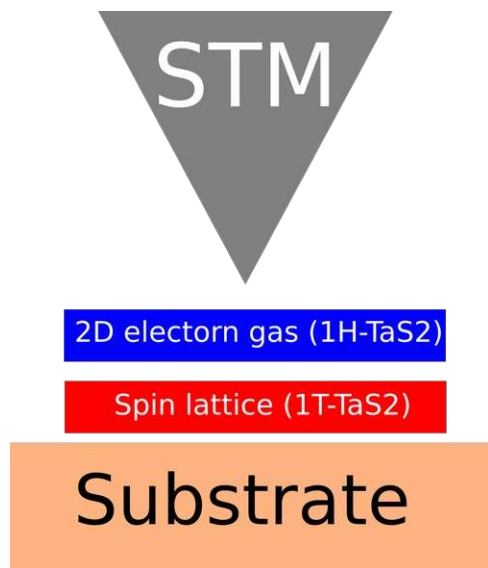
Heavy-fermion gap in the 2D electron gas



A heavy-fermion gap appears on the metal side

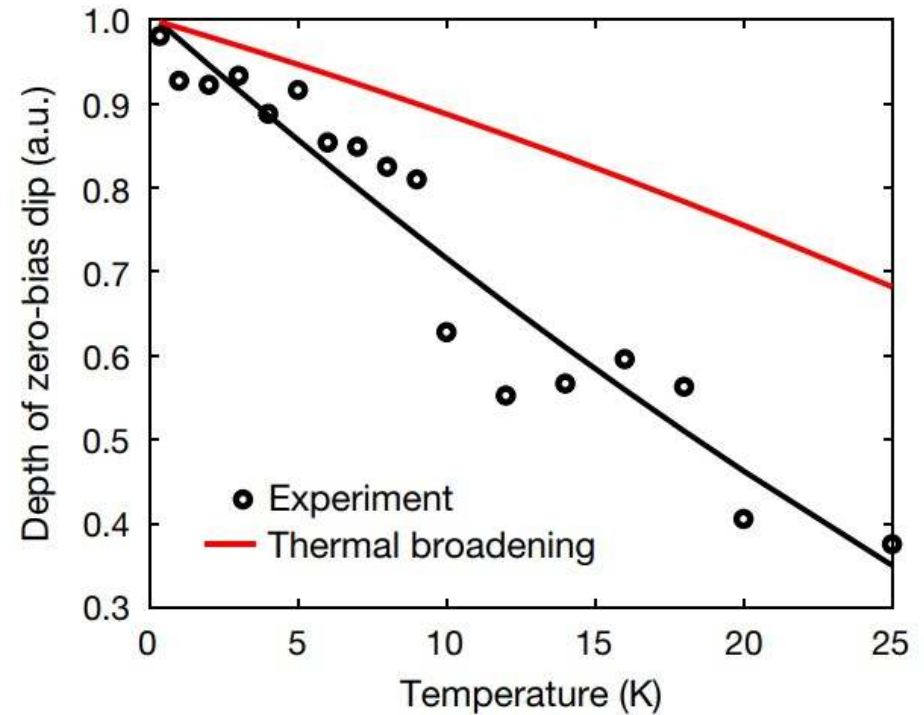
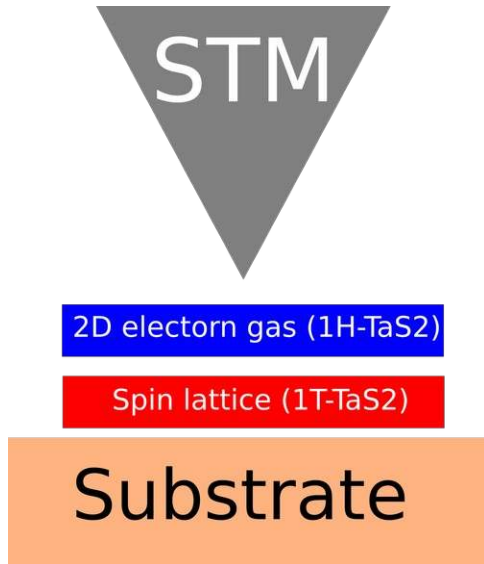
Temperature and field dependence consistent with a heavy-fermion gap

