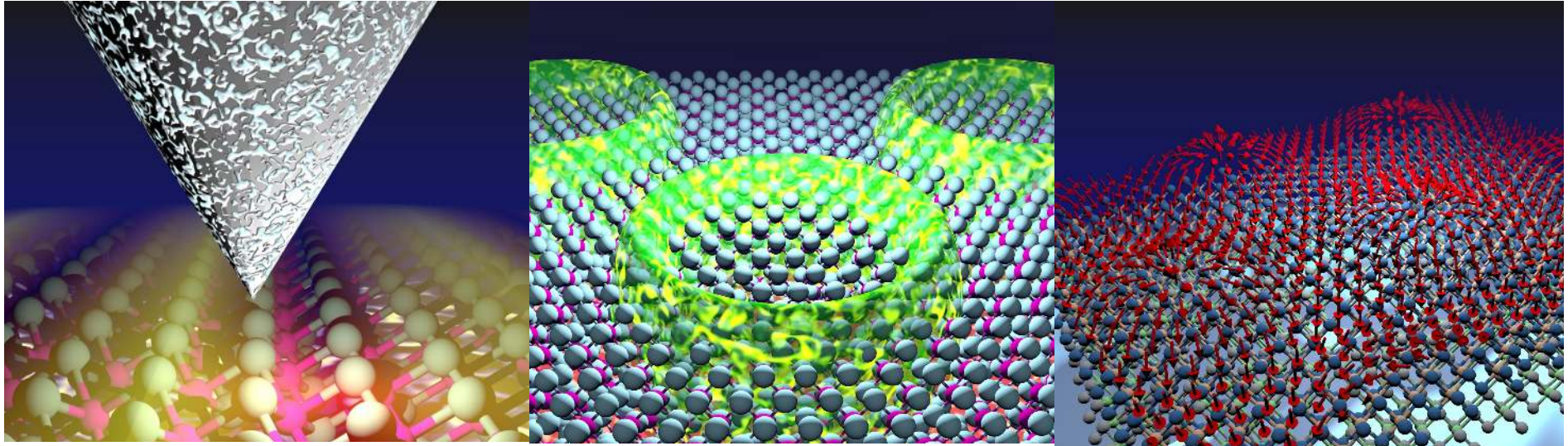


Artificial van der Waals multiferroics with twisted two-dimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



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

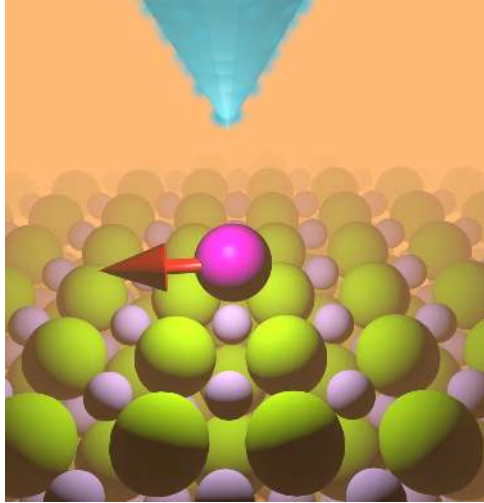
2D Materials 10 (2), 025026 (2023)

Nano Letters 24 (49), 15767–15773 (2024)

University of Hamburg, June 16th 2025

Building quantum matter with artificial lattices

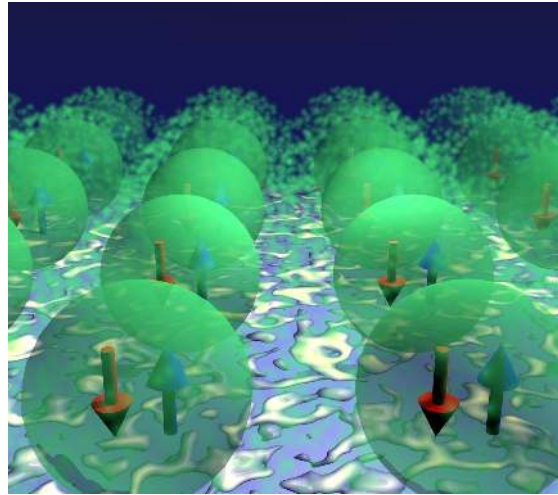
Atomic lattices



$a = 0.5 \text{ nm}$

Nature Physics 7 (9), 713-718 (2011)
Nature Physics 13 (7) 668 (2017)
Nature Nano 17 (4), 384-389 (2022)
Science 335 (6065), 196-199 (2012)

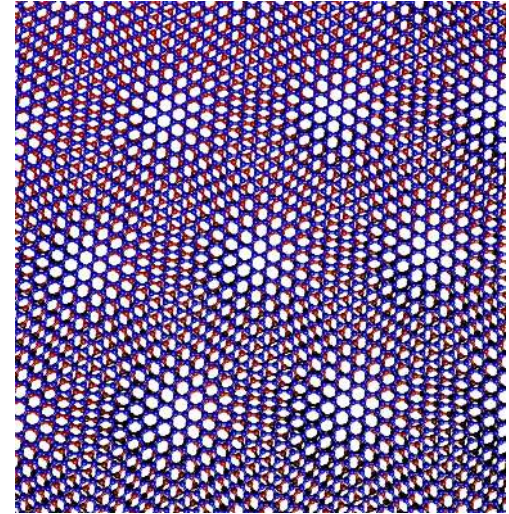
Molecular lattices



$a = 3 \text{ nm}$

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



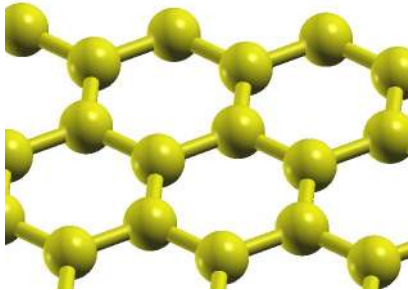
$a = 20 \text{ nm}$

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

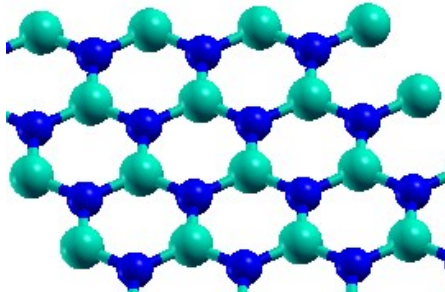
Semimetal

Graphene



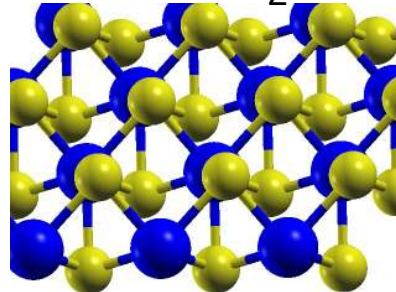
Insulator

BN



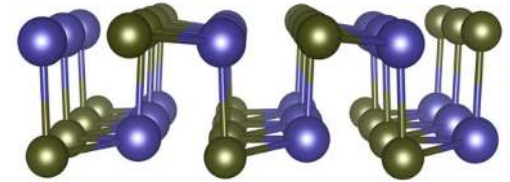
Superconductor

NbSe₂



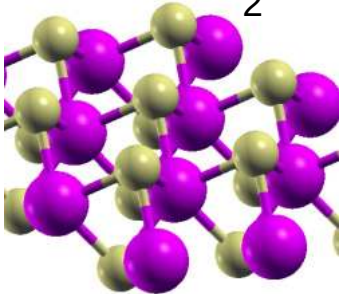
Ferroelectric

SnTe



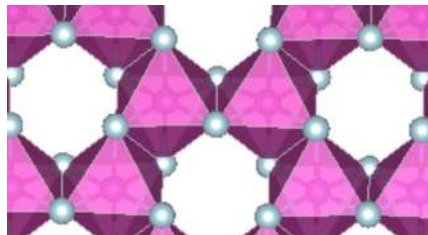
Semiconductor

WSe₂



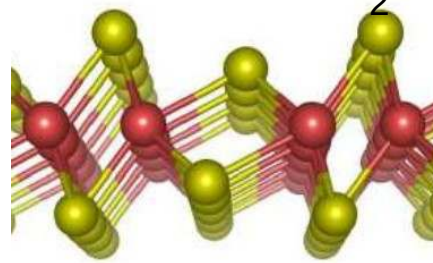
Ferromagnet

CrI₃



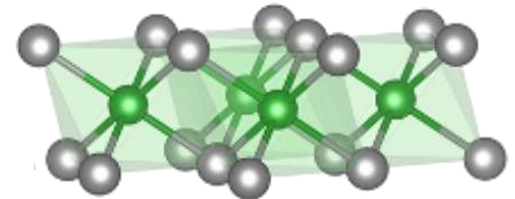
**Quantum spin
Hall insulator**

1T'-WTe₂



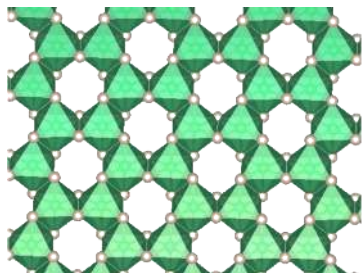
Multiferroic

NiI₂



Van der Waals magnetic materials

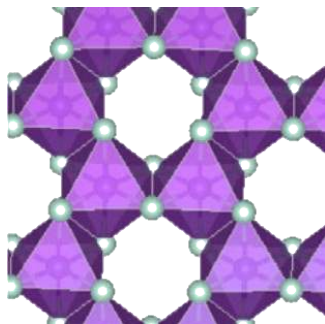
**Ferromagnet,
antiferromagnets**



CrI_3 , CrCl_3 , CrBr_3

Nature 546, 270–273 (2017)

**(proximal)
Quantum
spin-liquids**

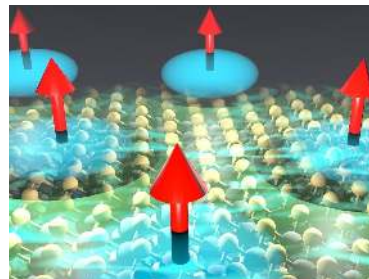


RuCl_3 , 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Break time-reversal

**Heavy-fermion
Kondo insulators**

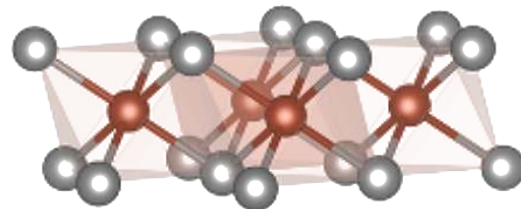


$1\text{T-TaS}_2/1\text{H-TaS}_2$

Nature 599, 582–586 (2021)

Do not break time-reversal

Multiferroics



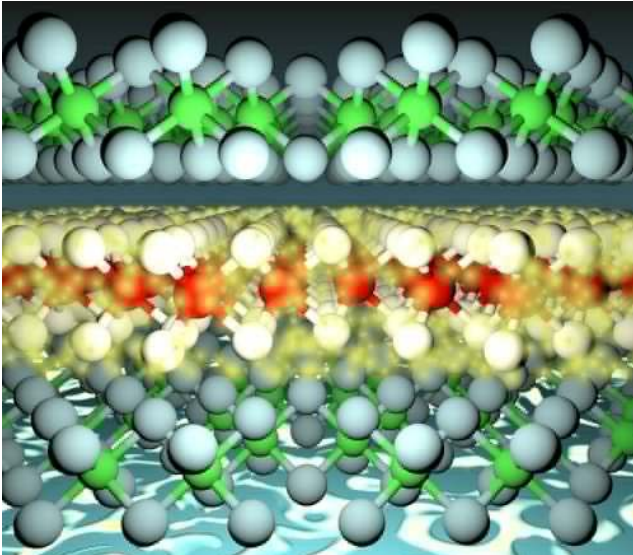
NiI_2

Nature 602, 601–605 (2022)

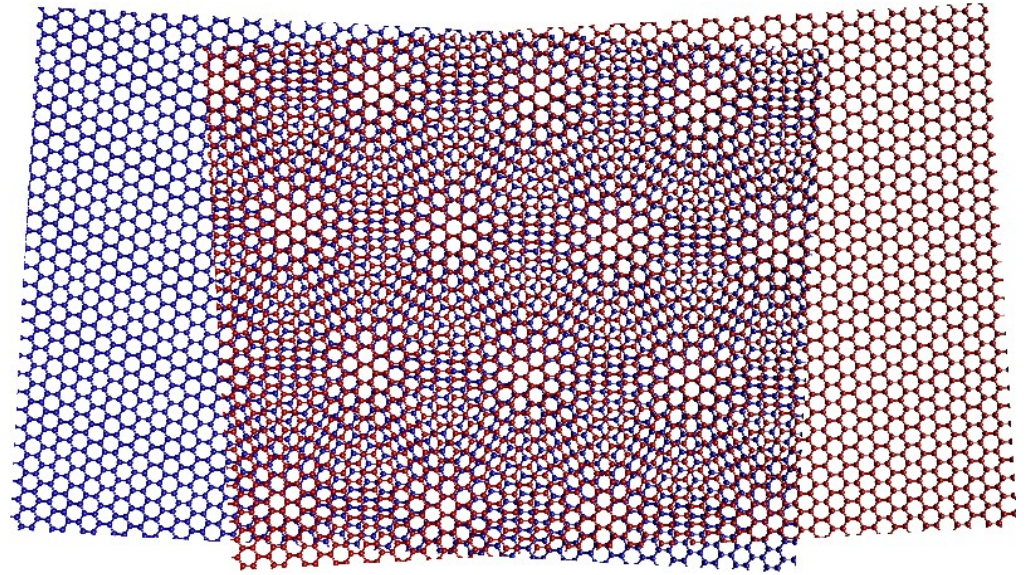
Break time-reversal
and inversion symmetry

The flexibility of two-dimensional materials

They can be stacked



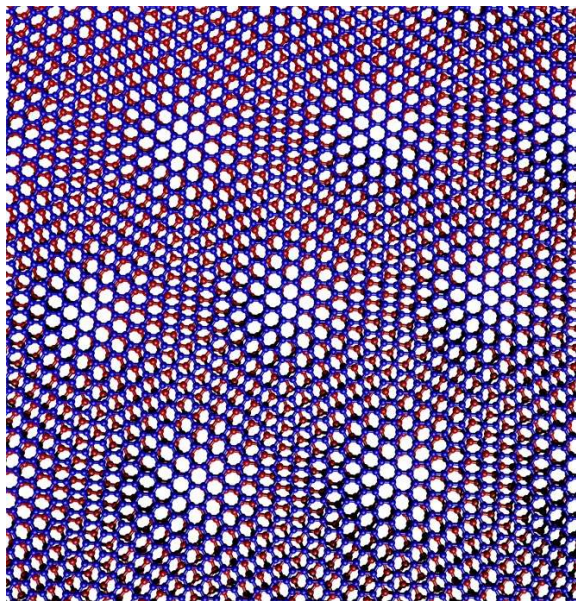
They can be rotated



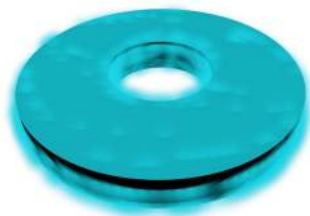
These are unique features of two-dimensional materials

The phases of twisted bilayer graphene

Twisted bilayer graphene

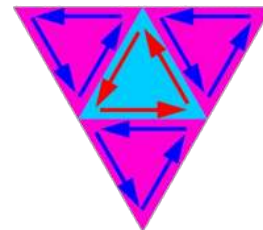


Superconductivity



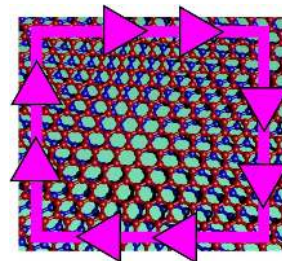
Nature 556, 43–50 (2018)

Topological networks



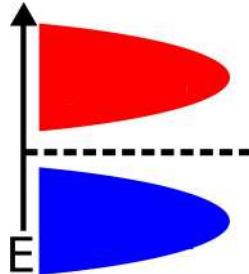
Nano Lett. 18, 11, 6725–6730 (2018)

Chern insulators



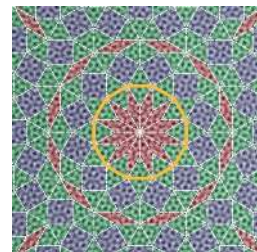
Science 365, 605–608 (2019)

Correlated insulators



Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782–786 (2018)

Proximal fractional Chern insulators



Nature 600, 439–443 (2021)

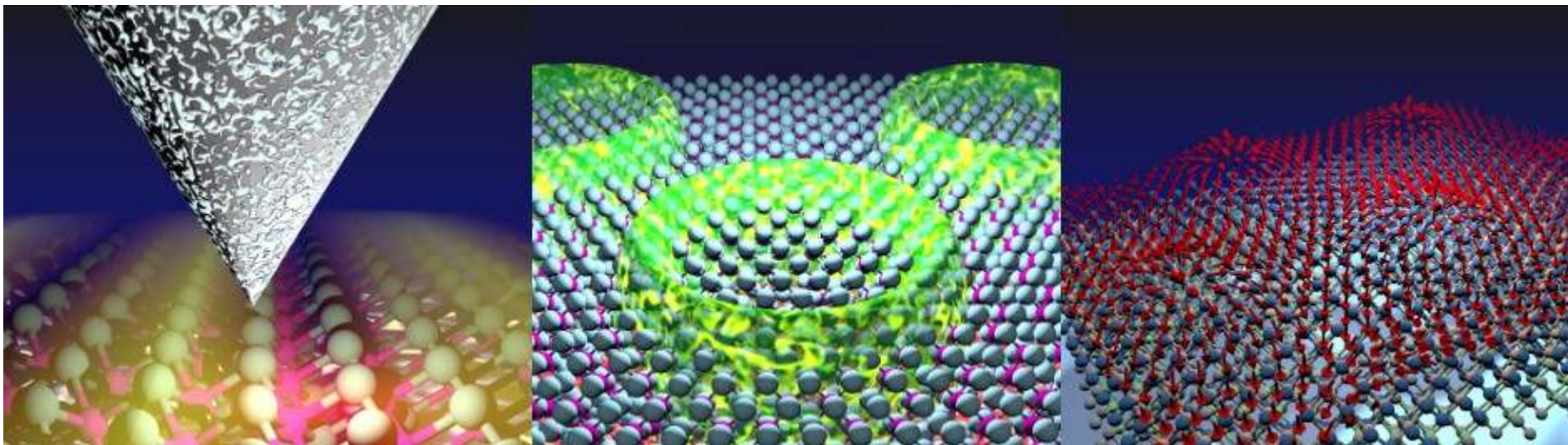
Which new phenomena could we get by twisting magnetic materials?

