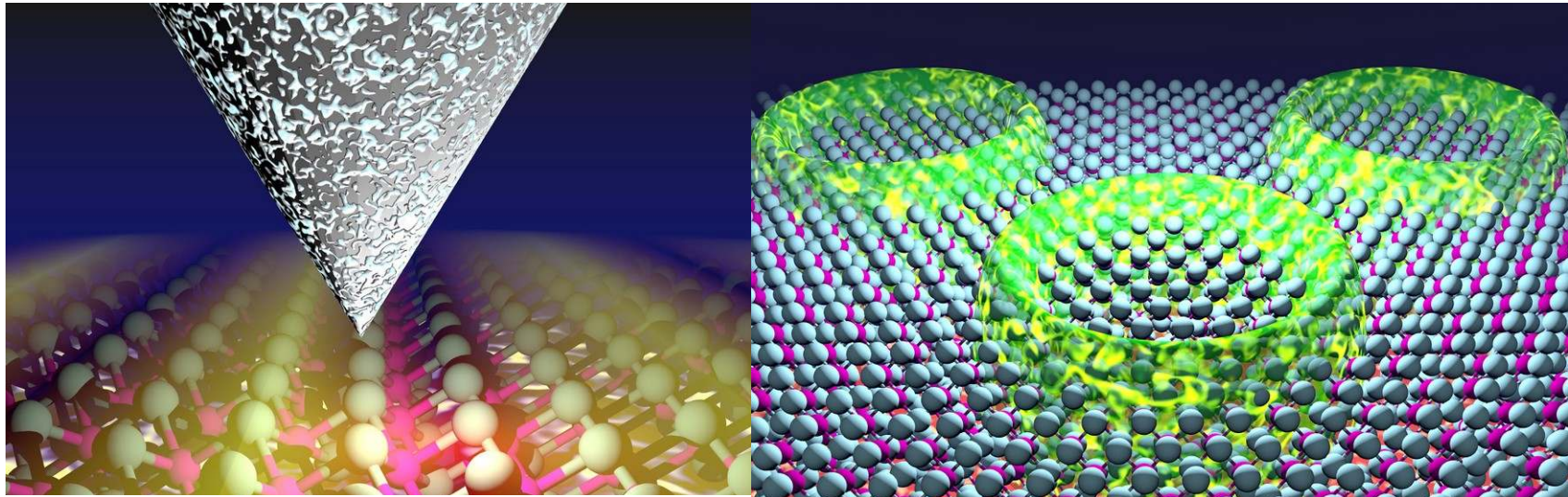


Artificial van der Waals multiferroics with twisted two-dimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



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

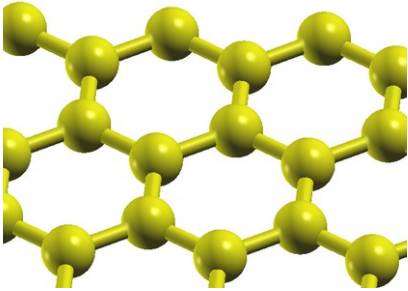
2D Materials 10 (2), 025026 (2023)

NT24, Fundamental properties symposia, MIT, Boston, June 27th 2024

The world of two-dimensional materials

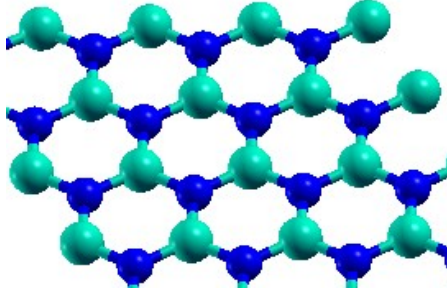
Semimetal

Graphene



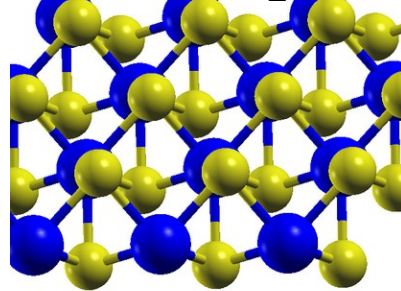
Insulator

BN



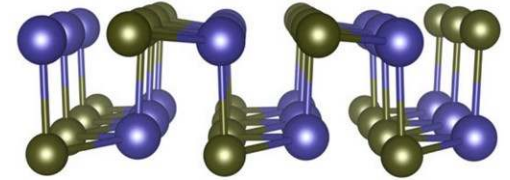
Superconductor

NbSe₂



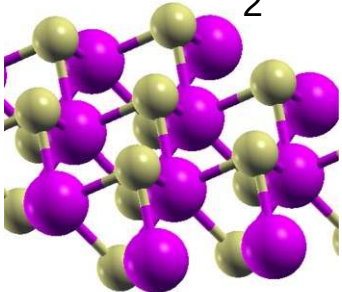
Ferroelectric

SnTe



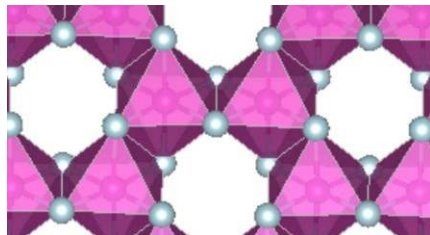
Semiconductor

WSe₂



Ferromagnet

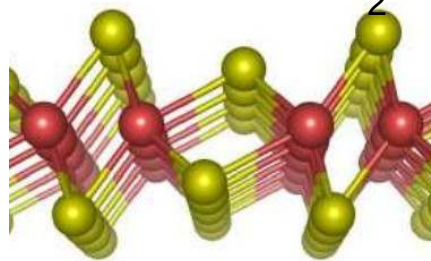
CrI₃



Quantum spin

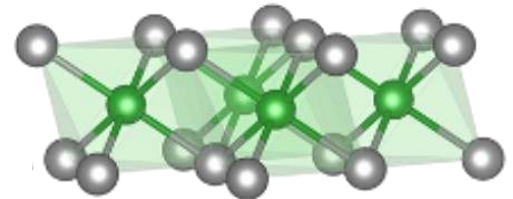
Hall insulator

1T'-WTe₂



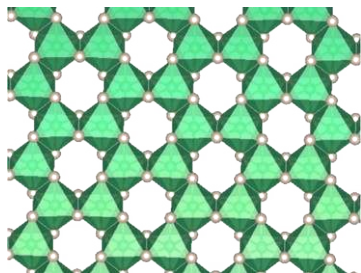
Multiferroic

NiI₂



Van der Waals magnetic materials

**Ferromagnet,
antiferromagnets**

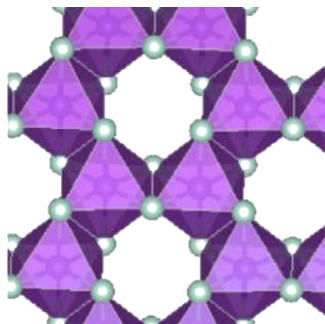


CrI_3 , CrCl_3 , CrBr_3

Nature 546, 270–273 (2017)

Break time-reversal

**(proximal)
Quantum
spin-liquids**

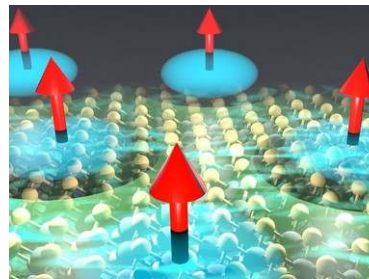


RuCl_3 , 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Do not break time-reversal

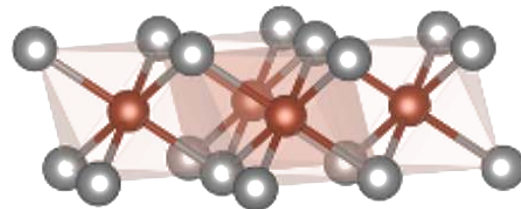
**Heavy-fermion
Kondo insulators**



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

Nature 599, 582–586 (2021)

Multiferroics



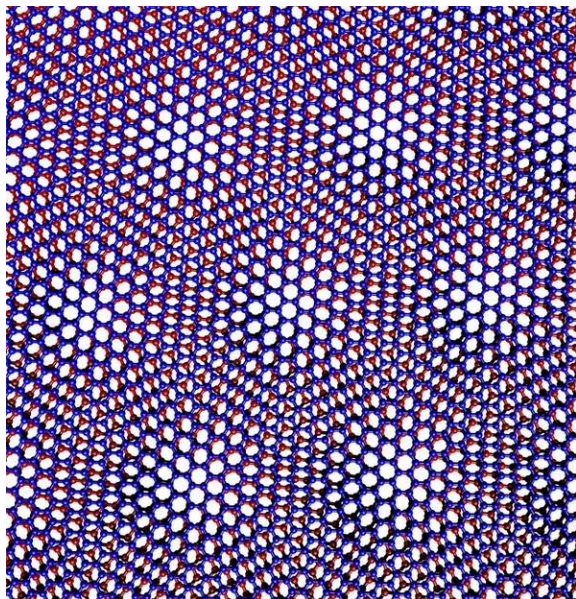
NiI_2

Nature 602, 601–605 (2022)

Break time-reversal
and inversion symmetry

The phases of twisted bilayer graphene

Twisted bilayer graphene

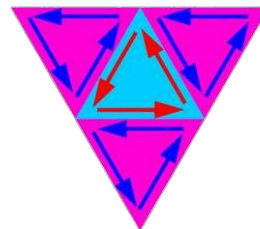


Superconductivity



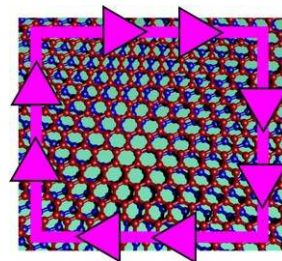
Nature 556, 43–50 (2018)

Topological networks



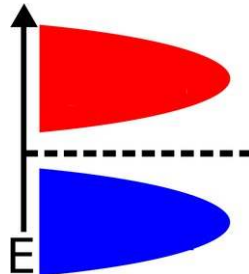
Nano Lett. 18, 11, 6725–6730 (2018)

Chern insulators



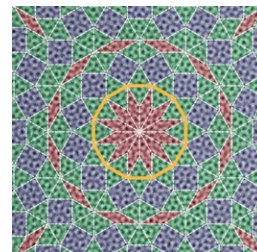
Science 365, 605–608 (2019)

Correlated insulators



Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782–786 (2018)

Proximal fractional Chern insulators

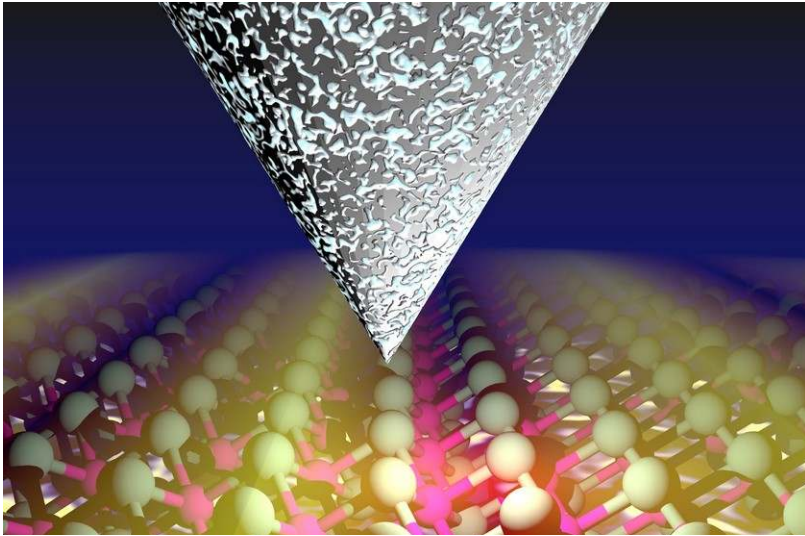


Nature 600, 439–443 (2021)

Which new phenomena could we get by twisting magnetic materials?