Heavy-fermion gap in the 2D electron gas



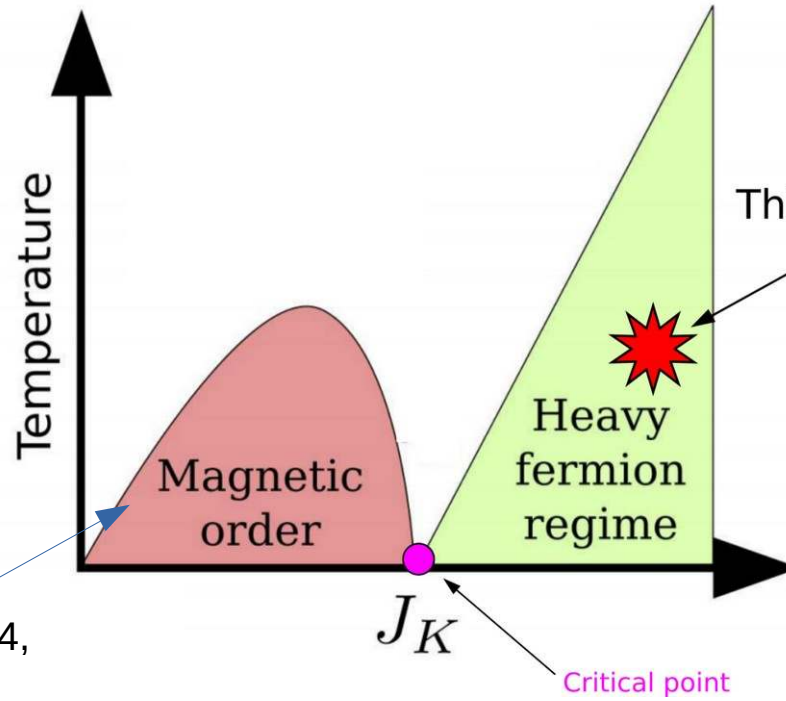
Heavy-fermion gap

Heavy-fermion gap in the 2D electron gas



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

The phase diagram van der Waals heavy-fermions



Nature Communications 14,
7005 (2023)

This experiment
Nature 599, 582–586 (2021)

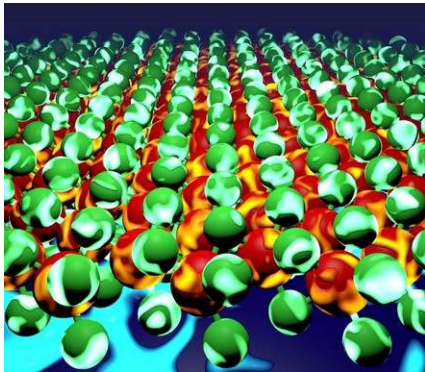
**Can we push this system to critical and different
unconventional superconducting regimes?**

Controlling the heavy-fermion state

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

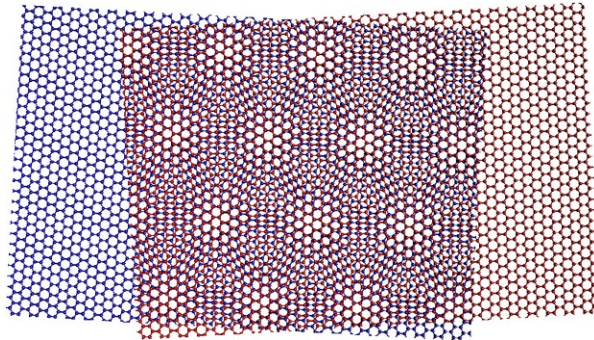
Most importantly, we can exploit the full tunability of van der Waals materials

Gating



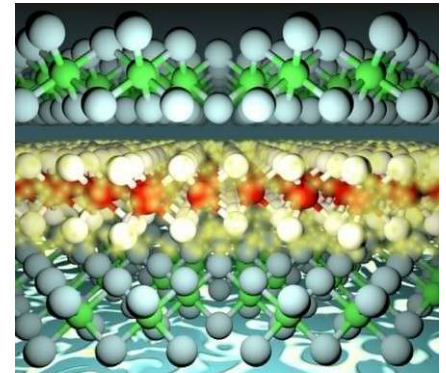
Science 306, 5696, 666-669 (2004)

Twist engineering



Nature Reviews Materials 6, 201–206 (2021)