Today's plan

**Origin and imaging of
multiferroicity in NiI_2**

**Artificial multiferroicity
by twisting CrBr_3**

**Topological multiferroicity
by twisting NiI_2**



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

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

Nano Letters 24 (49), 15767–15773 (2024)

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 and NiI_2 bilayers

(theory)

A. Fumega

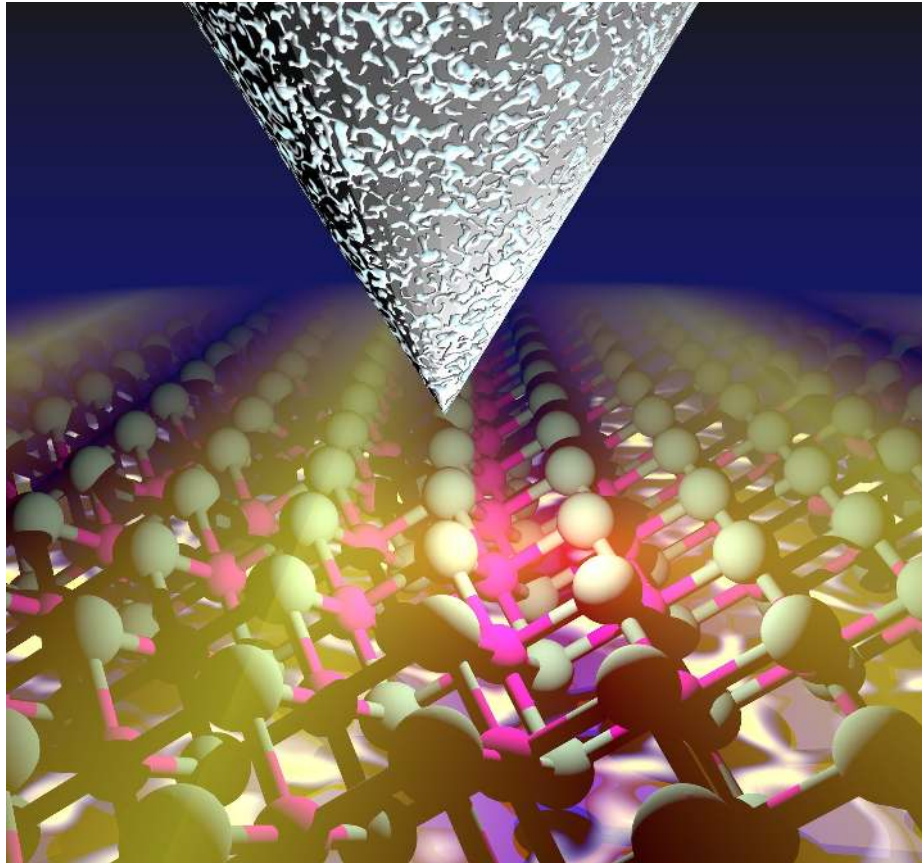


T. Antao



2D Materials 10 (2), 025026 (2023)
Nano Letters 24 (49), 15767–15773 (2024)

Origin and visualization of multiferroicity in NiI_2



V. Vaňo



M. Amini



A. Fumega



H. Gonzalez
Herrero



S. Kezilebieke



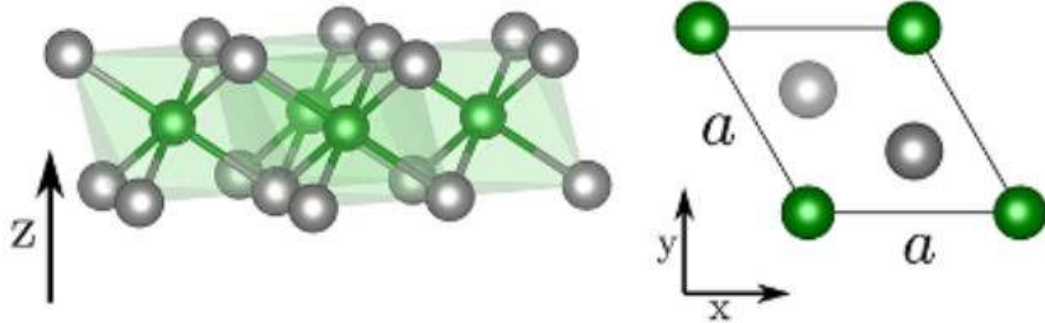
P. Liljeroth



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

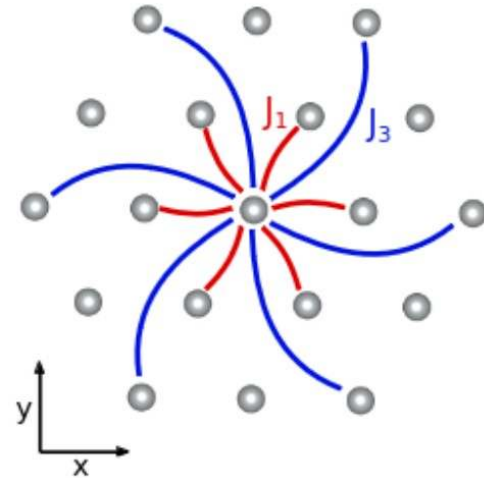
Monolayer NiI_2

Structure

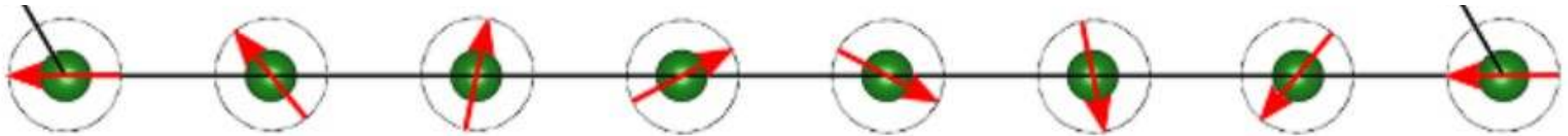


Nature 602, 601–605 (2022)

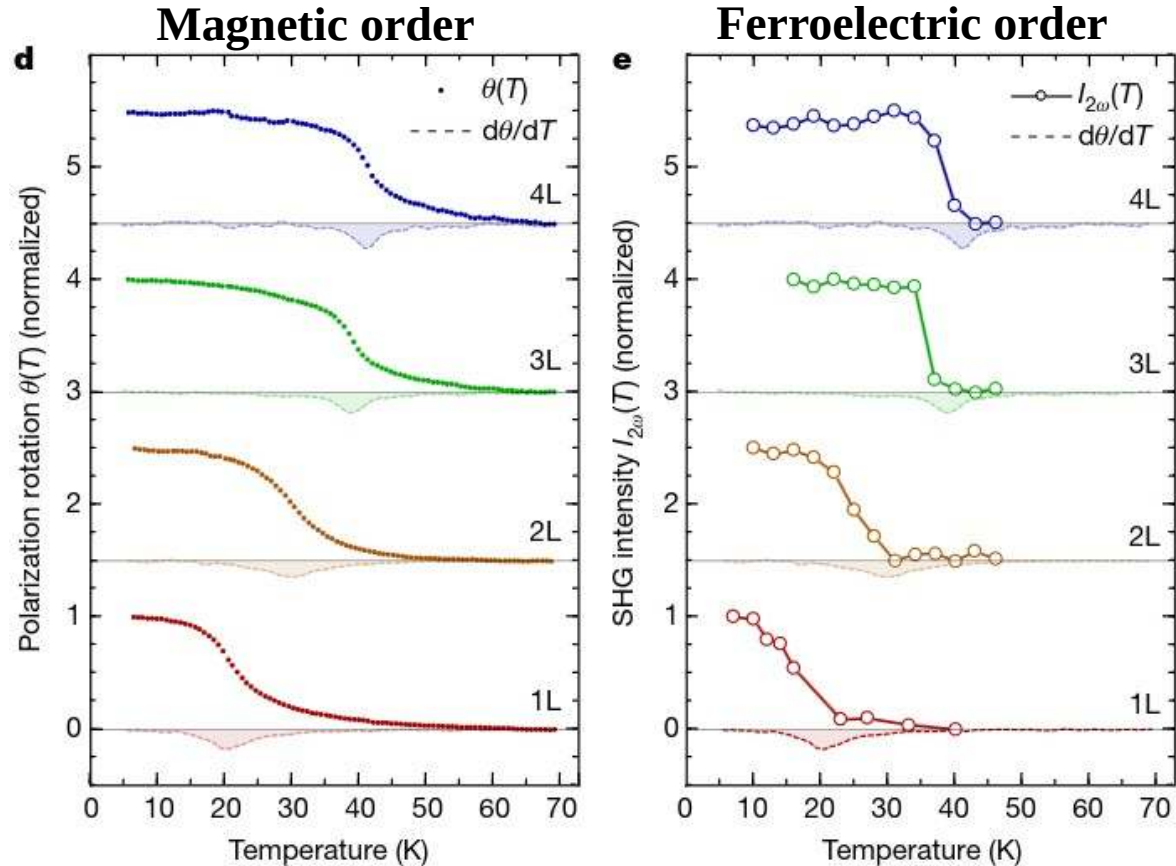
Frustrated magnetic
Heisenberg model



Magnetic structure



Multiferroicity in monolayer NiI_2



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

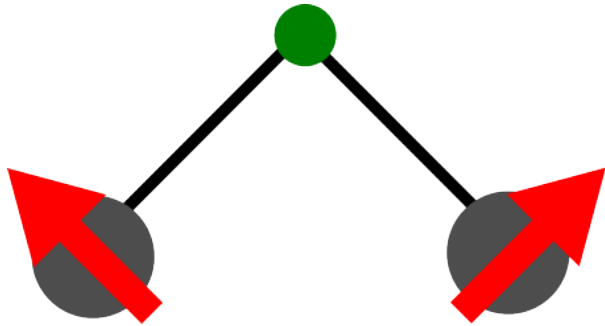
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

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

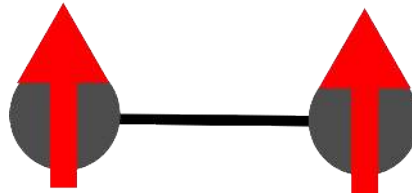
Antisymmetric exchange

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



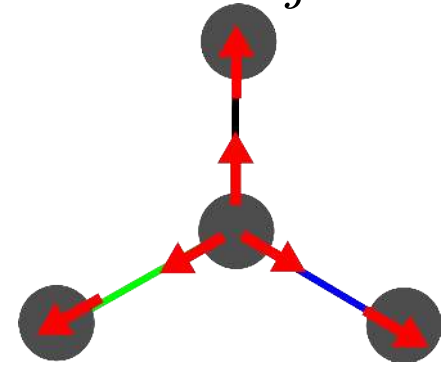
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

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



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

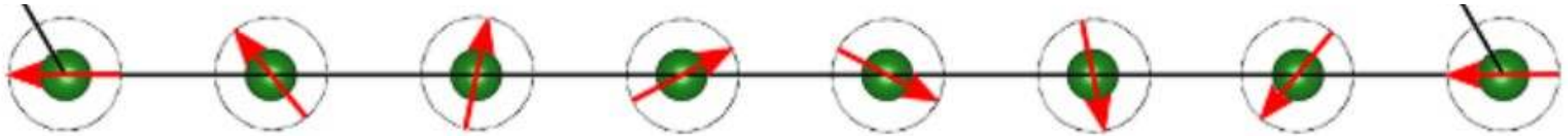
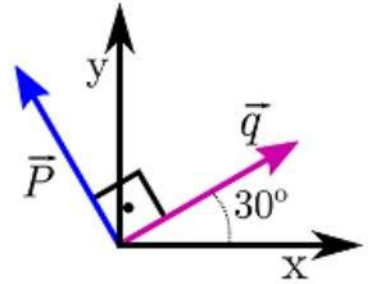
In NiI_2 , the ferroelectricity is created by inverse antisymmetric exchange

The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

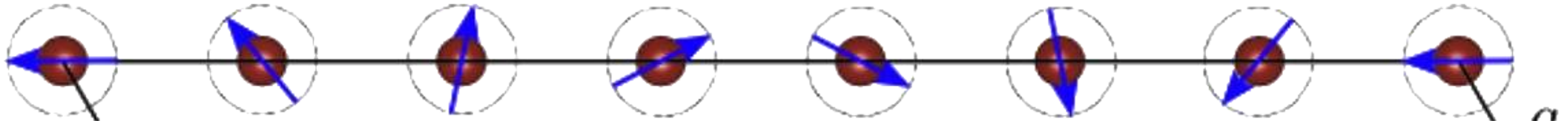
$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$$

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



The emergence of non-collinear order drives ferroelectricity

The origin of ferroelectricity in NiI_2



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

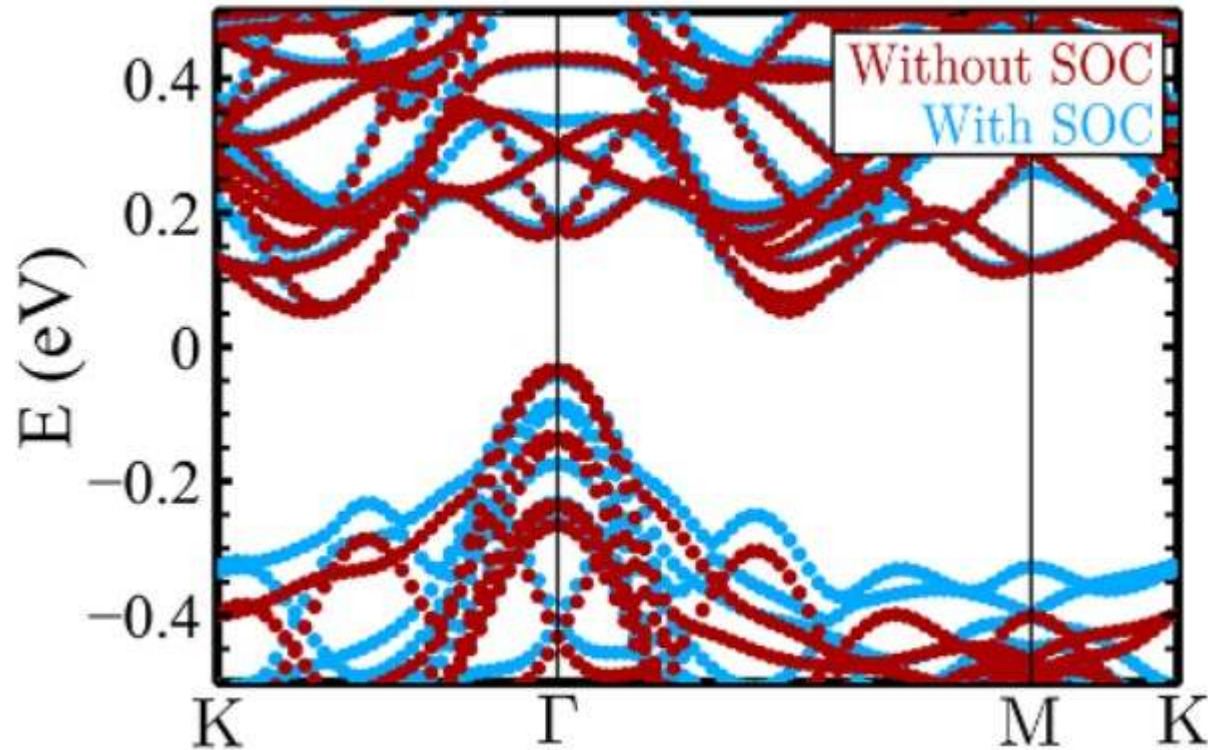
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

