Artificial van der Waals multiferroics with twisted two-dimensional materials

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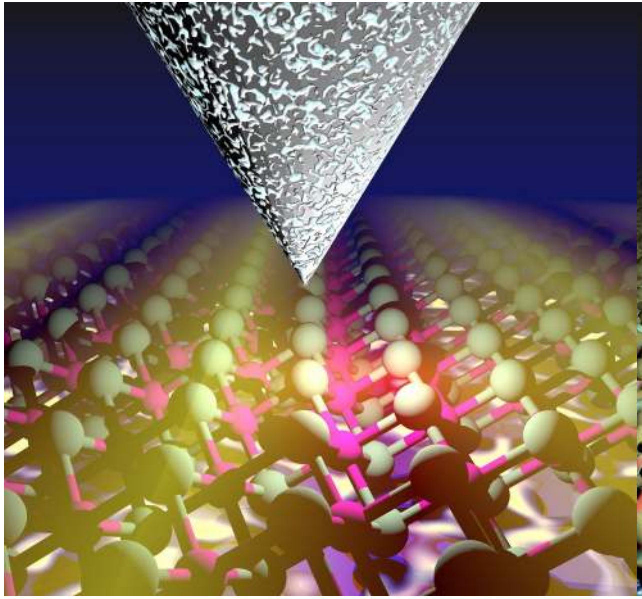
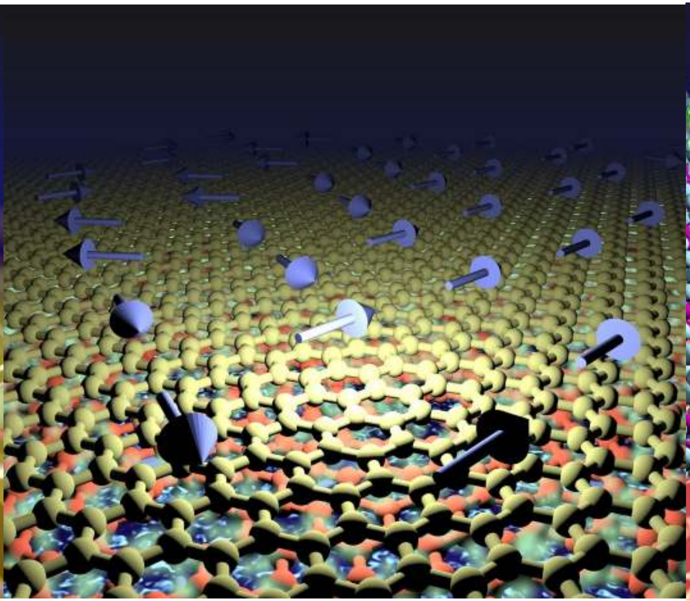
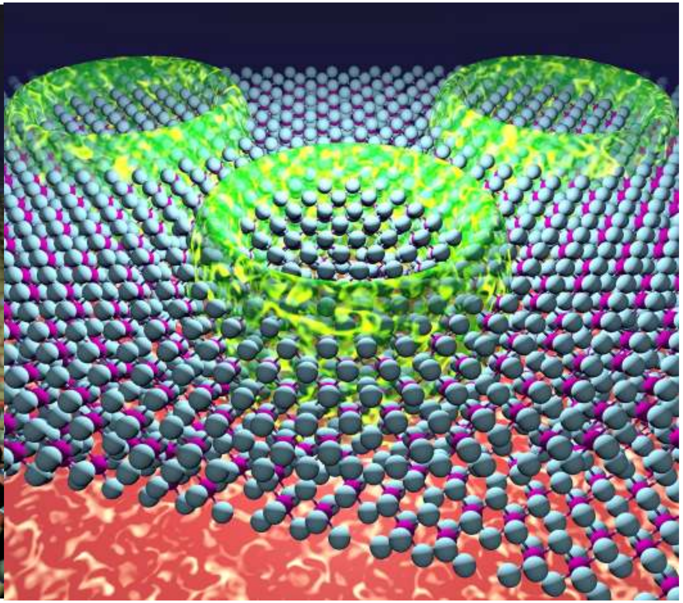


2D Materials 9 (2), 025010 (2022)
arXiv:2309.11217 (2023)



2D Materials, 9 (2), 024002 (2022)



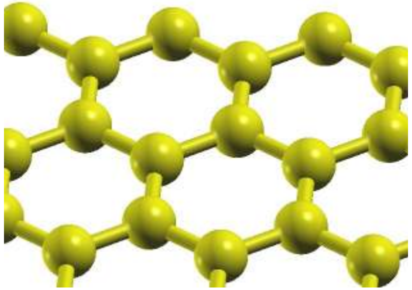
2D Materials 10 (2), 025026 (2023)

Workshop on Twistronics and Moiré Materials: Bridging Theory and Experiments, ICTP Trieste
January 19th 2024

Plenty of room for twisting

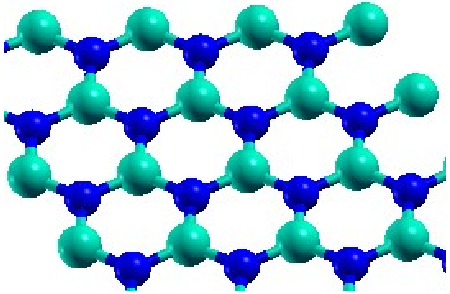
Semimetal

Graphene



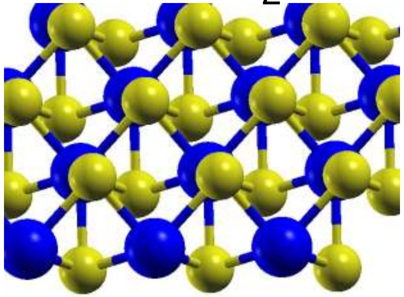
Insulator

BN



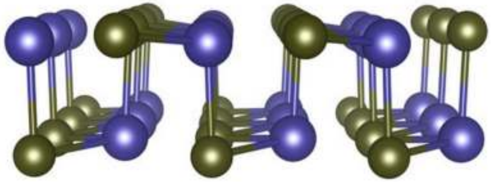
Superconductor

$NbSe_2$



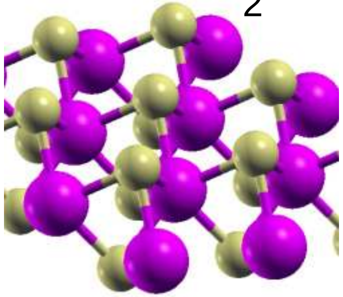
Ferroelectric

SnTe



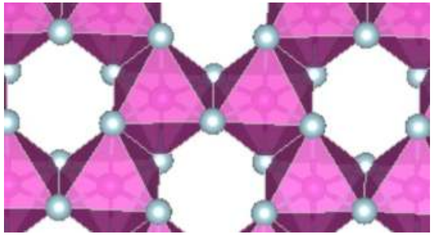
Semiconductor

WSe_2



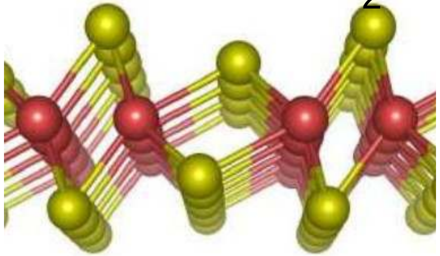
Ferromagnet

CrI_3



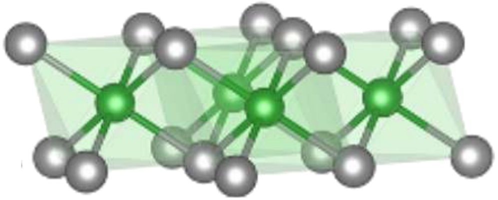
Quantum spin Hall insulator

1T’- WTe_2



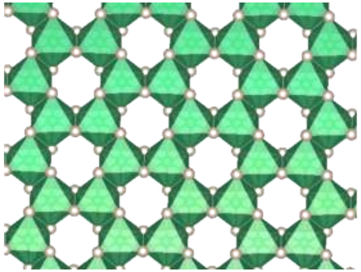
Multiferroic

NiI_2



Van der Waals magnetic materials

Ferromagnet, antiferromagnets

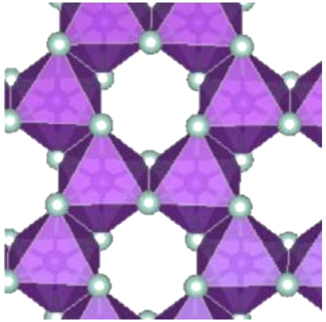


CrI_3, $CrCl_3$, $CrBr_3$

Nature 546, 270–273 (2017)

Break time-reversal

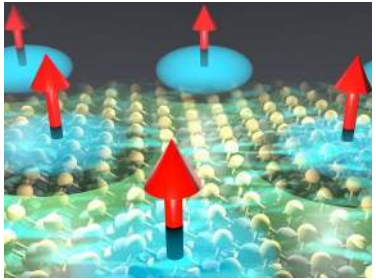
(proximal) Quantum spin-liquids



$RuCl_3$, 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Heavy-fermion Kondo insulators

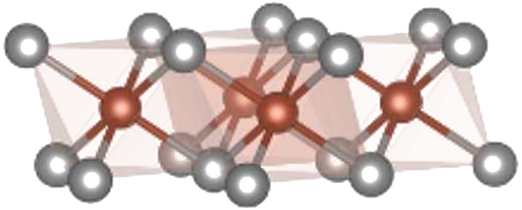


1T-TaS_2/1H-TaS_2

Nature 599, 582–586 (2021)

Do not break time-reversal

Multiferroics



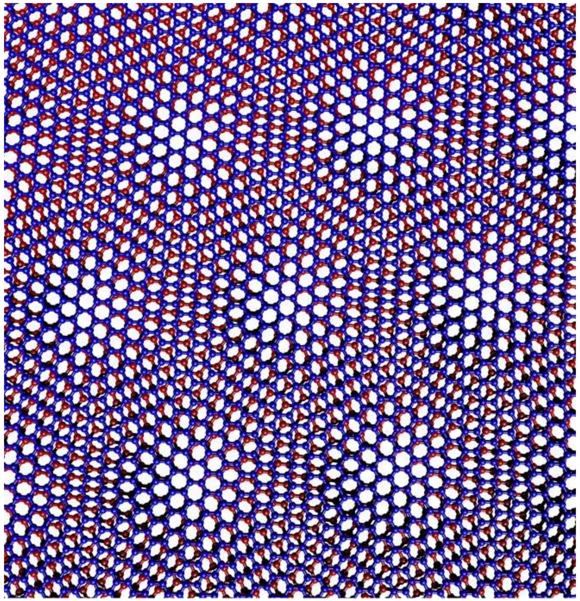
NiI_2

Nature 602, 601–605 (2022)

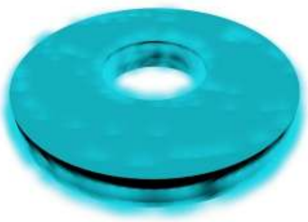
Break time-reversal and inversion symmetry

The phases of twisted bilayer graphene

Twisted bilayer graphene



Superconductivity

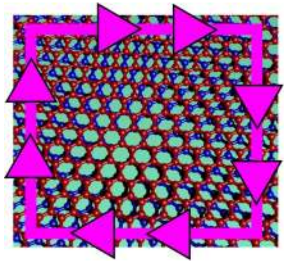


Nature 556, 43–50 (2018)

Topological networks

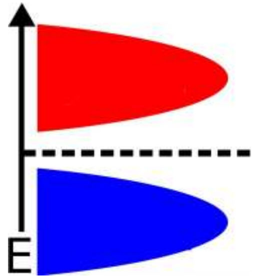
Nano Lett. 18, 11, 6725-6730 (2018)

Chern insulators



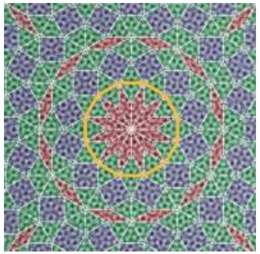
Science 365, 605-608 (2019)

Correlated insulators



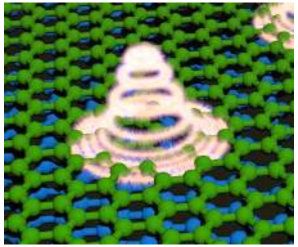
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782-786 (2018)

Proximal fractional Chern insulators

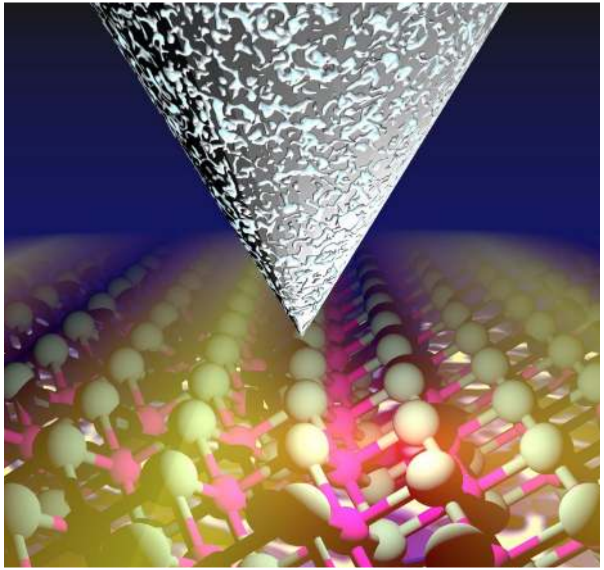


Nature 600, 439–443 (2021)

Which new phenomena could we get by twisting magnetic materials?

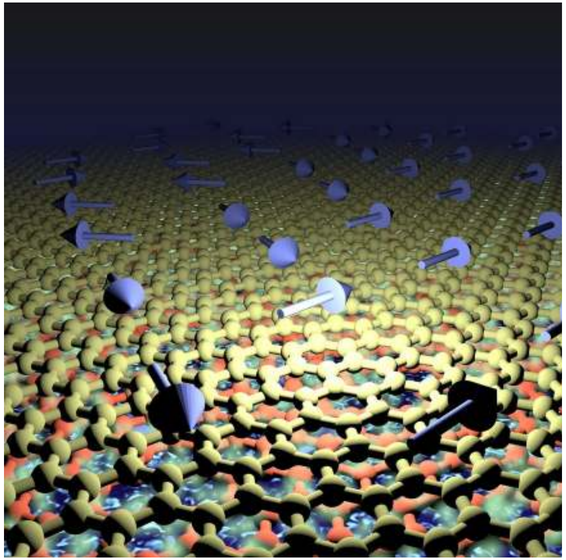
Today's plan

Multiferroicity and the built-in moire of NiI_2



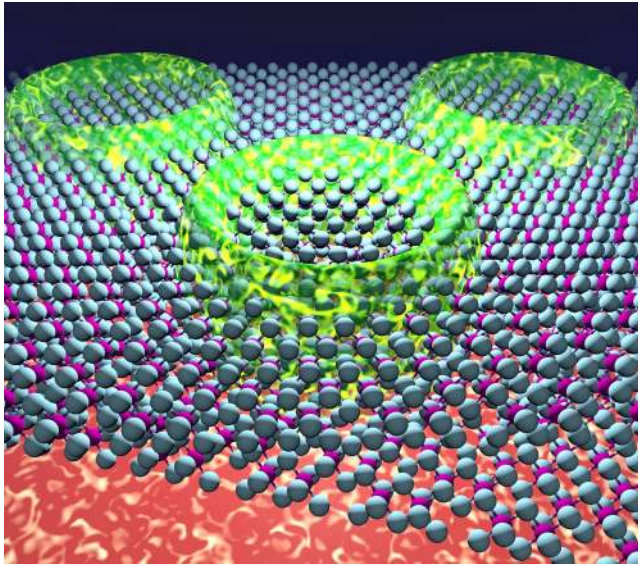
2D Materials 9 (2), 025010 (2022)
arXiv:2309.11217 (2023)

The impact of multiferroic NiI_2 proximity in TBG



2D Materials 9 024002 (2022)

Artificial multiferroicity by twisting 2D magnets



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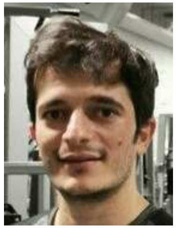
Behind the scenes

Intrinsic multiferroics, NiI_2

(theory and experiment)

V. Vaňo

M. Amini



A. Fumega

H. Gonzalez Herrero

S. Kezilebieke

P. Liljeroth

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arXiv:2309.11217 (2023)

