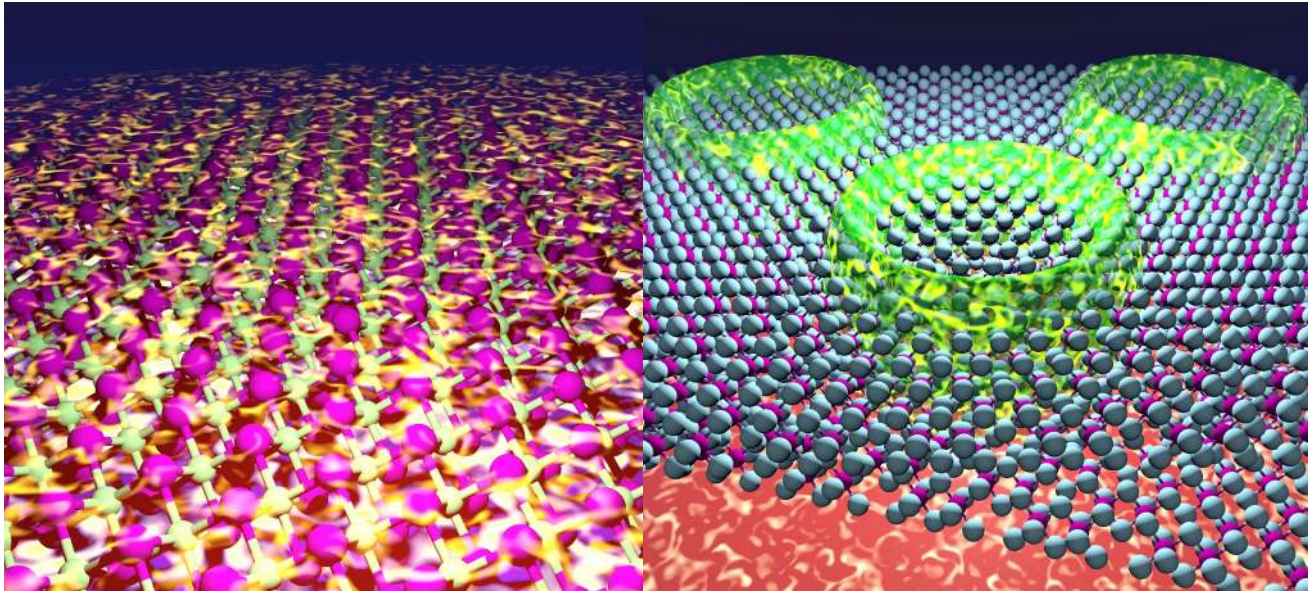
Van der Waals multiferroics

Jose Lado

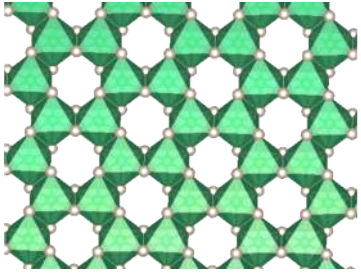
Department of Applied Physics, Aalto University, Finland



Quantum Magnetic Materials, Lorentz Center, Leiden, October 17th 2023

Van der Waals magnetic materials

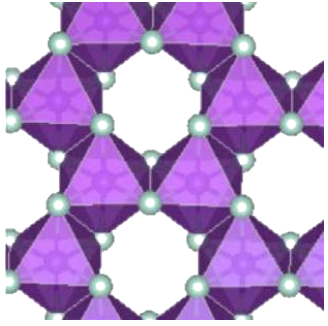
Ferromagnet, antiferromagnets



CrI_3, $CrCl_3$, $CrBr_3$

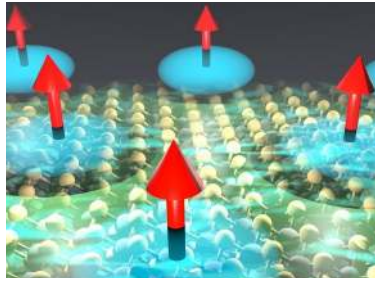
Nature 546, 270–273 (2017)

(proximal) Quantum spin-liquids



$RuCl_3$, 1T-TaS_2

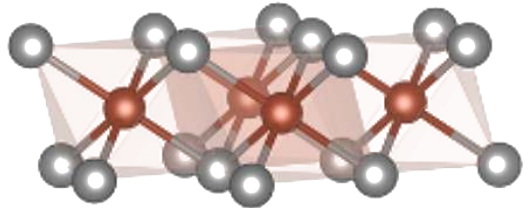
Heavy-fermion Kondo insulators



1T-TaS_2/1H-TaS_2

Nature 599, 582–586 (2021)

Multiferroics



NiI_2

Nature 602, 601–605 (2022)

Nature Physics 17, 1154–1161 (2021)

Break time-reversal

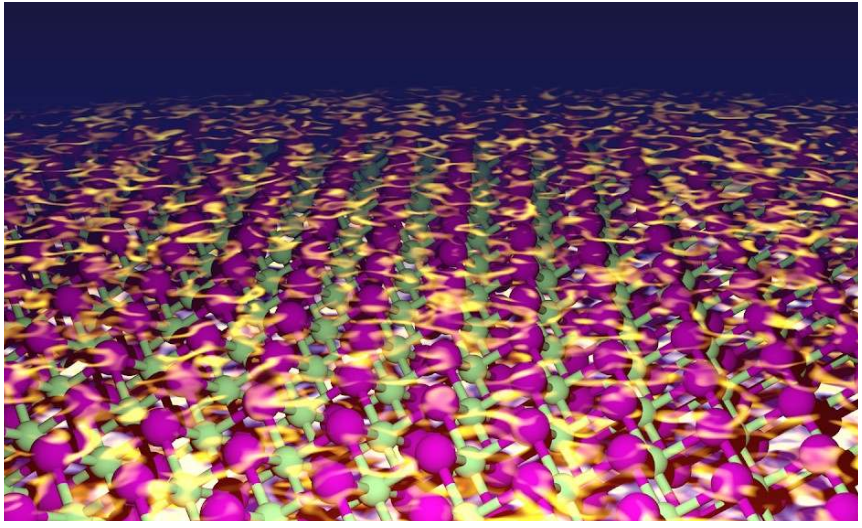
Do not break time-reversal

Break time-reversal and inversion symmetry

Two strategies for multiferroicity

Intrinsic multiferroics, NiI_2

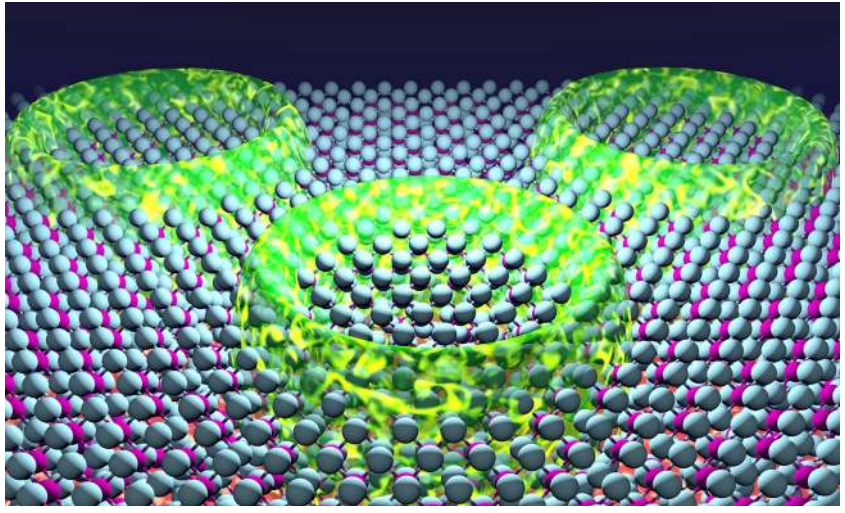
(theory and experiments)



Nature 602, 601–605 (2022)
2D Materials 9 (2), 025010 (2022)
arXiv:2309.11217 (2023)

Artificial multiferroics, twisted $CrBr_3$ bilayers

(theory, and potential from experiments)



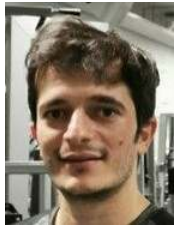
Nature Nano 17, 143–147 (2022)
ArXiv:2204.01636 (2022)
Nature Elec 6, 434–442 (2023)
2D Materials 10 (2), 025026 (2023)

Two strategies for multiferroicity

Intrinsic multiferroics, NiI_2
(theory and experiment)