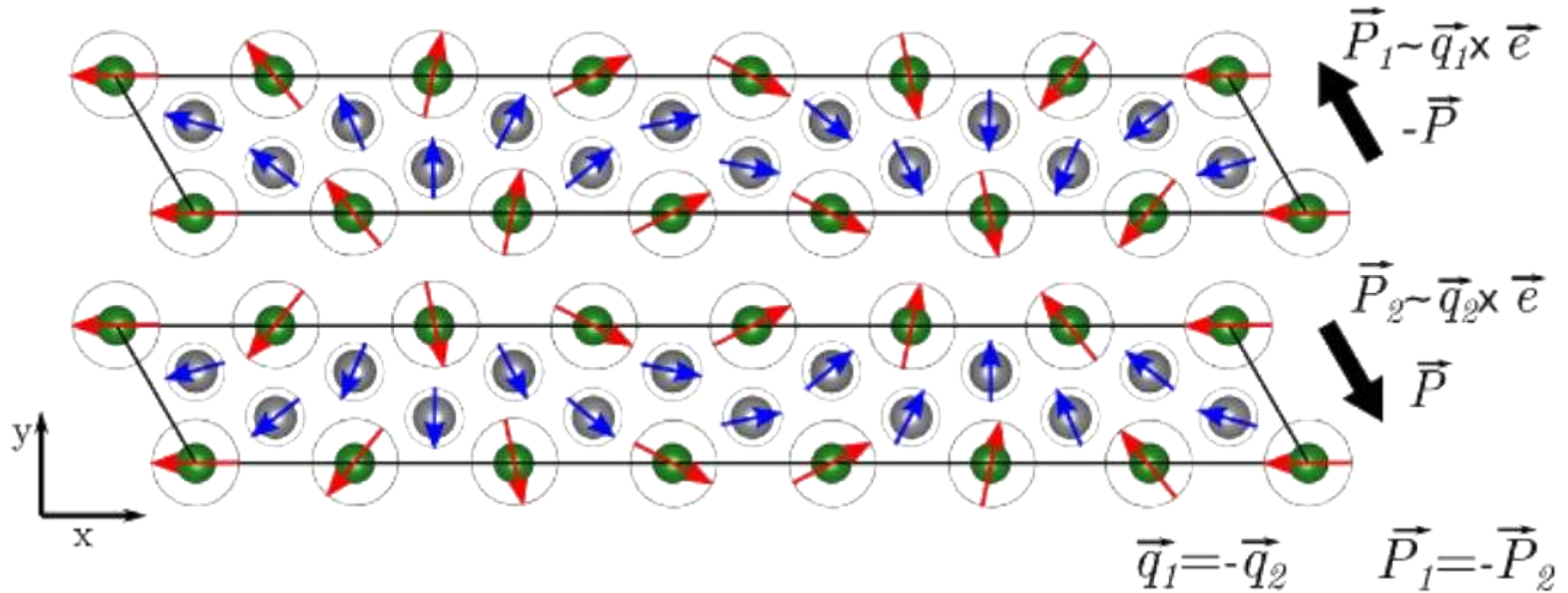
The impact of spin-orbit coupling in the electronic structure



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

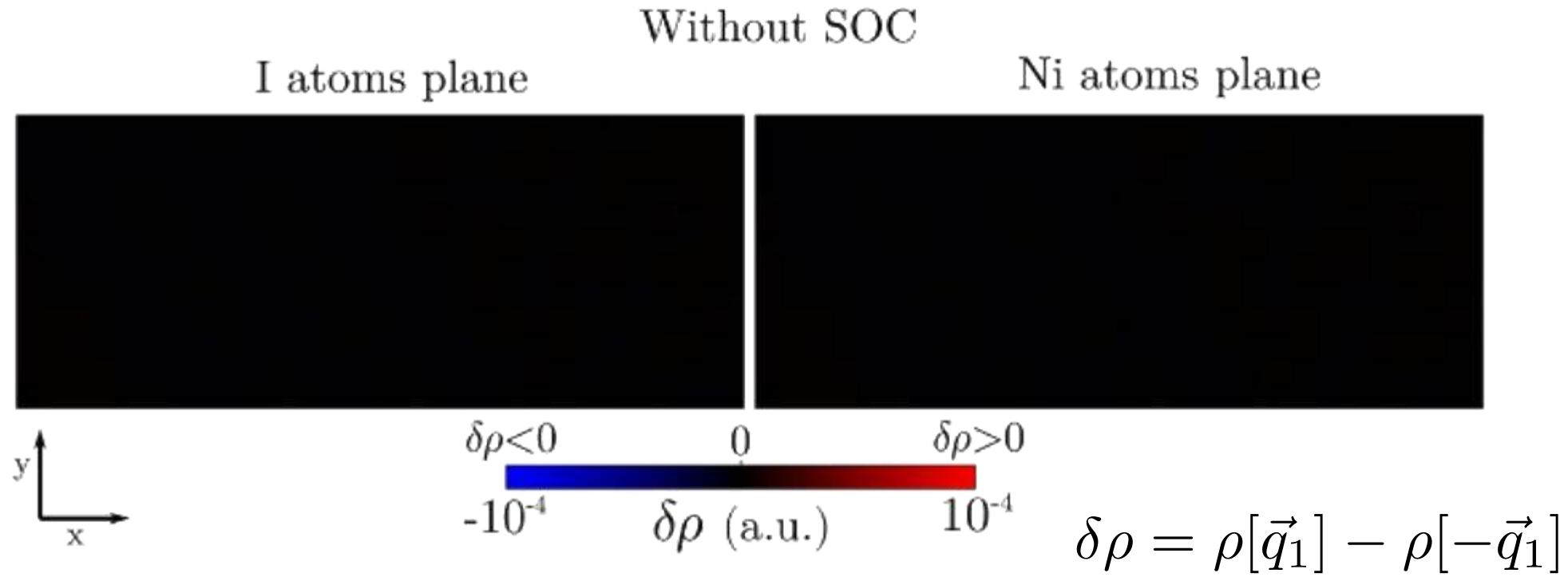
Ferroelectric correction to the electronic density



We will examine the ferroelectric density correction

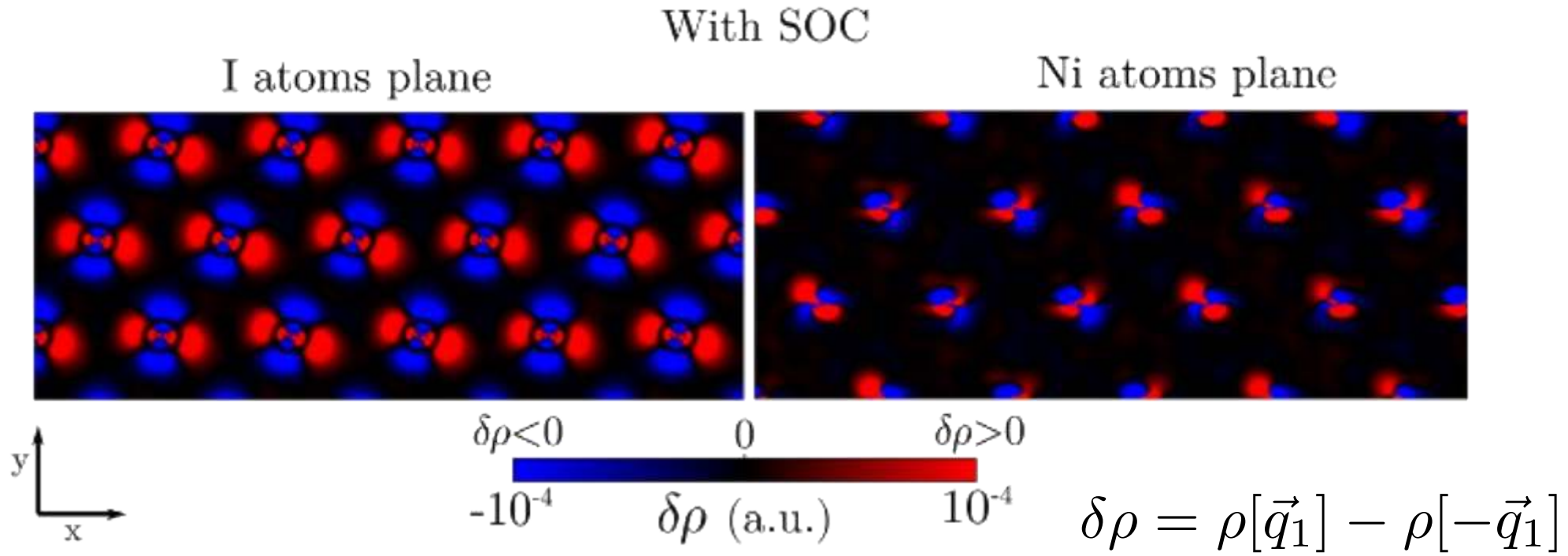
$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

Ferroelectric correction to the electronic density



In the absence of SOC, no ferroelectric density appears

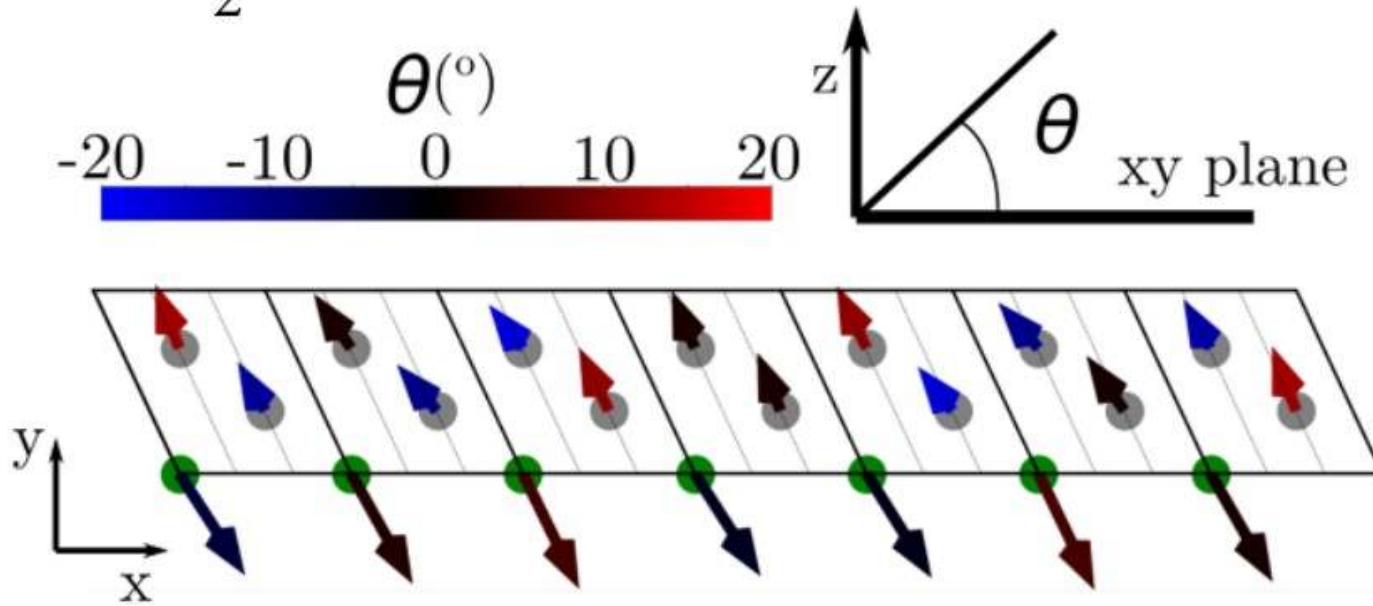
Ferroelectric correction to the electronic density



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

The ferroelectric force

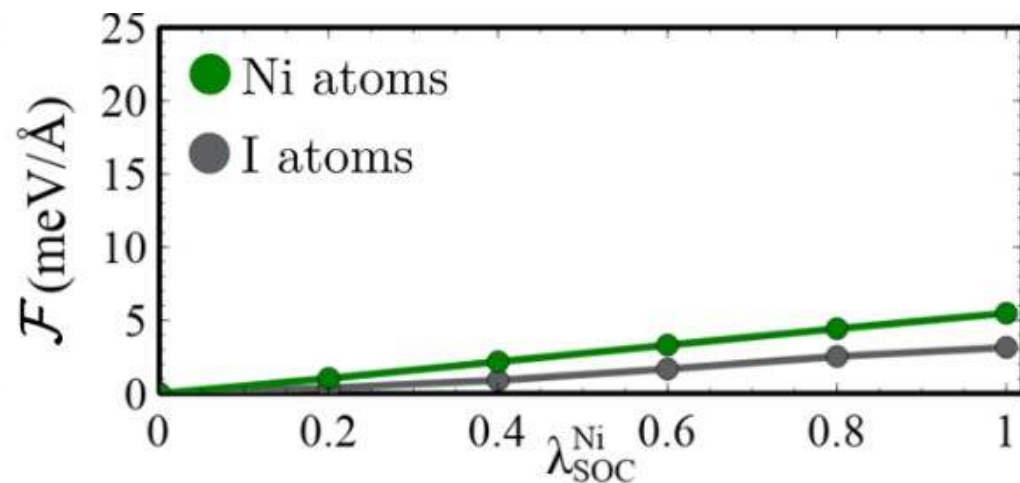
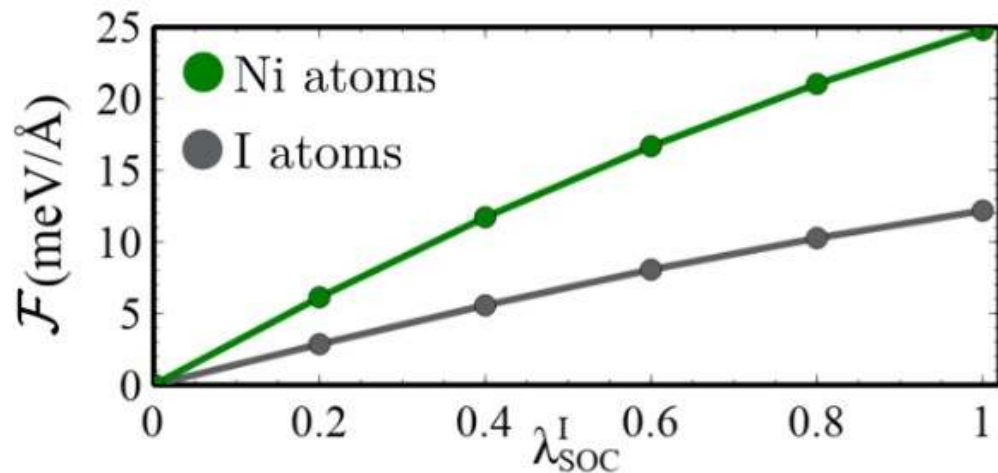
$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$