Controlling moire states with a multiferroic

(theory)

Guangze Chen

Maryam Khosravian

Aline Ramires

2D Materials 9 (2) 024002 (2022)

Artificial multiferroics, twisted $CrBr_3$ bilayers

(theory)

A. Fumega

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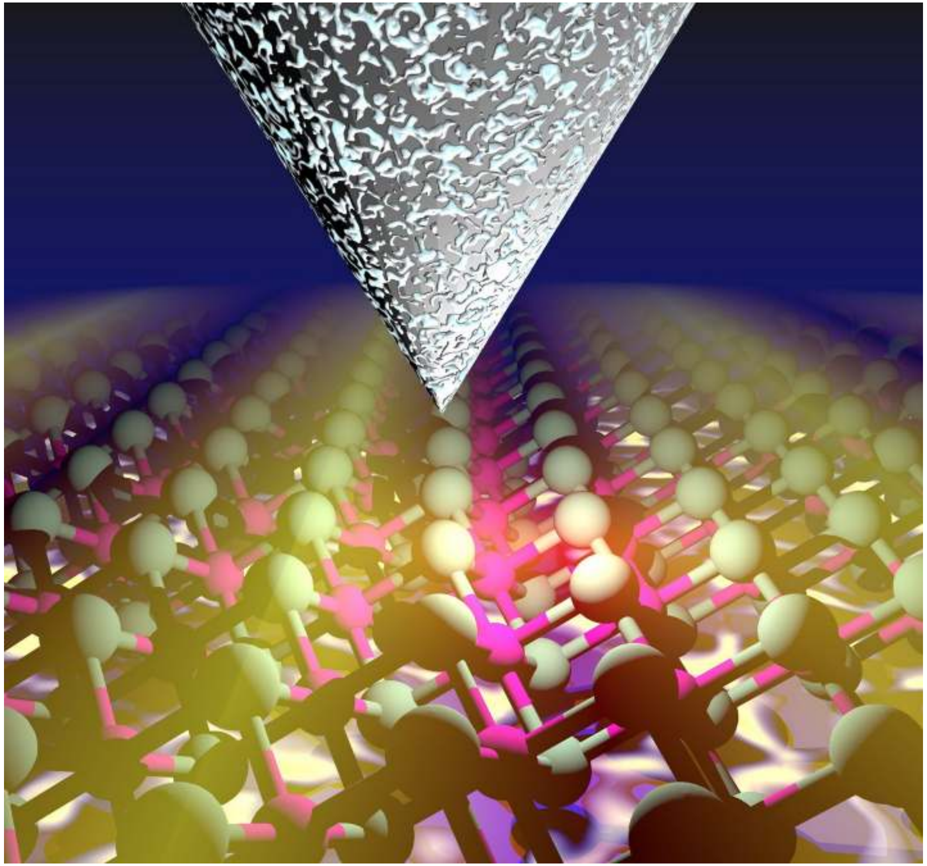
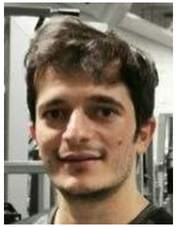
Origin and visualization of multiferroicity in NiI_2



V. Vaňo

M. Amini



A. Fumega

H. Gonzalez Herrero

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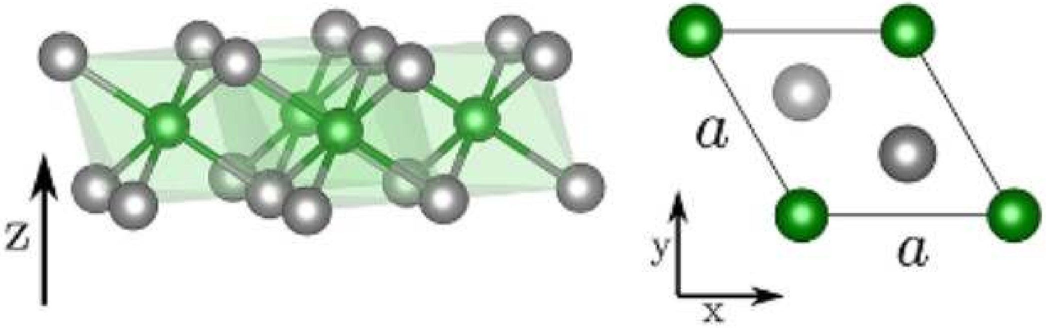
P. Liljeroth

2D Materials 9 (2), 025010 (2022)
arXiv:2309.11217 (2023)

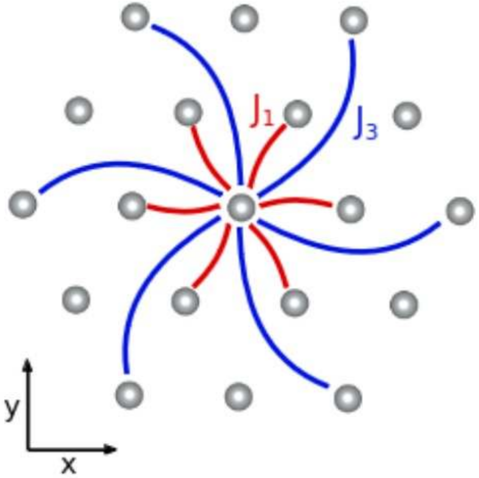
Monolayer NiI_2

Structure

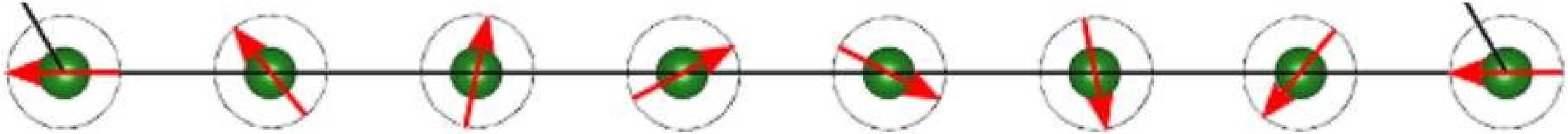


Nature 602, 601–605 (2022)

Frustrated magnetic Heisenberg model

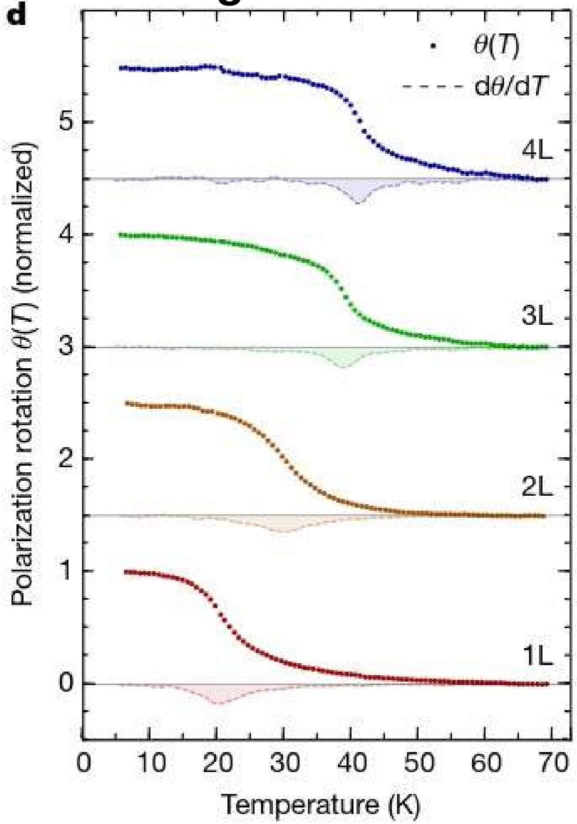


Magnetic structure

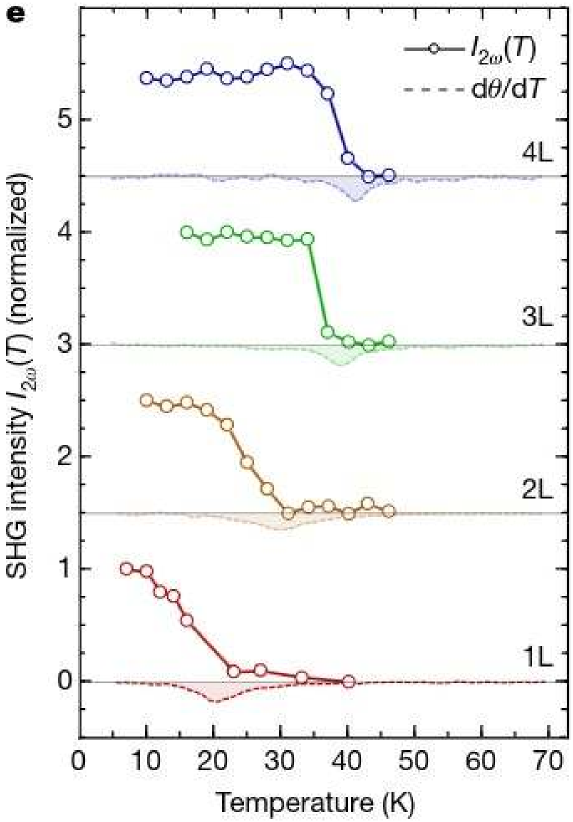


Multiferroicity in monolayer NiI_2

Magnetic order



Ferroelectric order



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

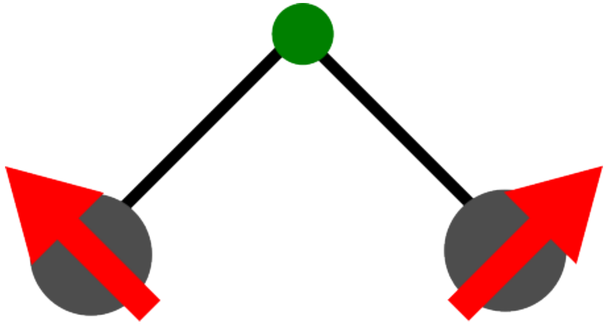
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

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

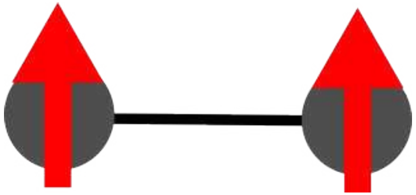
Antisymmetric exchange

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



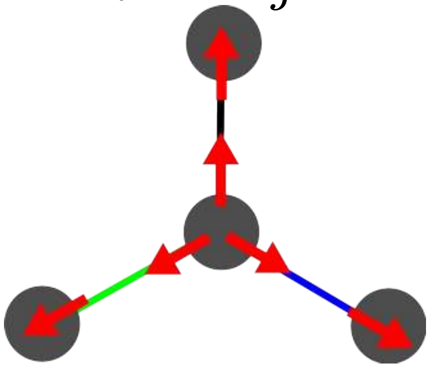
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

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



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

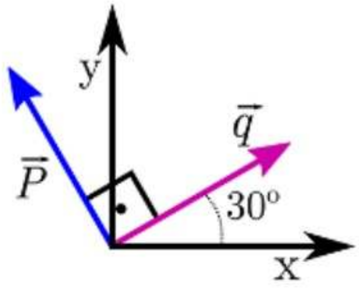
In NiI_2, the ferroelectricity is created by inverse antisymmetric exchange

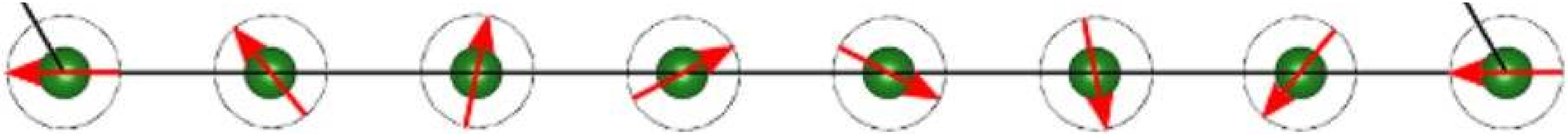
The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

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

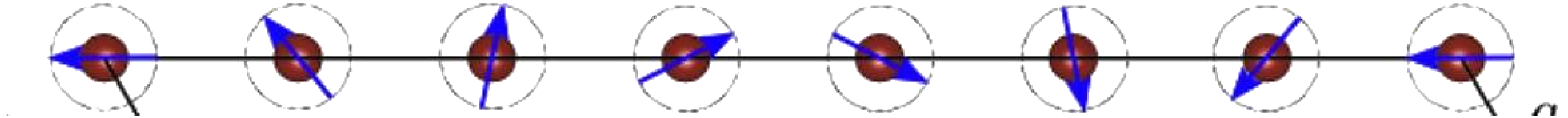
$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e}\cdot\mathbf{S})(\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}_0))}\mathbf{M}(\mathbf{r}_0)$$





The emergence of non-collinear order is drives ferroelectricity

The origin of ferroelectricity in NiI_2



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

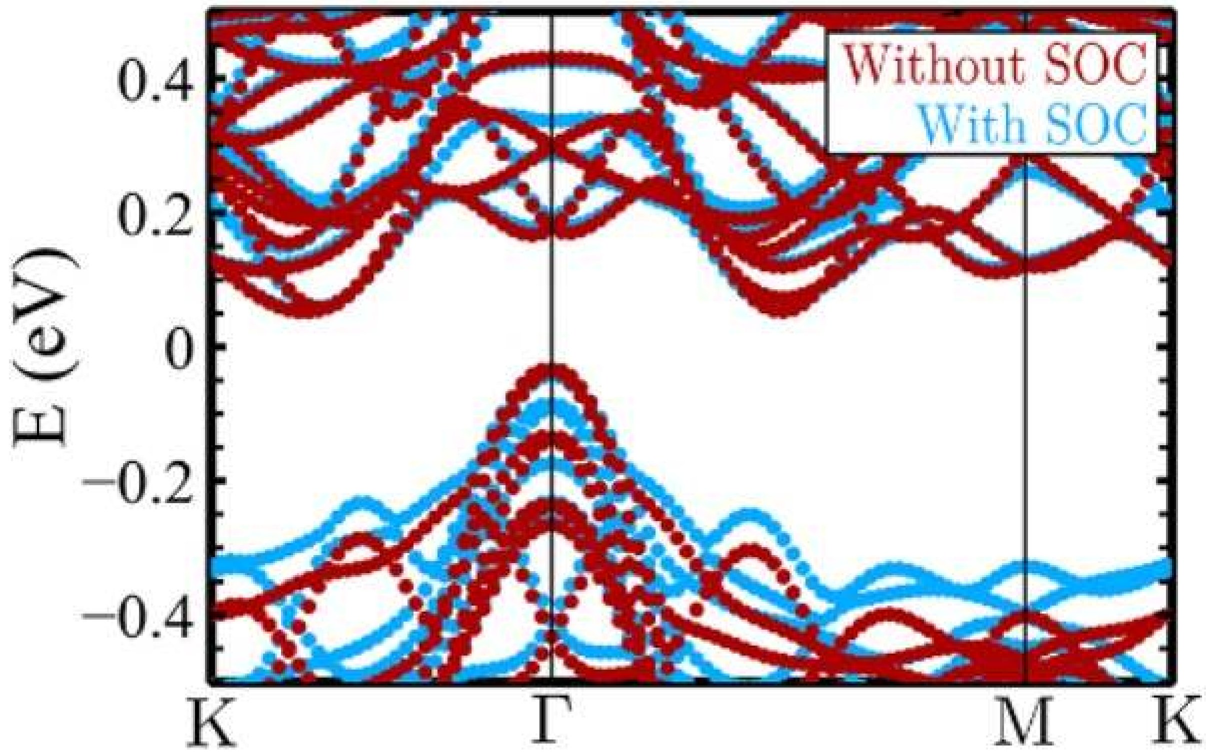
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