Materials engineering

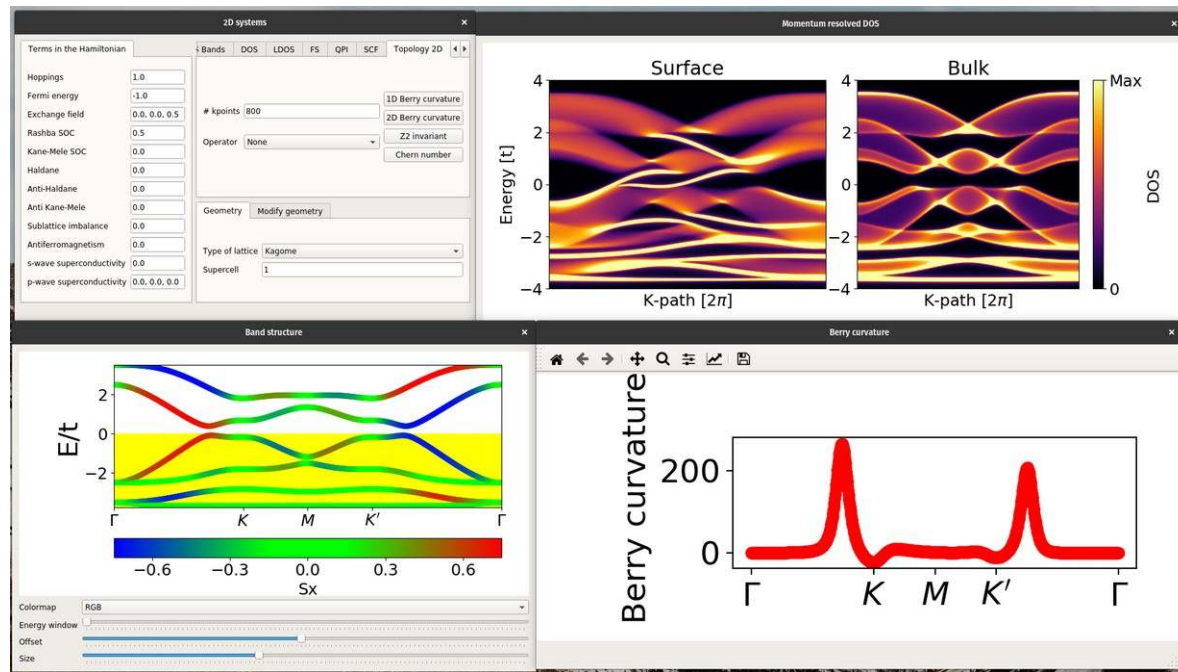
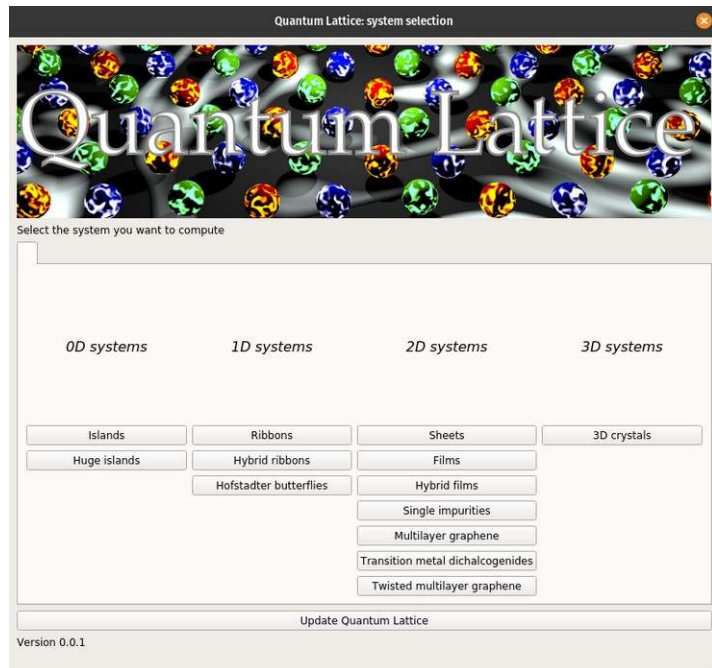


Nature 499, 419–425 (2013)

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

Open-source software for
van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