V. Vaňo

M. Amini



A. Fumega

H. Gonzalez Herrero

S. Kezilebieke

P. Liljeroth

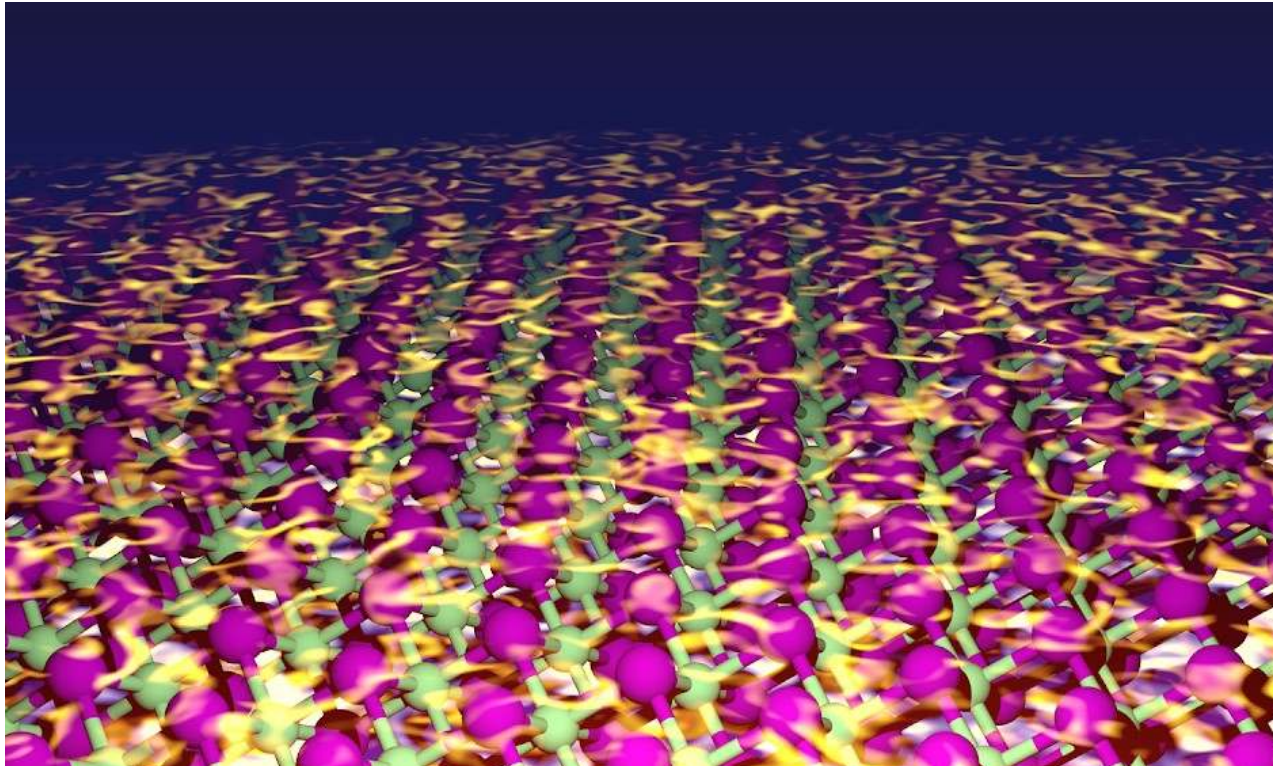
2D Materials 9 (2), 025010 (2022)
arXiv:2309.11217 (2023)

Artificial multiferroics, twisted $CrBr_3$ bilayers
(theory)

A. Fumega

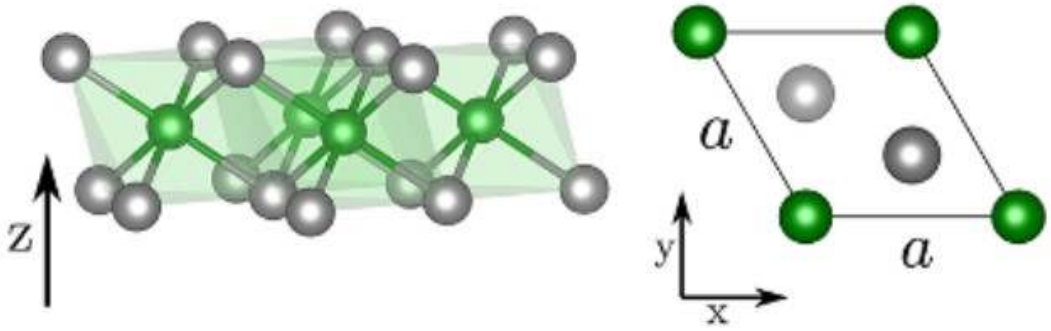
2D Materials 10 (2), 025026 (2023)

Intrinsic multiferroics, NiI_2



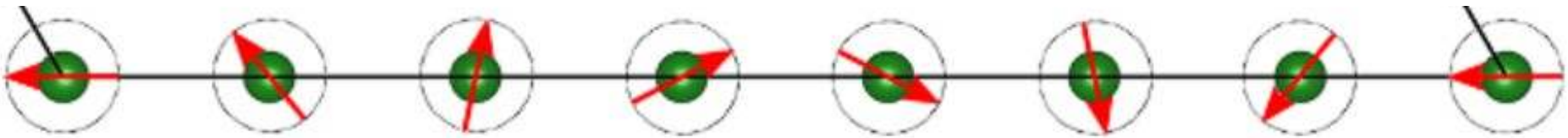
Monolayer NiI_2

Structure



Nature 602, 601–605 (2022)

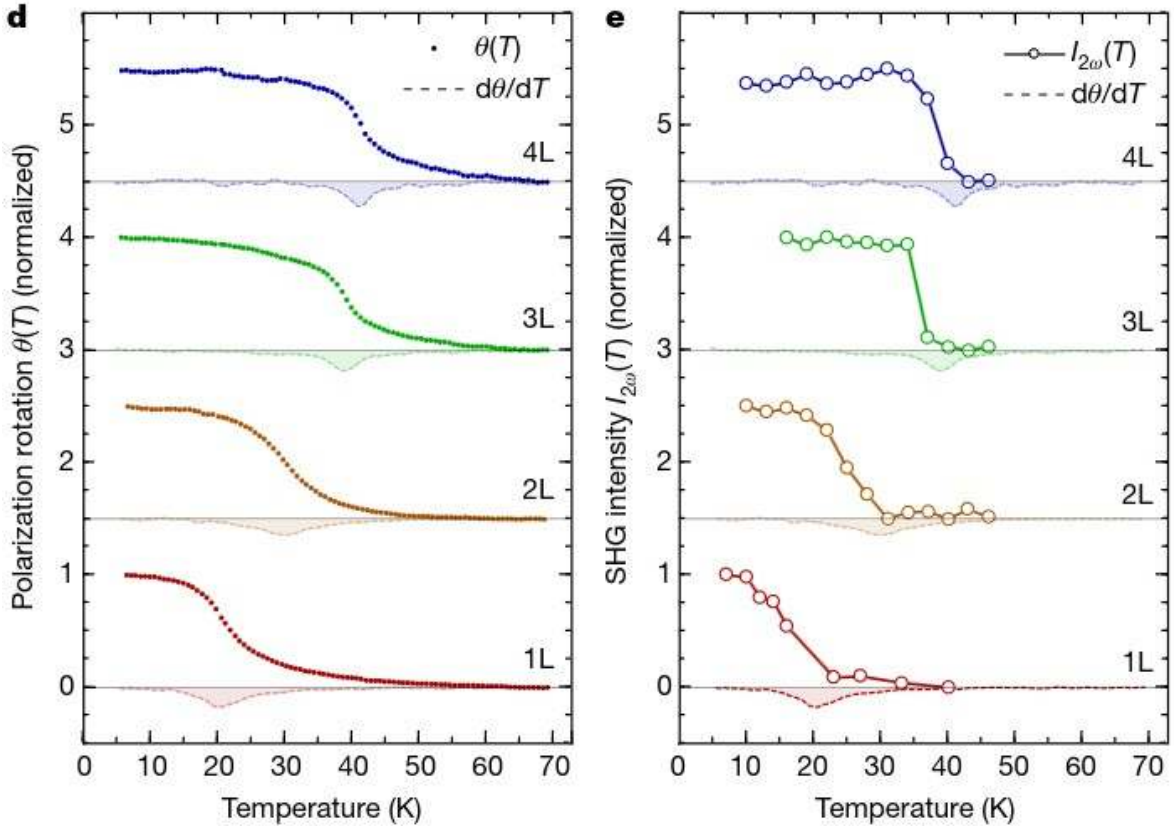
Magnetic structure



Multiferroicity in monolayer NiI_2

Magnetic order`

Ferroelectric order`



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

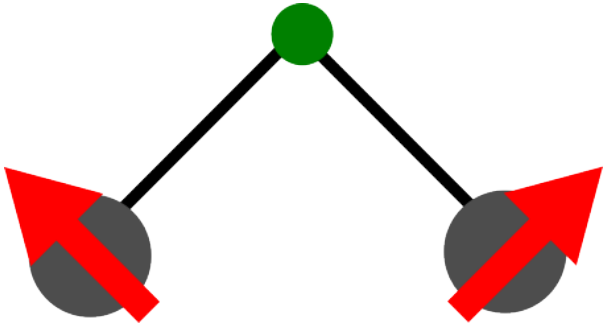
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

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

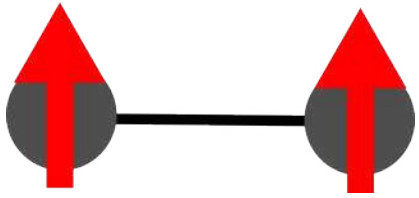
Antisymmetric exchange

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



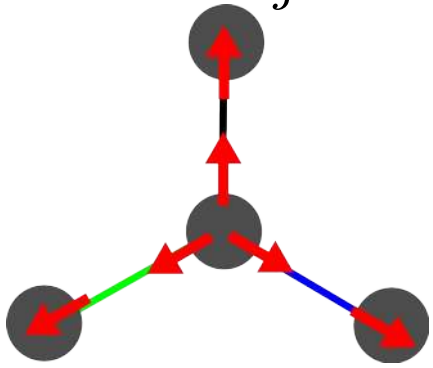
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

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



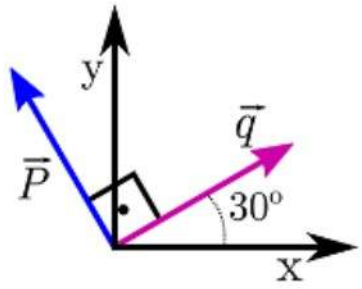
In NiI_2, the non-collinear order is promoted by isotropic exchange

In NiI_2, the ferroelectricity is created by inverse antisymmetric exchange

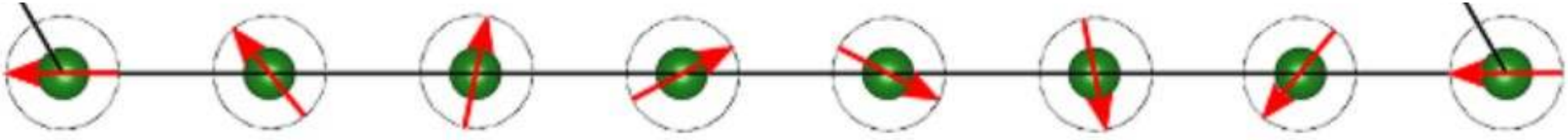
The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