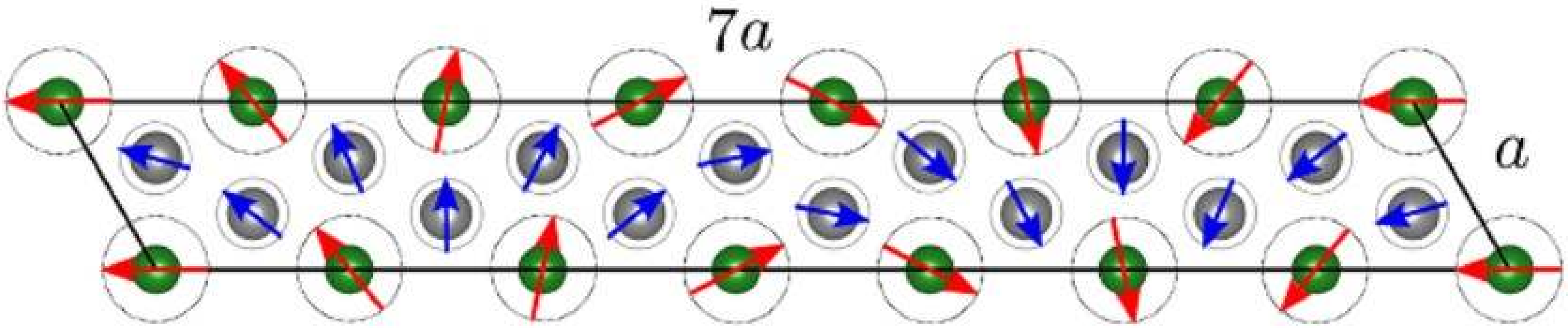
The impact of spin-orbit coupling in the electronic structure



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

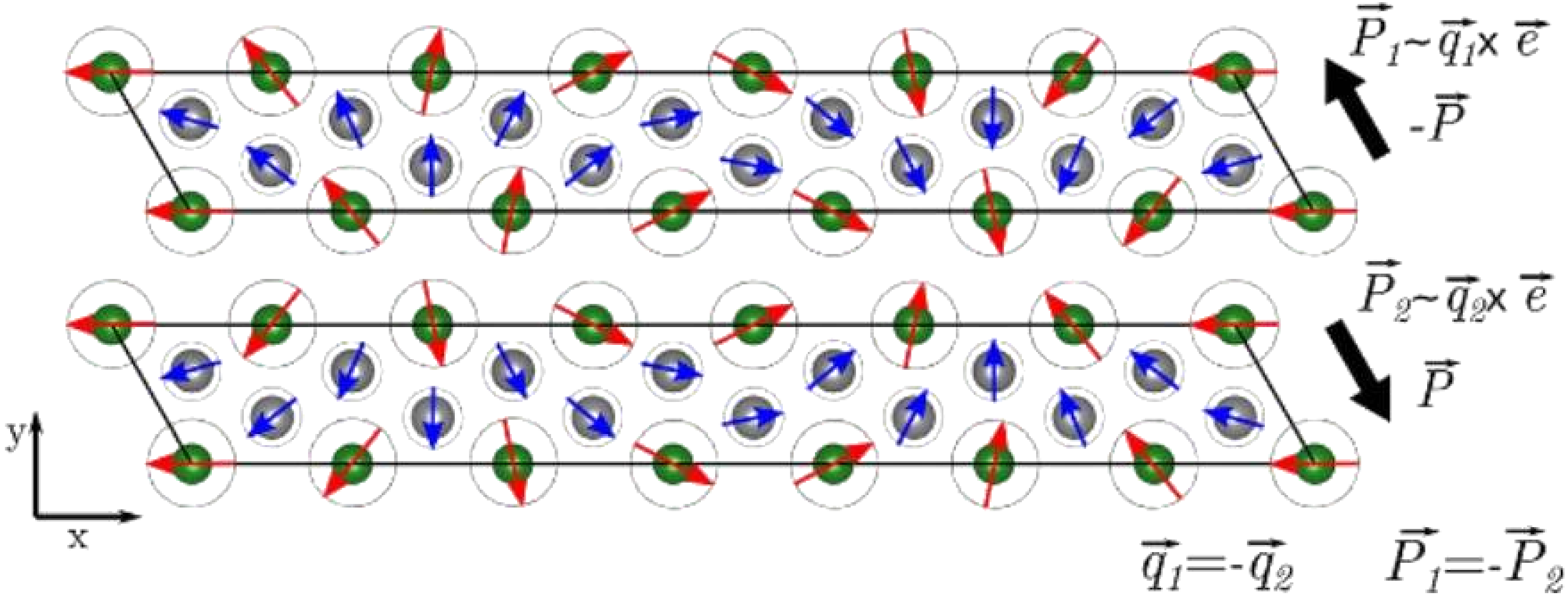
There is a major role of the halide close to the Fermi level

The interplay between magnetism and halide electronic structure



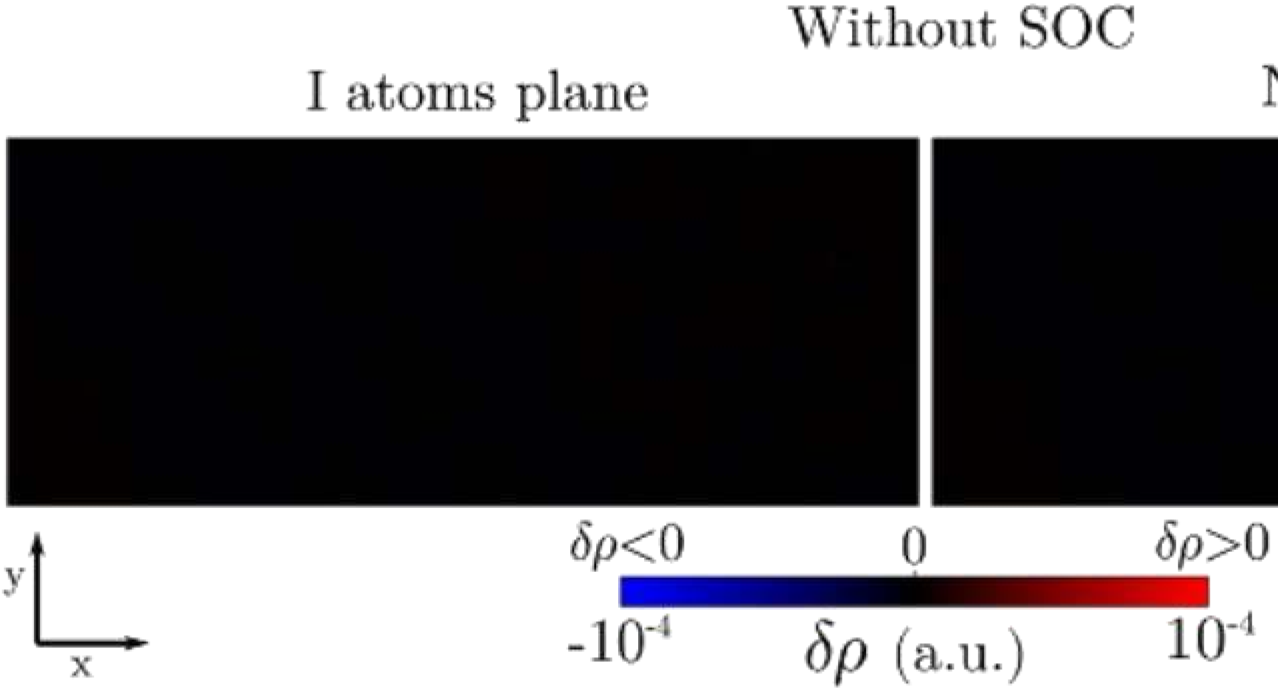
Magnetism not only appears in Ni, but a sizable value merges also in I

Ferroelectric correction to the electronic density



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

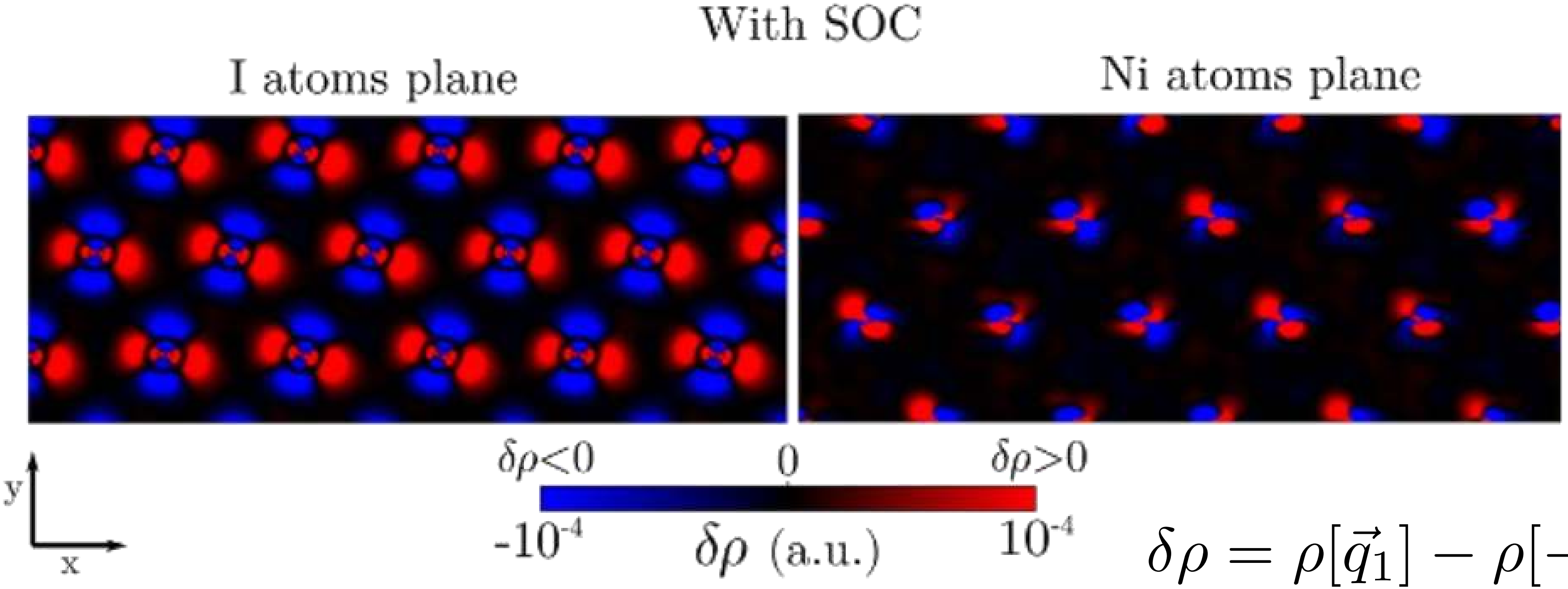
Ferroelectric correction to the electronic density



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

In the absence of SOC, no ferroelectric density appears

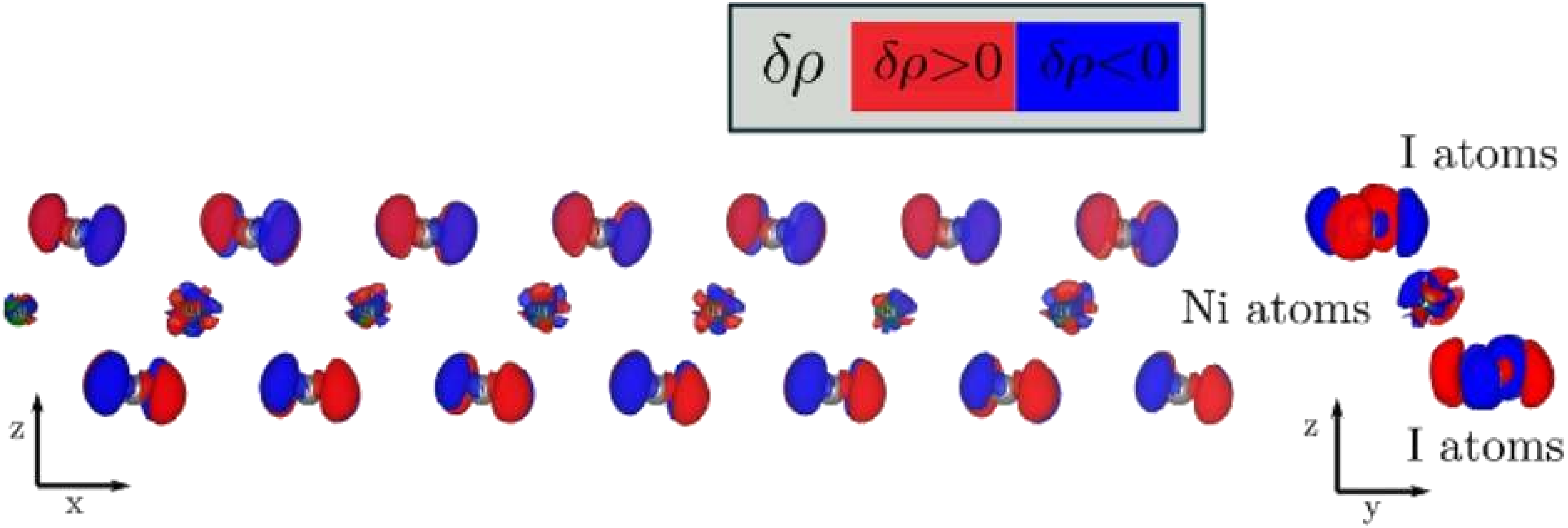
Ferroelectric correction to the electronic density



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

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

Ferroelectric correction to the electronic density

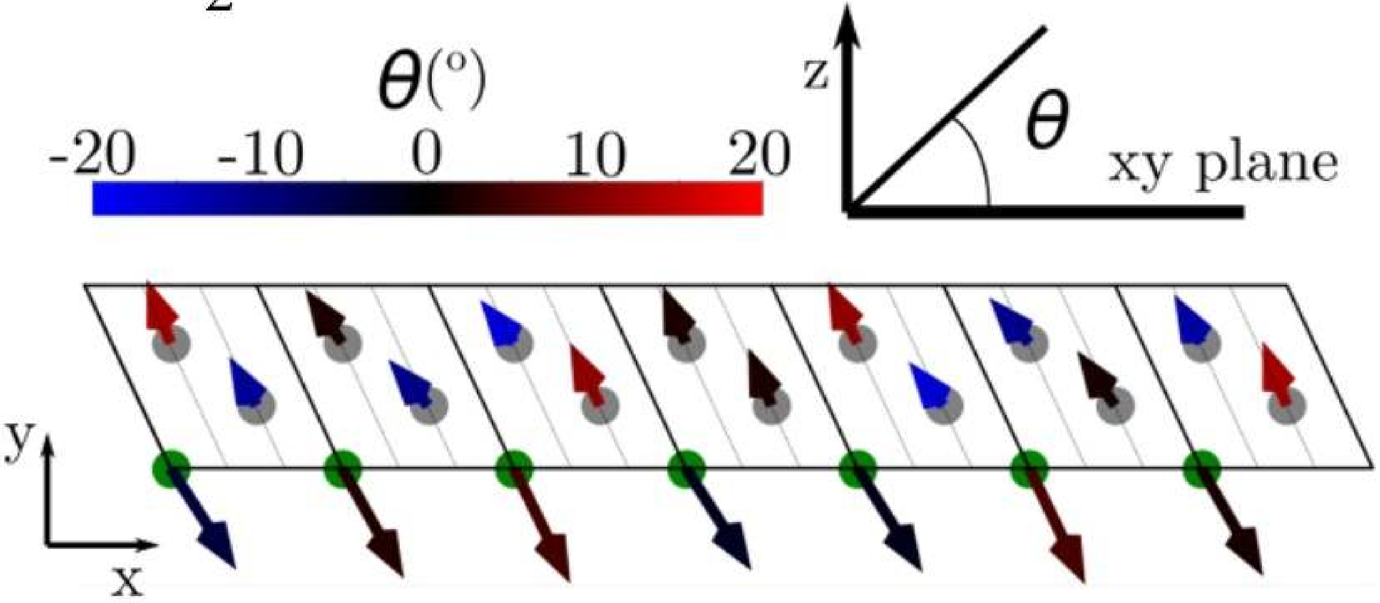


$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

The ferroelectric density has comparable magnitudes in Ni and I

The ferroelectric force

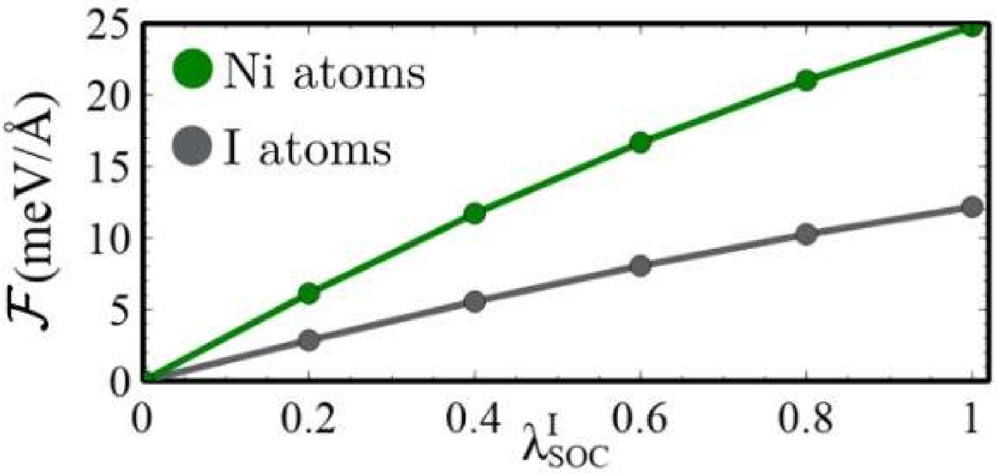
$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$