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

The ferroelectric force

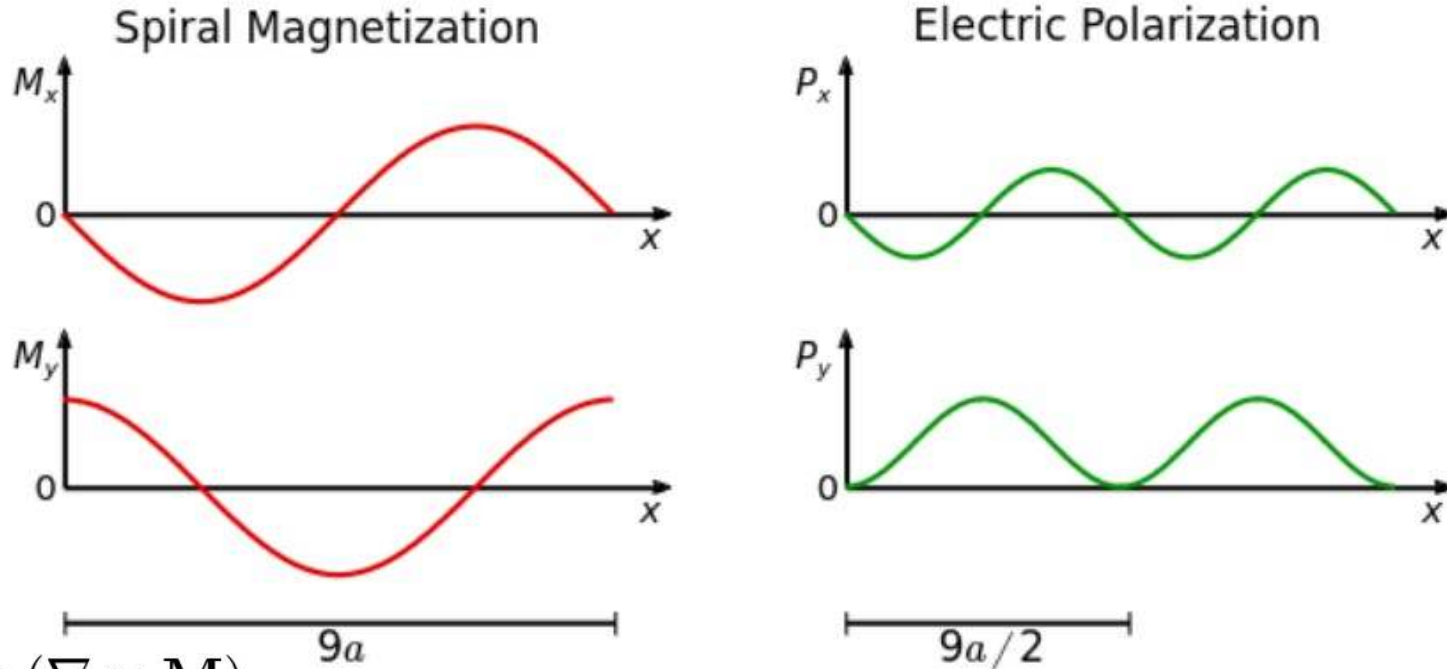
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\text{SOC}}^{\text{Ni}} \mathcal{H}_{\text{SOC}}^{\text{Ni}} + \lambda_{\text{SOC}}^{\text{I}} \mathcal{H}_{\text{SOC}}^{\text{I}},$$



The dominant contribution to the ferroelectric force comes from iodine

Modulated multiferroicity in monolayer NiI_2

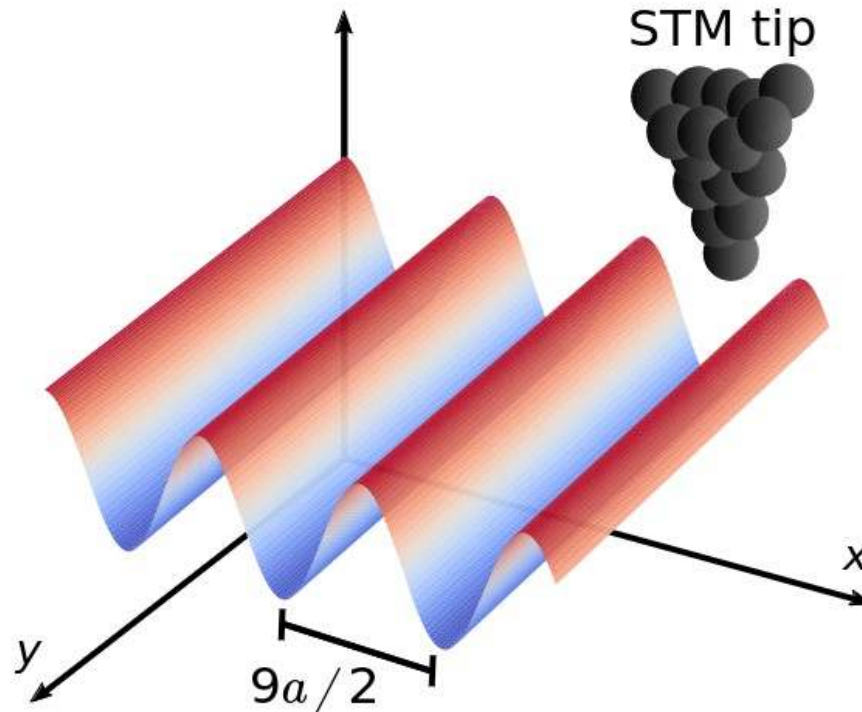
The ferroelectric order has actually a modulation in real space



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

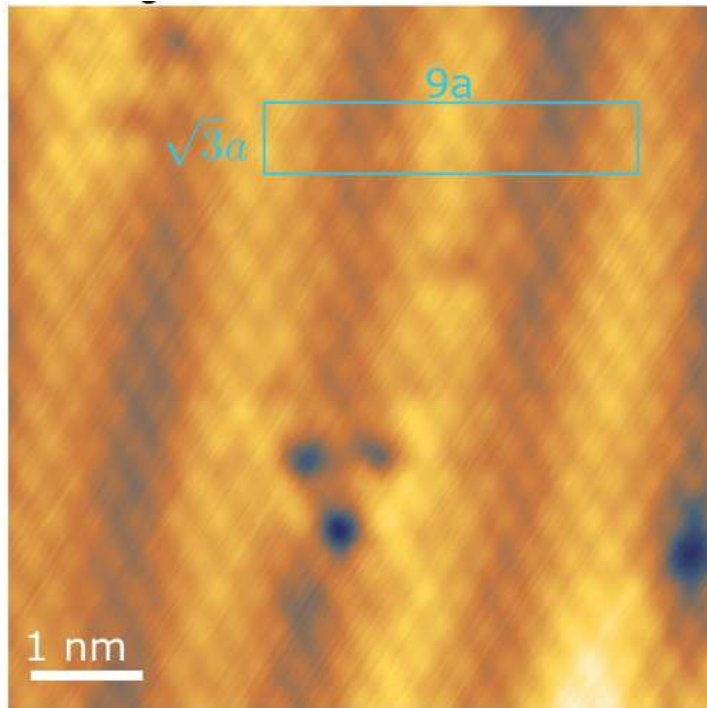
Visualizing multiferroicity in monolayer NiI_2

Modulated electrostatic potential

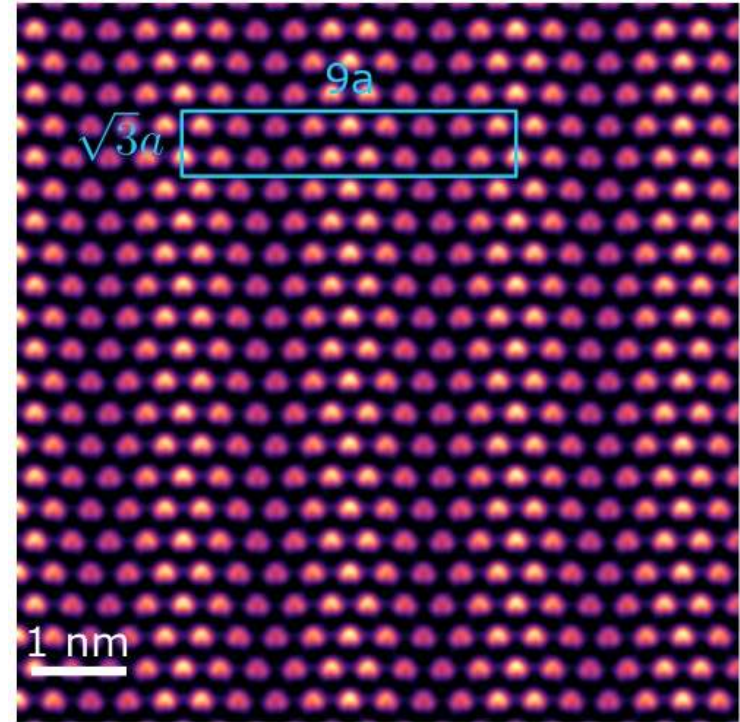


Visualizing multiferroicity in monolayer NiI_2

Experiment



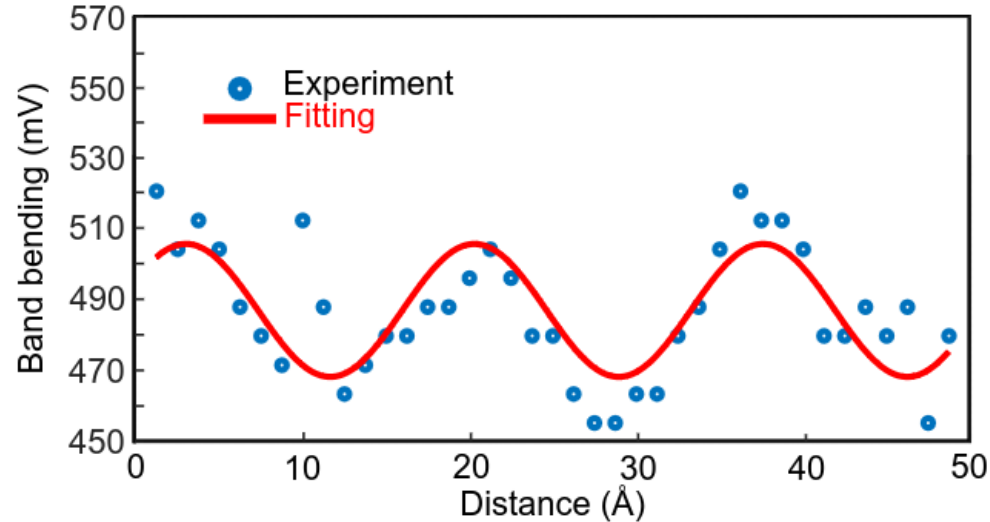
Theory



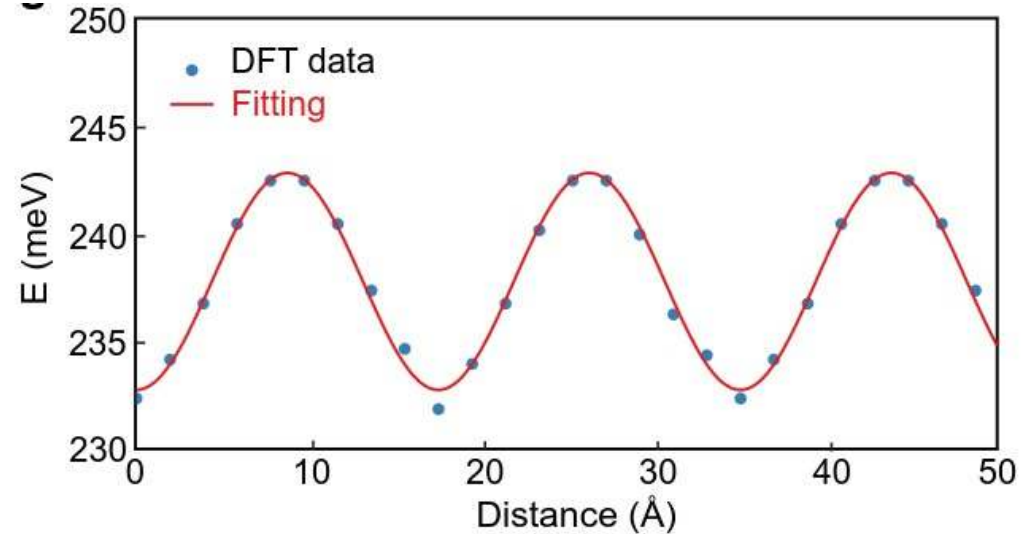
$$\Lambda_{\mathbf{q}}(-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

Ferroelectricity from modulated band offset

Band offset (experiment)

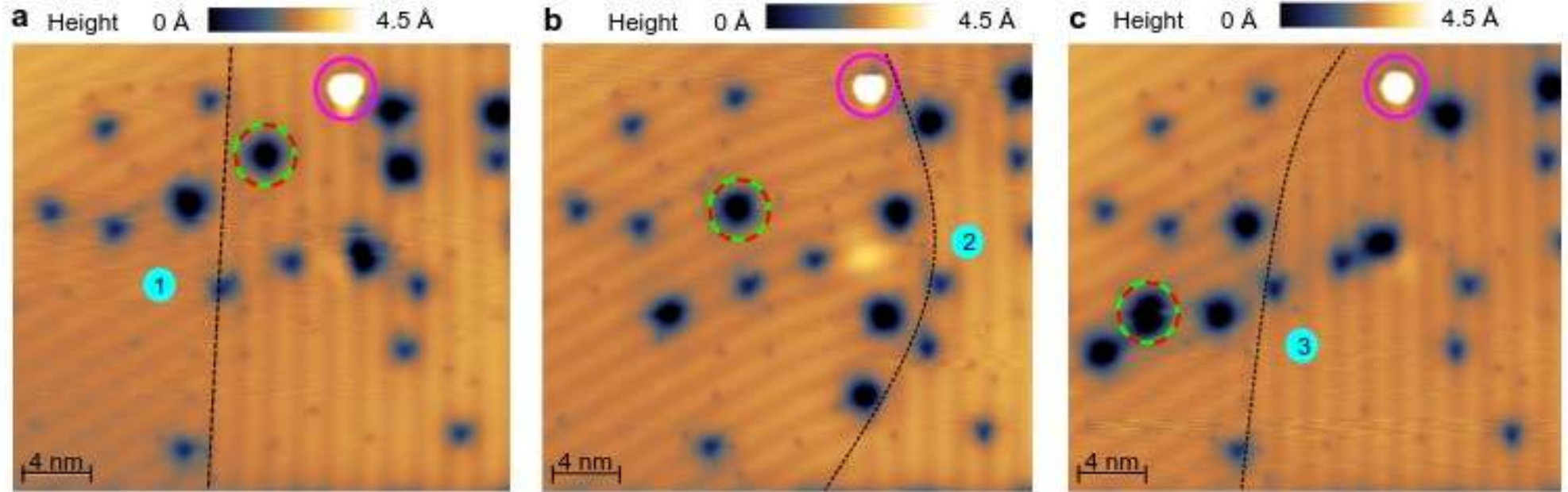


Band offset (theory)



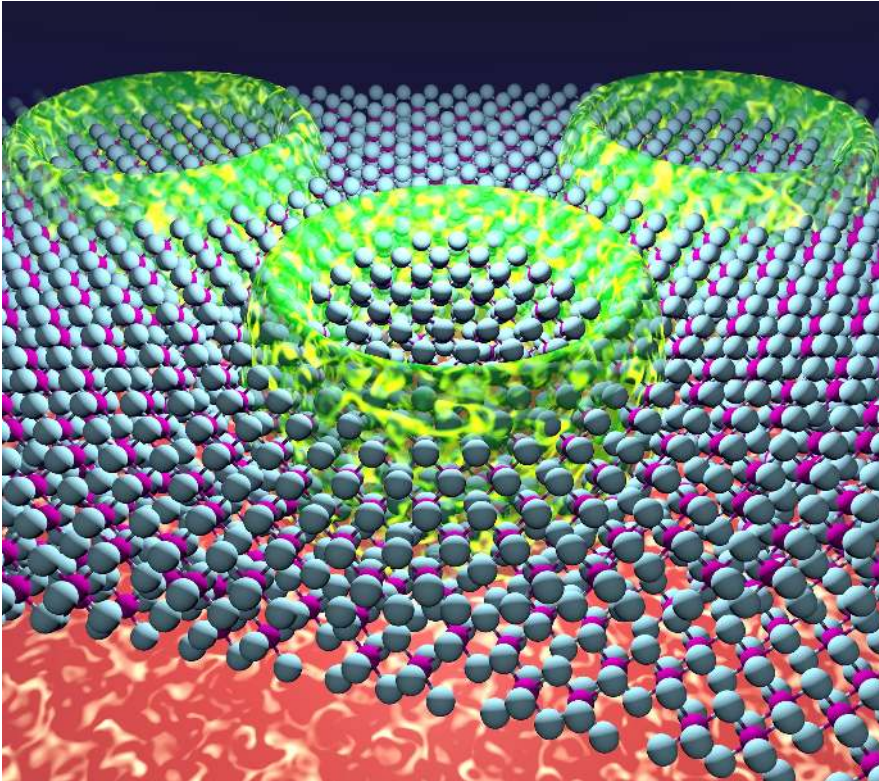
$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{M^2} = \Lambda q (-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