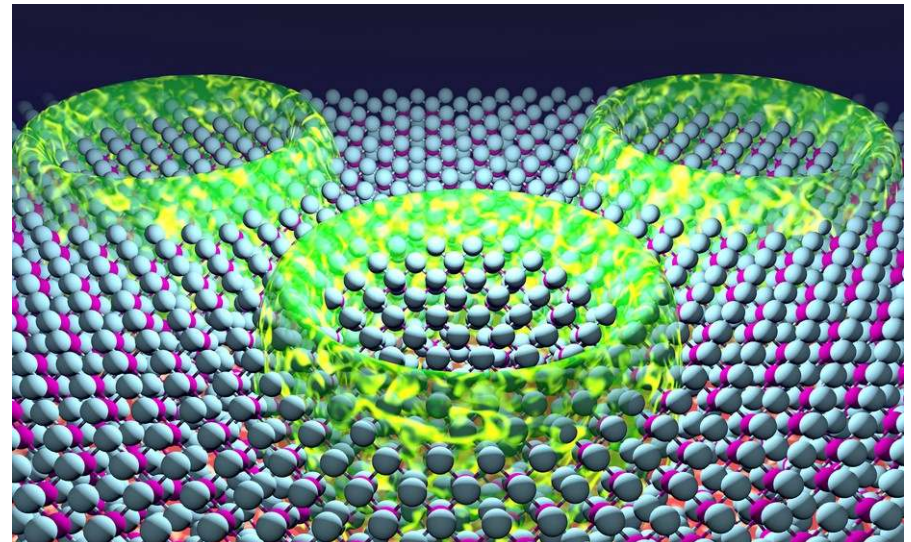
Today's plan

Origin and imaging of 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. Vaño



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

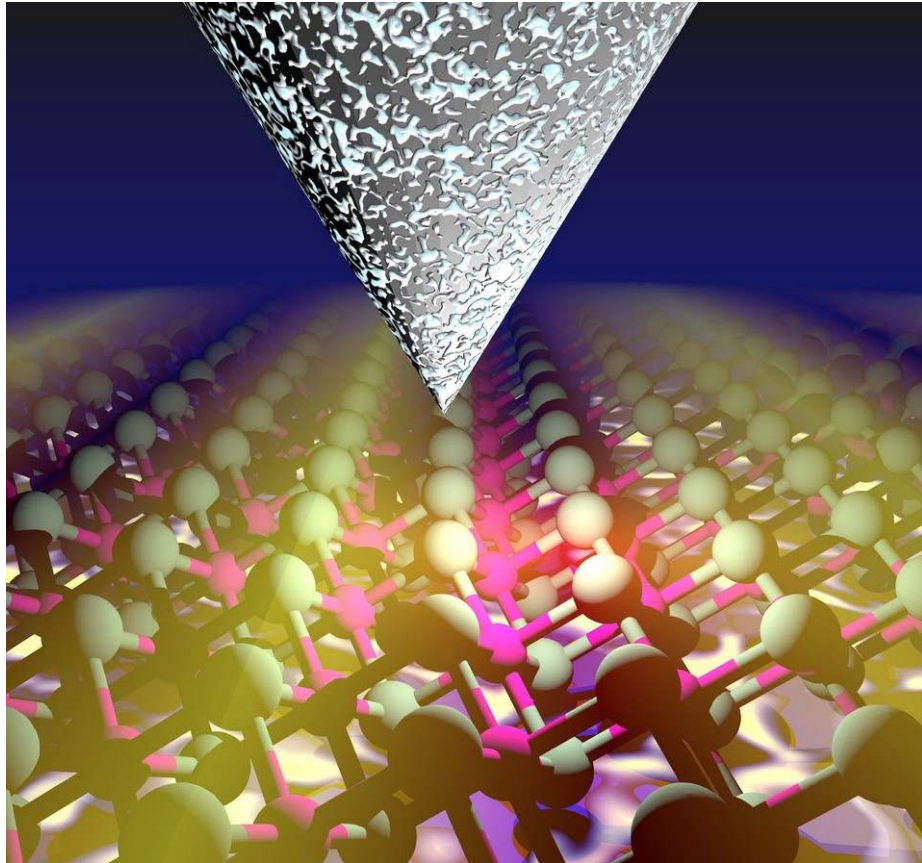
(theory)

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



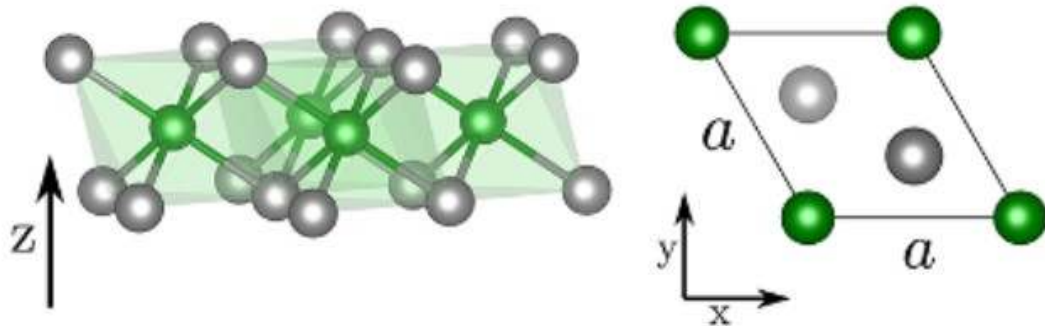
P. Liljeroth



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

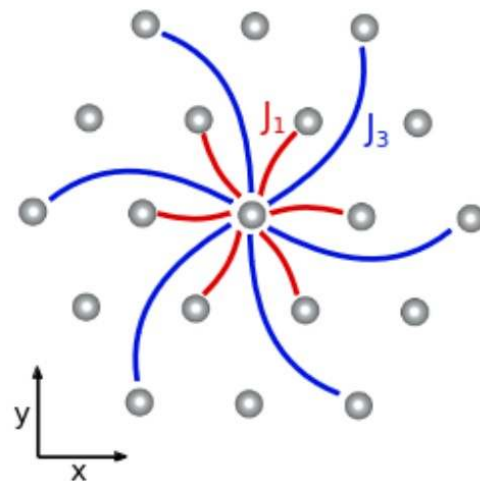
Monolayer NiI_2

Structure

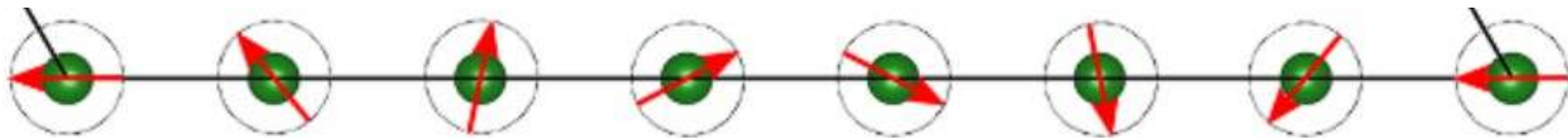


Nature 602, 601–605 (2022)

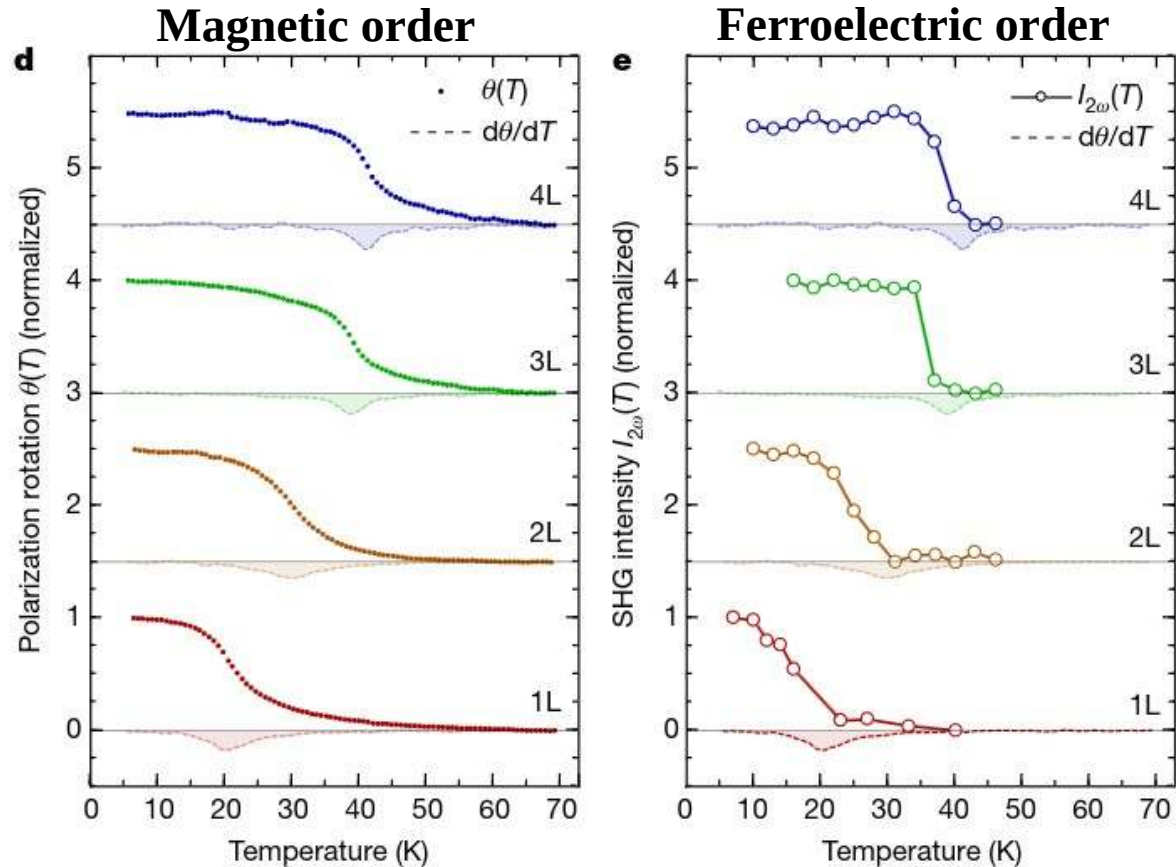
Frustrated magnetic
Heisenberg model



Magnetic structure



Multiferroicity in monolayer NiI_2



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

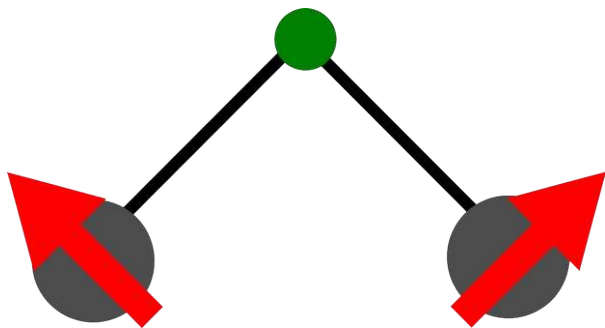
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

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

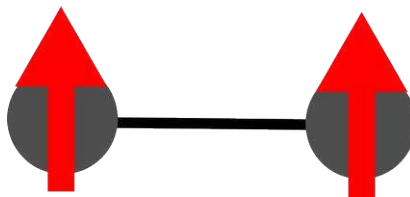
Antisymmetric exchange

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



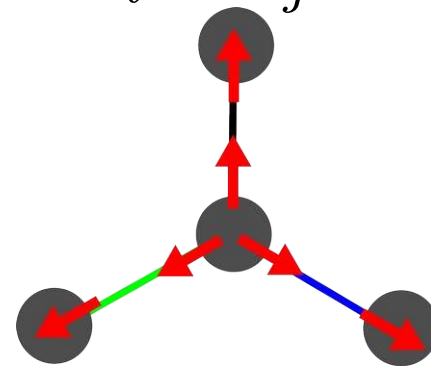
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