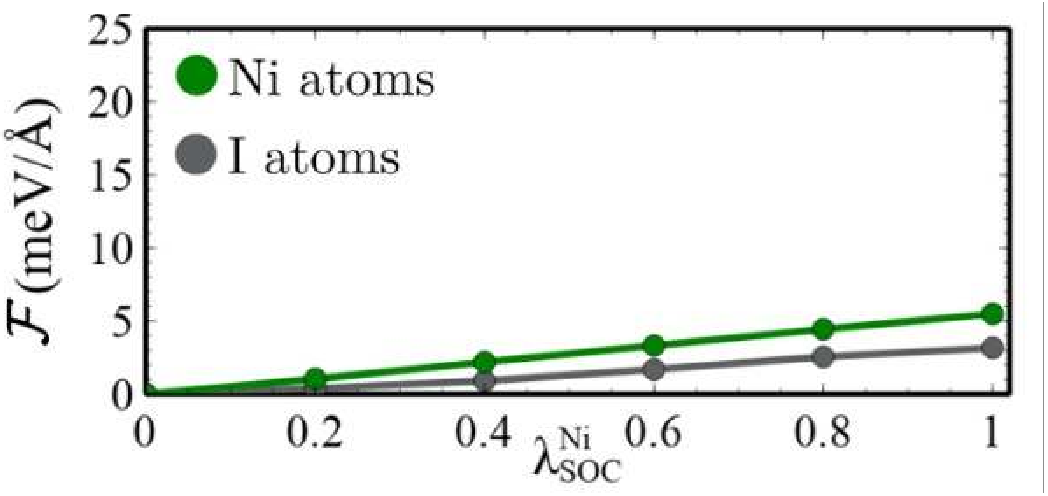


The forces between the two magnetic configuration account for the ferroelectric displacement

The ferroelectric force

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

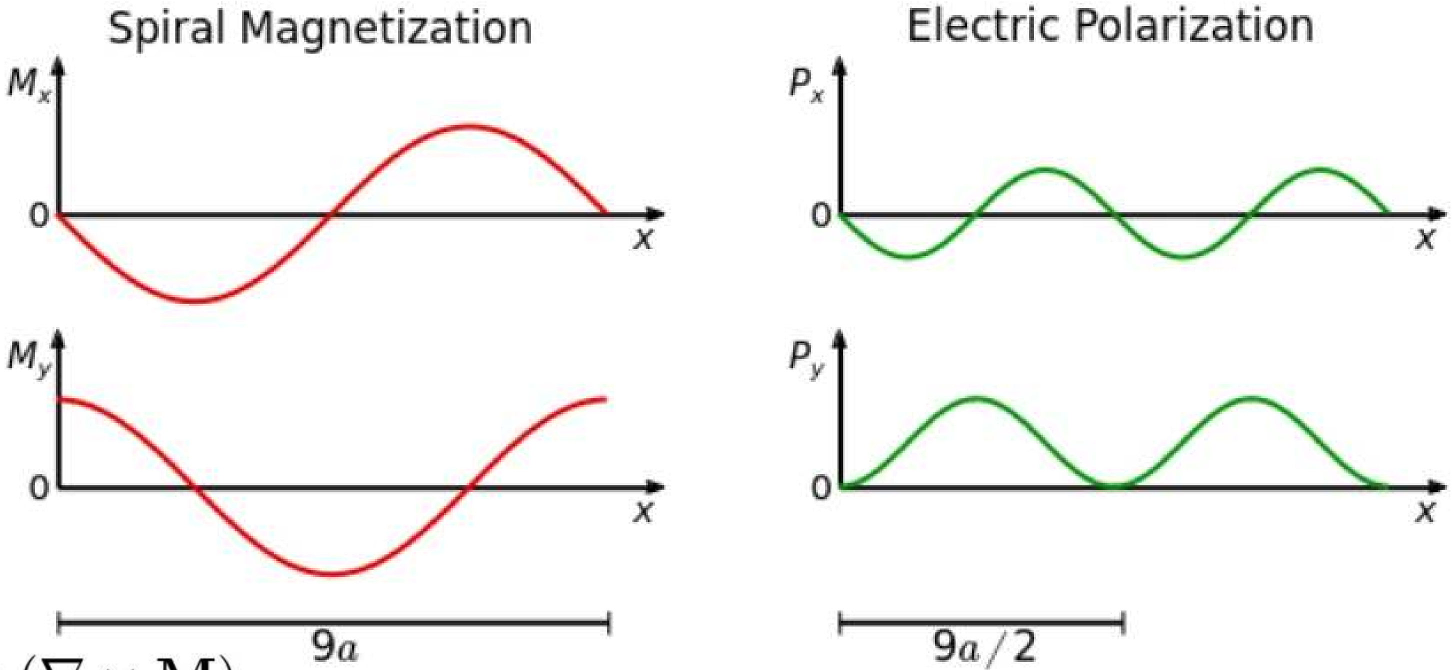




The dominant contribution to the ferroelectric force comes from iodine

Modulated multiferroicity in monolayer NiI_2

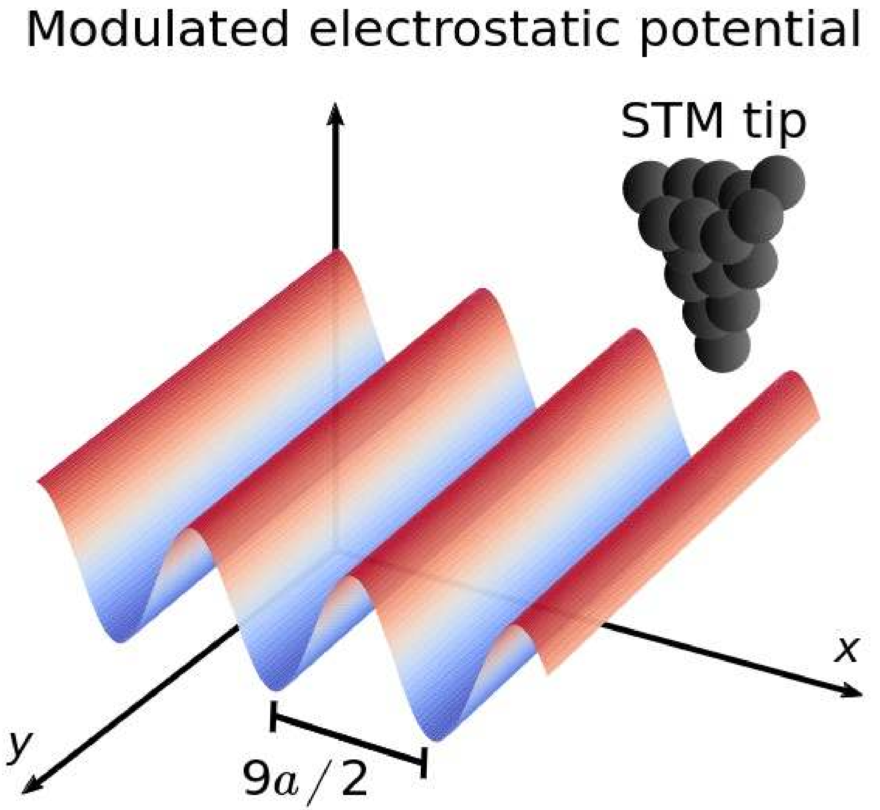
The ferroelectric order has actually a modulation in real space



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

arXiv:2309.11217 (2023)

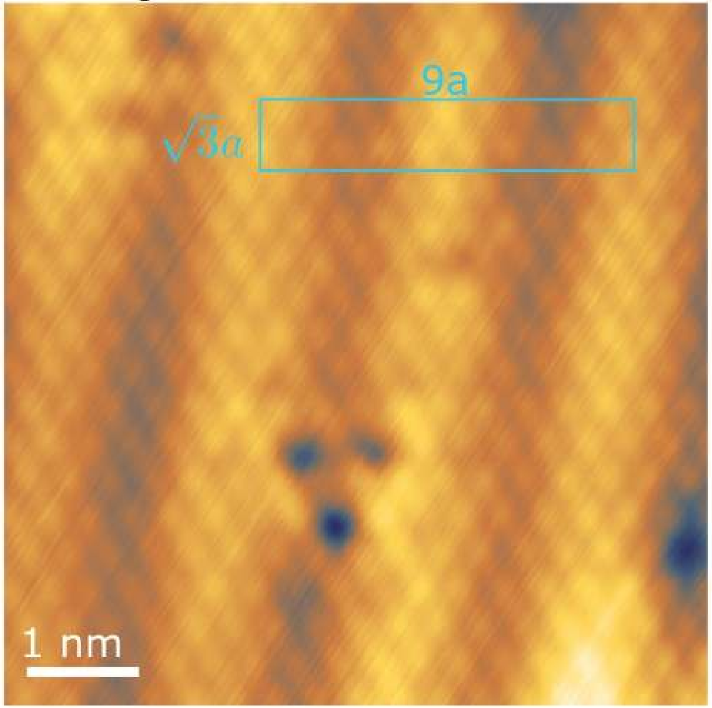
Visualizing multiferroicity in monolayer NiI_2



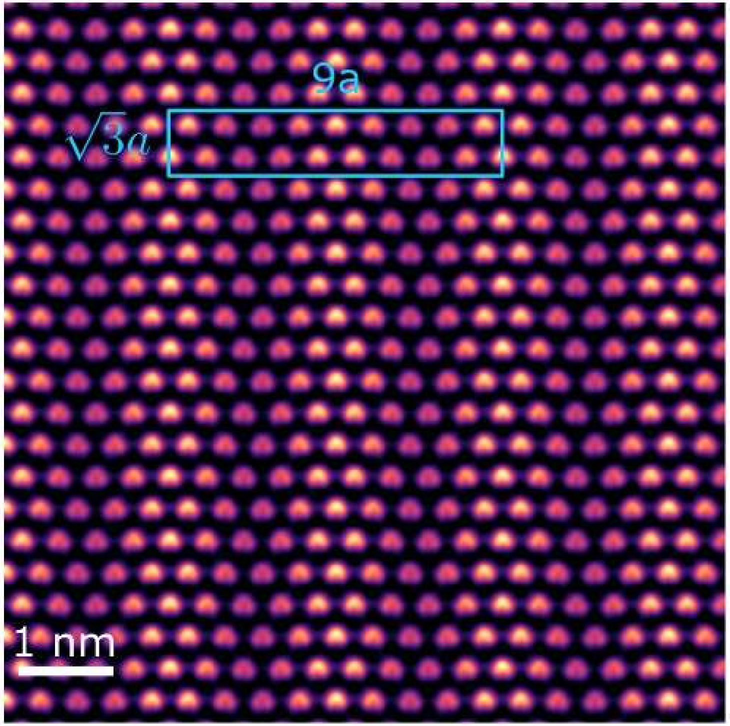
arXiv:2309.11217 (2023)

Visualizing multiferroicity in monolayer NiI_2

Experiment



Theory

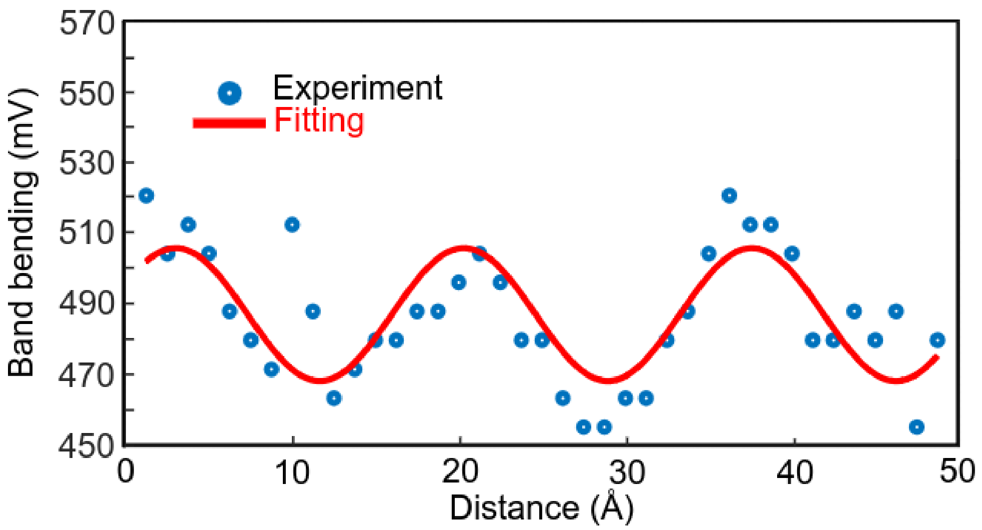


$$\Lambda \mathrm{q}(-\sin(2\mathbf{q}\cdot\mathbf{r}), \sin^2(\mathbf{q}\cdot\mathbf{r}), 0)$$

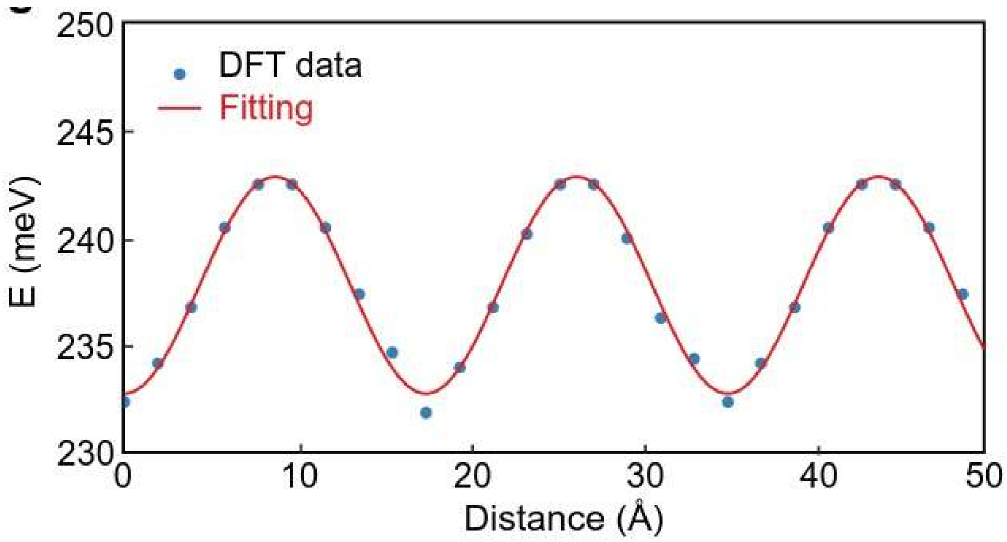
arXiv:2309.11217 (2023)

Ferroelectricity from modulated band offset

Band offset (experiment)



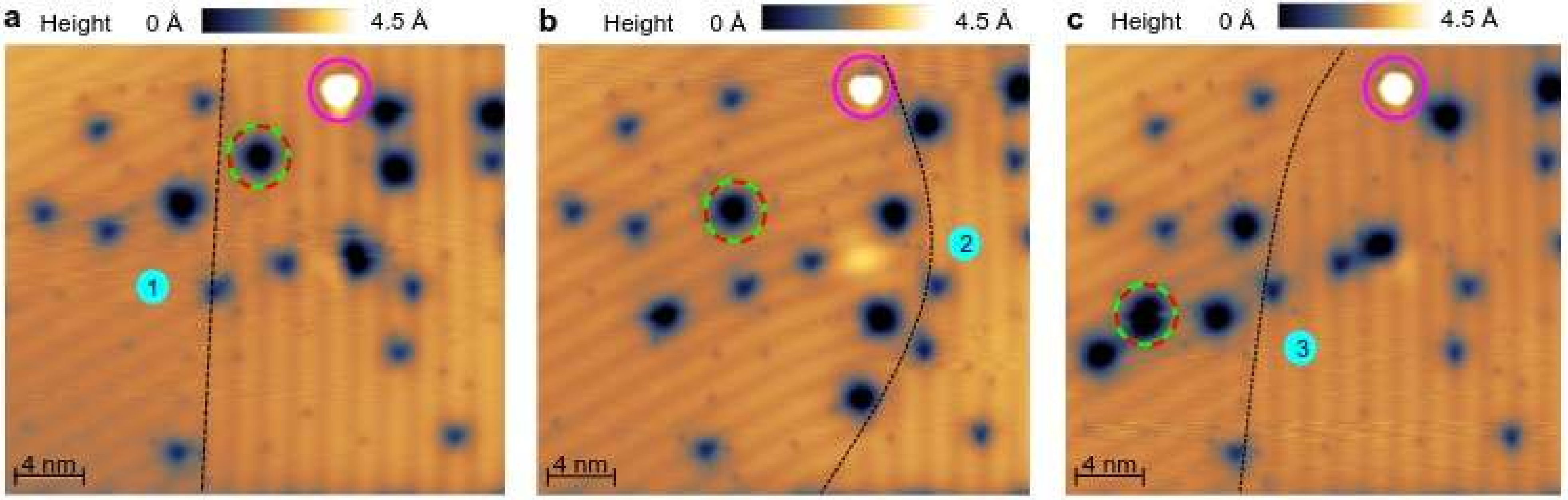
Band offset (theory)