Controlling multiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

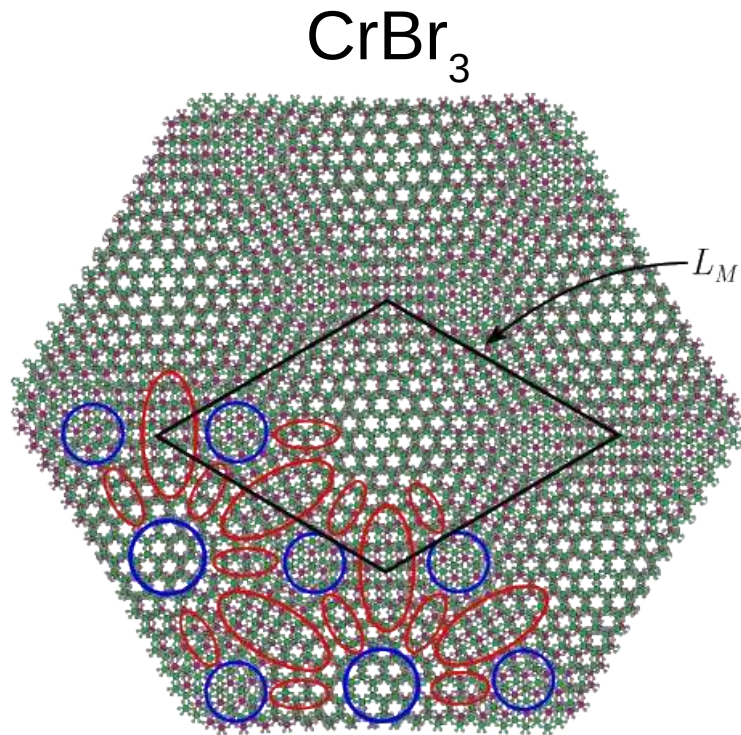
Engineering artificial multiferroics with twisted van der Waals materials



A. Fumega

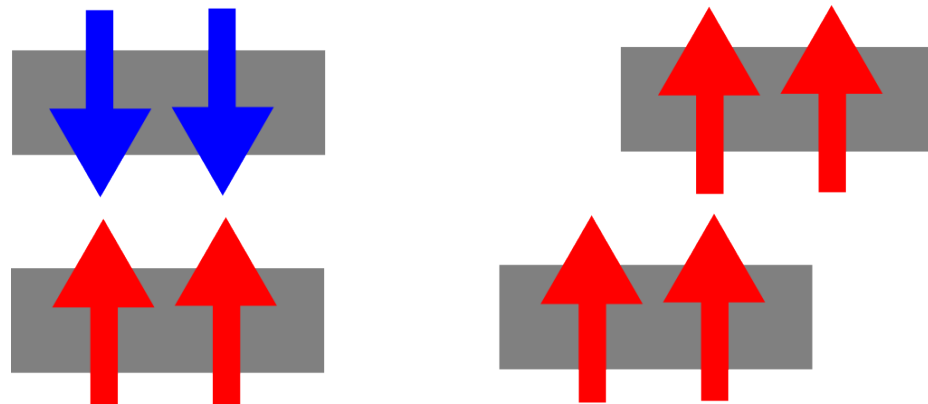


Twisted 2D trihalides



Bilayer Stacking

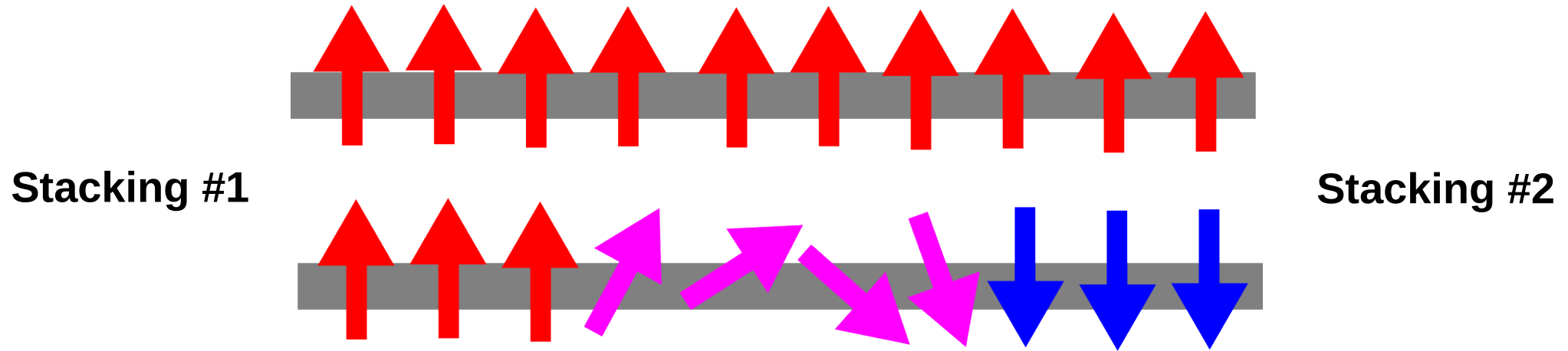
(M)	Monoclinic
(R)	Rhombohedral



The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Mixed FE/AF domains

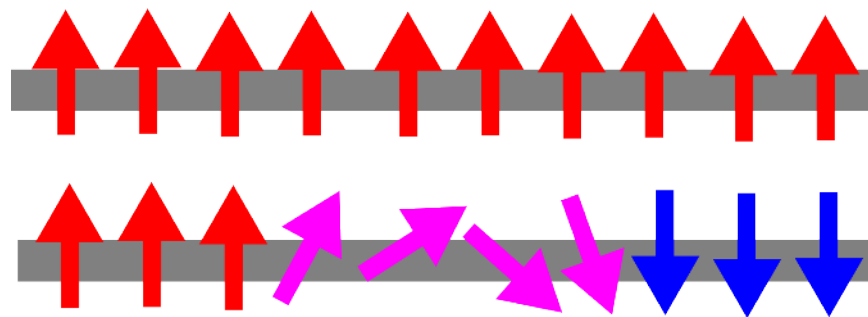
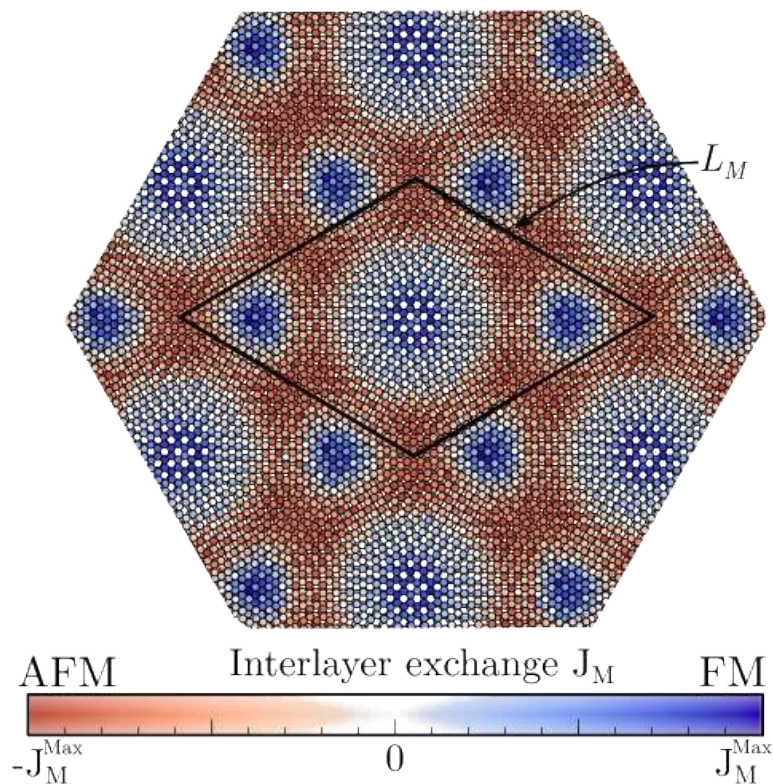
Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism

Nature Physics 19, 1150–1155 (2023)

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

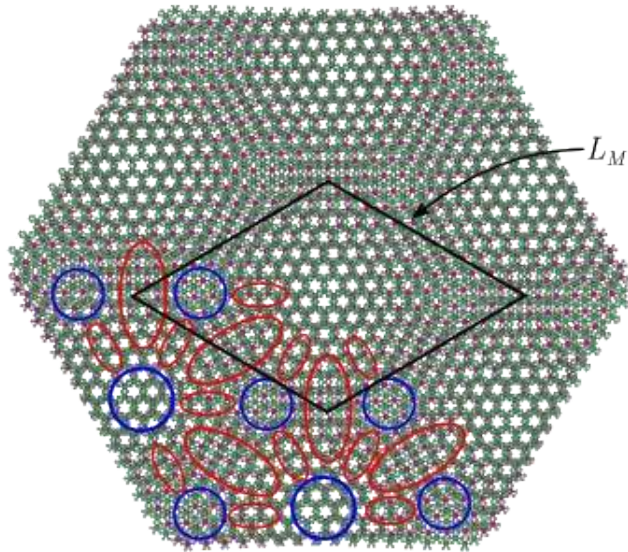
SOC driven

Spin rotation axis

Wavevector of the spin spiral

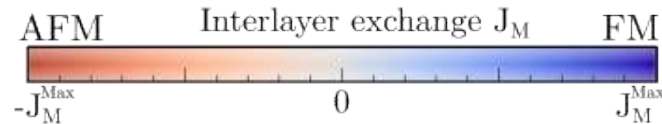
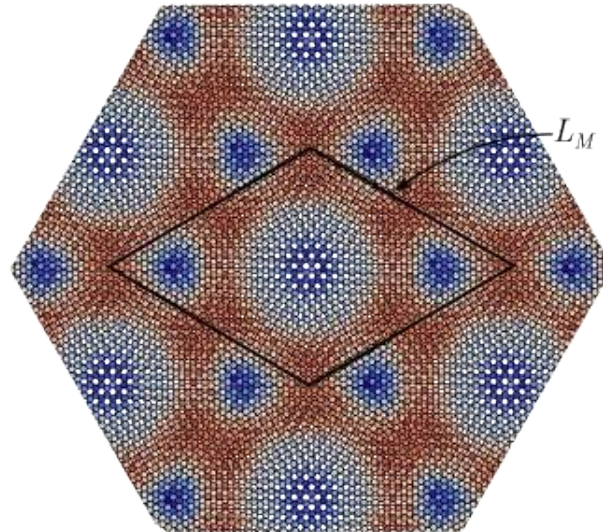
Stacking, magnetic order and electric polarization

Structure

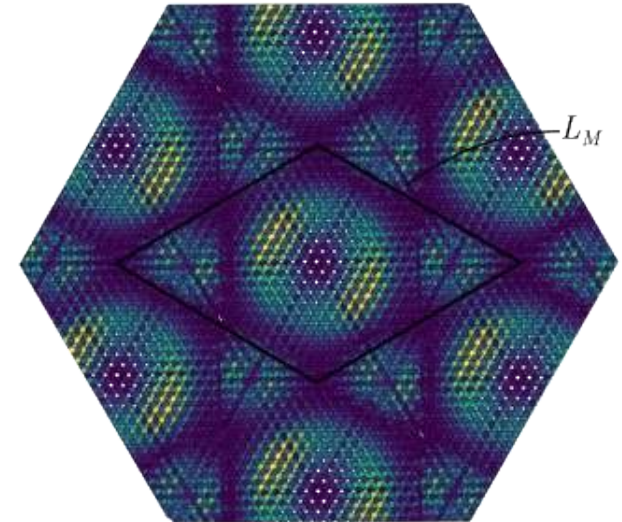


Bilayer Stacking $\left\{ \begin{array}{l} \textcircled{M} \text{ Monoclinic} \\ \textcircled{R} \text{ Rhombohedral} \end{array} \right.$

Magnetic order



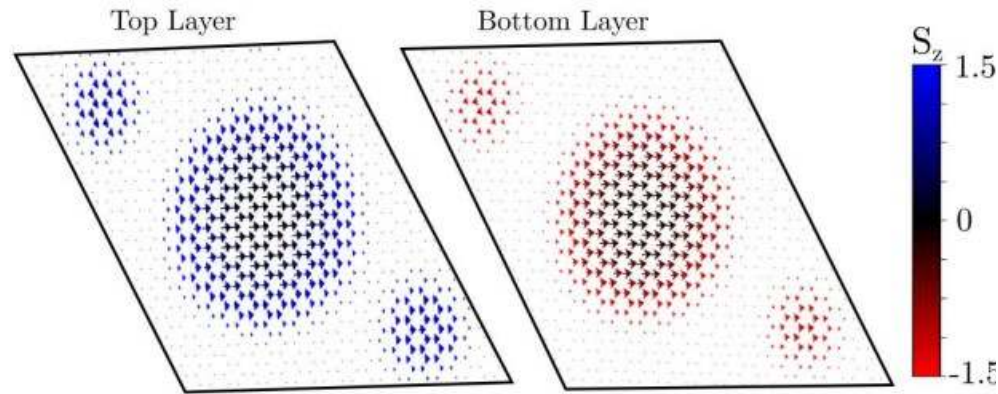
Ferroelectric order



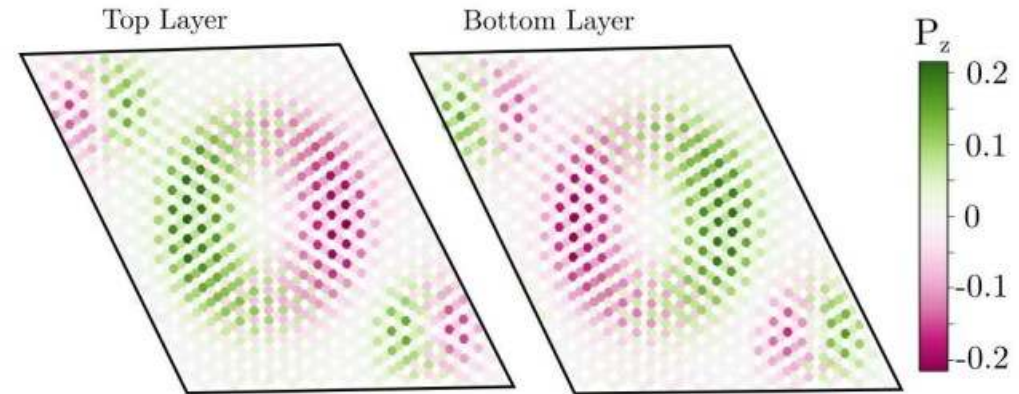
Stacking, magnetic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