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

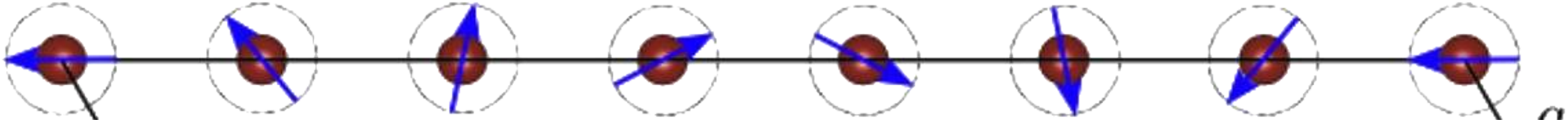


$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e}\cdot\mathbf{S})(\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}_0))}\mathbf{M}(\mathbf{r}_0)$$



The emergence of non-collinear order is driives ferroelectricity

The origin of ferroelectricity in NiI_2



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

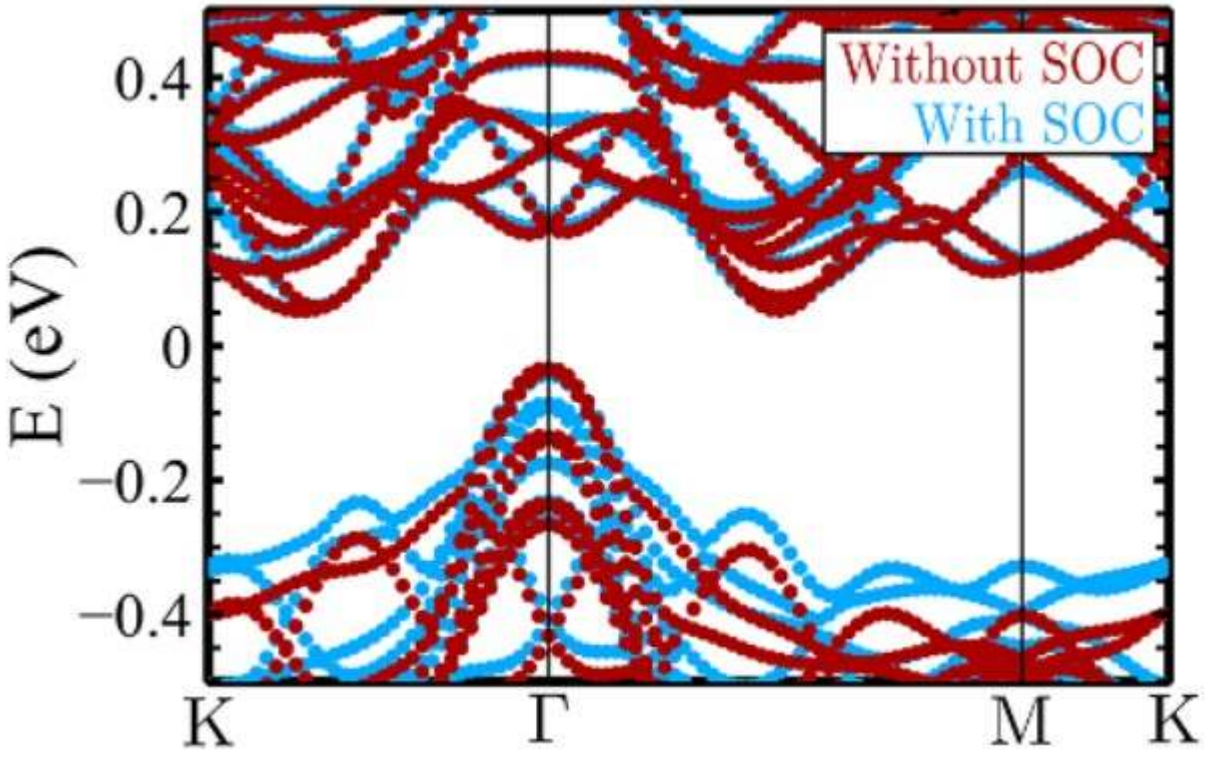
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

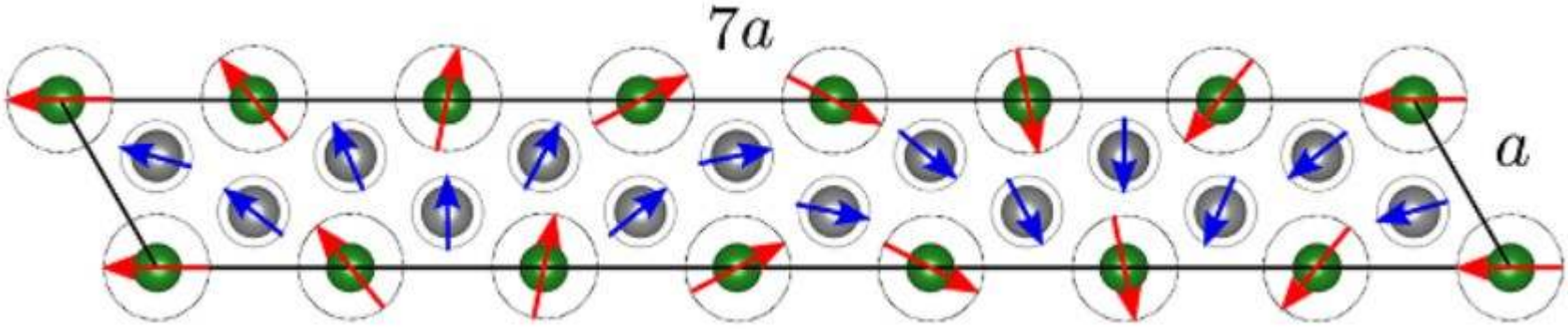
The impact of spin-orbit coupling in the electronic structure



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

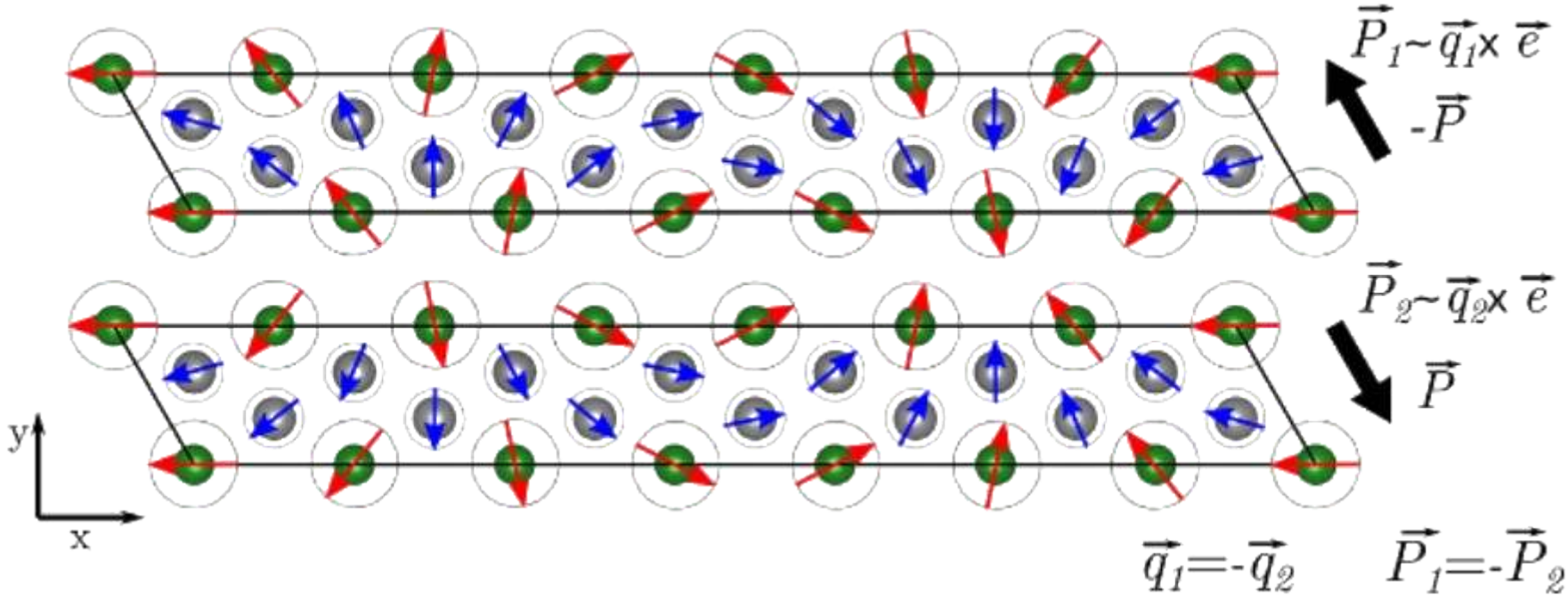
There is a major role of the halide close to the Fermi level