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

arXiv:2309.11217 (2023)

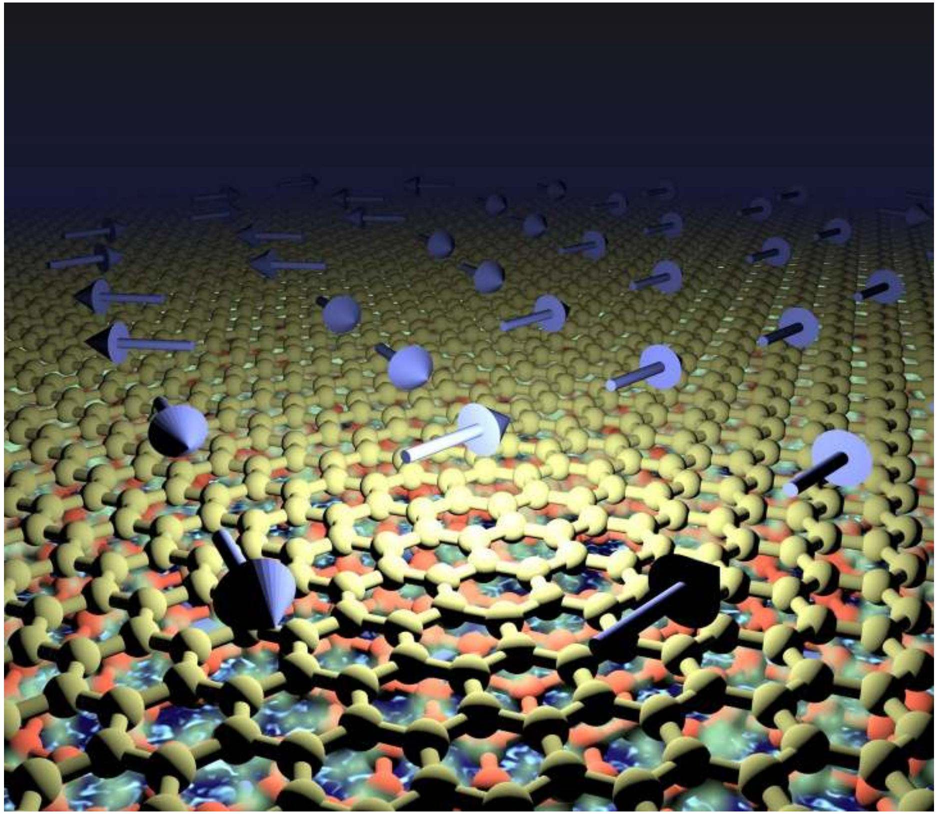
Controlling muultiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

arXiv:2309.11217 (2023)

Imprinting spin spirals in twisted bilayer via proximity



Guangze Chen

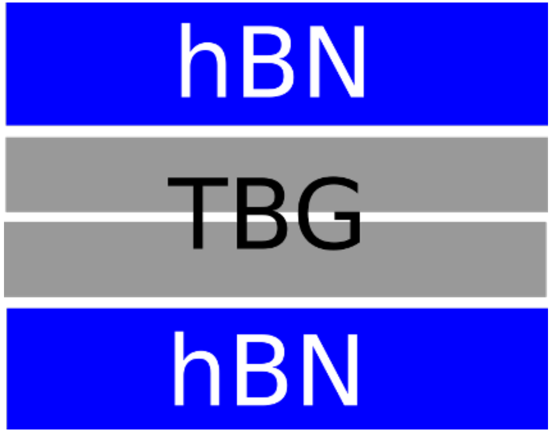
Maryam Khosravian

Aline Ramires

2D Materials 9 (2) 024002 (2022)

The role of encapsulation in twisted bilayer graphene

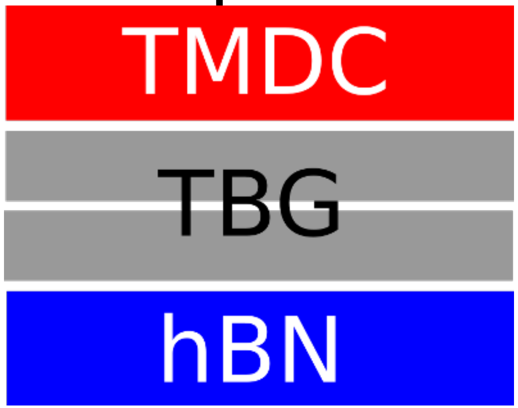
Aligned hBN encapsulation



Chern insulators

Science, 367(6480), 900-903 (2020)

Transition metal dichalcogenide encapsulation

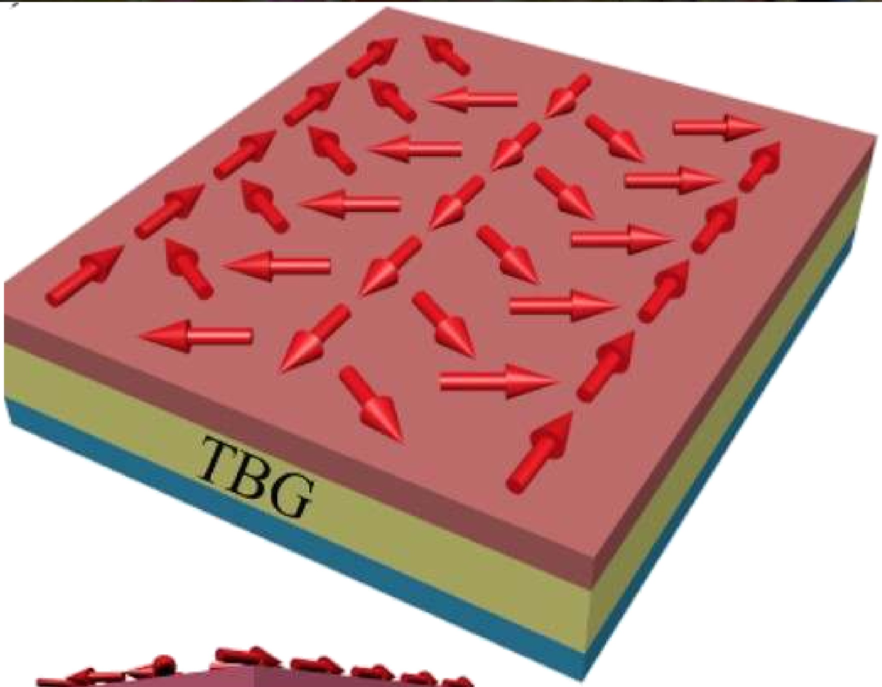


Spin-orbit coupling driven ferromagnetism

Science, 375(6579), 437-441 (2022)

Can we control the electronic structure with a multiferroic NiI_2 encapsulation?

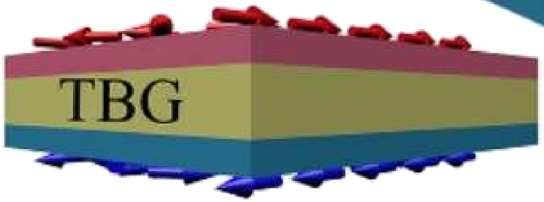
Twisted bilayer graphene encapsulated between NiI_2



$$H = H_0 + H_J$$

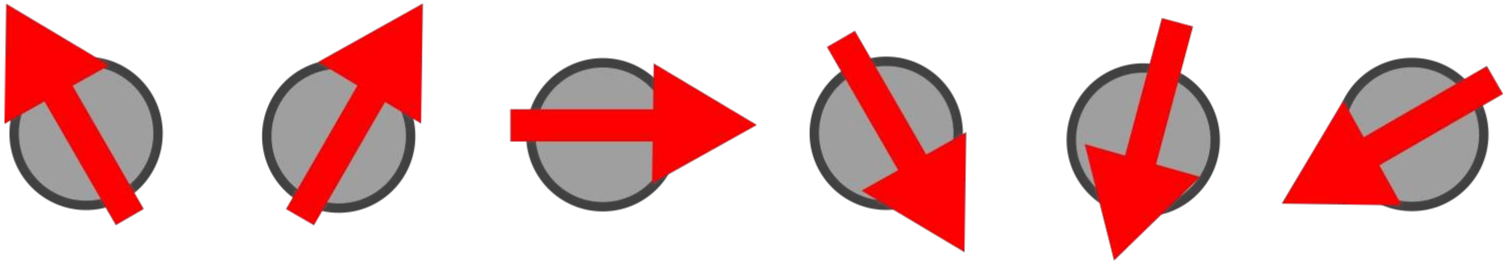
TBG Hamiltonian

Spin-spiral exchange proximity effect



NiI_2
Twisted bilayer graphene
NiI_2

The impact of a spin spiral in the electronic structure



Generalized Bloch's theorem (symmetry is translation and spin rotation)

$$\Psi(\mathbf{r}+\mathbf{R}) = e^{i\mathbf{q}\cdot\mathbf{R}\hat{S}_z} e^{i\mathbf{k}\cdot\mathbf{R}} \Psi(\mathbf{r})$$

$\mathbf{q}$ Spin-spiral wavevector

$\mathbf{k}$ Bloch wavevector

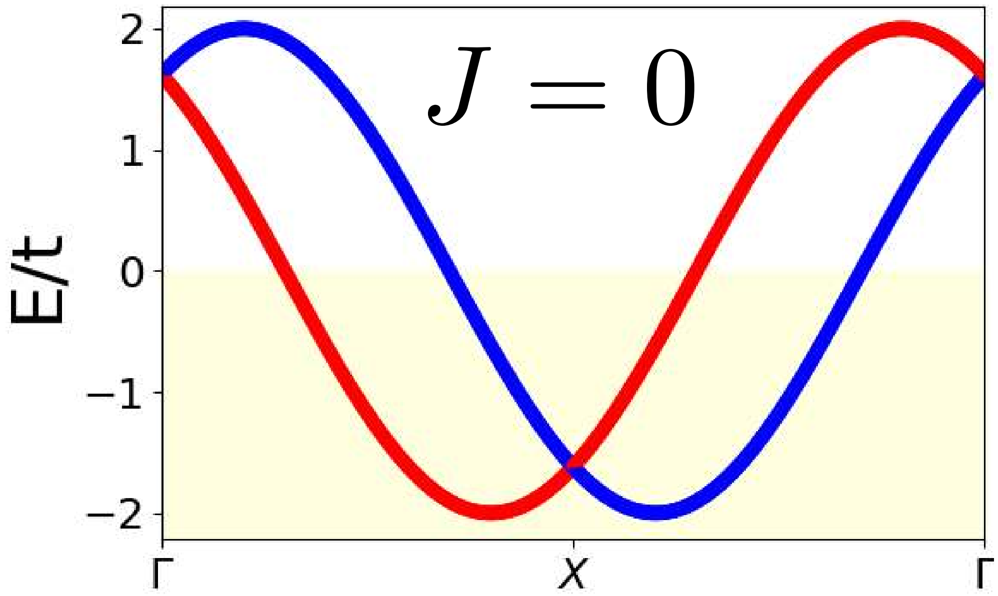
$\mathbf{q}+k$ For spin $\uparrow$

$\mathbf{q}-k$ For spin $\downarrow$

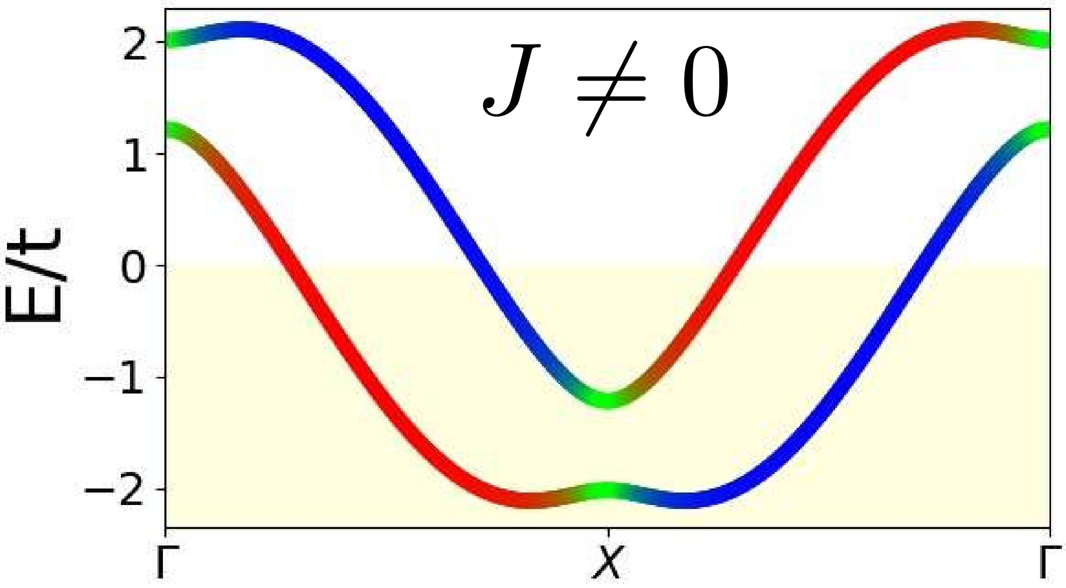
A spin-spiral acts as an artificial spin-orbit coupling (momentum-dependent spin splitting)

Spin spiral in a 1D chain

No exchange (just spiral boost)



Finite spiral exchange

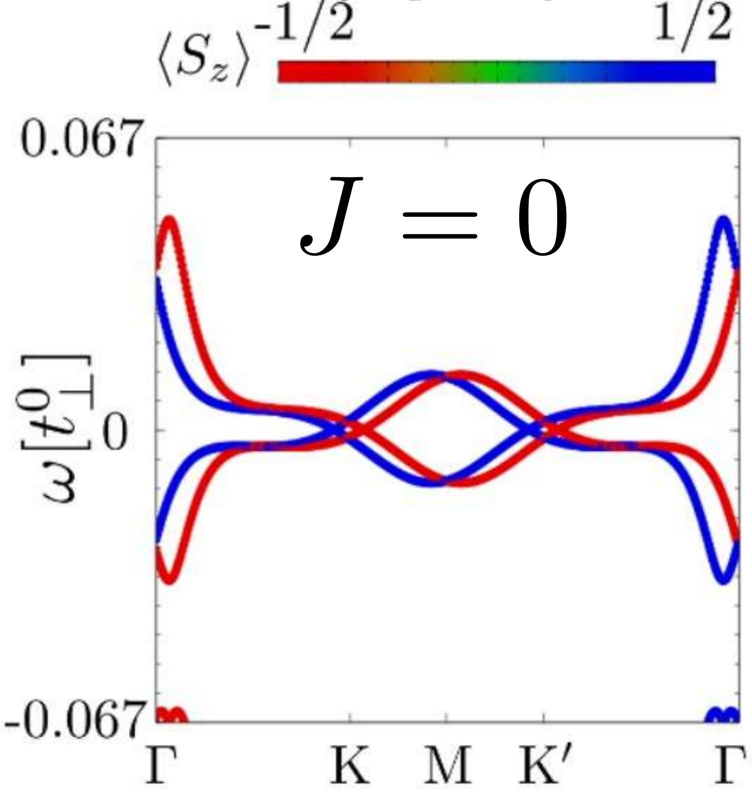


A spin spiral exchange creates spin-momentum locking and spin anticorossings

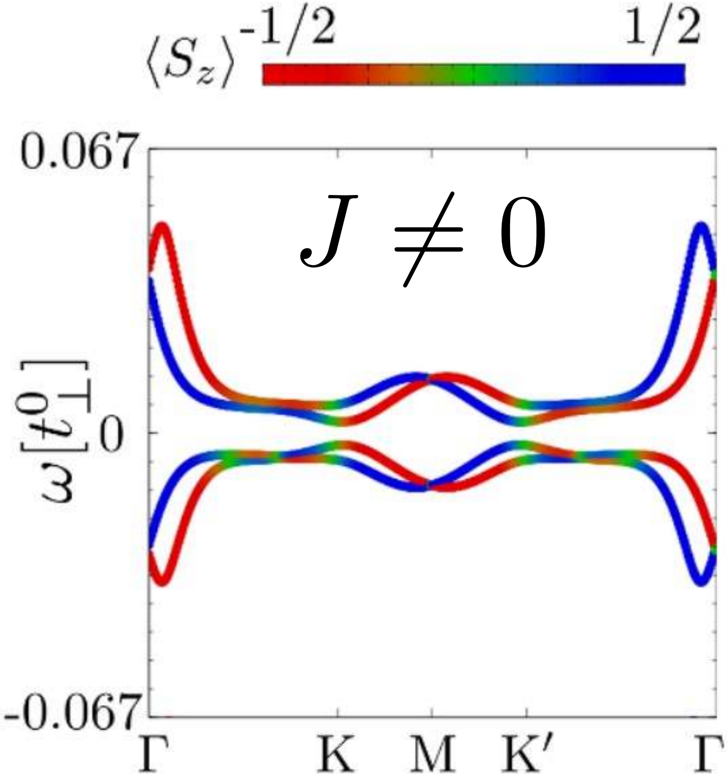
Equivalent to a synthetic spin-orbit coupling and a net exchange fiield