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



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

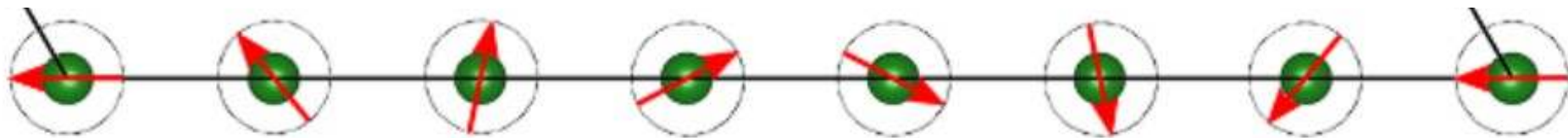
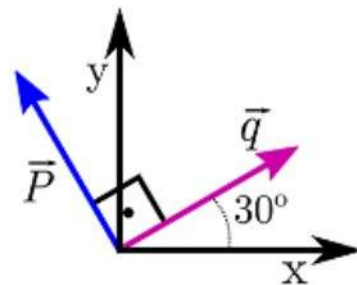
In NiI_2 , the ferroelectricity is created by inverse antisymmetric exchange

The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

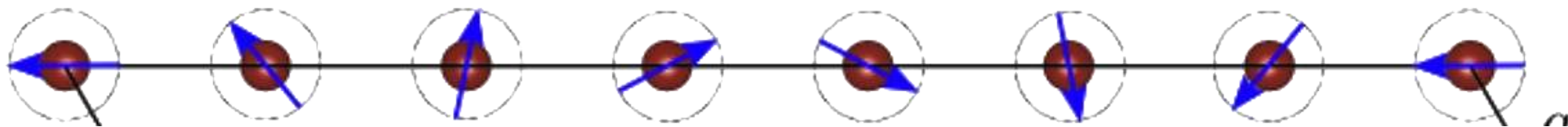
$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$$

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



The emergence of non-collinear order drives ferroelectricity

The origin of ferroelectricity in NiI_2



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

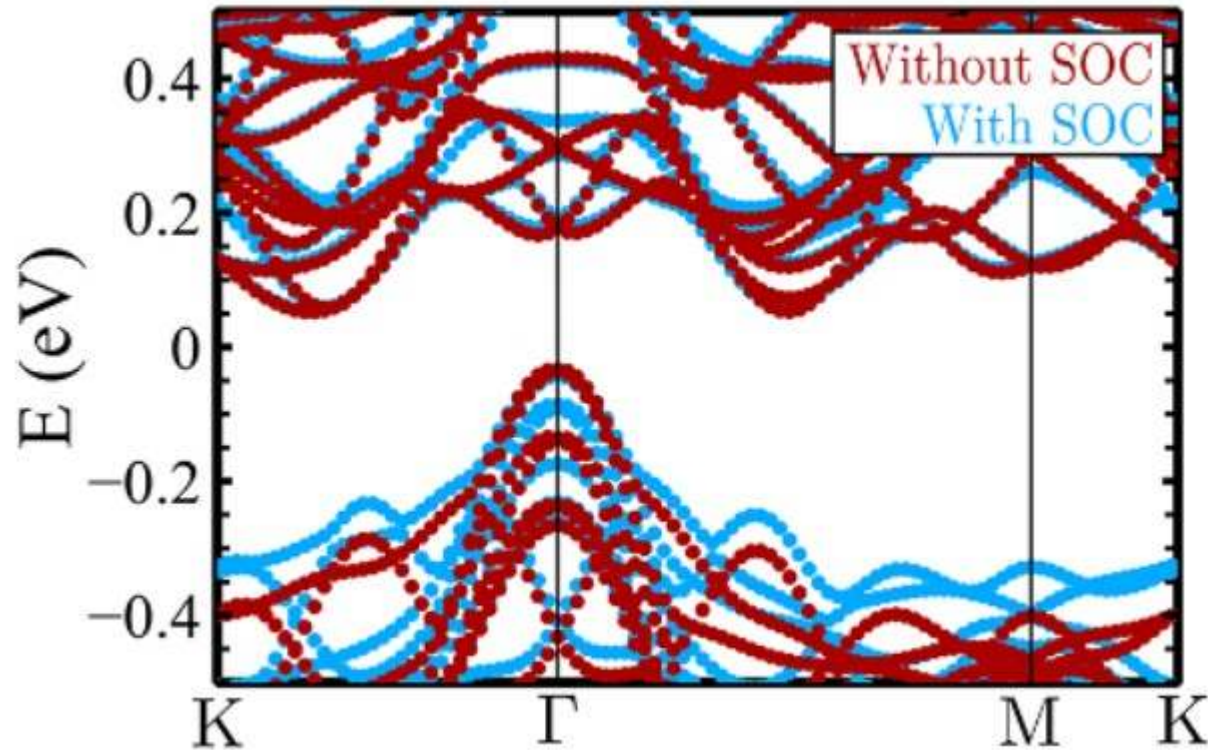
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

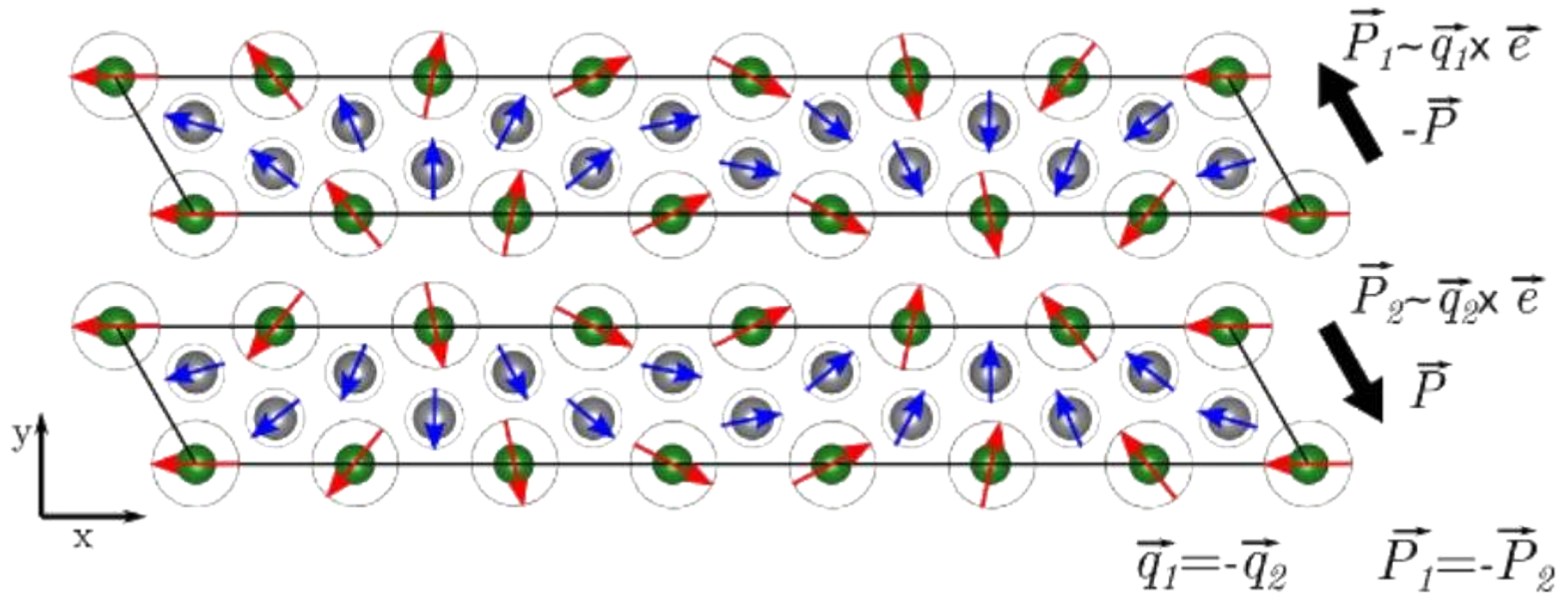
The impact of spin-orbit coupling in the electronic structure



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

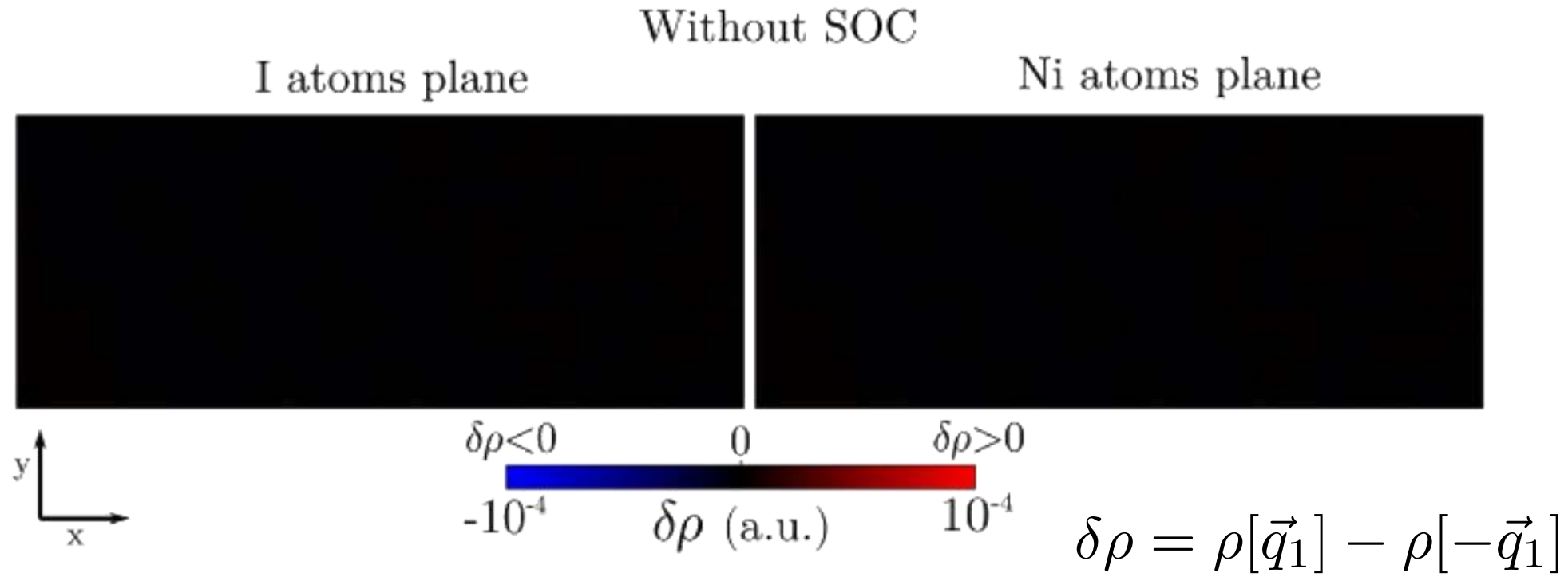
Ferroelectric correction to the electronic density