The interplay between magnetism and halide electronic structure



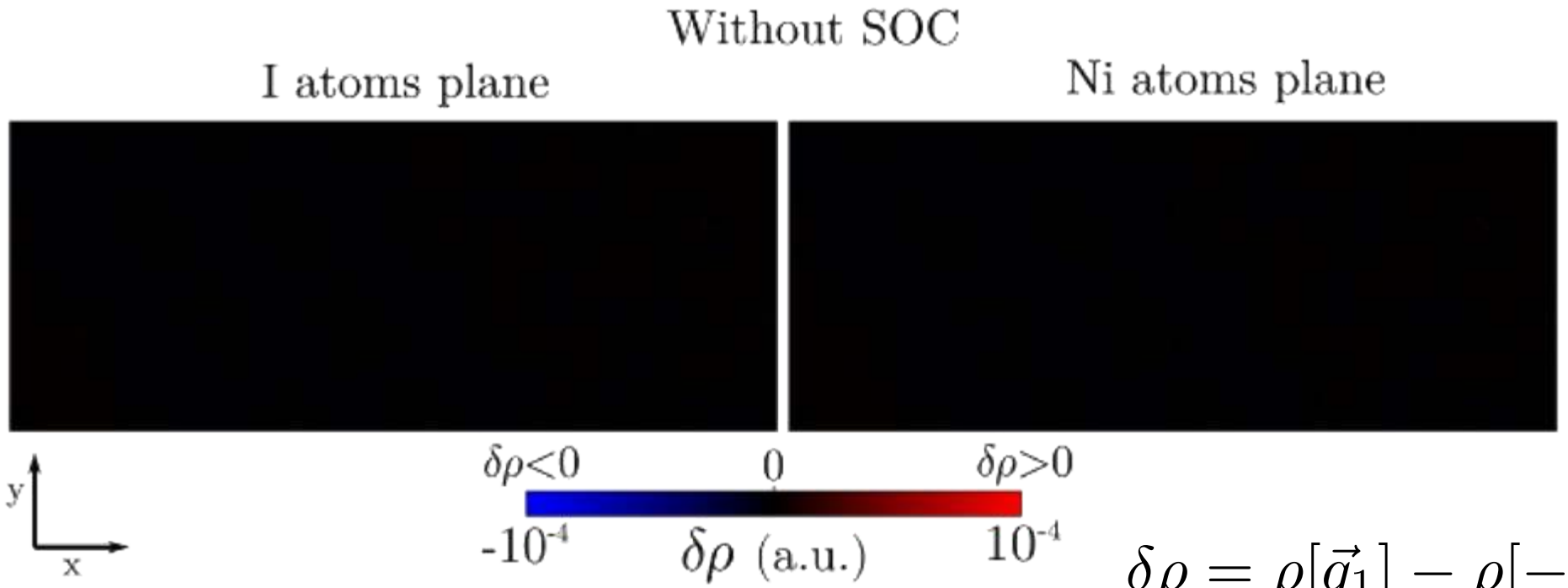
Magnetism not only appears in Ni, but a sizable value merges also in I

Ferroelectric correction to the electronic density



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

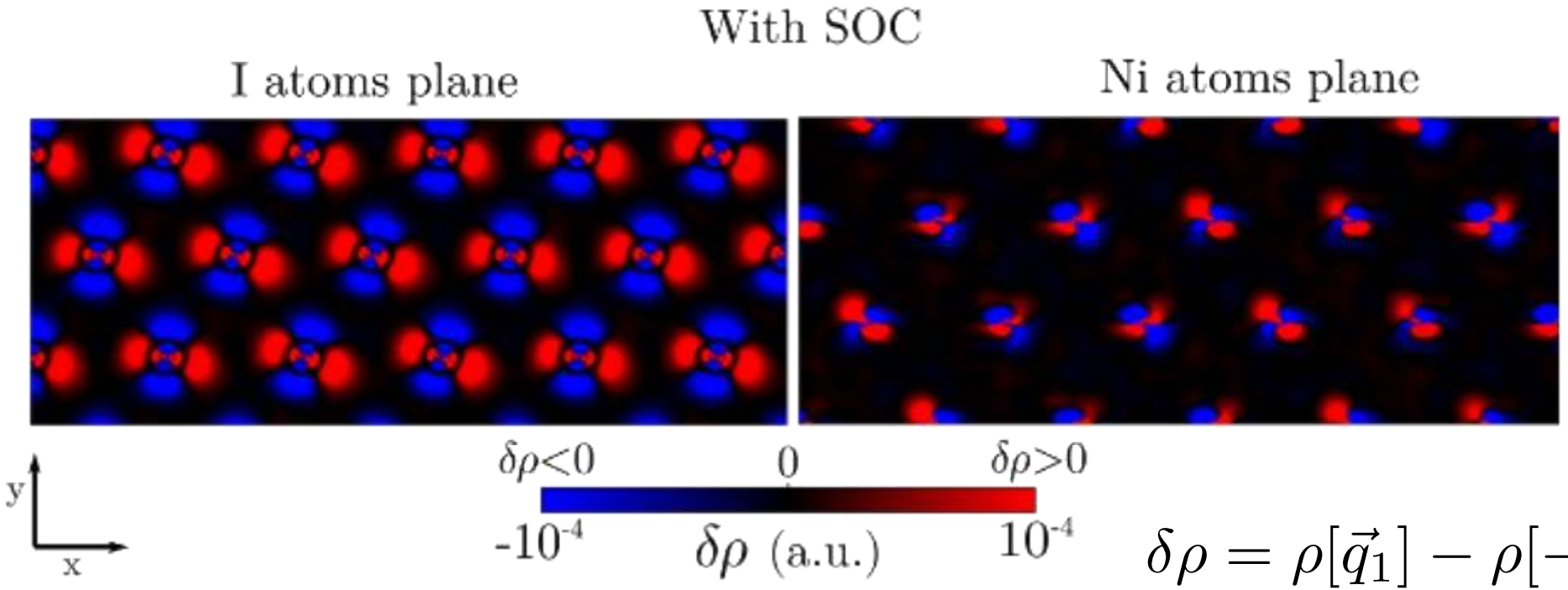
Ferroelectric correction to the electronic density



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

In the absence of SOC, no ferroelectric density appears

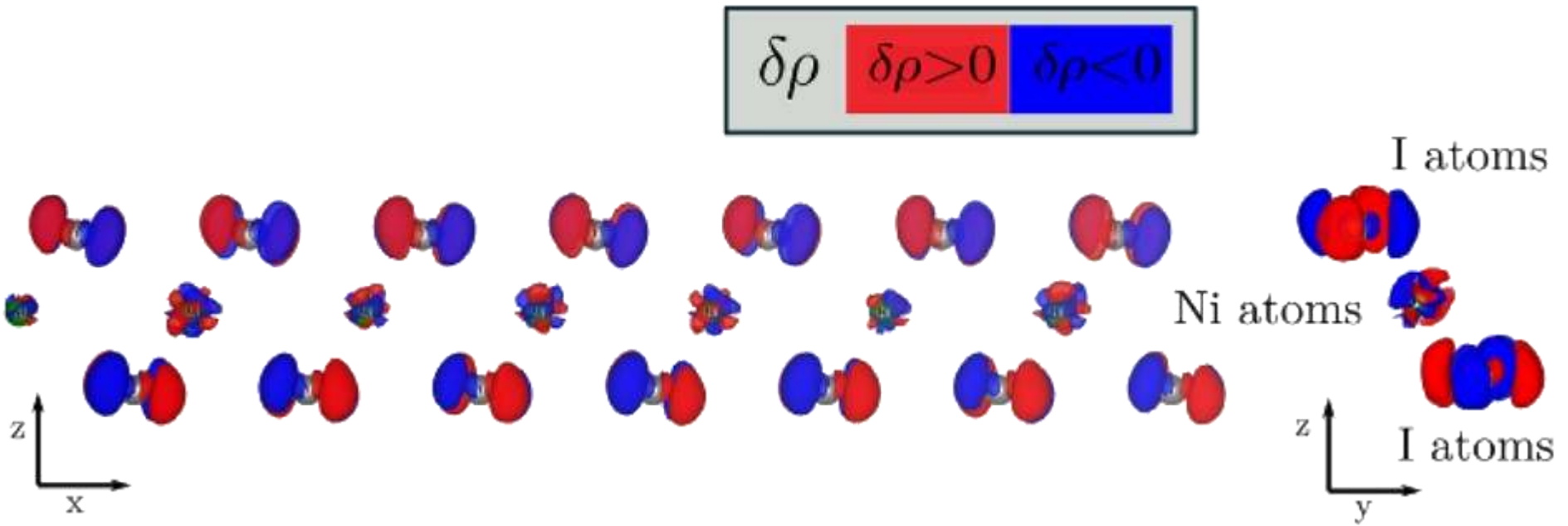
Ferroelectric correction to the electronic density