Spiral exchange in twisted bilayer graphene

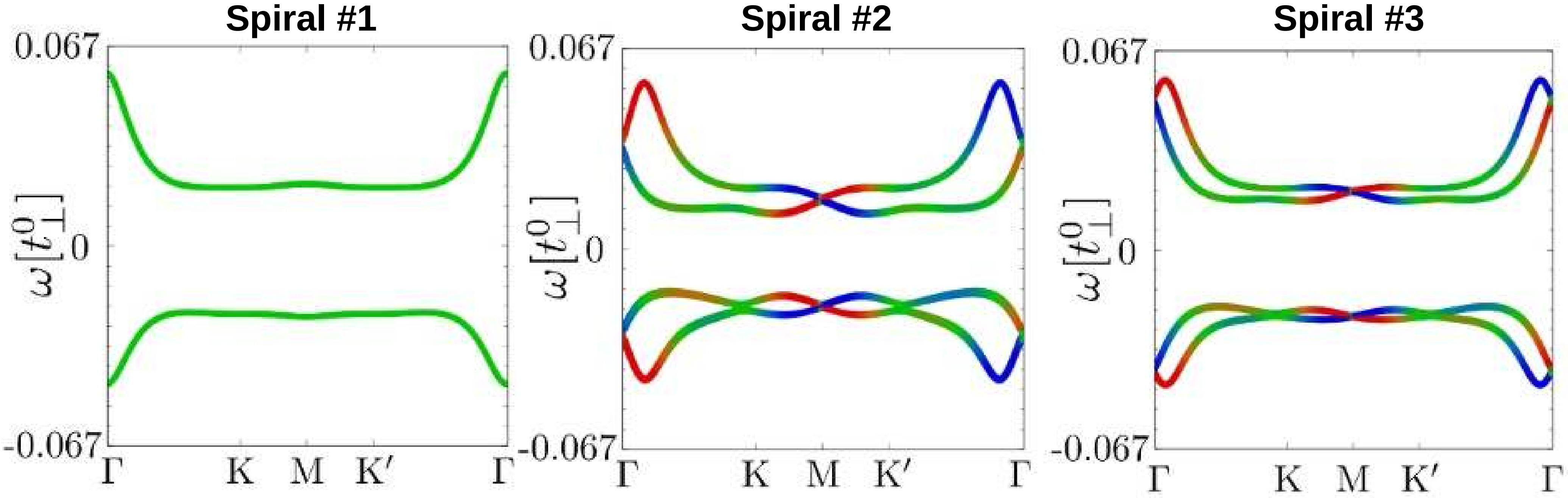
No exchange (just spiral boost)



Finite spiral exchange



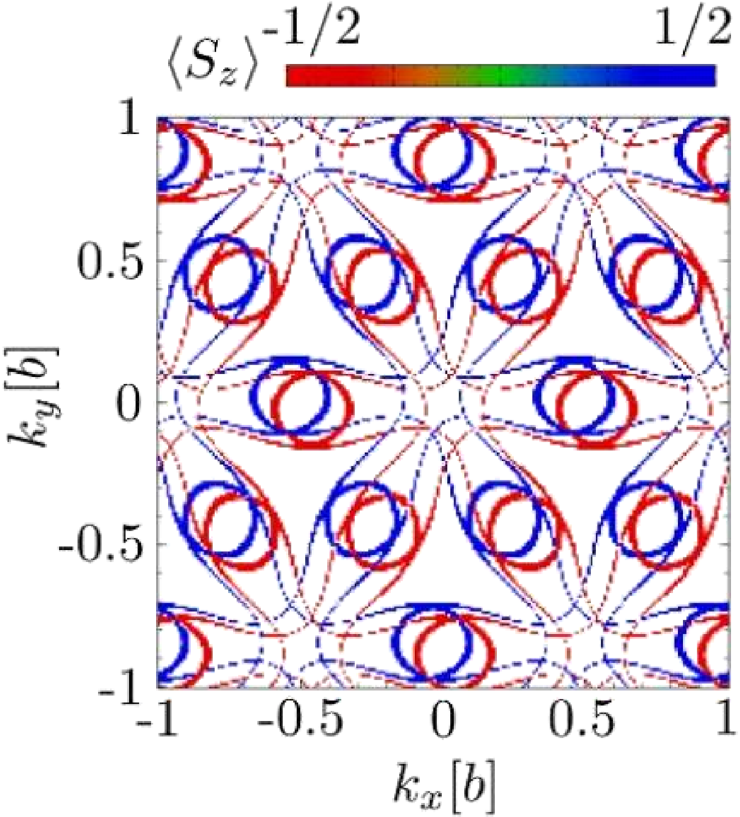
Spiral exchange in twisted bilayer graphene



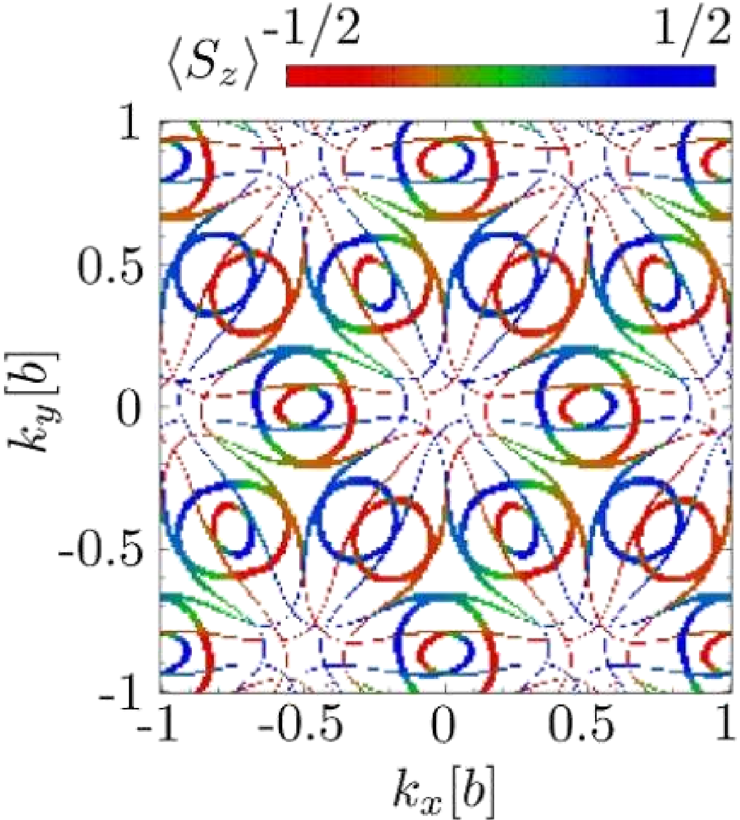
The electronic dispersion is strongly tuned depending on the exact spin spiral

Fermi surface control via spiral proximity in twisted bilayer

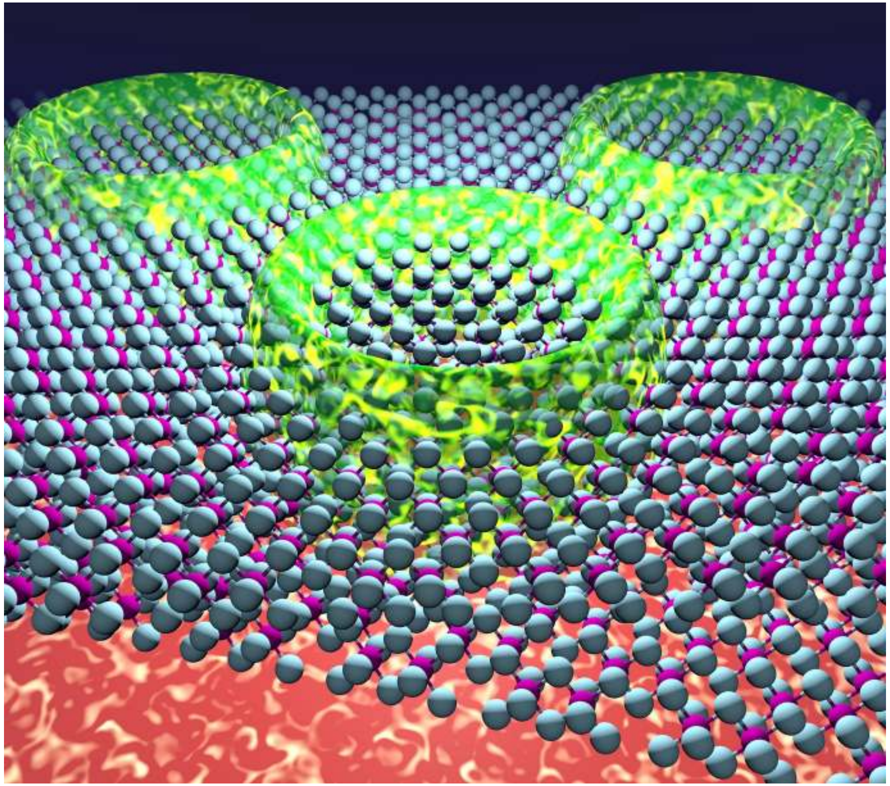
No exchange (just spiral boost)



Finite spiral exchange



Engineering artificial multiferroics with twisted van der Waals materials

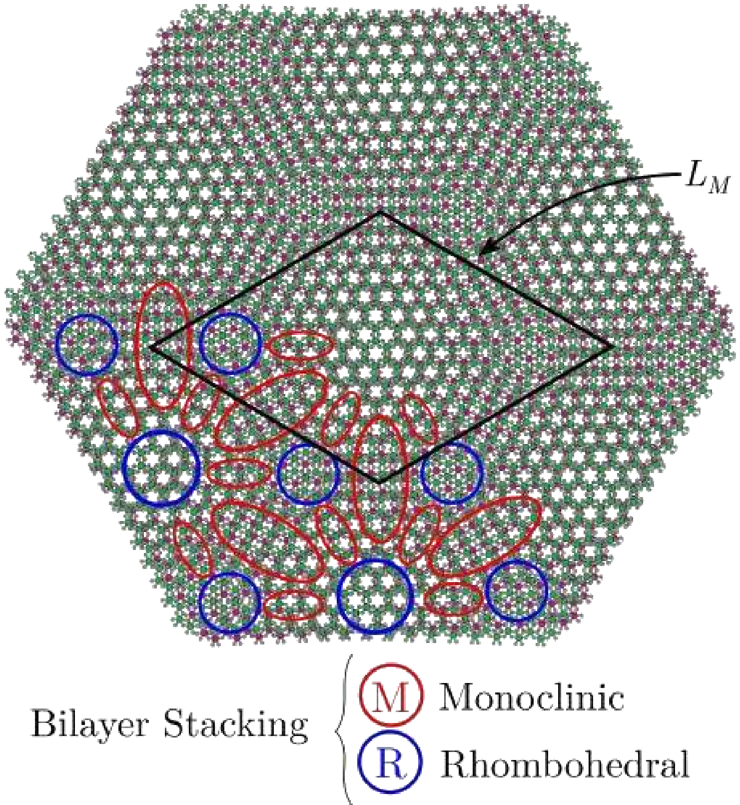


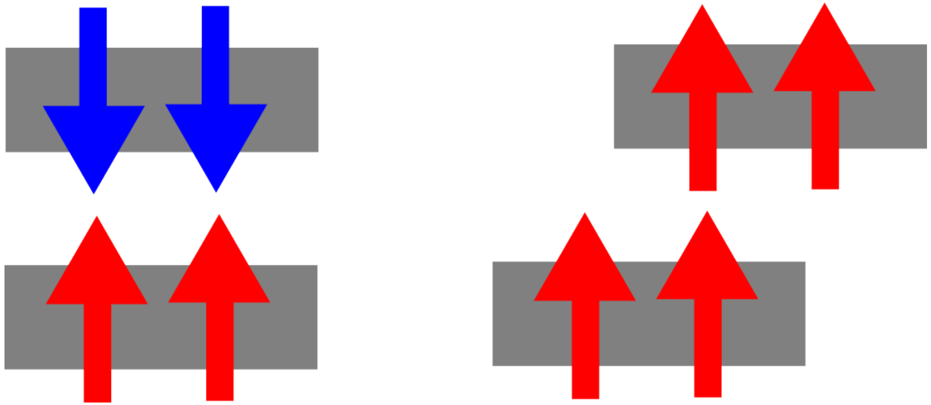
A. Fumega

2D Materials 10 025026 (2023)

Twisted 2D trihalides

$CrBr_3$

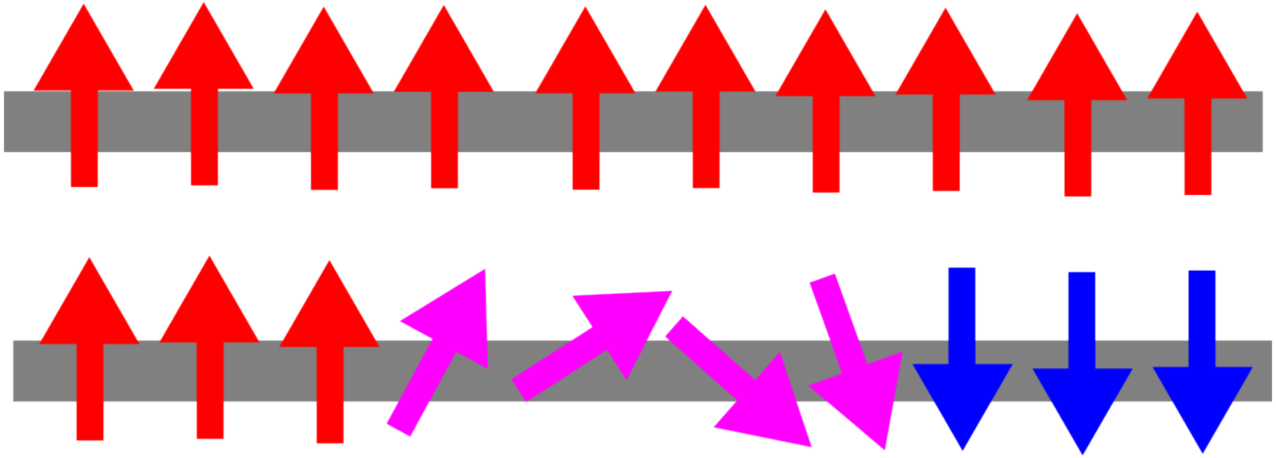




The local stacking determines the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Stacking #1