We will examine the ferroelectric density correction

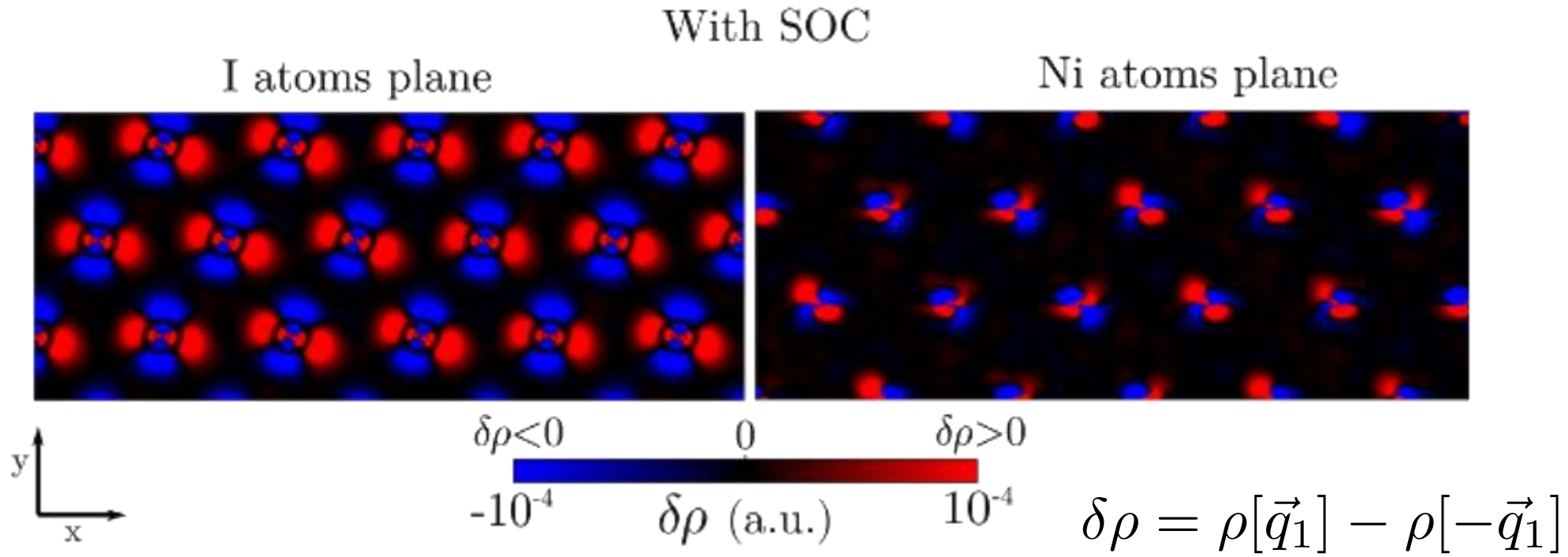
$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

Ferroelectric correction to the electronic density



In the absence of SOC, no ferroelectric density appears

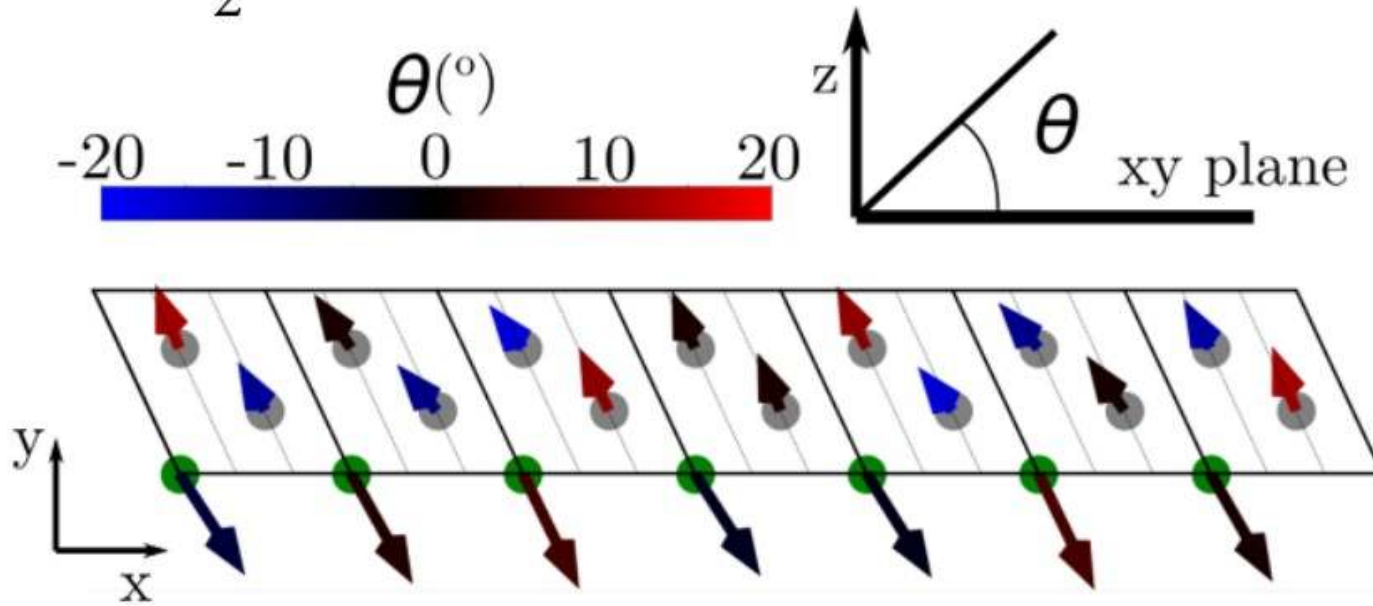
Ferroelectric correction to the electronic density



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

The ferroelectric force

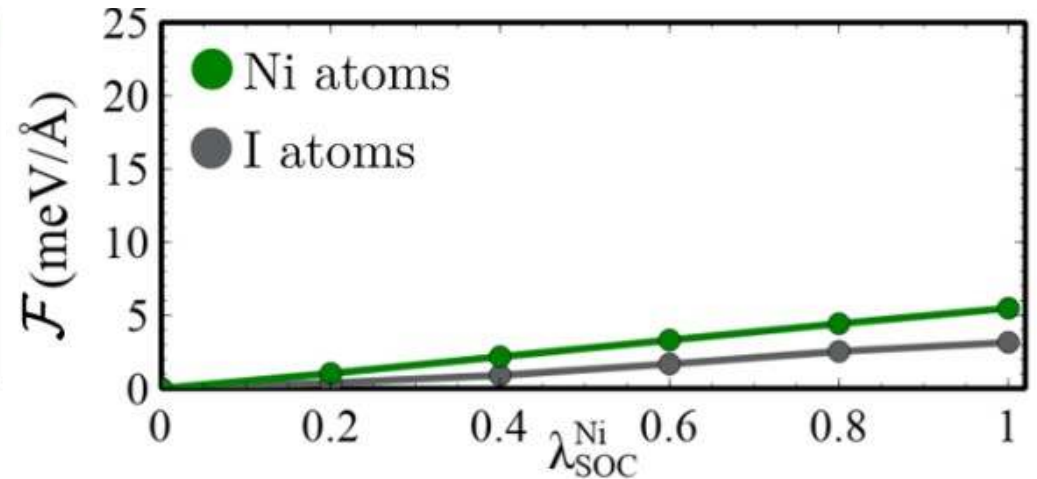
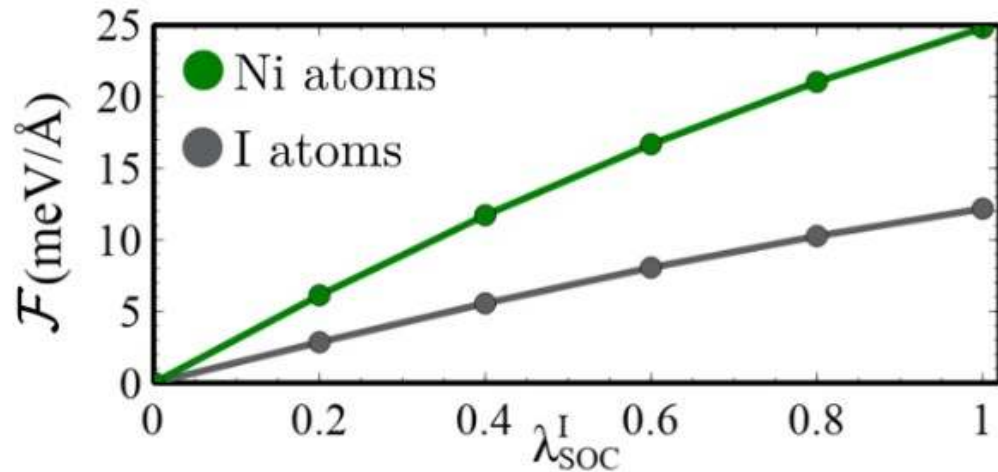
$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$



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

The ferroelectric force

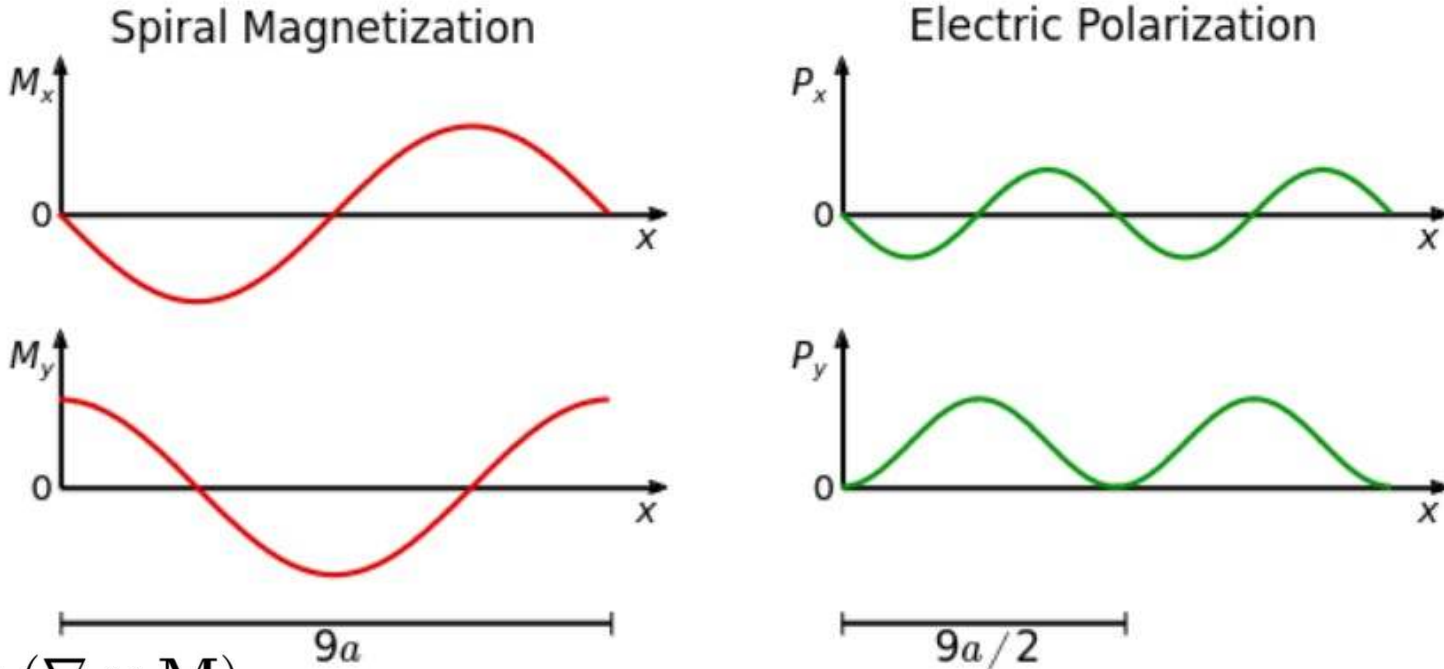
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\text{SOC}}^{\text{Ni}} \mathcal{H}_{\text{SOC}}^{\text{Ni}} + \lambda_{\text{SOC}}^{\text{I}} \mathcal{H}_{\text{SOC}}^{\text{I}},$$