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

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

Ferroelectric correction to the electronic density

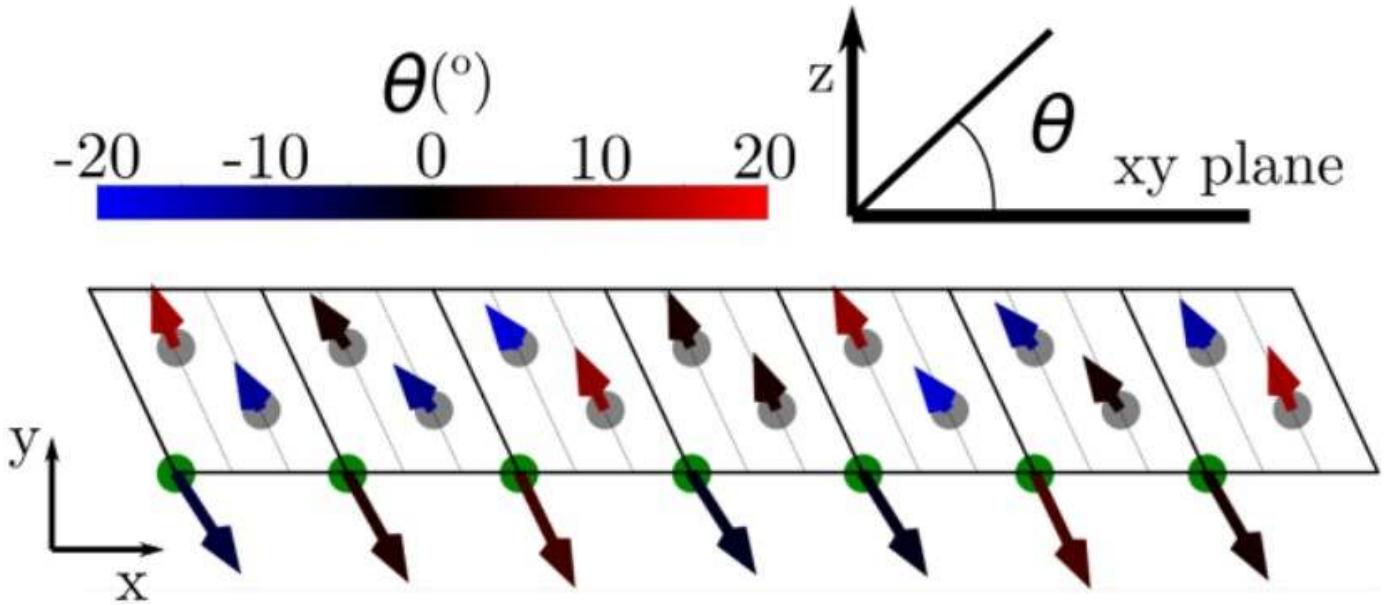


$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

The ferroelectric density has comparable magnitudes in Ni and I

The ferroelectric force

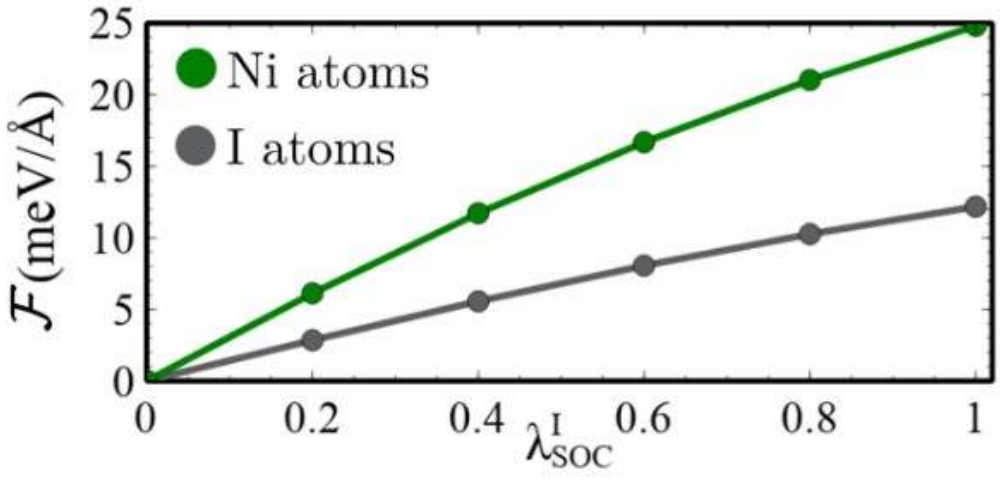
$$\mathcal{F}_\alpha = \frac{1}{2}(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]),$$

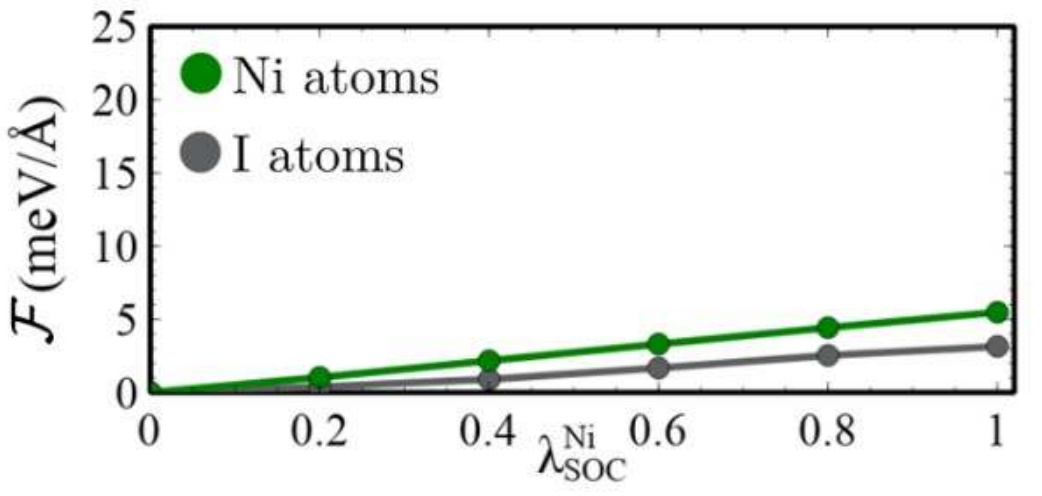


The forces between the two magnetic configuration account for the ferroelectric displacement

The ferroelectric force

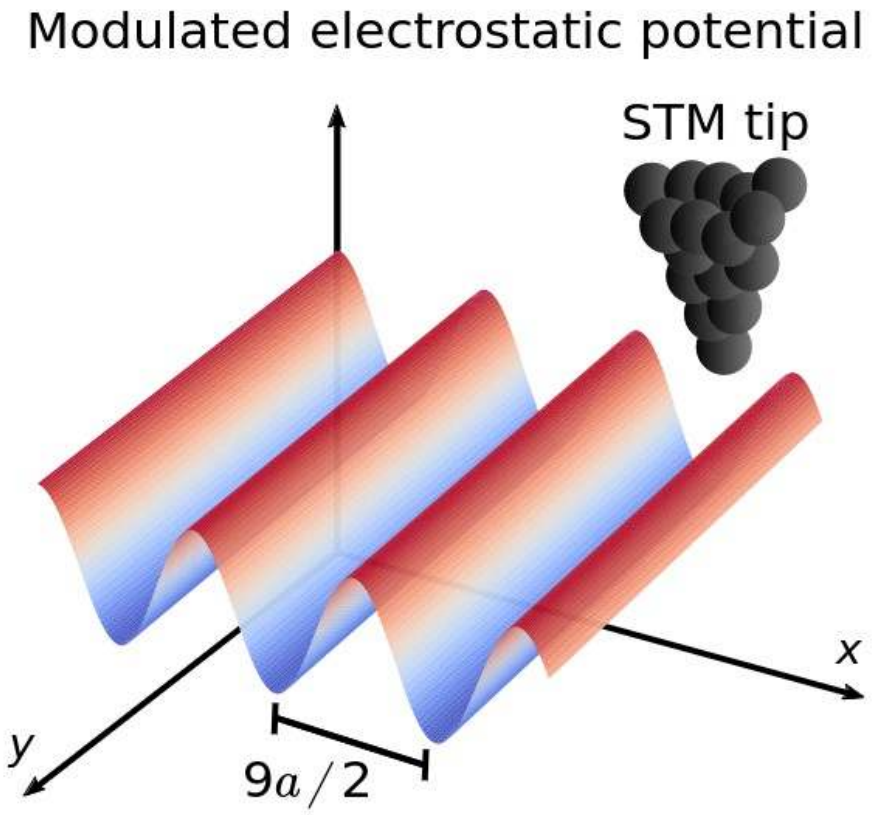
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\mathrm{SOC}}^{\mathrm{Ni}} \mathcal{H}_{\mathrm{SOC}}^{\mathrm{Ni}} + \lambda_{\mathrm{SOC}}^{\mathrm{I}} \mathcal{H}_{\mathrm{SOC}}^{\mathrm{I}},$$





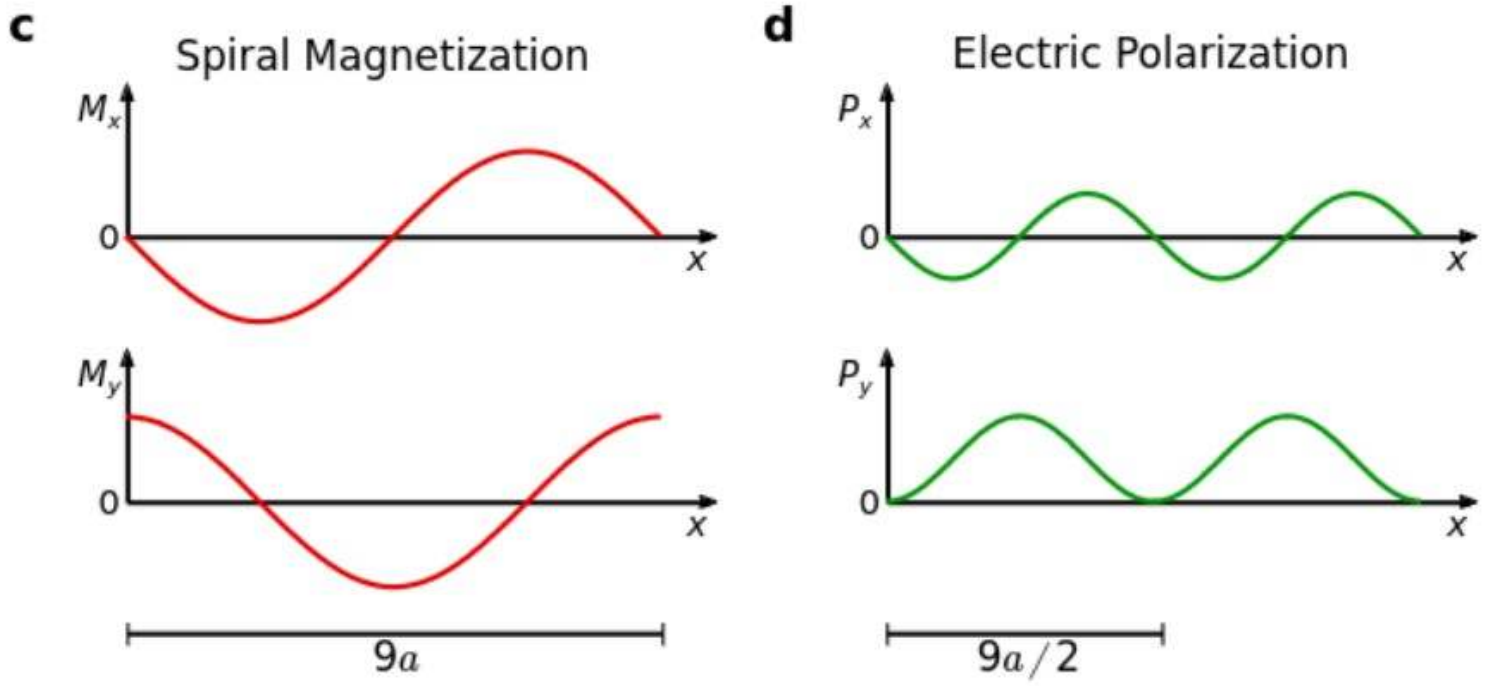
The dominant contribution to the ferroelectric force comes from iodine