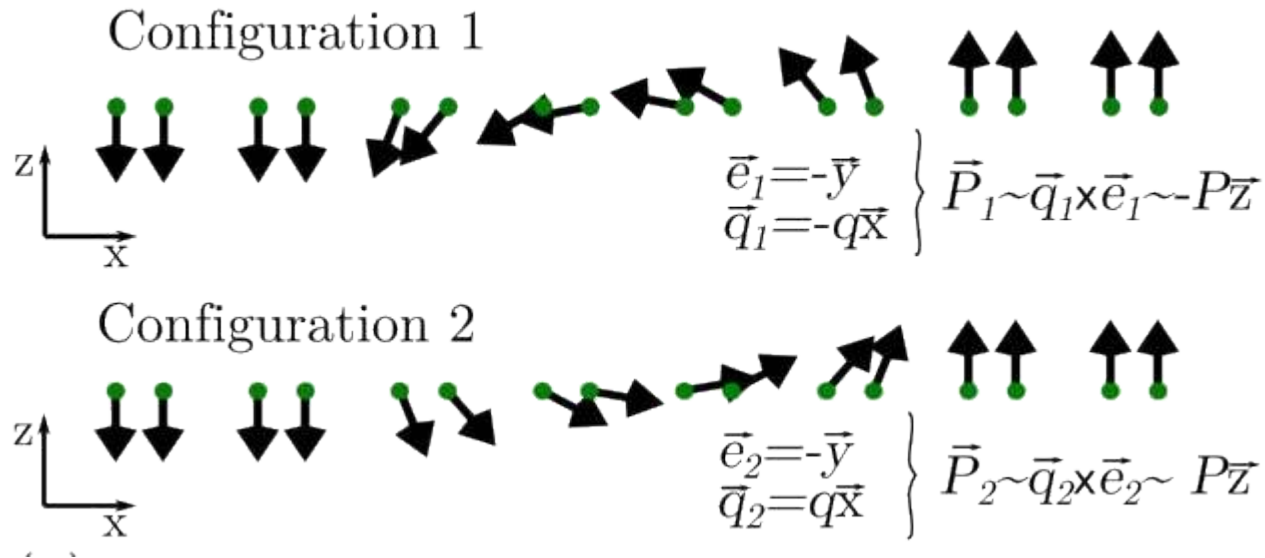


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



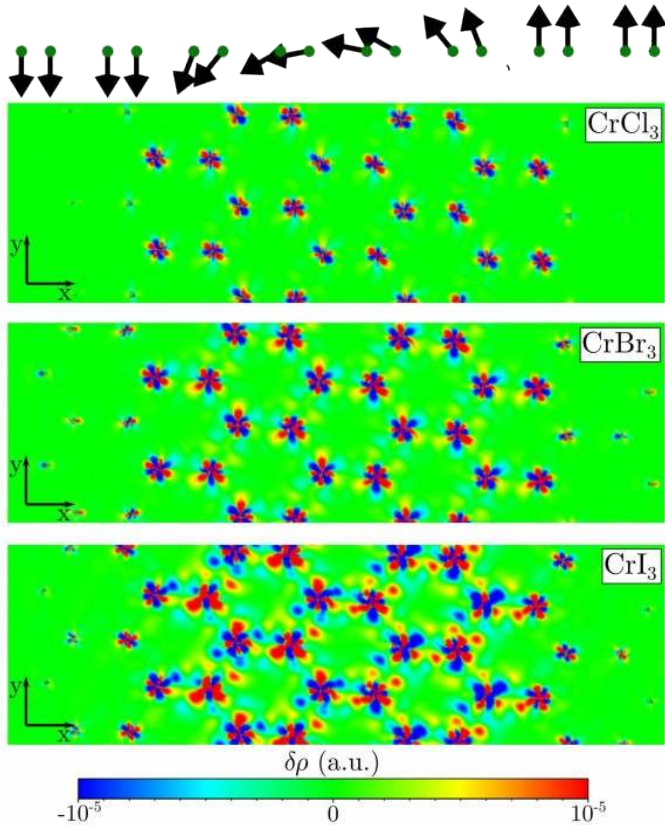
Ferroelectric electronic density

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

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

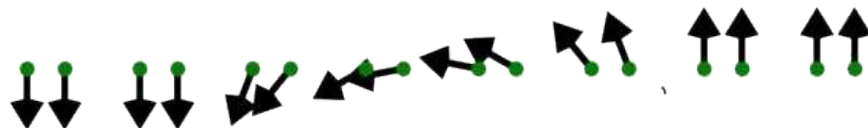
$$\delta\rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

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

It becomes stronger the heavier the halide

The ferroelectric force

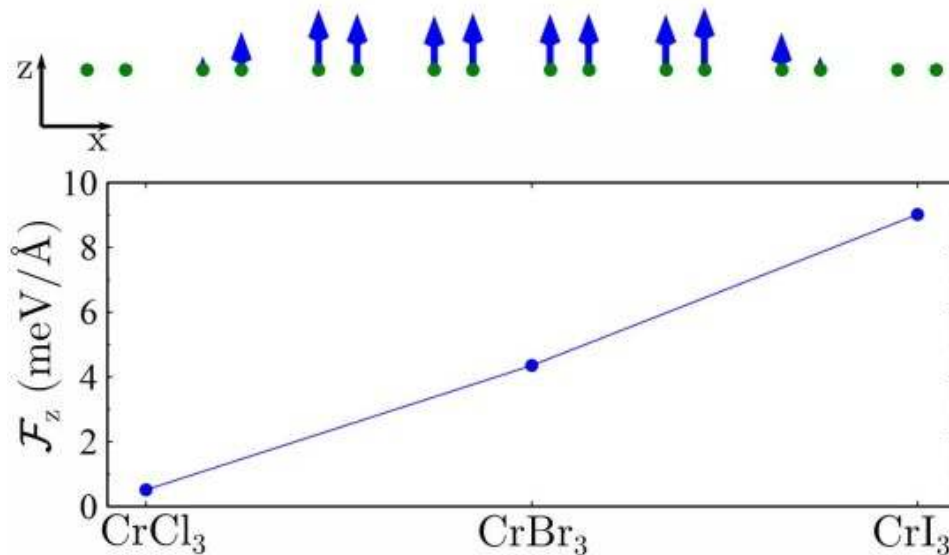
Magnetic configuration



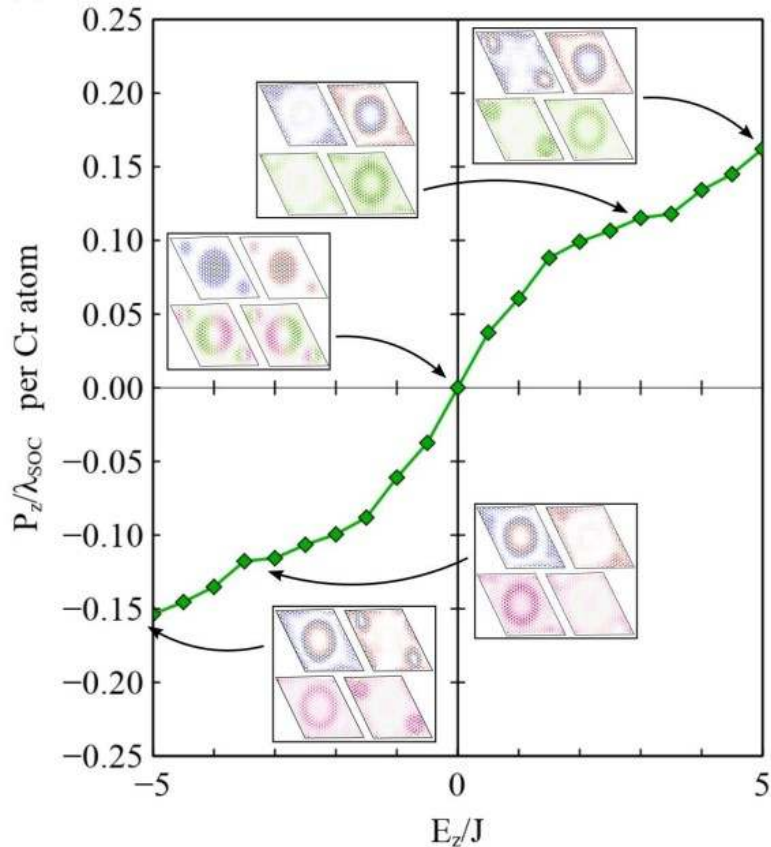
Ferroelectric force

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

The ferroelectric force
becomes
Stronger for heavier halides



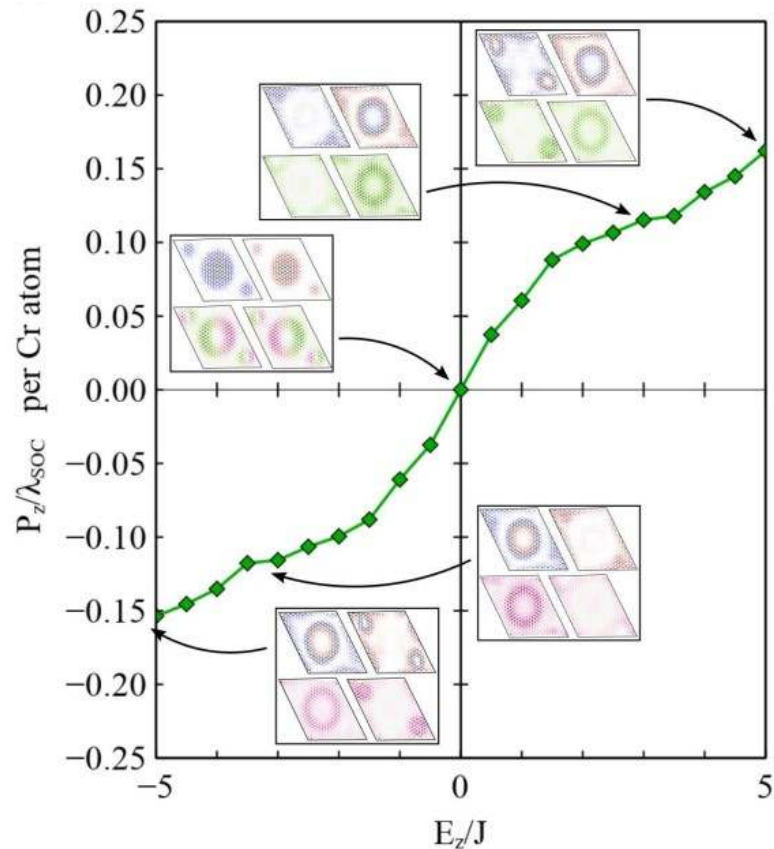
Electrically controllable skyrmions in twisted magnets



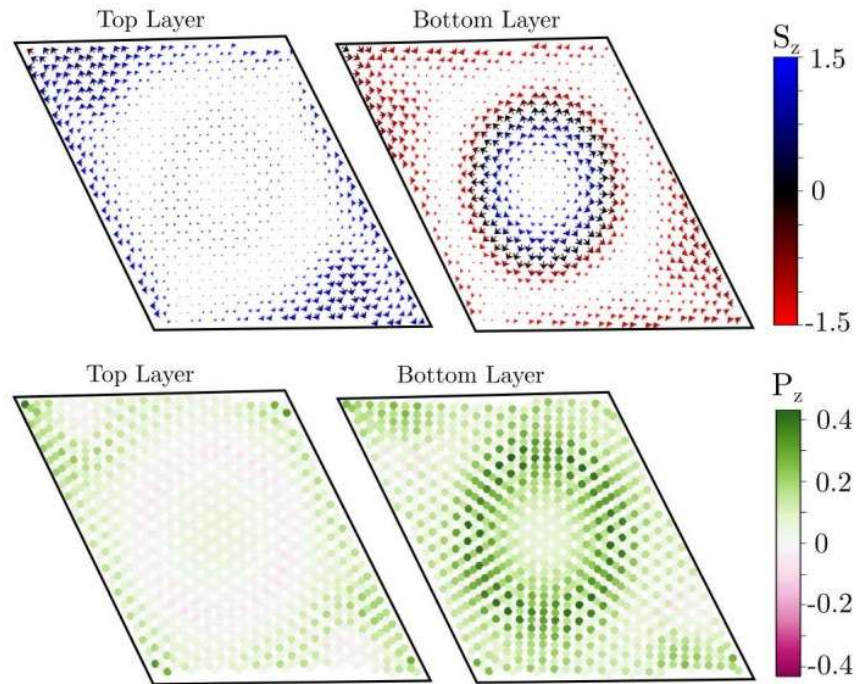
The electric polarization directly couples to an out-of plane electric field

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

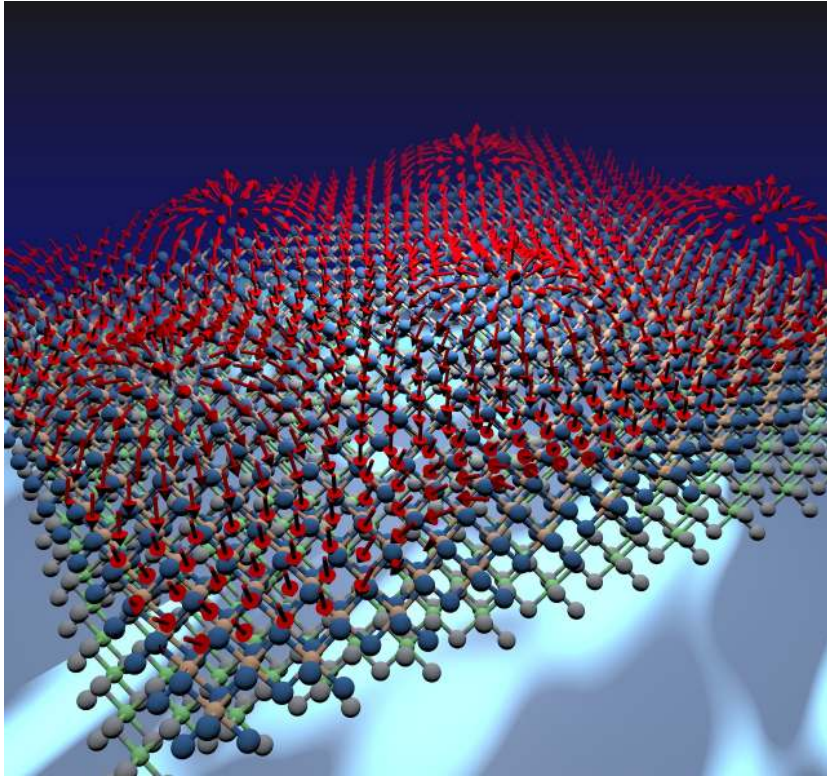
Electrically controllable skyrmions in twisted magnets



Skyrmions at finite interlayer bias



Electric control of moiré skyrmions in twisted multiferroic NiI_2 bilayers



A. Fumega



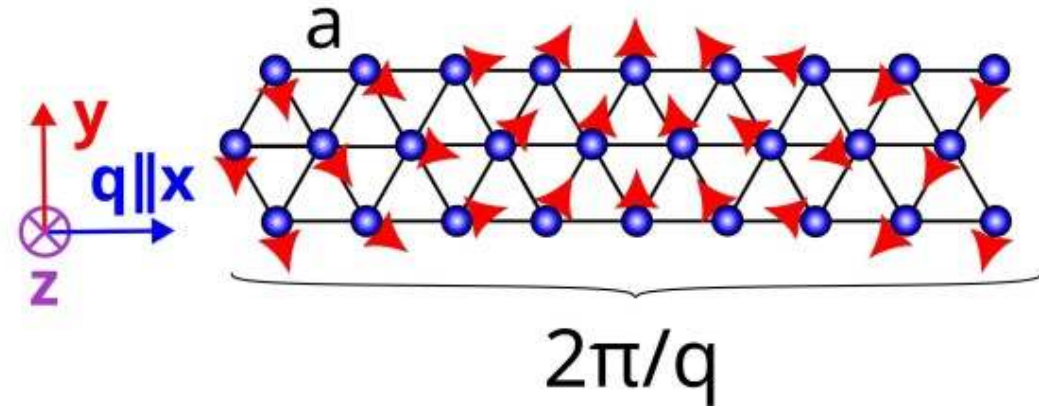
T. Antao



Nano Letters 24 (49), 15767–15773 (2024)