Stacking #2

Mixed FE/AF domains

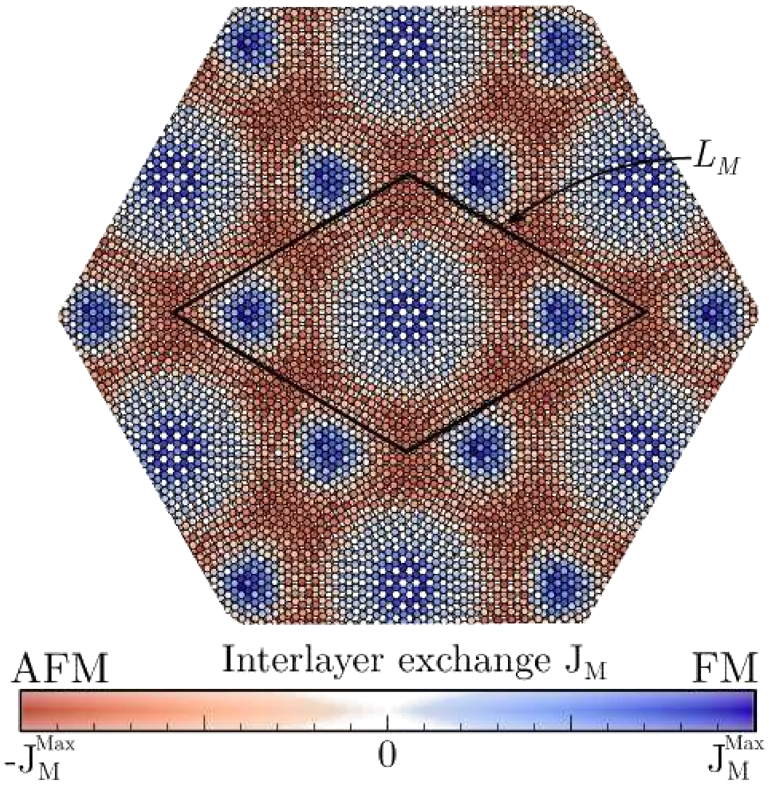
Nature Nanotechnology 17, 143–147 (2022)

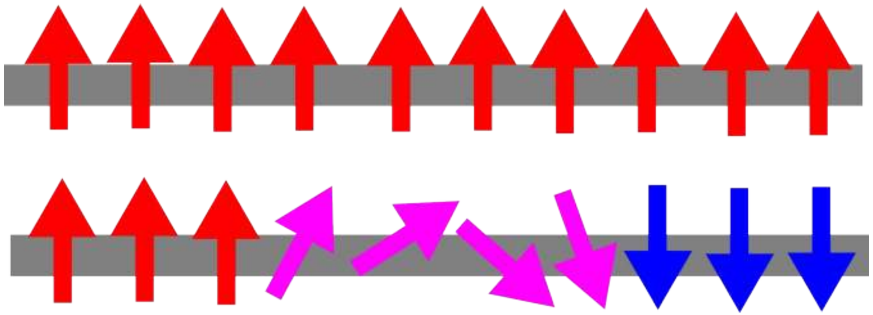
Non-collinear magnetism

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

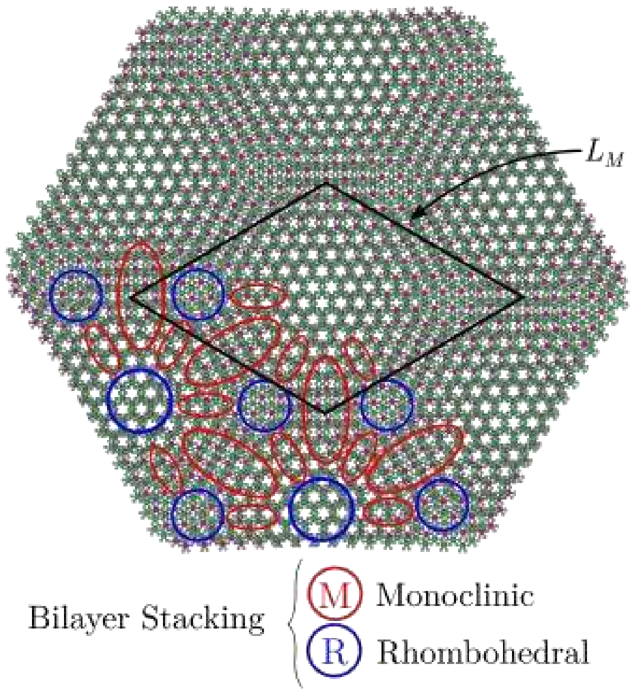
SOC driven

Wavevector of the spin spiral

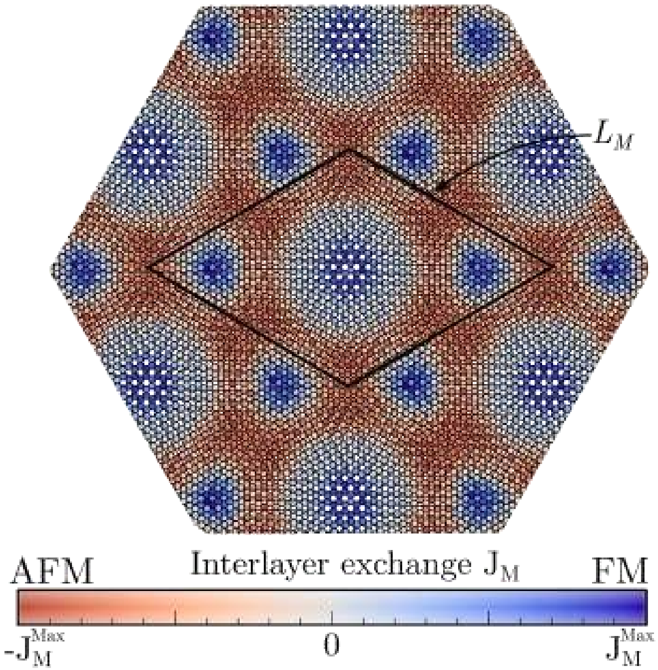
Spin rotation axis

Stacking, magnetic order and electric polarization

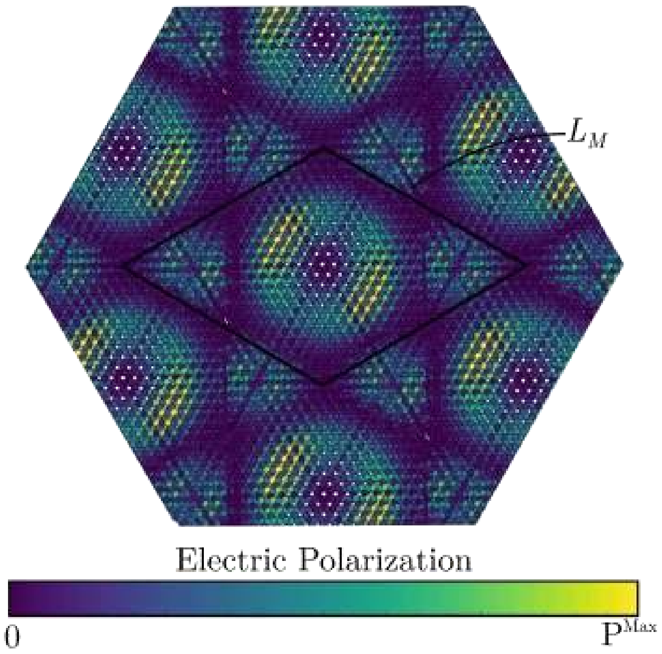
Structure



Magnetic order



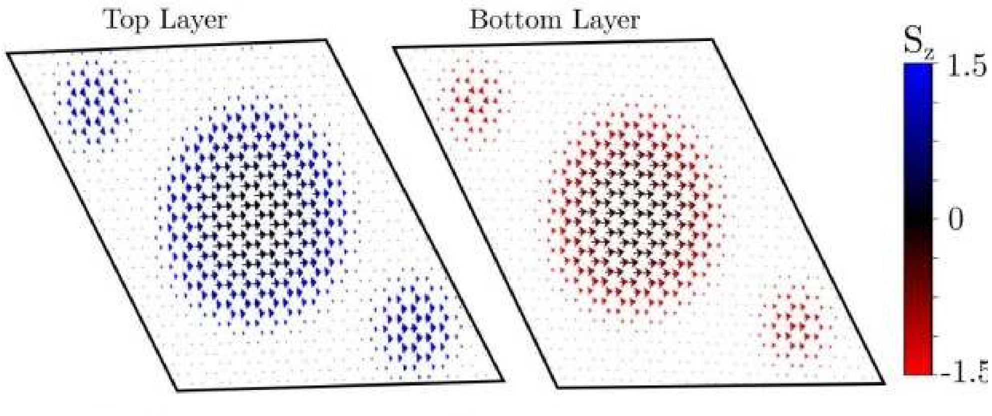
Ferroelectric order



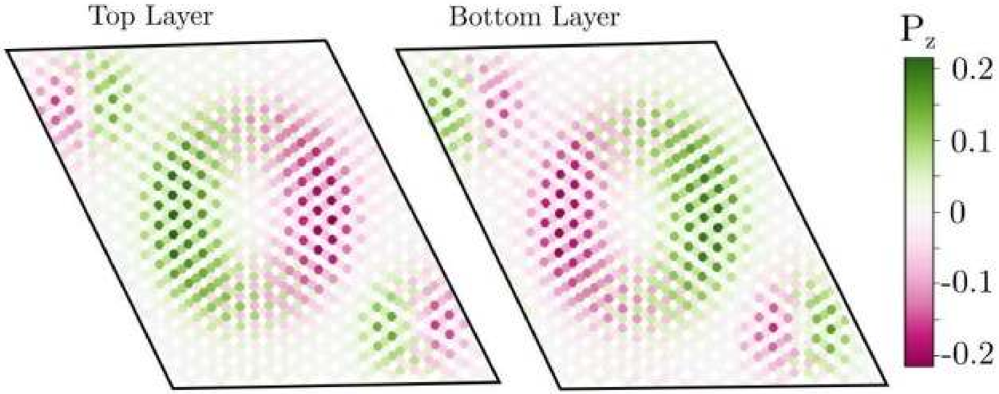
Stacking, magntic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

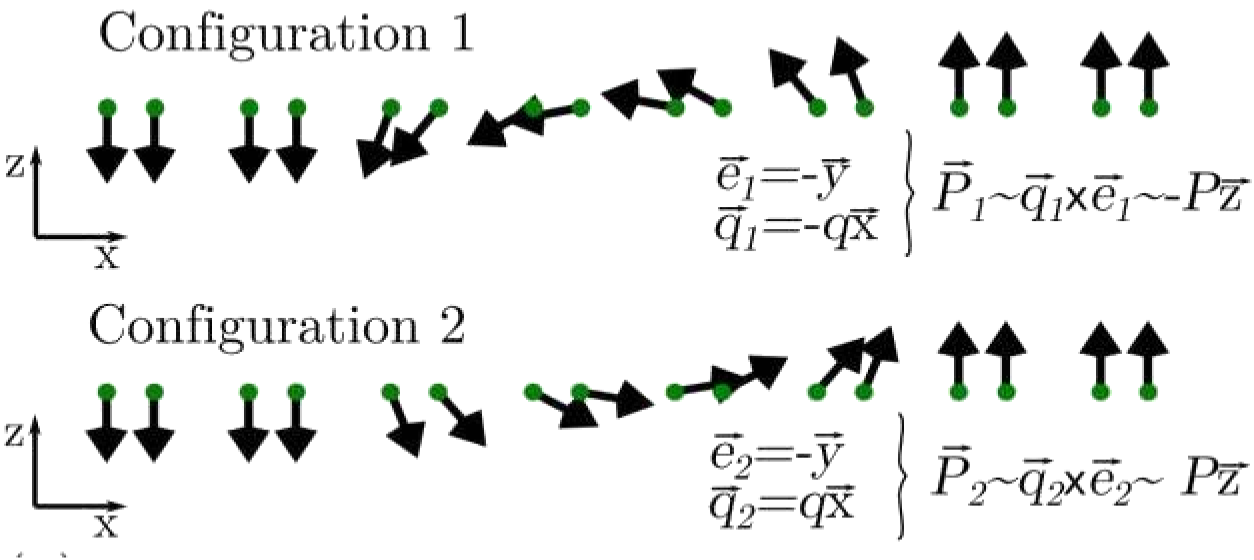


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



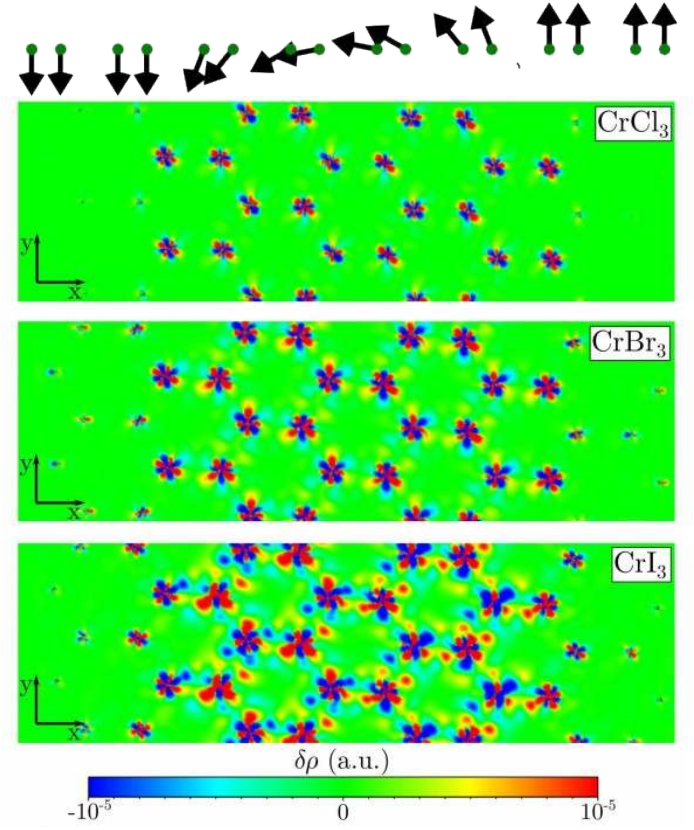
Ferroelectric electronic density

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

Ferroelectric force

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

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

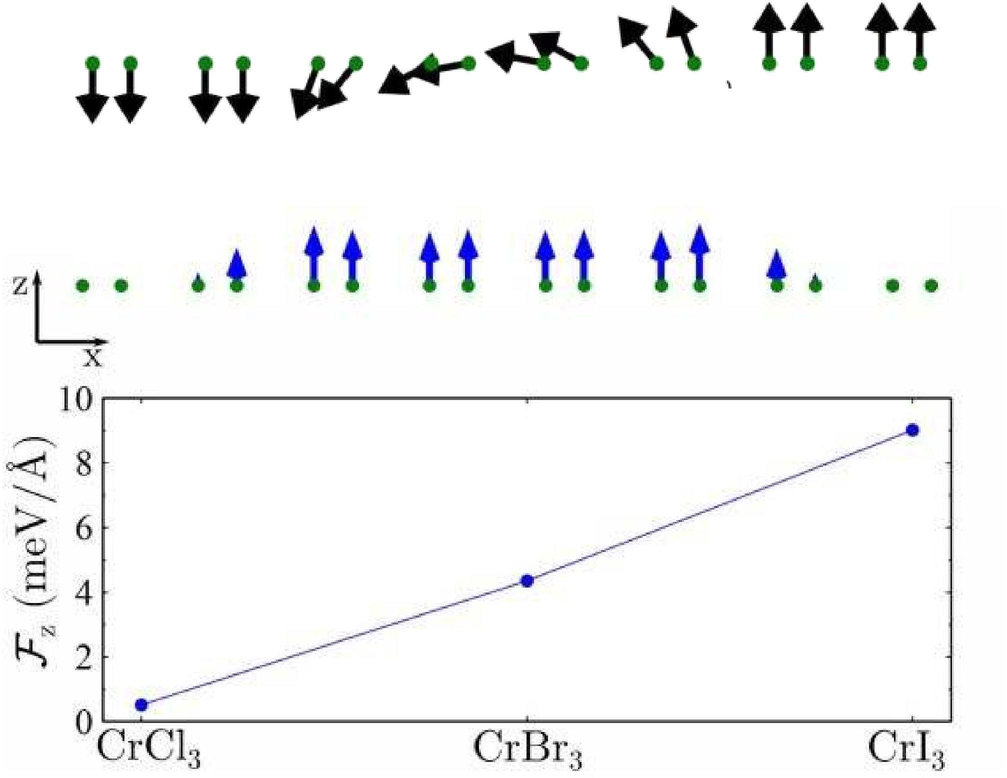
The ferroelectric force

Magnetic configuration

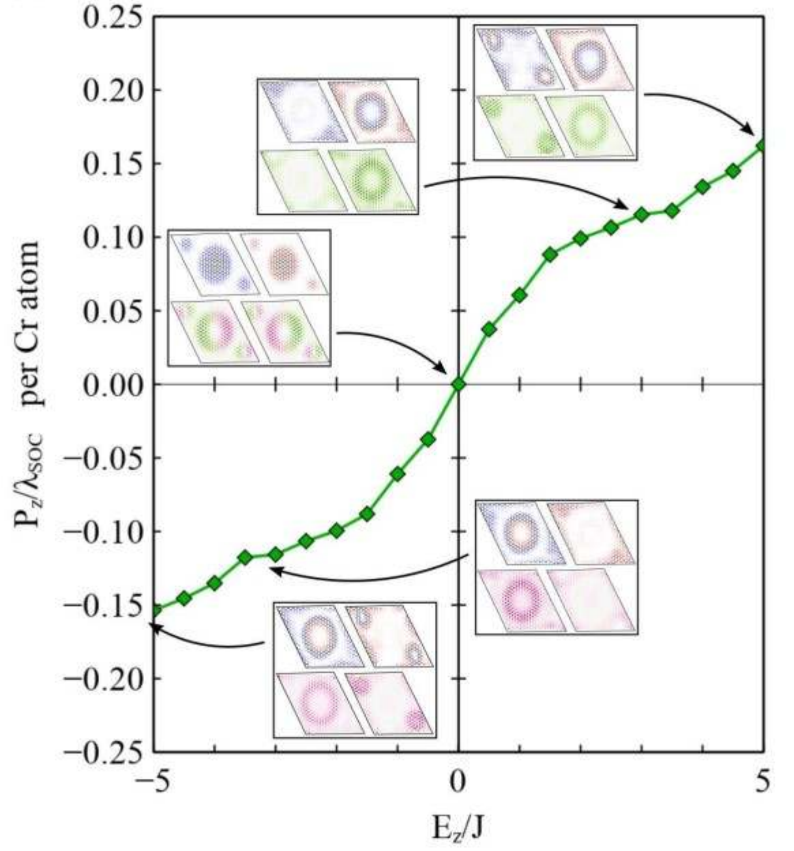
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