The dominant contribution to the ferroelectric force comes from iodine

Modulated multiferroicity in monolayer NiI_2

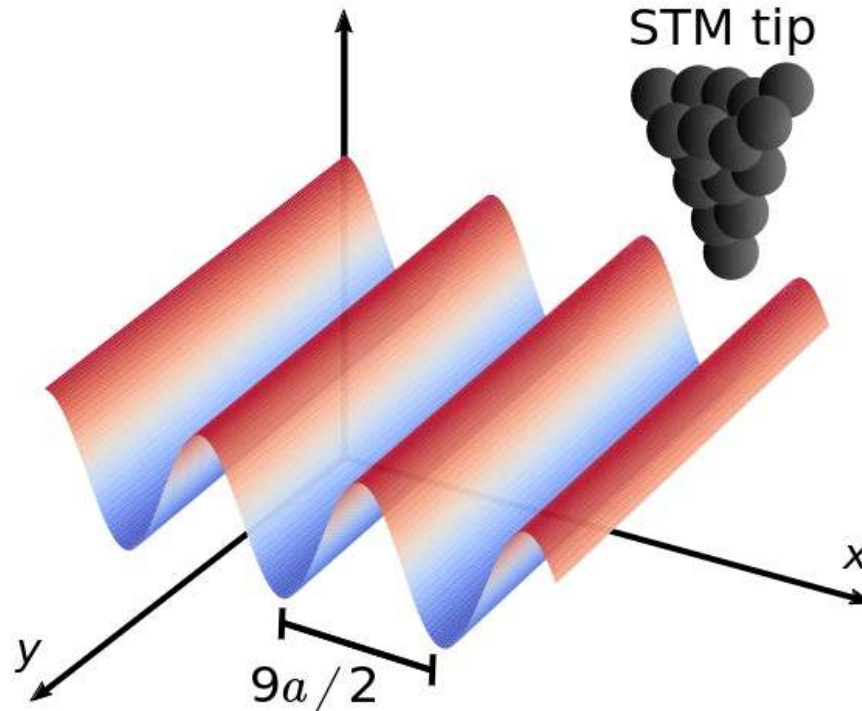
The ferroelectric order has actually a modulation in real space



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

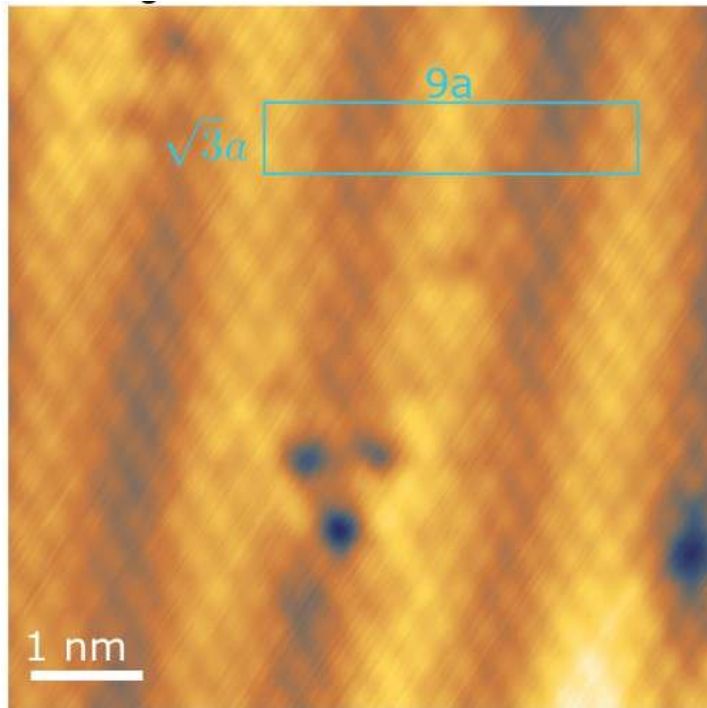
Visualizing multiferroicity in monolayer NiI_2

Modulated electrostatic potential

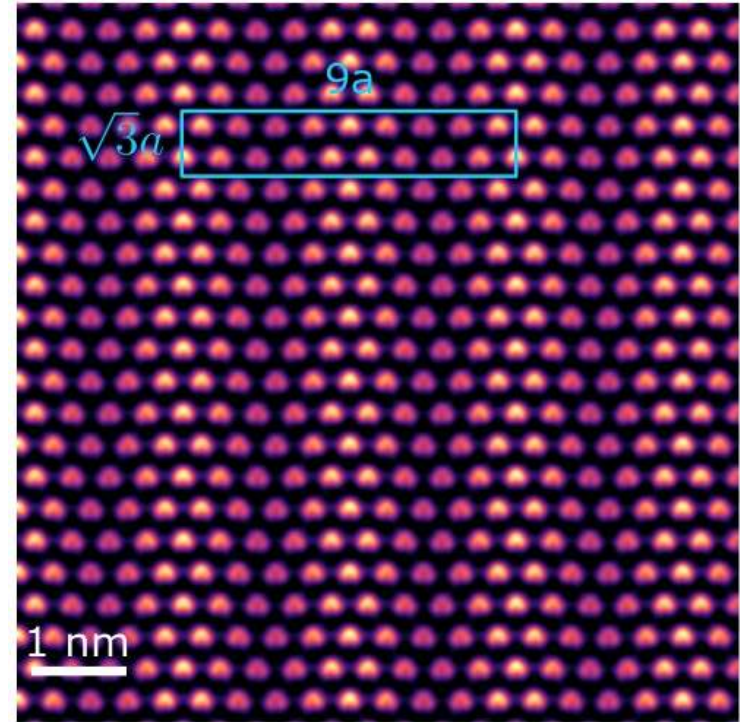


Visualizing multiferroicity in monolayer NiI_2

Experiment



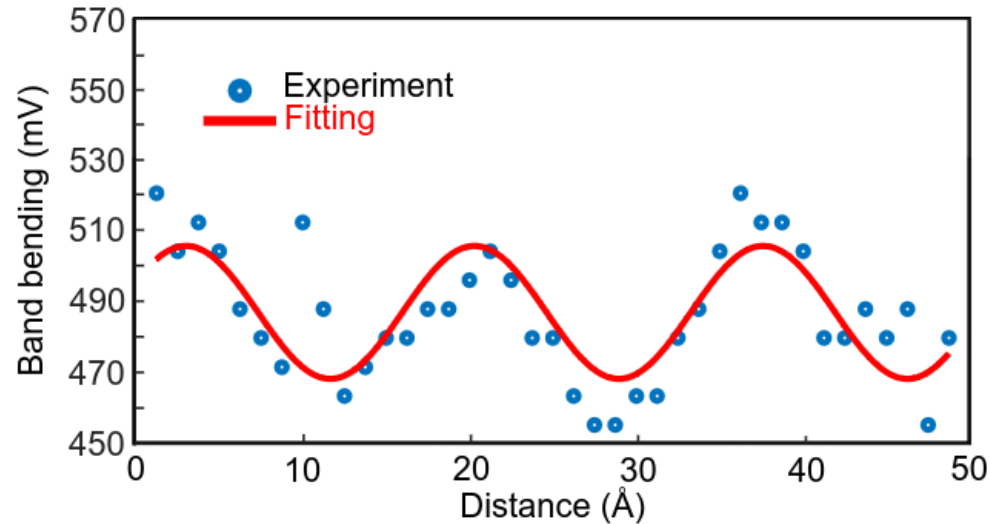
Theory



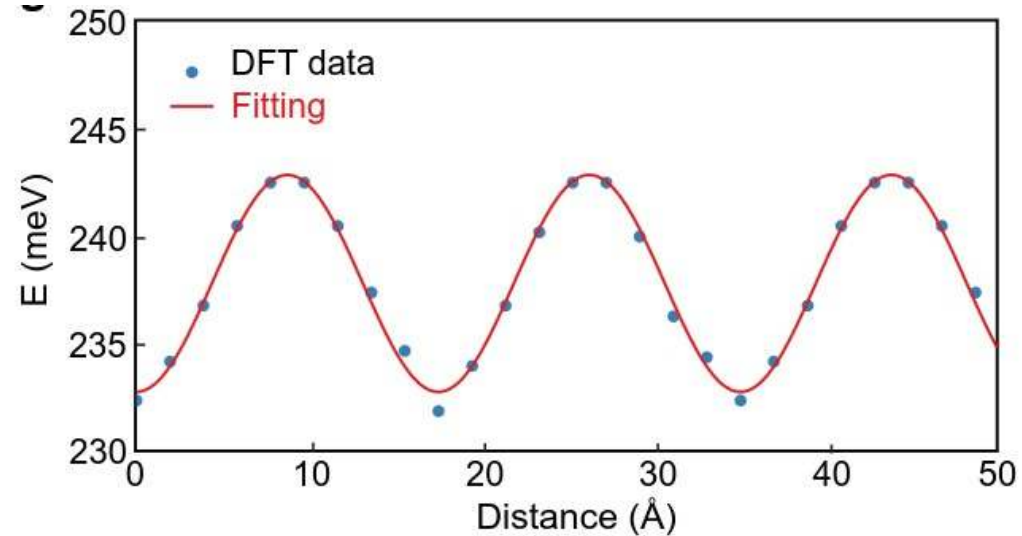
$$\Lambda_{\mathbf{q}}(-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

Ferroelectricity from modulated band offset

Band offset (experiment)

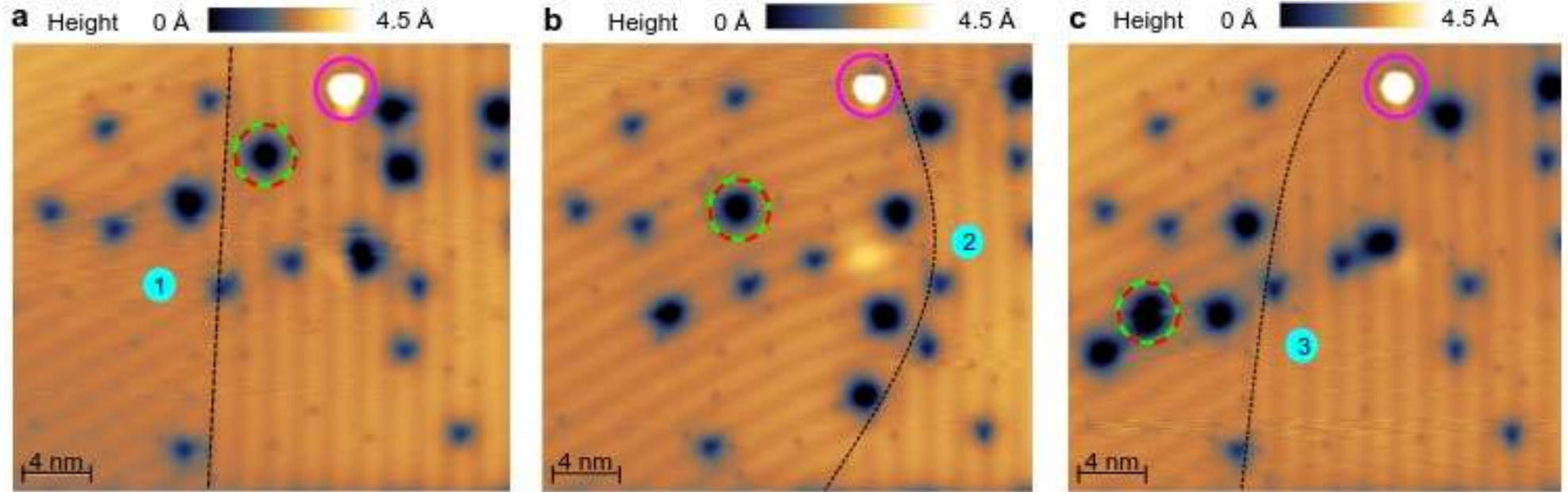


Band offset (theory)