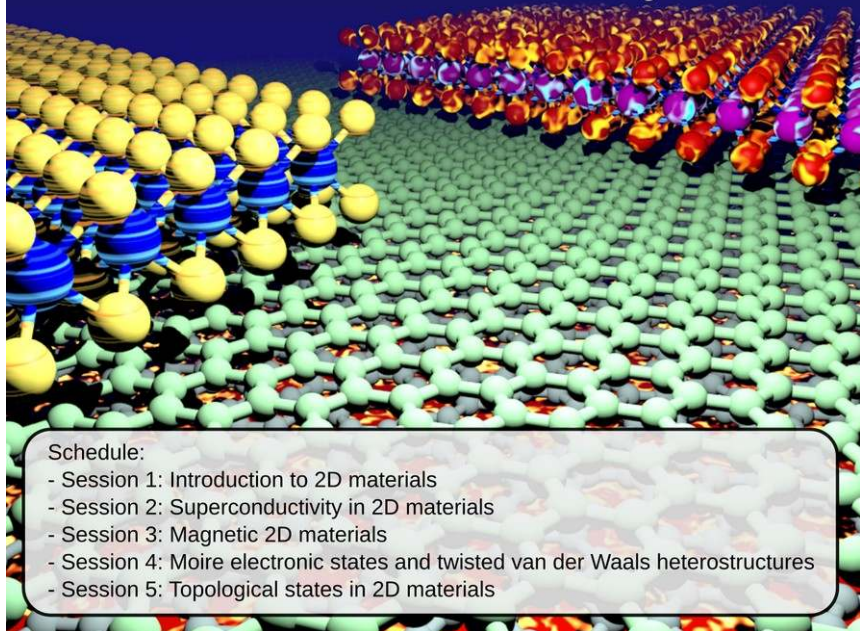


Quantum Lattice: open source interactive interface for tight binding modeling

<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

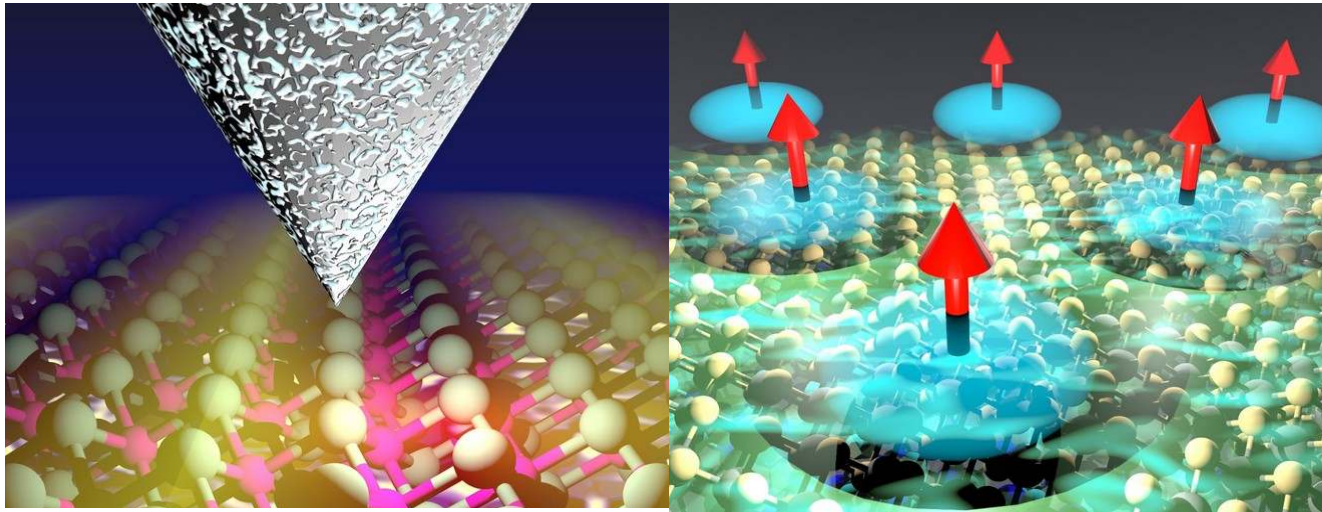


<https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ>

- 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
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Take home

Van der Waals materials provide a new platform to engineer exotic matter, including multiferroics and heavy-fermion Kondo lattices



2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)
Advanced Materials, 2311342 (2024)

Phys. Rev. Lett. 127, 026401 (2021)
Nature 599, 582–586 (2021)
Nano Letters 24 (14), 4272–4278 (2024)

Thank you

Funding from