Visualizing multiferroicity in monolayer NiI_2



arXiv:2309.11217 (2023)

Visualizing multiferroicity in monolayer NiI_2

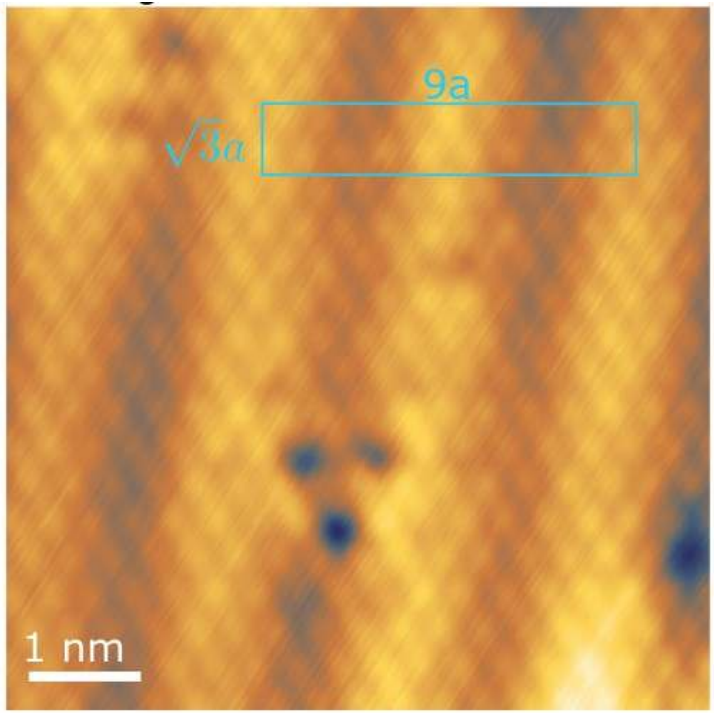


$$\mathbf{P} = \Lambda \frac{\mathbf{M} \times (\nabla \times \mathbf{M})}{\mathrm{M}^2} = \Lambda \mathrm{q}(-\sin(2\mathbf{q}\cdot\mathbf{r}), \sin^2(\mathbf{q}\cdot\mathbf{r}), 0)$$

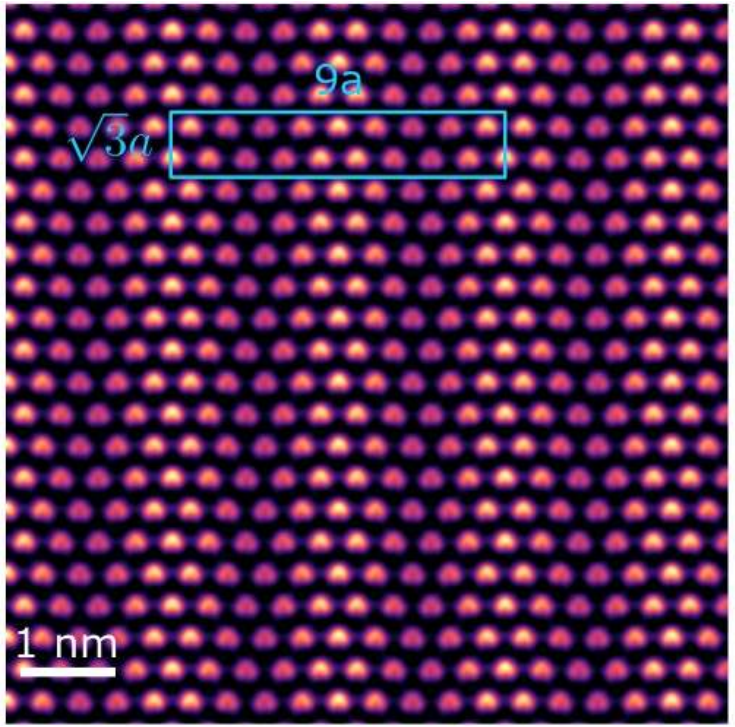
arXiv:2309.11217 (2023)

Visualizing multiferroicity in monolayer NiI_2

Experiment



Theory

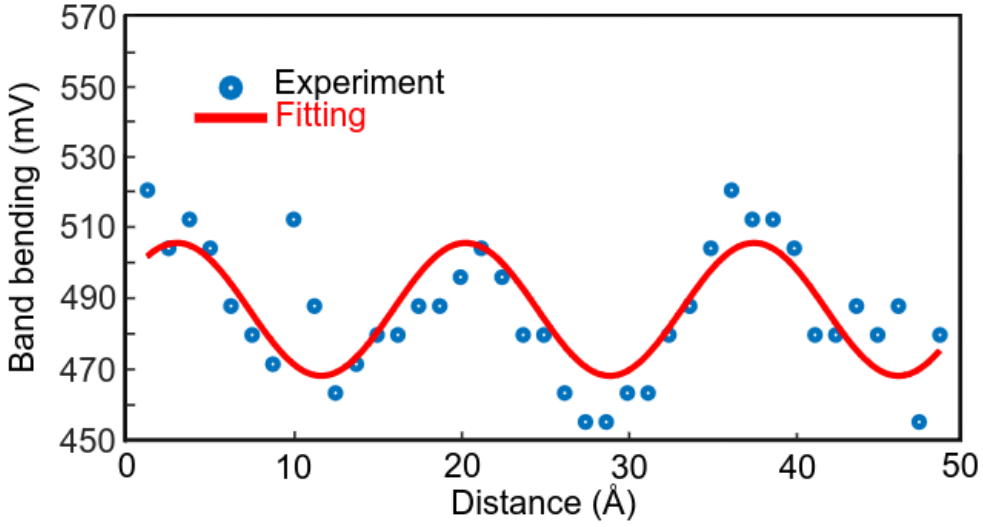


$$\Lambda \mathbf{q}(-\sin(2\mathbf{q}\cdot\mathbf{r}), \sin^2(\mathbf{q}\cdot\mathbf{r}), 0)$$

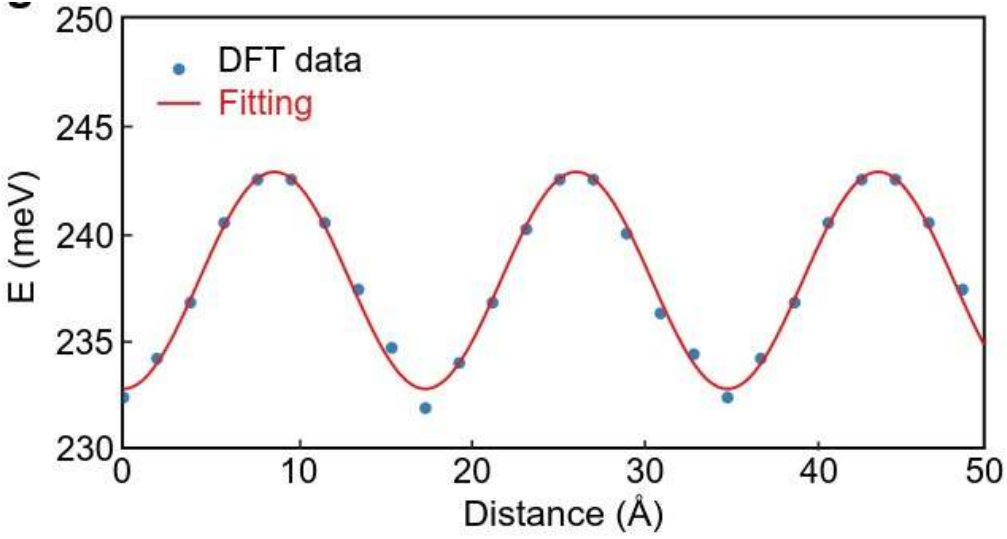
arXiv:2309.11217 (2023)

Ferroelectricity from modulated band offset

Band offset (experiment)