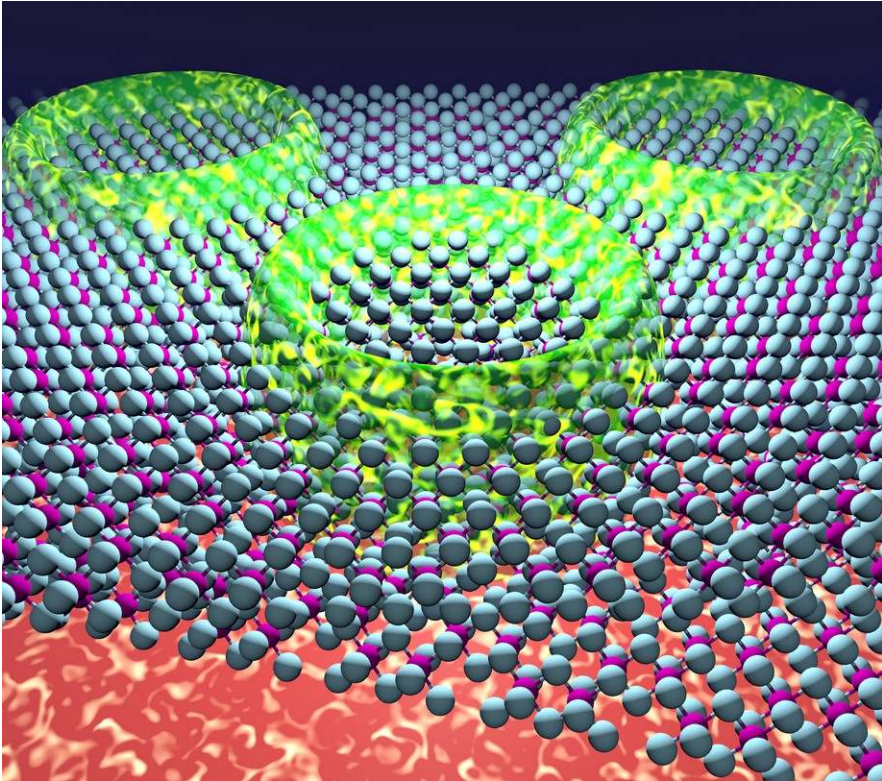
$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{M^2} = \Lambda q (-\sin(2\mathbf{q} \cdot \mathbf{r}), \sin^2(\mathbf{q} \cdot \mathbf{r}), 0)$$

Controlling multiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

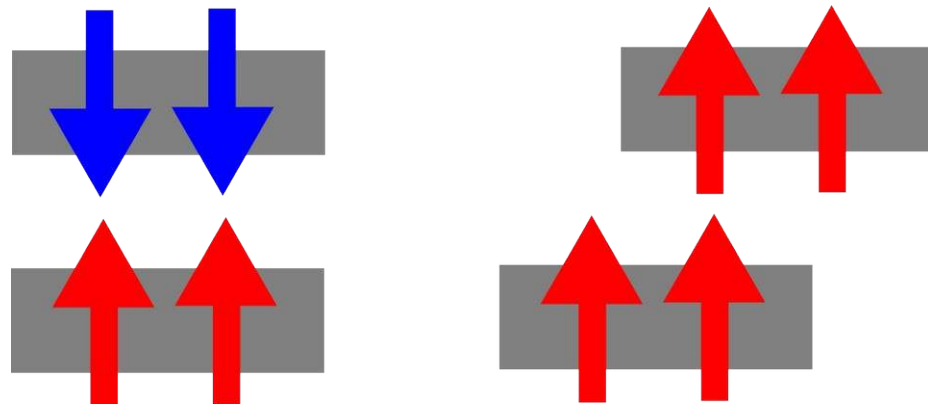
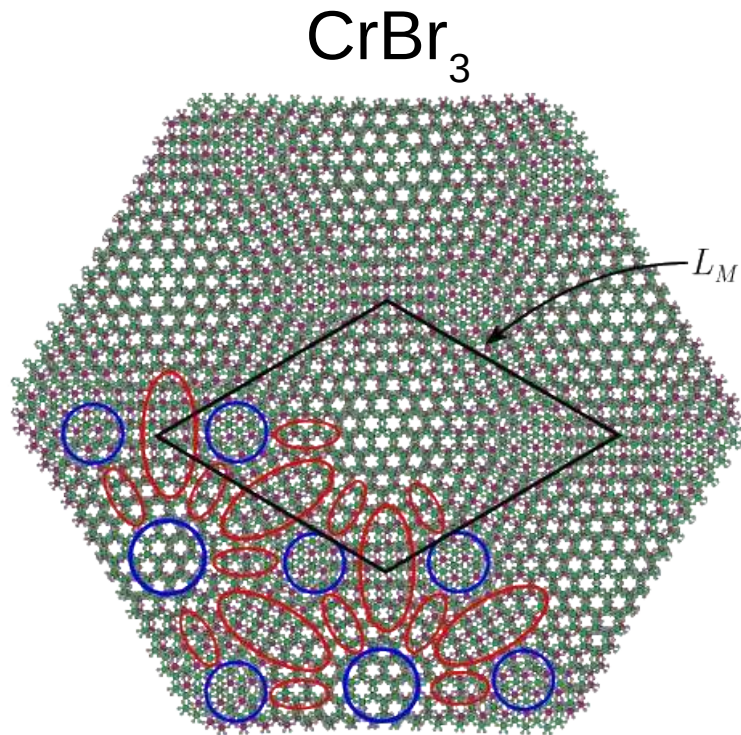
Engineering artificial multiferroics with twisted van der Waals materials



A. Fumega



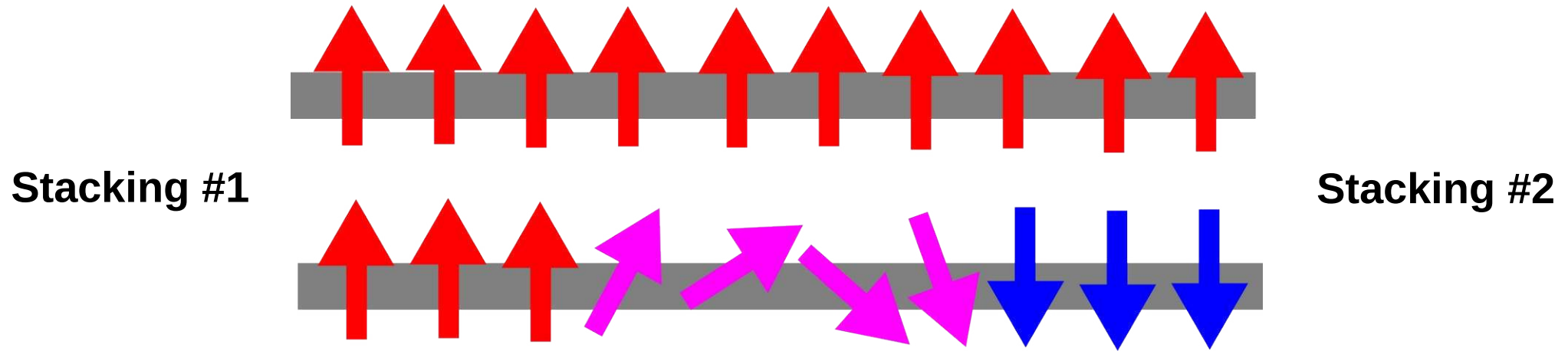
Twisted 2D trihalides



The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Mixed FE/AF domains

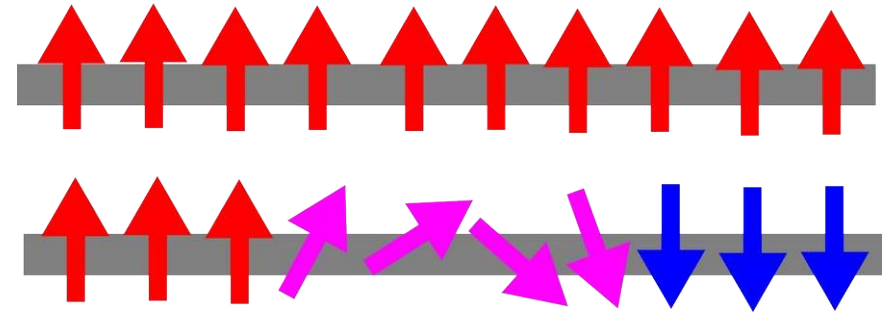
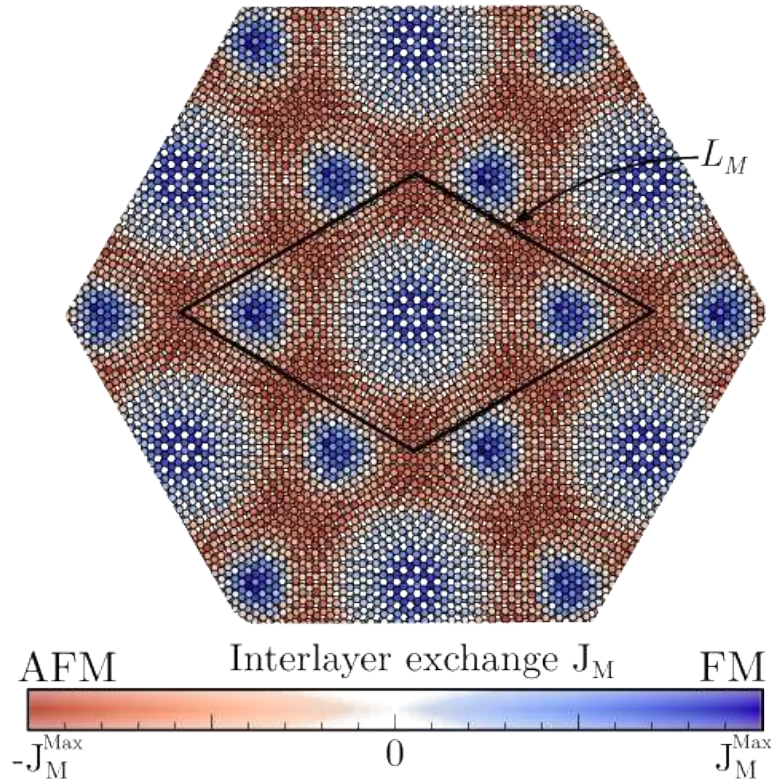
Nature Nanotechnology 17, 143–147 (2022)

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

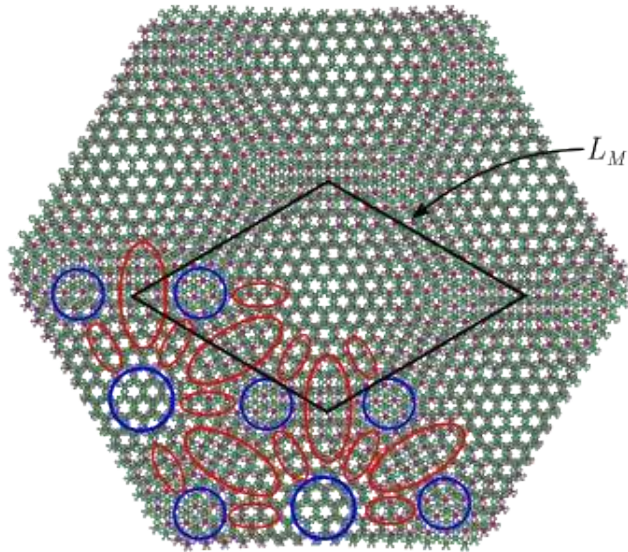
SOC driven

Spin rotation axis

Wavevector of the spin spiral

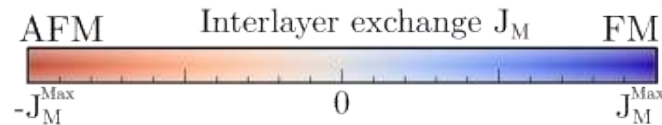
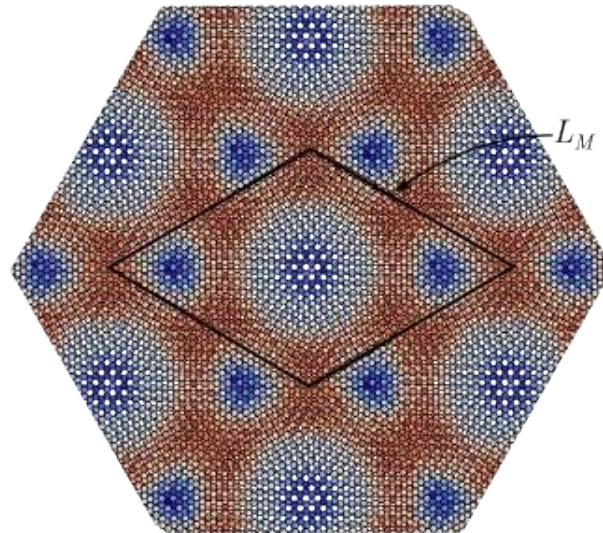
Stacking, magnetic order and electric polarization