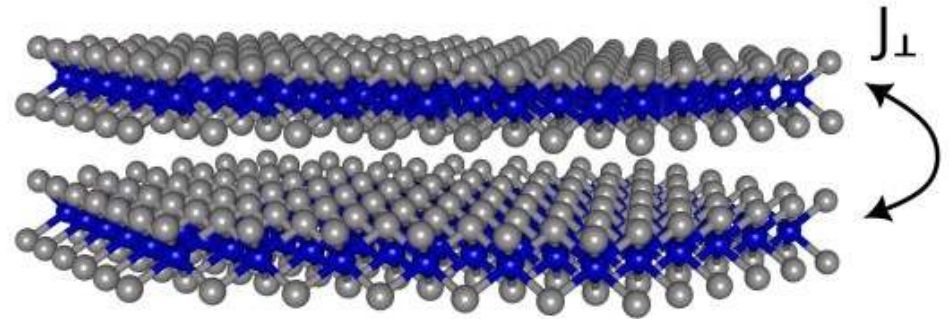
From monolayer NiI2 to twisted bilayer

NiI2 magnetic order



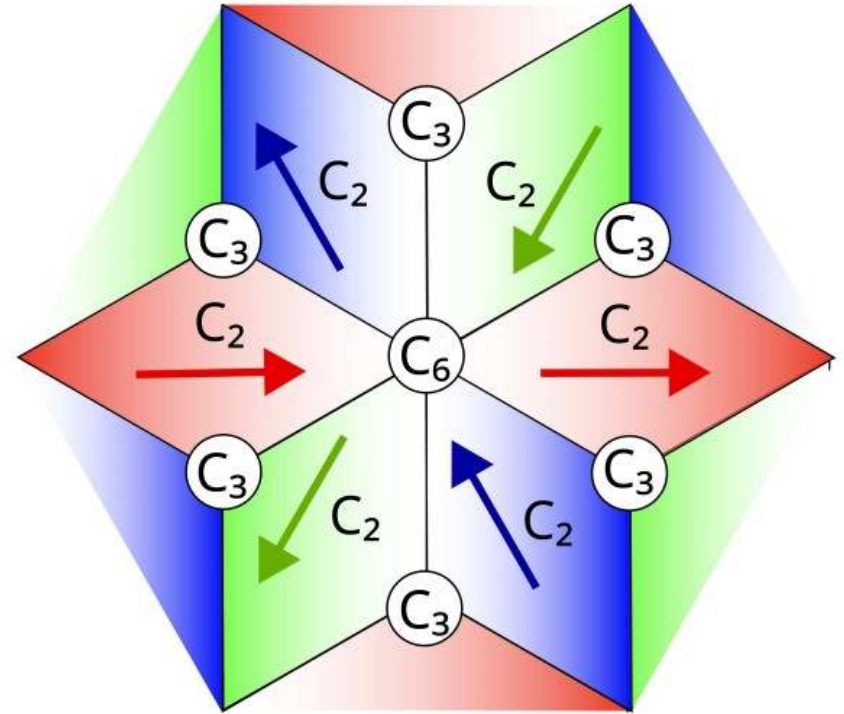
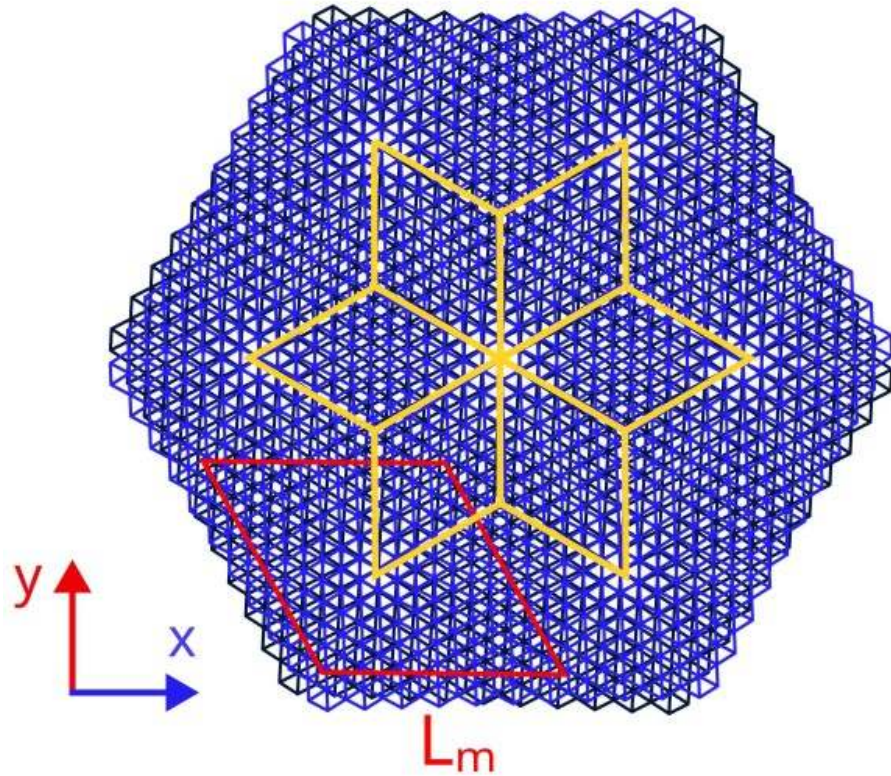
Monolayer NiI2 shows a frustrated magnetic order

Twisted bilayer NiI2



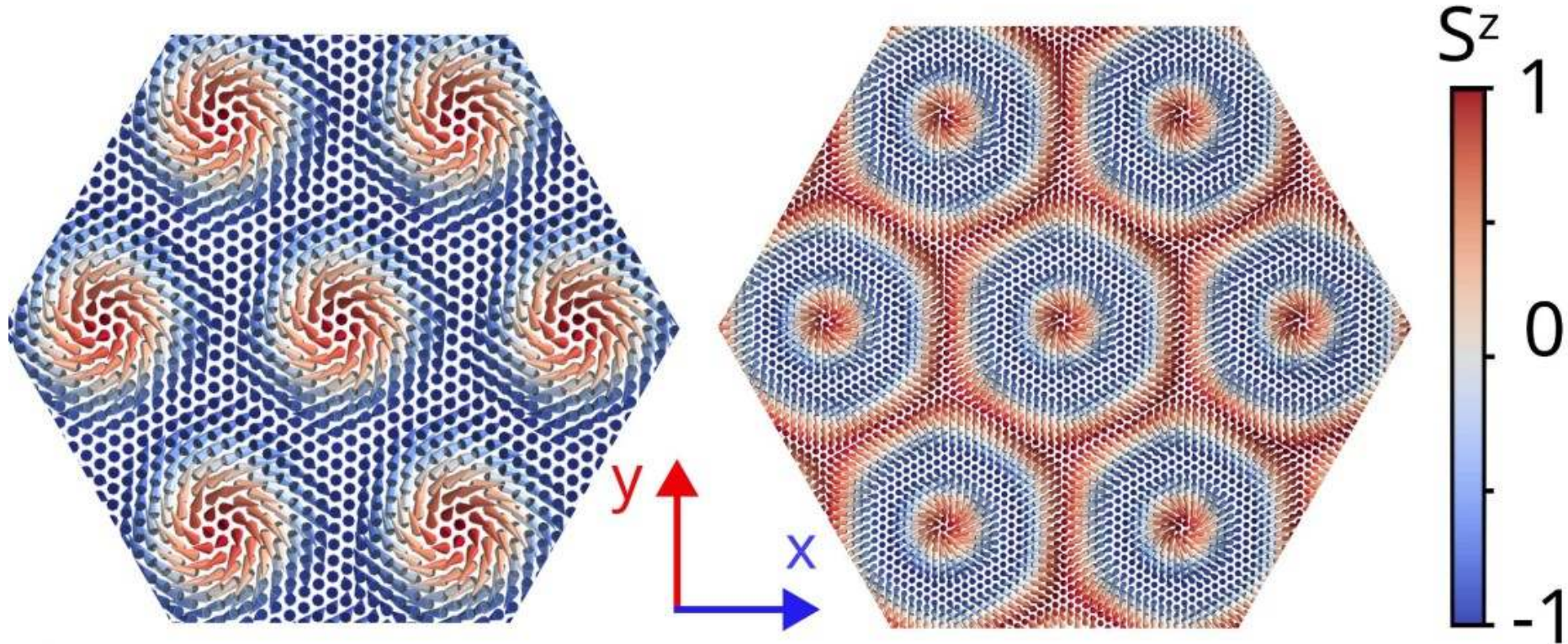
Twisted NiI2 shows additional frustration from the interlayer exchange

Structure and spiral mosaic in twisted NiI_2



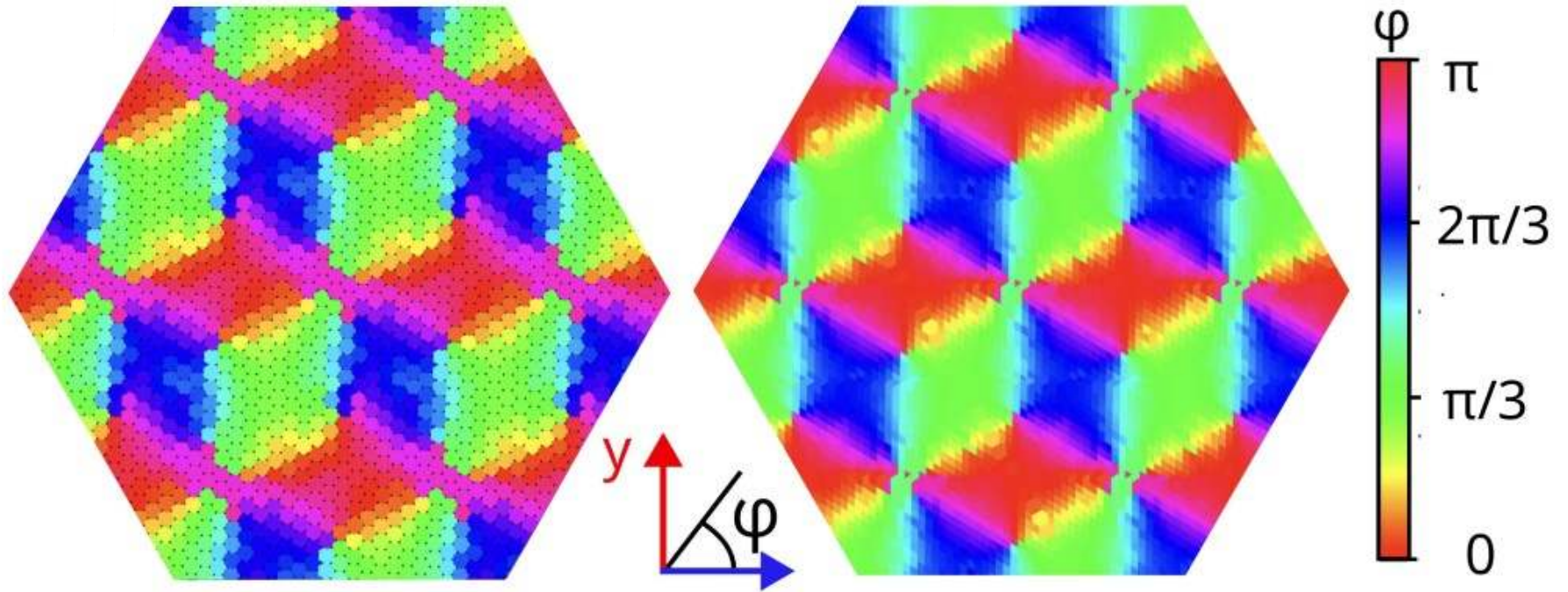
The local different stacking in twisted NiI_2 promotes a mosaic of spin spirals

Emergent moire topological magnetism



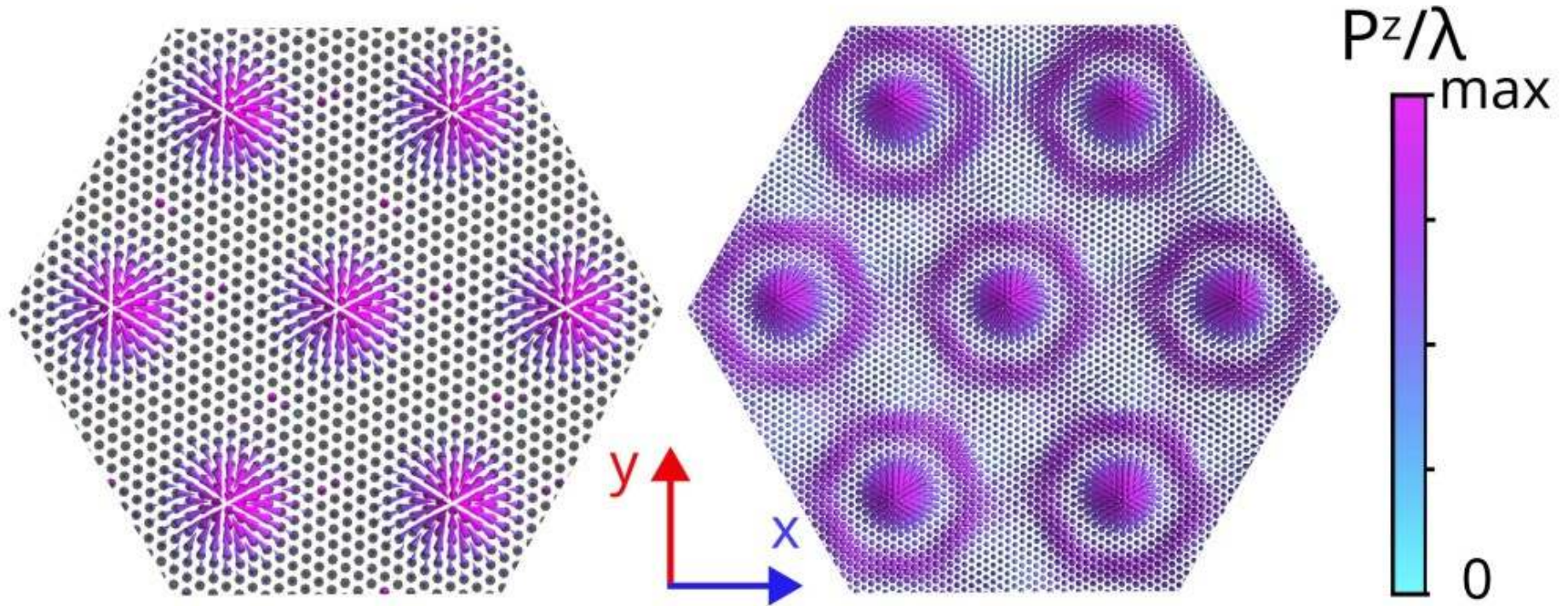
The competition between intralayer and inter/layer exchange leads to moire magnetic skyrmions

Spin spiral moire mosaic



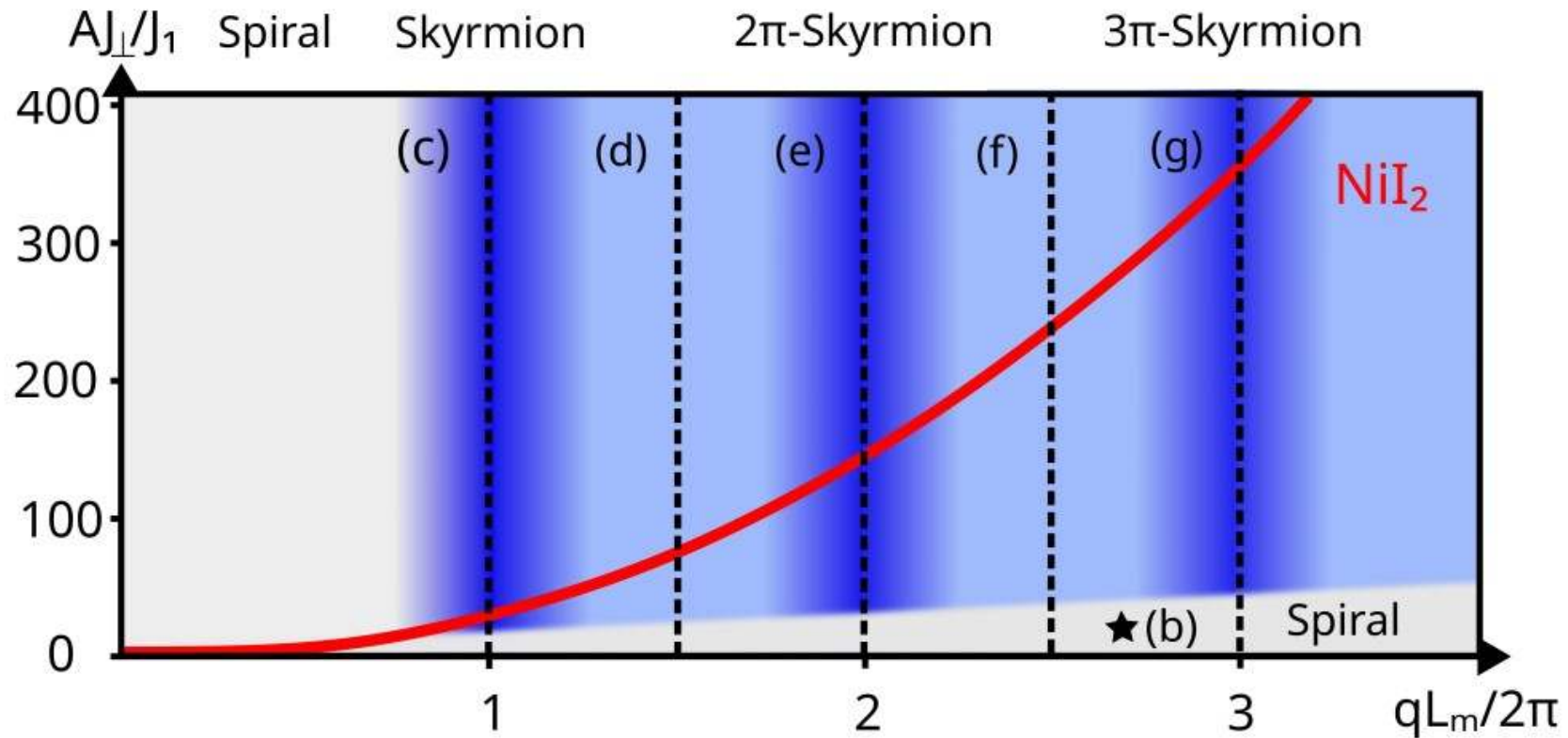
The moire stacking pattern promote different q-vectors in different regions of space