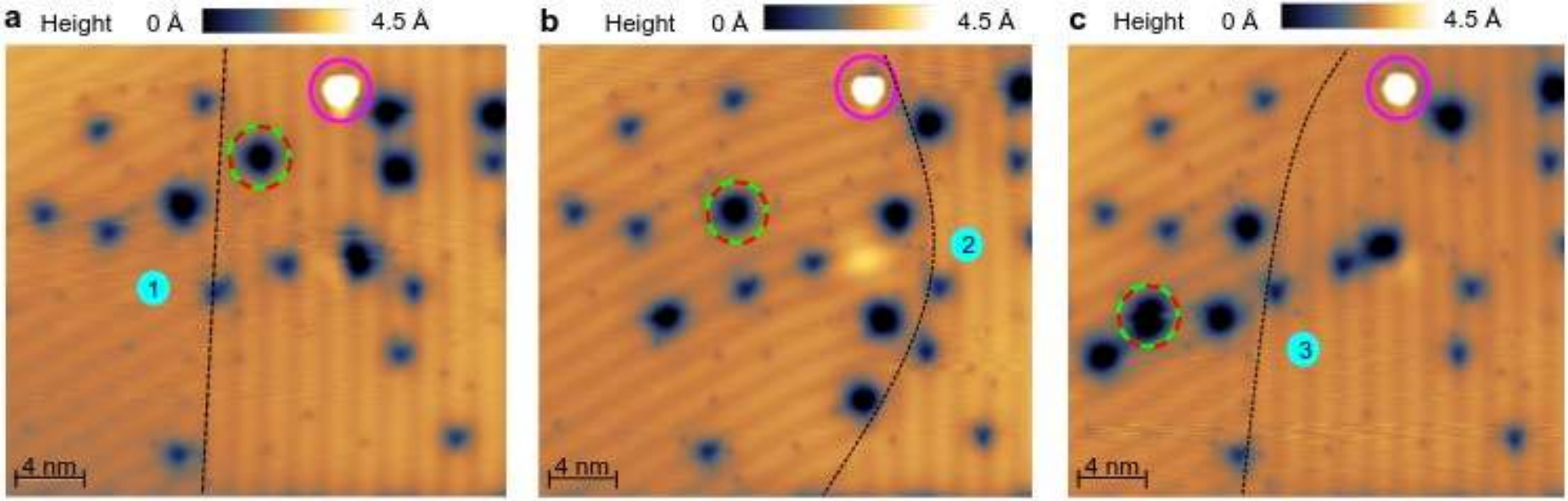
Band offset (theory)



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

arXiv:2309.11217 (2023)

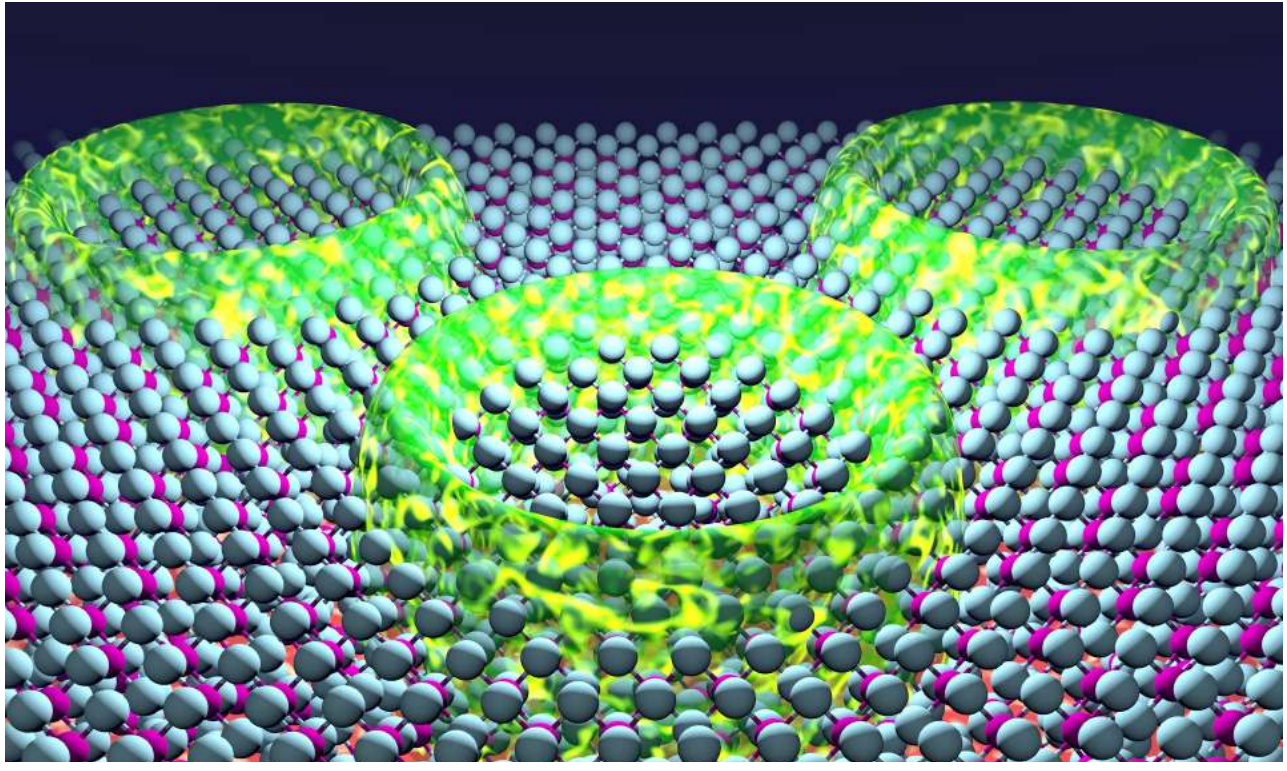
Controlling muultiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

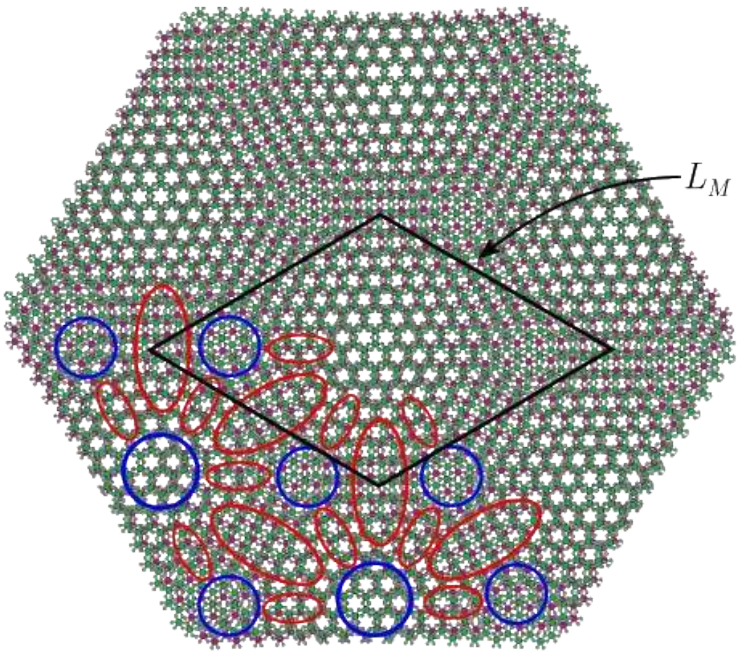
arXiv:2309.11217 (2023)

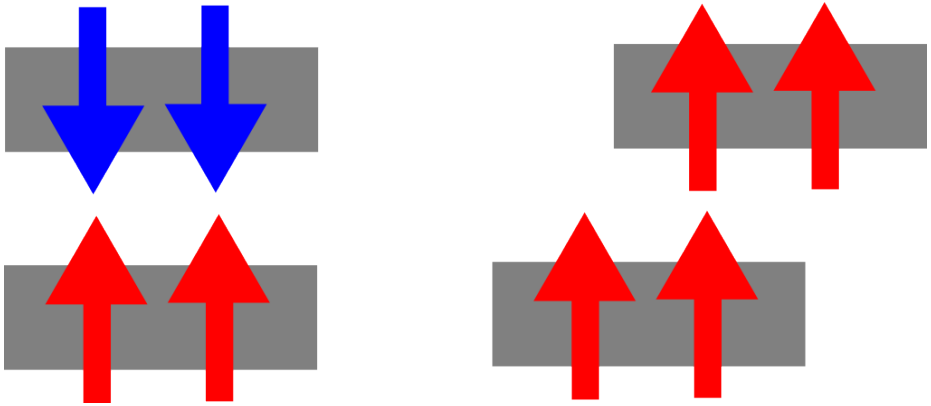
Artificial multiferroics, twisted $CrBr_3$ bilayers