Structure

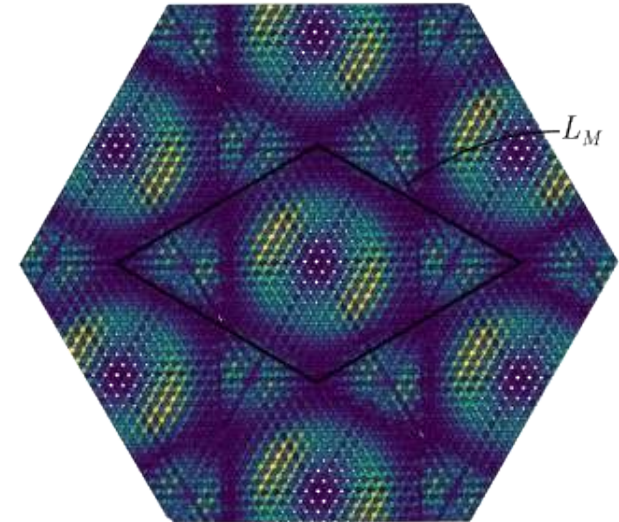


Bilayer Stacking $\left\{ \begin{array}{l} \text{(M)} \text{ Monoclinic} \\ \text{(R)} \text{ Rhombohedral} \end{array} \right.$

Magnetic order



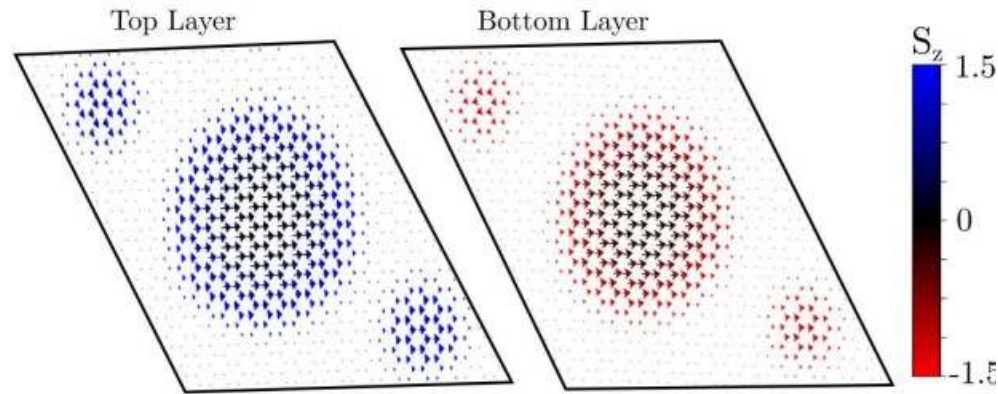
Ferroelectric order



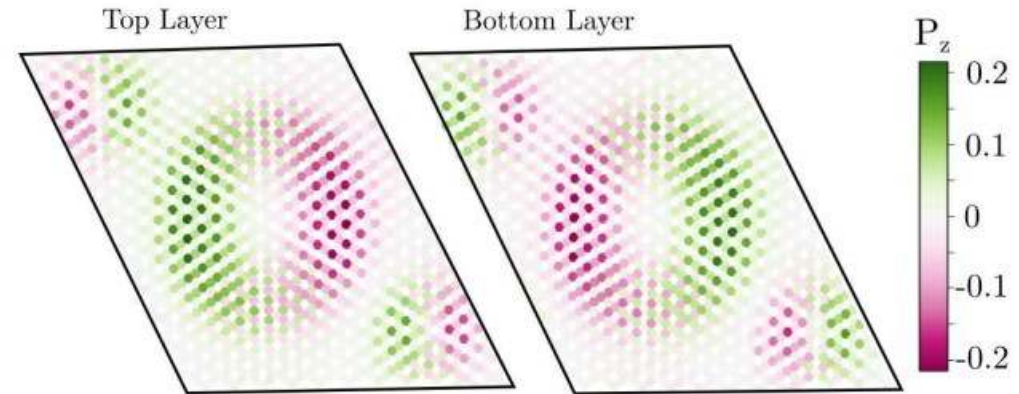
Stacking, magnetic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

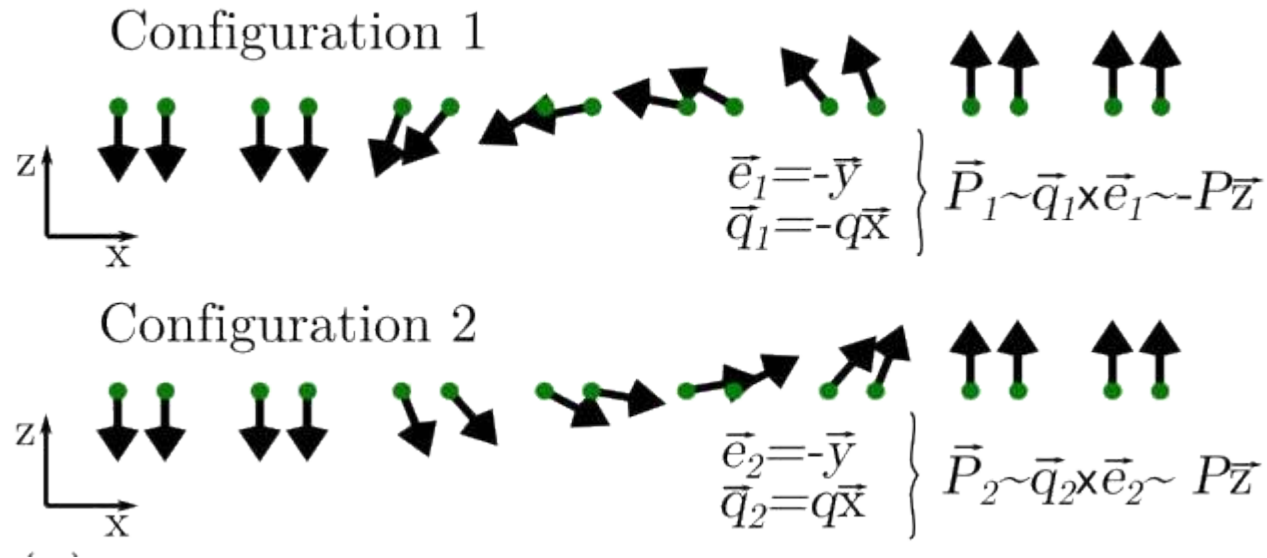


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



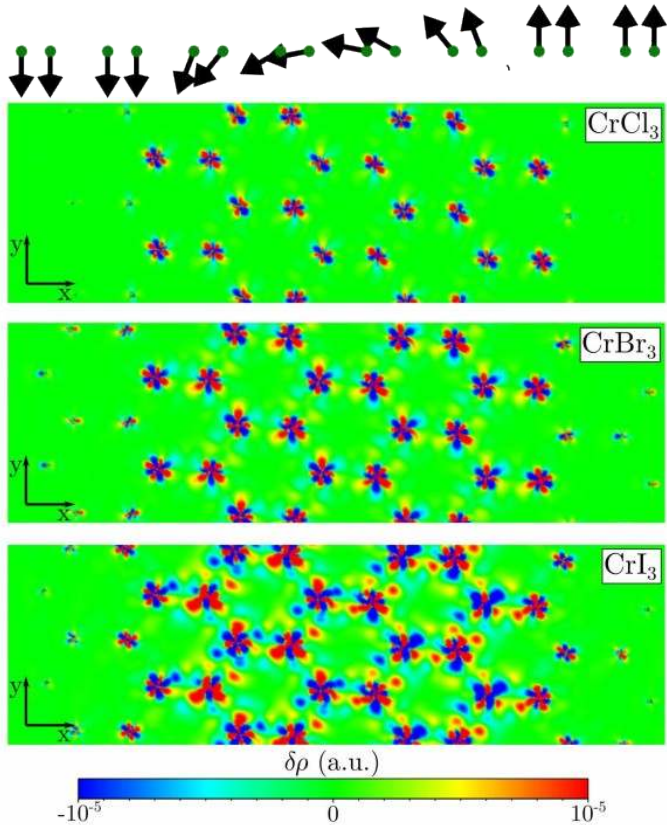
Ferroelectric electronic density

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

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

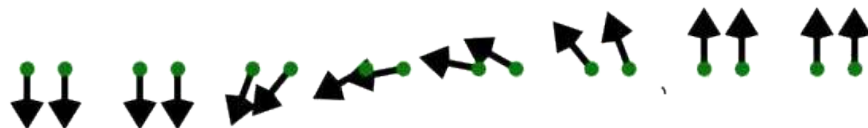
$$\delta\rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

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

It becomes stronger the heavier the halide

The ferroelectric force

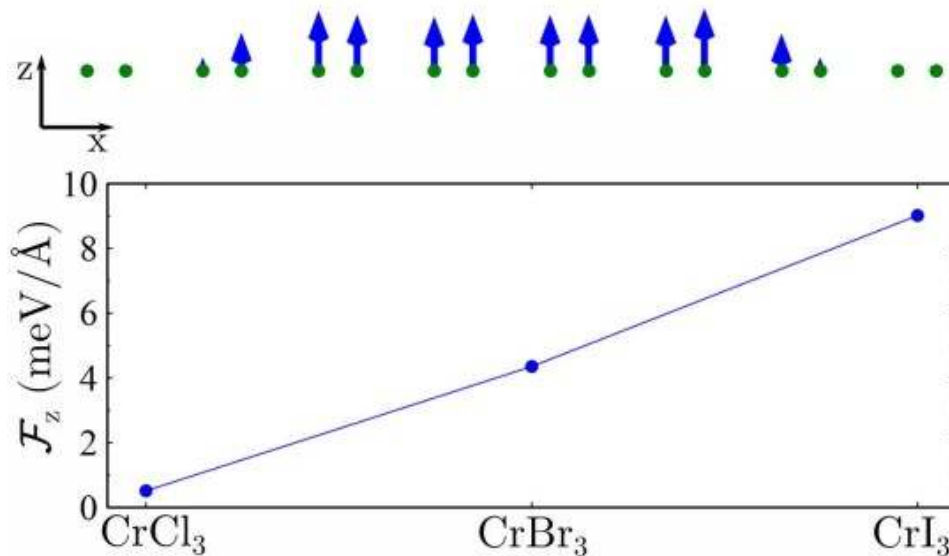
Magnetic configuration



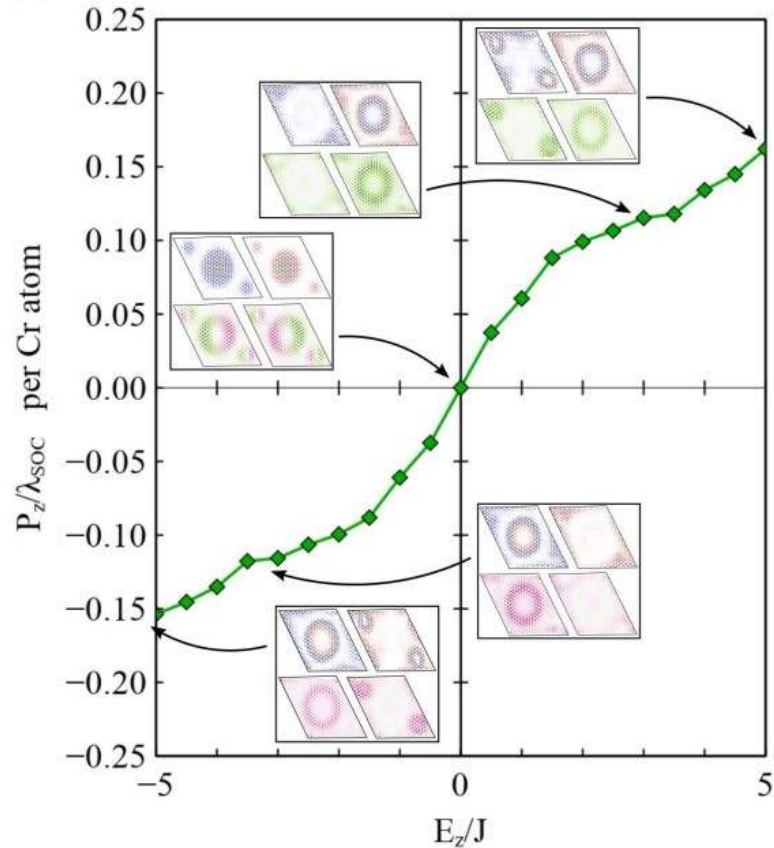
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