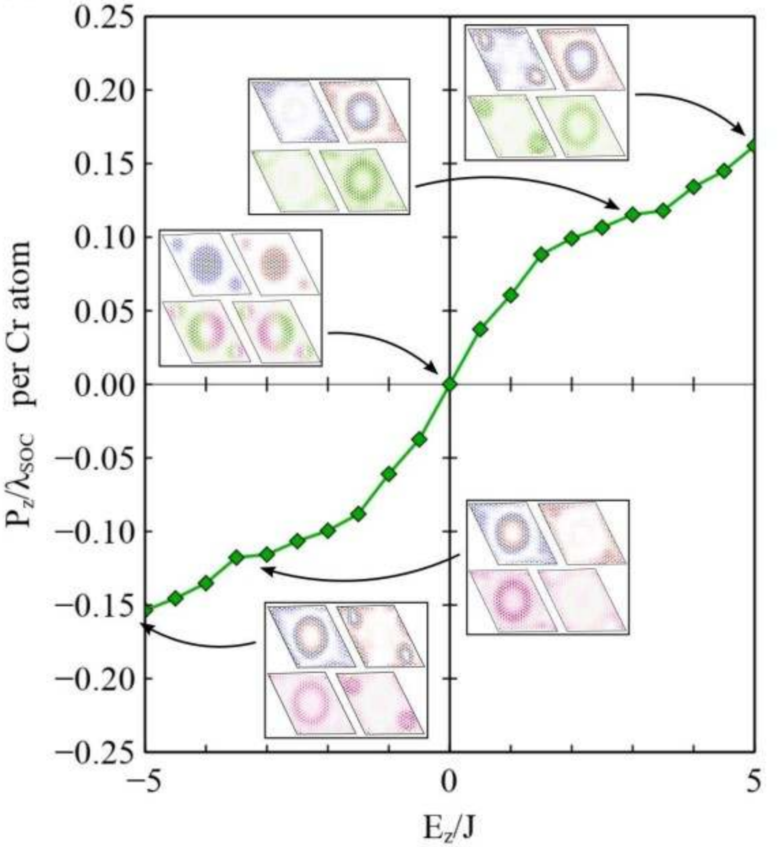
Electrically controllable skyrmions in twisted magnets



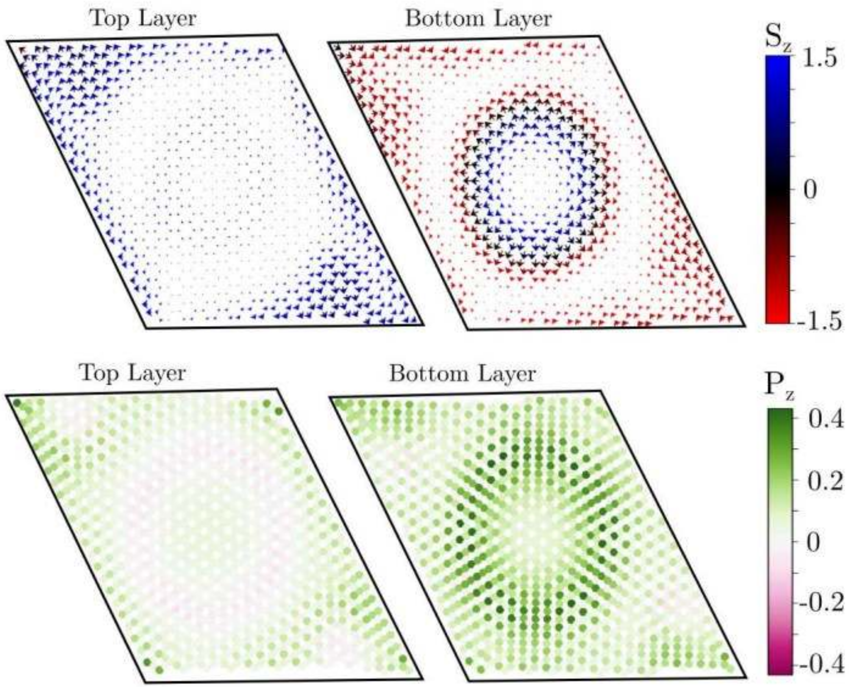
The electric polarization directly couples to an out-of plane electric field

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

Electrically controllable skyrmions in twisted magnets

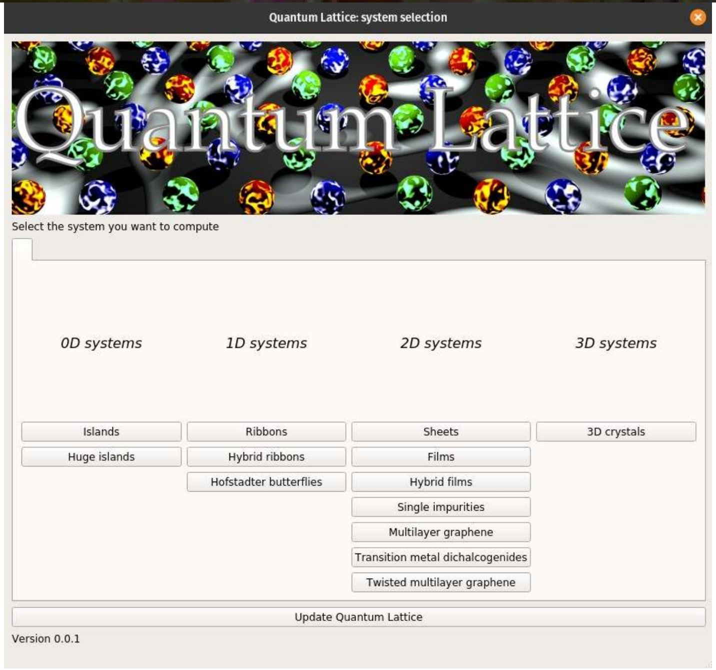


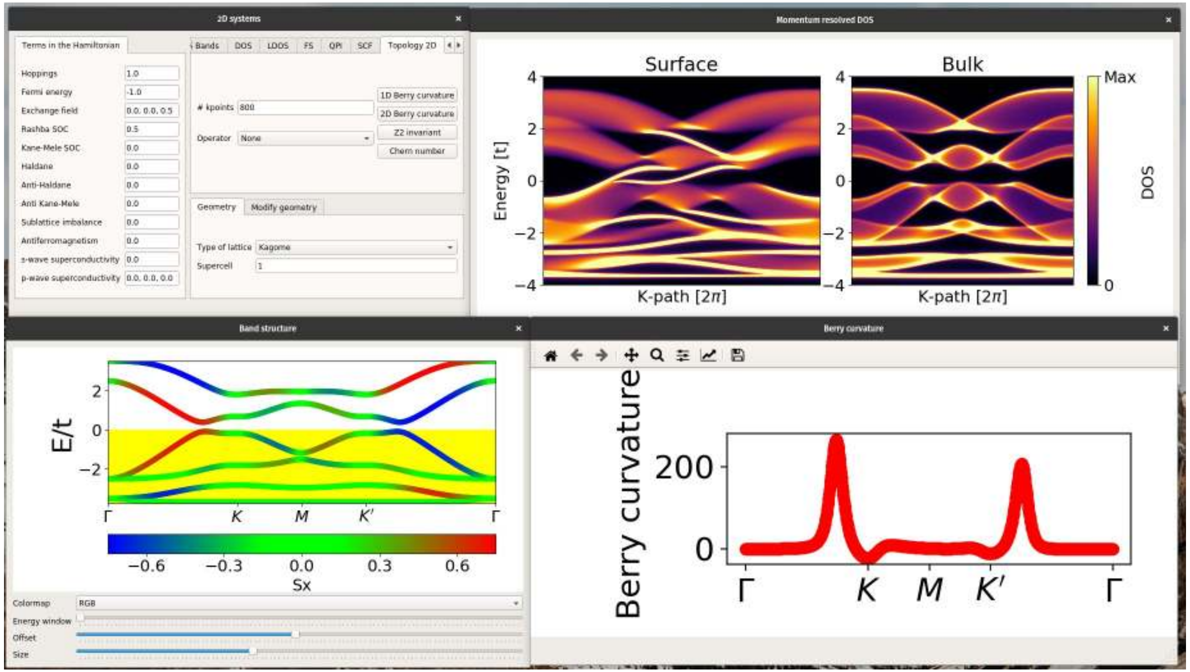
Skyrmions at finite interlayer bias



Open-source software for van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

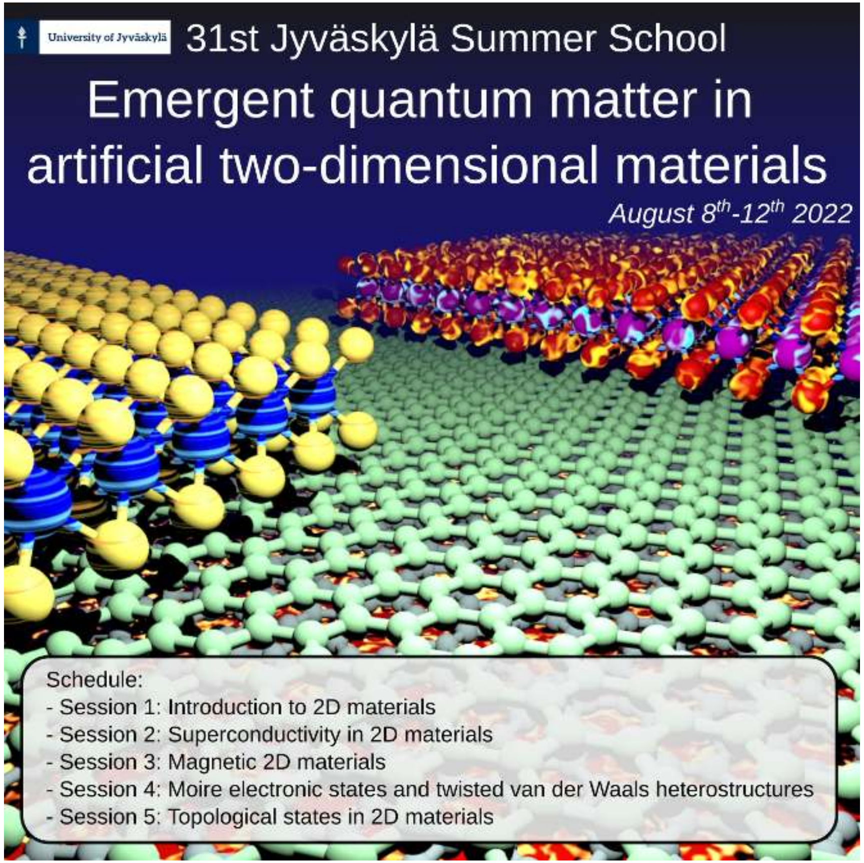




Quantum Lattice: open source interactive interface for tight binding modeling

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

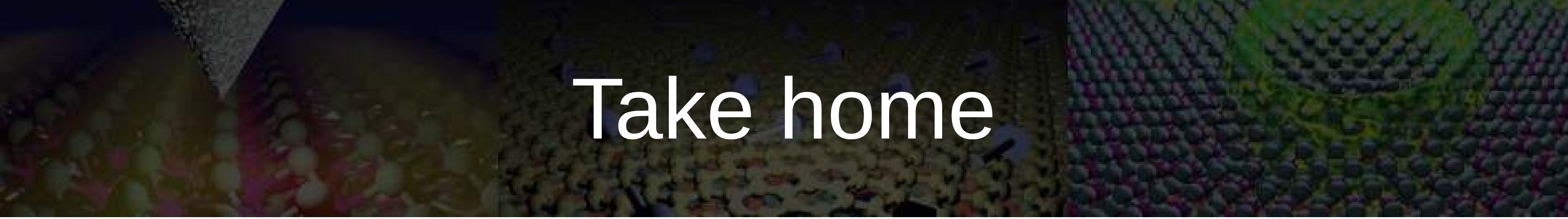
Quantum matter in van der Waals materials school



Recordings available at

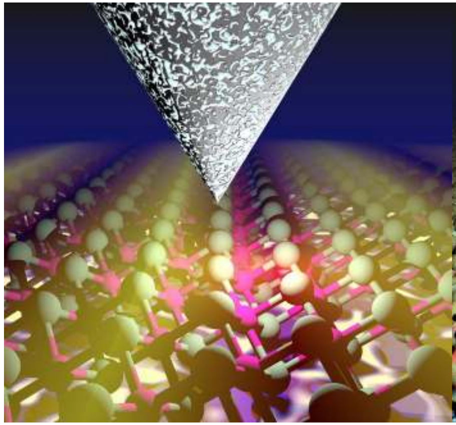
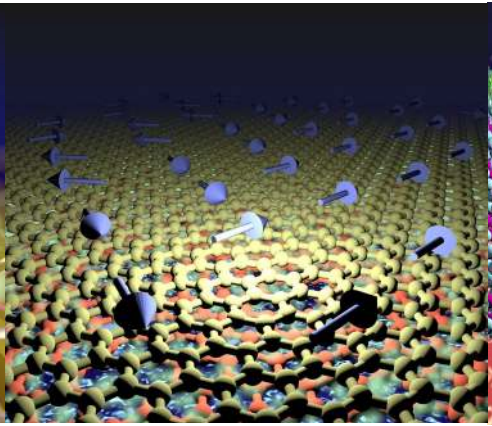
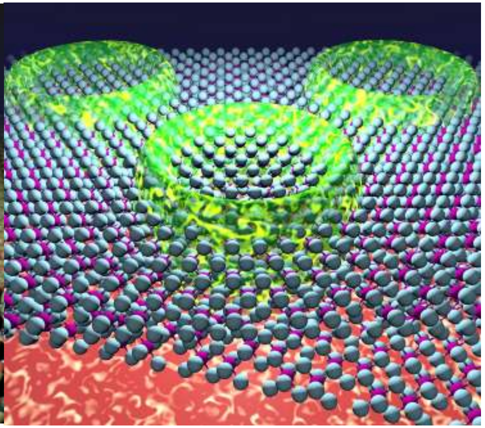
https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ

- 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 multiferroics offer a new playground to explore unconventional phenomena in moire heterostructures



2D Materials, 9 (2), 024002 (2022)
2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)
arXiv:2309.11217 (2023)

Thank you

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SUOMEN AKATEMIA