Emergent moire ferroelectricity



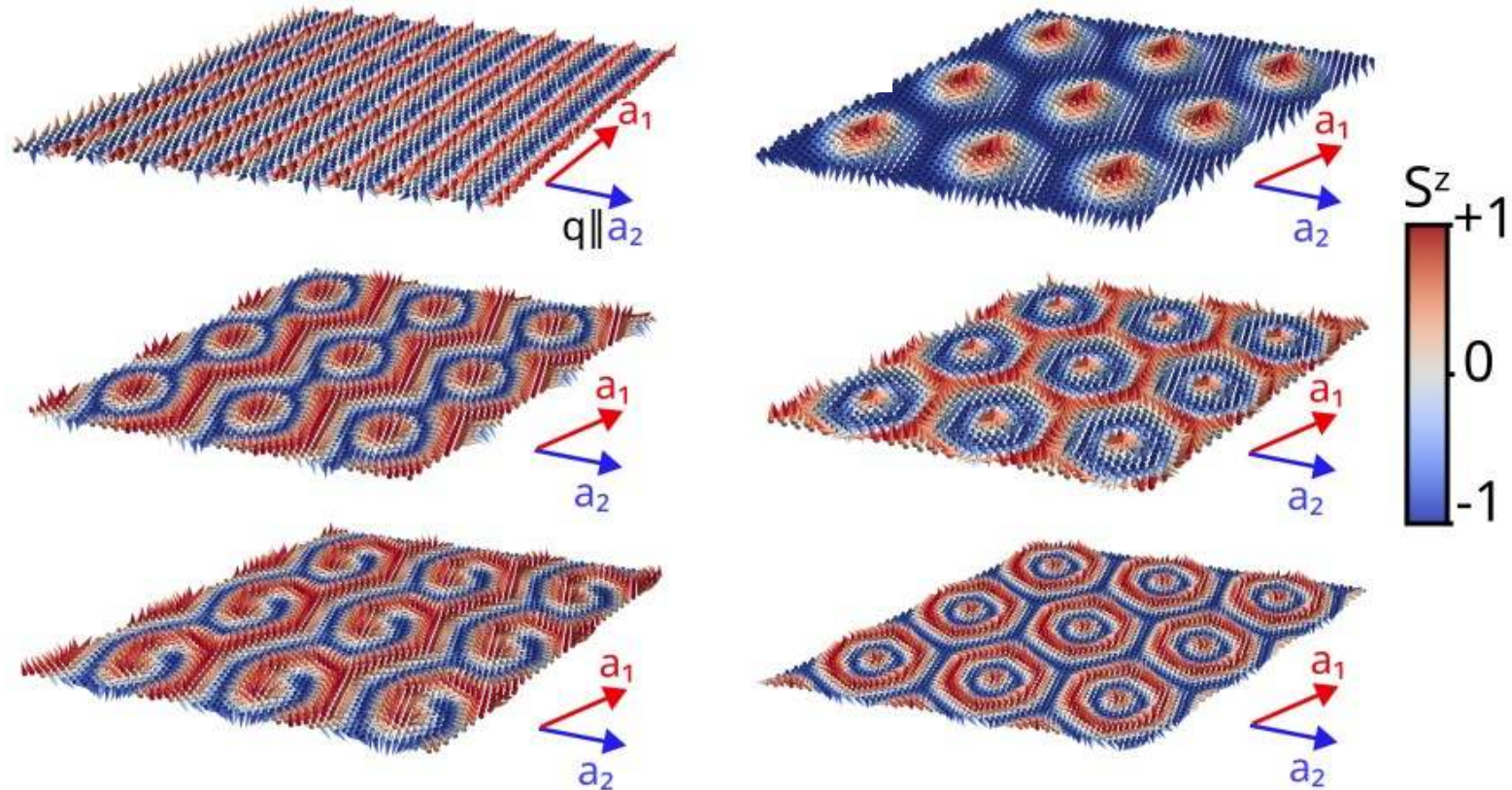
Out-of plane ferroelectricity emerges associated with the magnetic skyrmions

Skyrmion phases as a function of twist angle



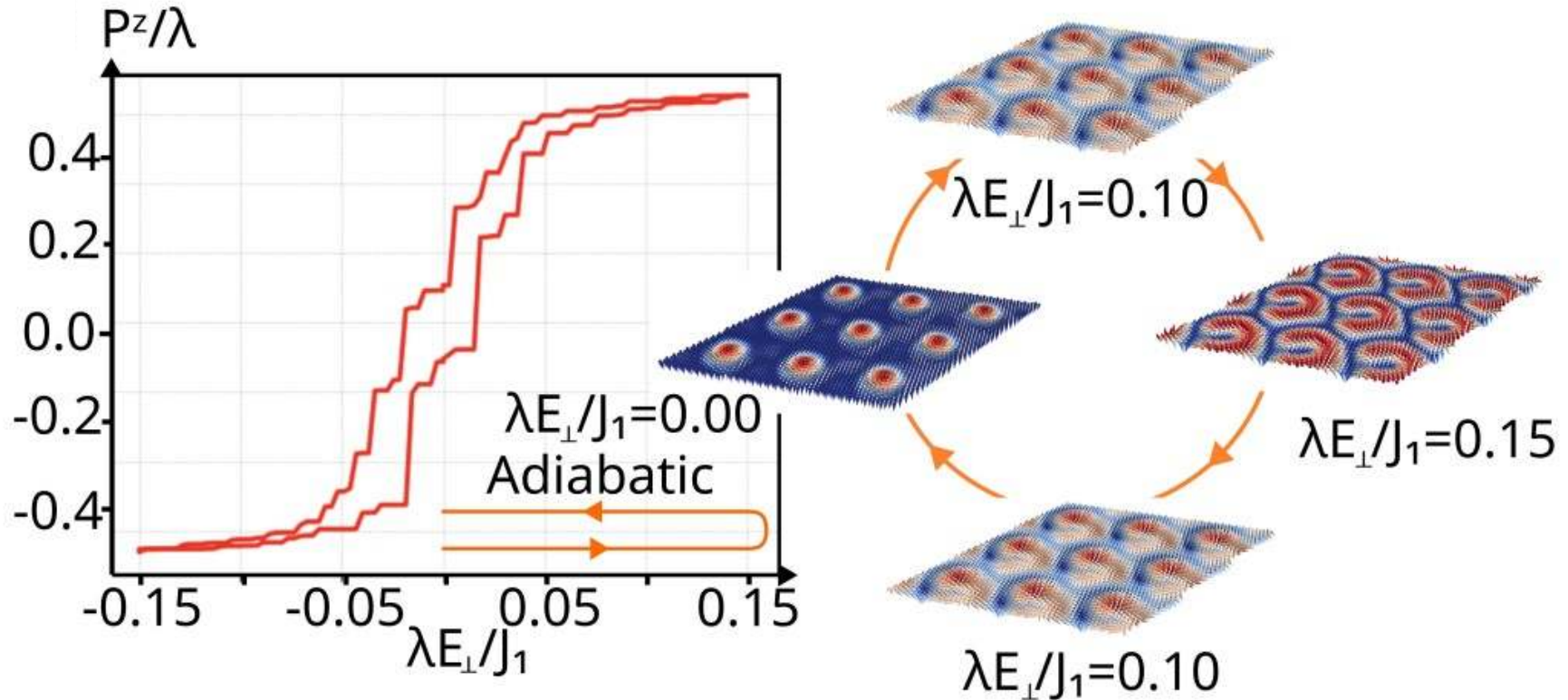
The twist angle controls the magnetic phase emerging in the twisted bilayer

Skyrmion phases as a function of twist angle



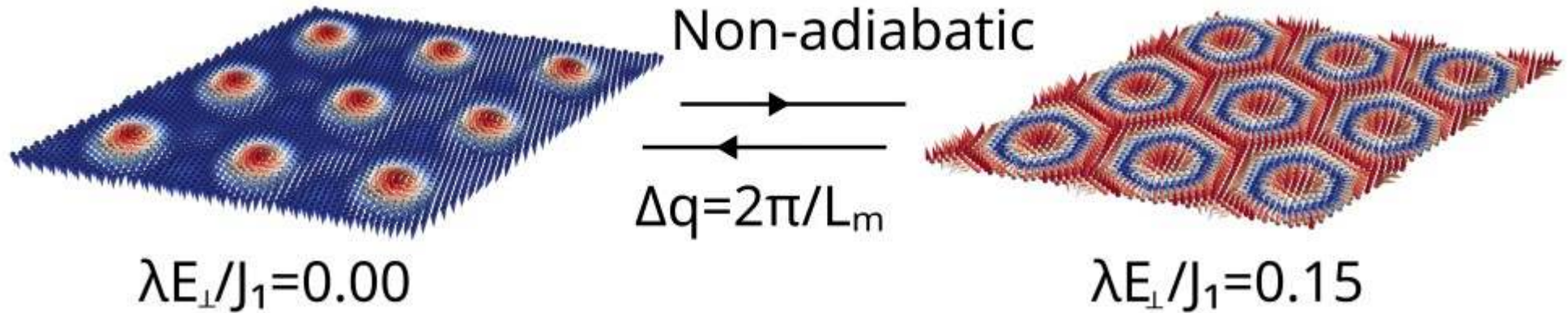
As the twist angle is changed, skyrmion lattices of different windings appear

Controlling magnetic order with an electric interlayer bias



Interlayer electric biases lead to electrically controllable topological magnetic order

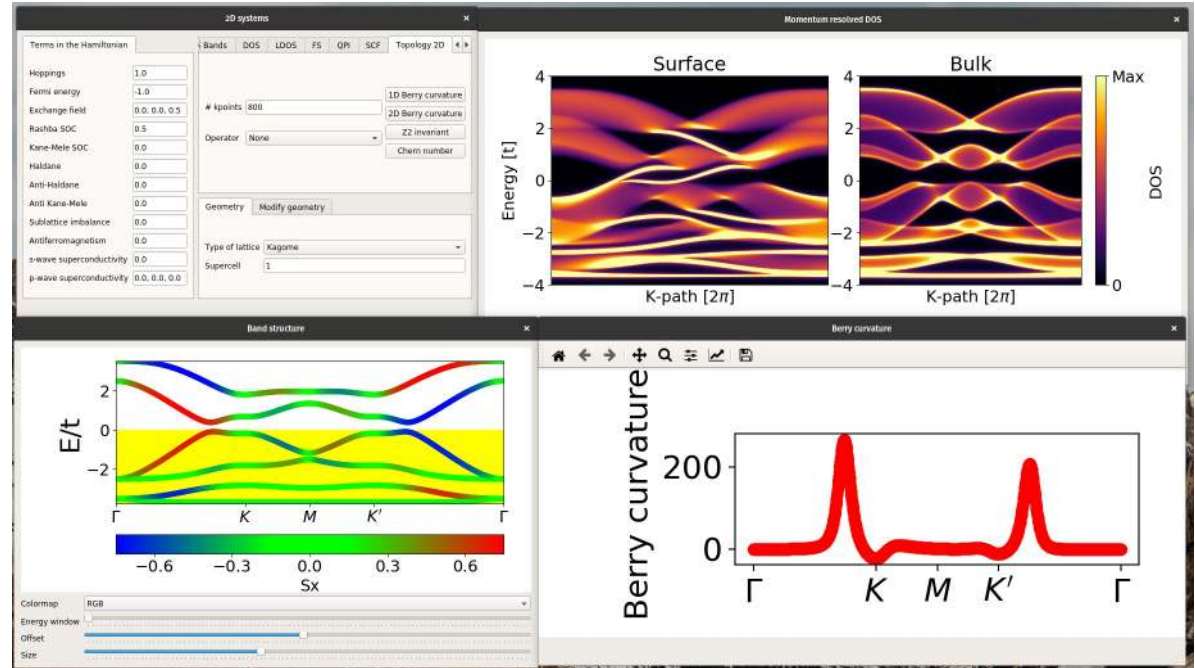
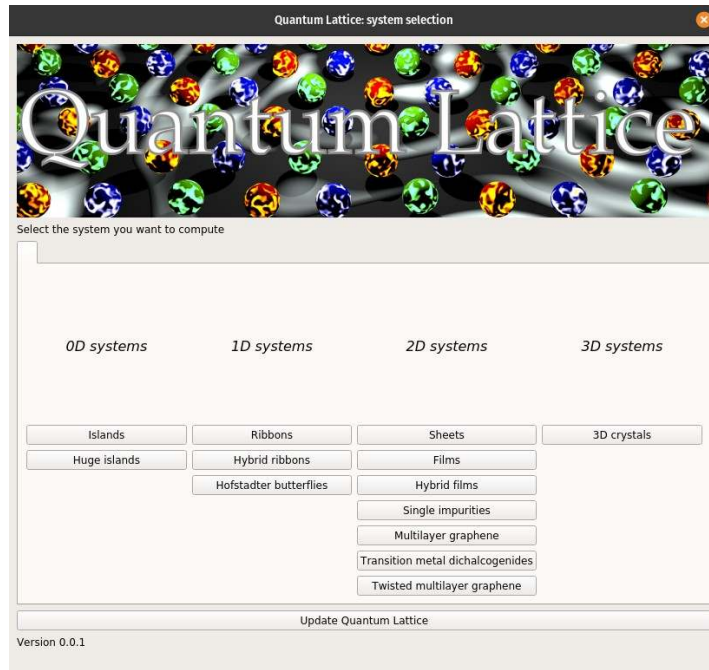
Controlling magnetic order with an electric interlayer bias



**Adiabatic and non-adiabatic changes of the electric bias
stabilize different magnetic orders**

Open-source software for
van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

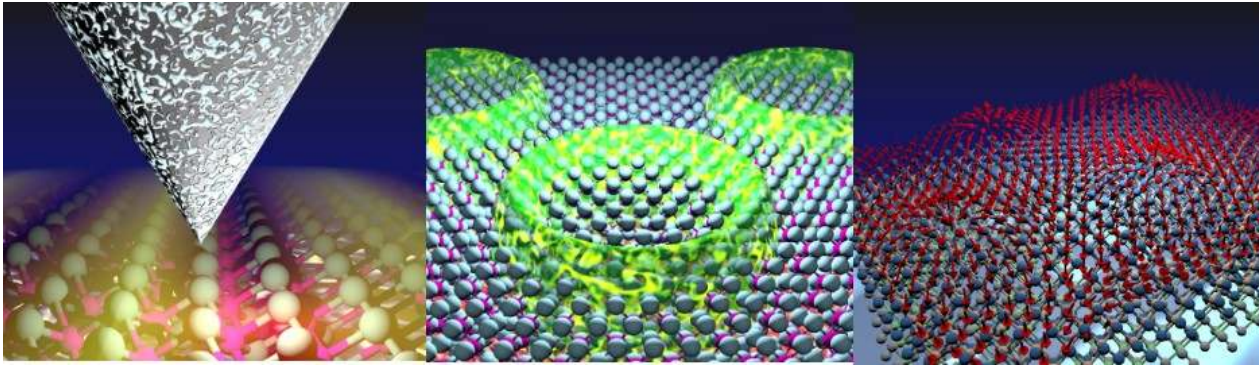


Quantum Lattice: open source interactive interface for tight binding modeling

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

Take home

Van der Waals multiferroics offer a new playground to explore unconventional phenomena in moire heterostructures



2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)
Advanced Materials, 2311342 (2024)
Nano Letters 24 (49), 15767–15773 (2024)

Funding from

Finnish Quantum Flagship