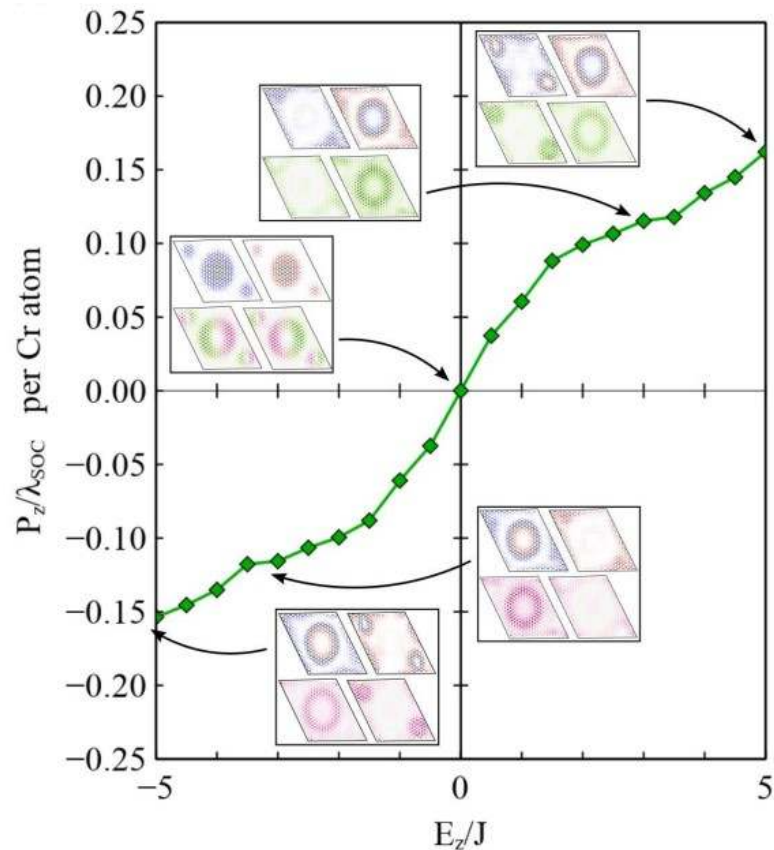
Electrically controllable skyrmions in twisted magnets



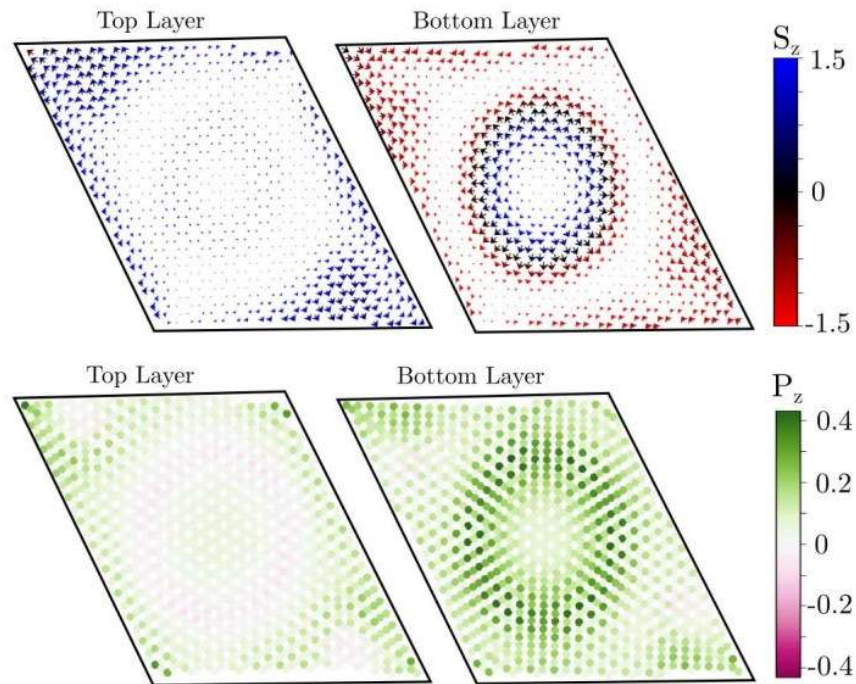
The electric polarization directly couples to an out-of plane electric field

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

Electrically controllable skyrmions in twisted magnets

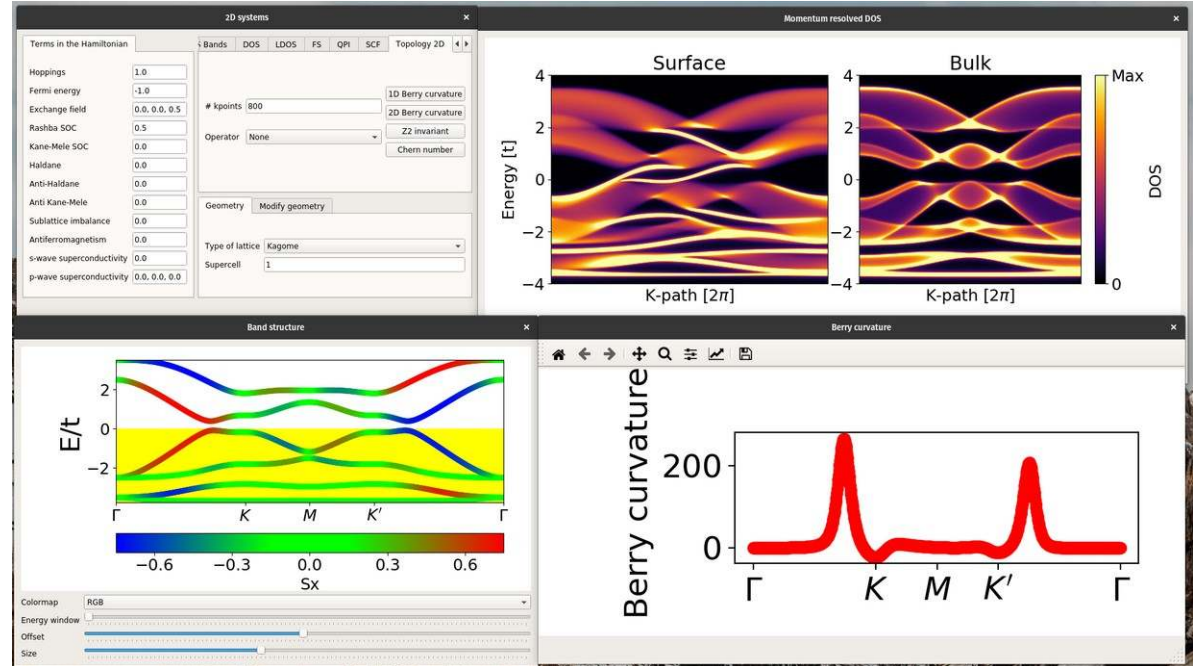
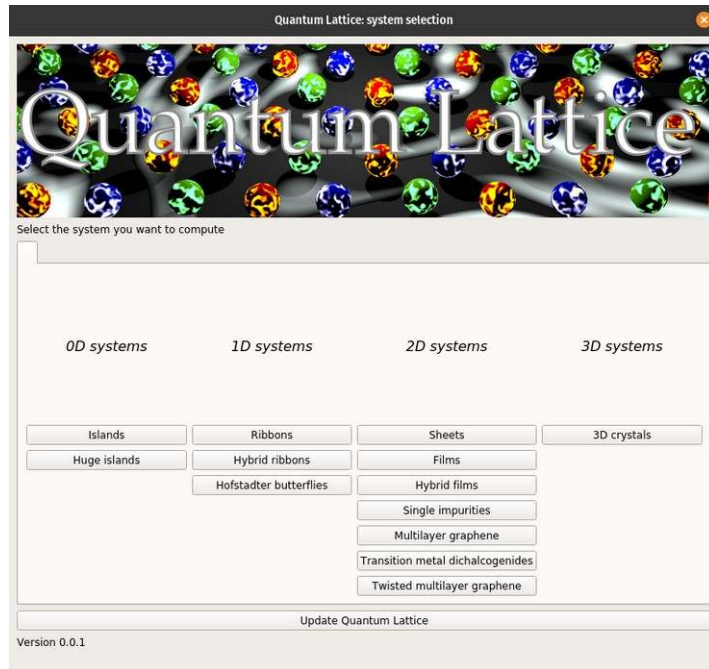


Skyrmions at finite interlayer bias



Open-source software for
van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

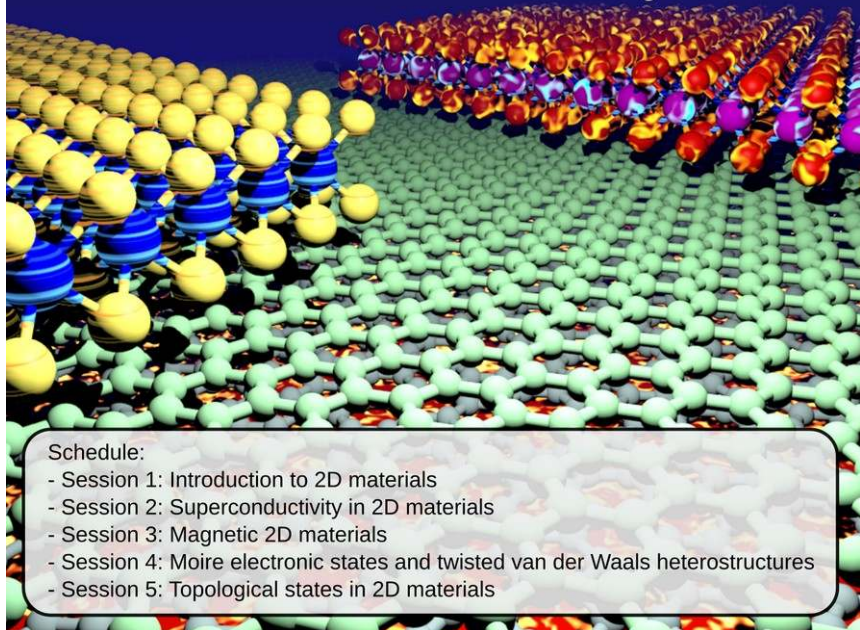


Quantum Lattice: open source interactive interface for tight binding modeling

<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

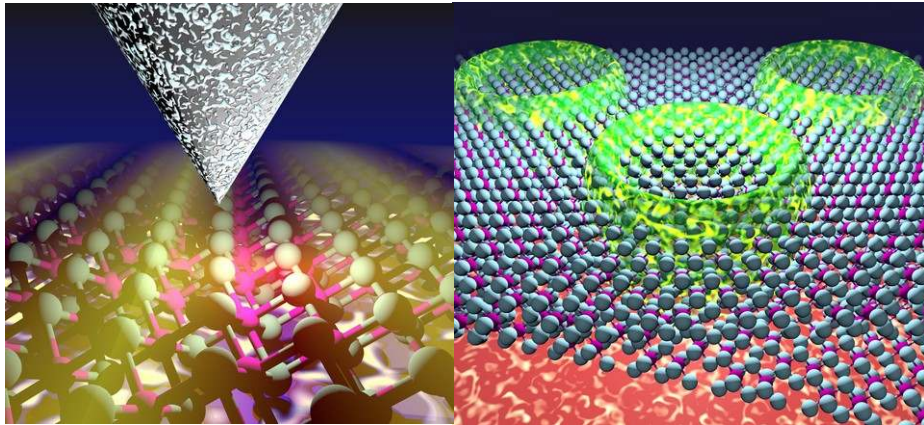


<https://www.youtube.com/channel/UCgnB-4CcqRQnvTi7P0cUakQ>

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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)

Thank you

Funding from