Twisted 2D trihalides

$CrBr_3$

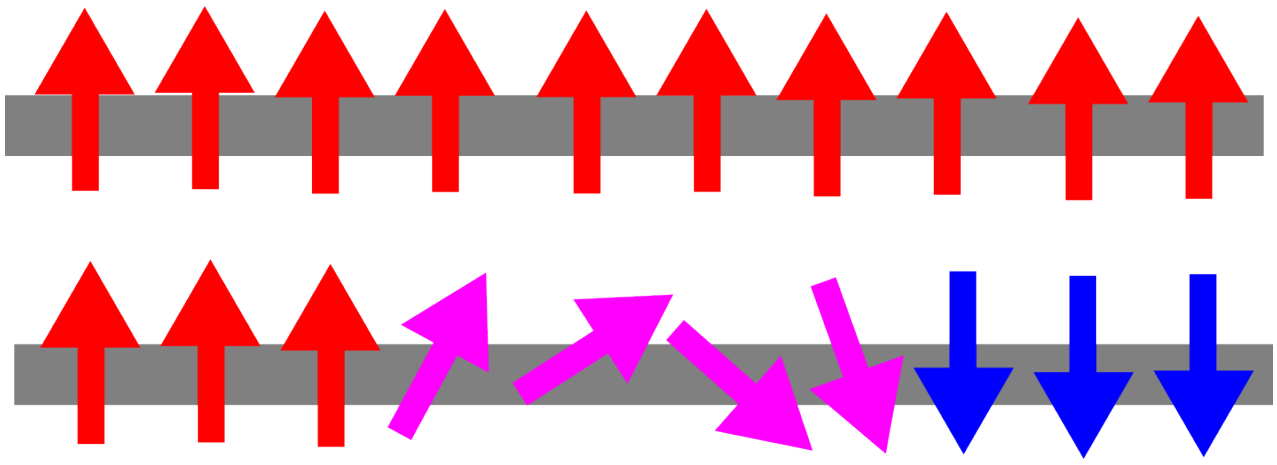




The local stacking determines
the coupling between layers

Nature Nano 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Stacking #1

Stacking #2

Mixed FE/AF domains

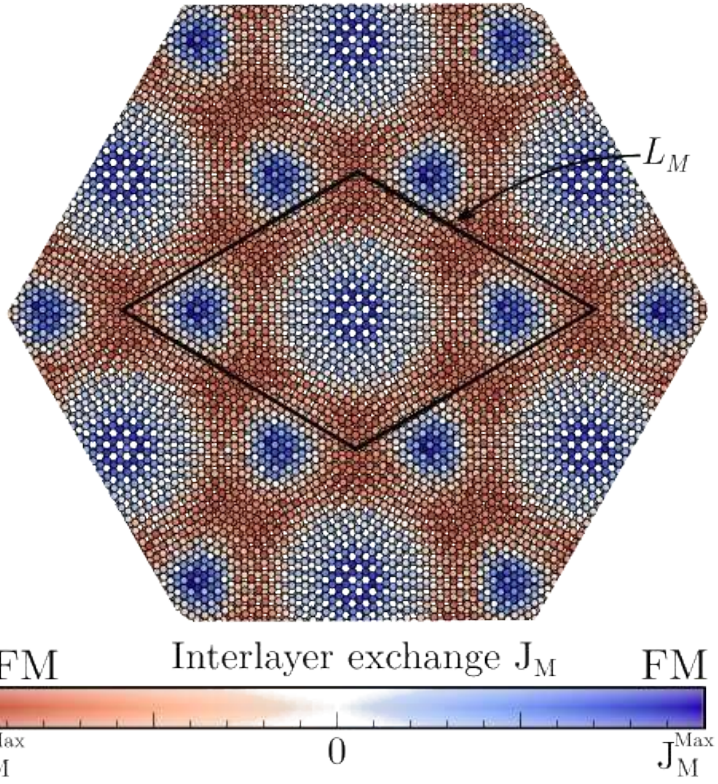
Nature Nanotechnology 17, 143–147 (2022)

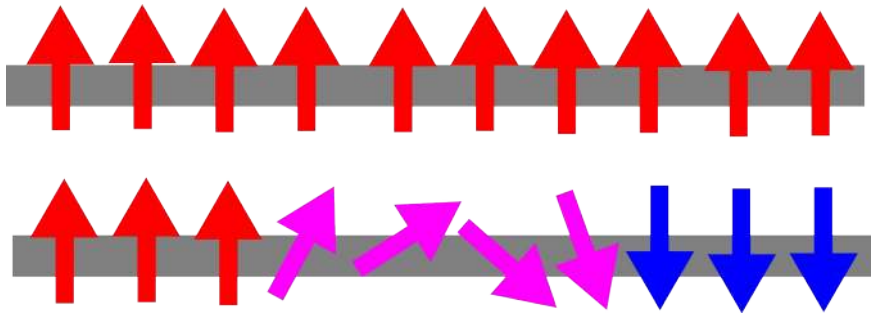
Non-collinear magnetism

arXiv:2204.01636
Nature Elec 6, 434–442 (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

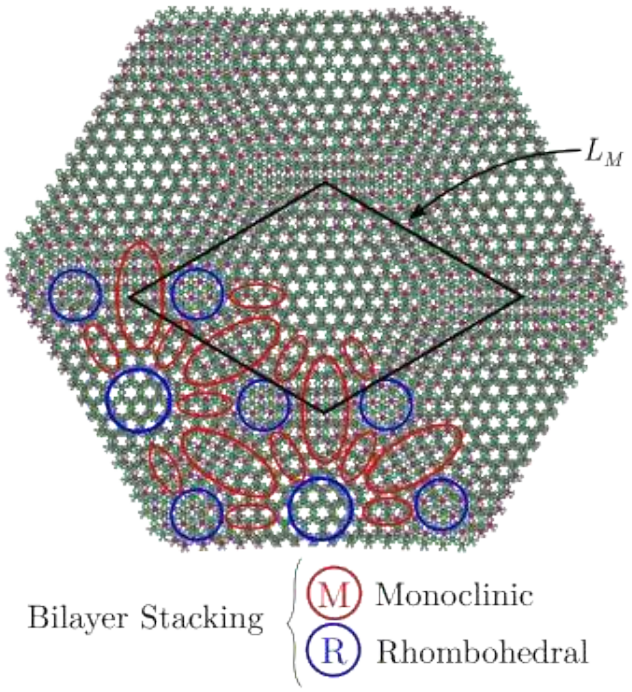
SOC driven

Wavevector of the spin spiral

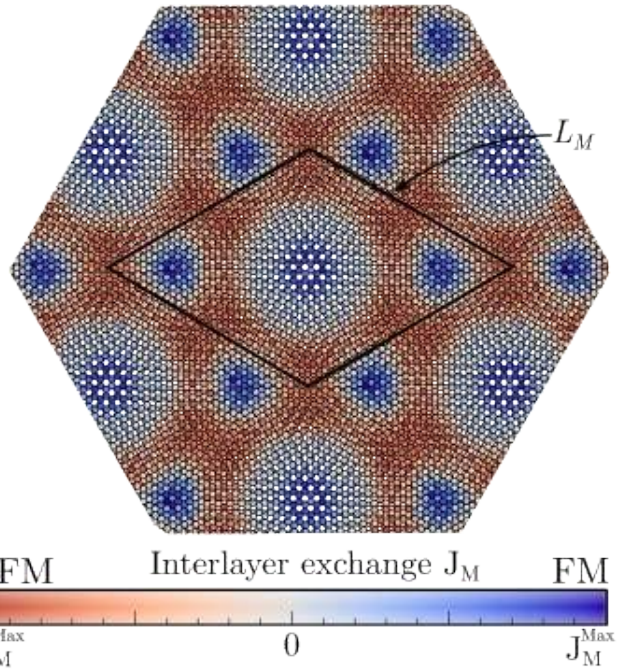
Spin rotation axis

Stacking, magnetic order and electric polarization

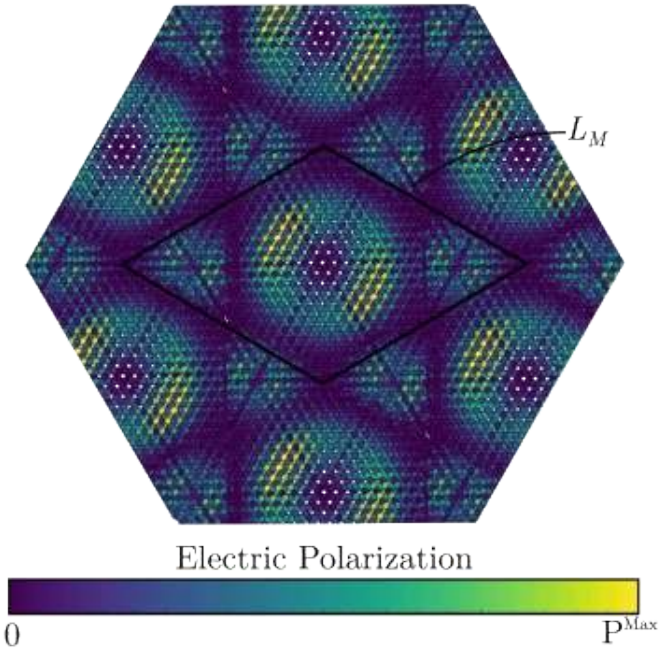
Structure



Magnetic order



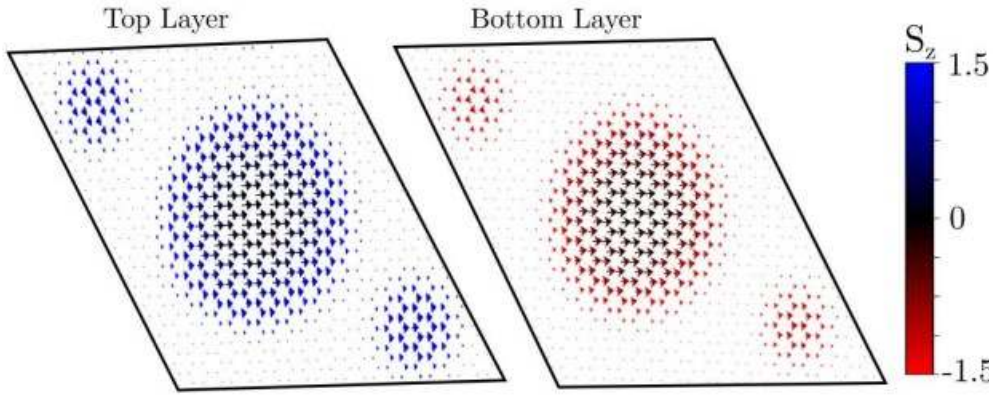
Ferroelectric order



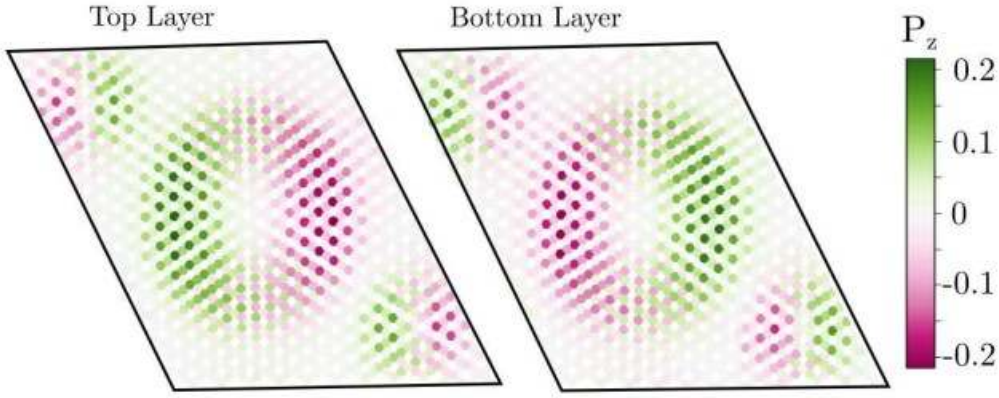
Stacking, magntic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

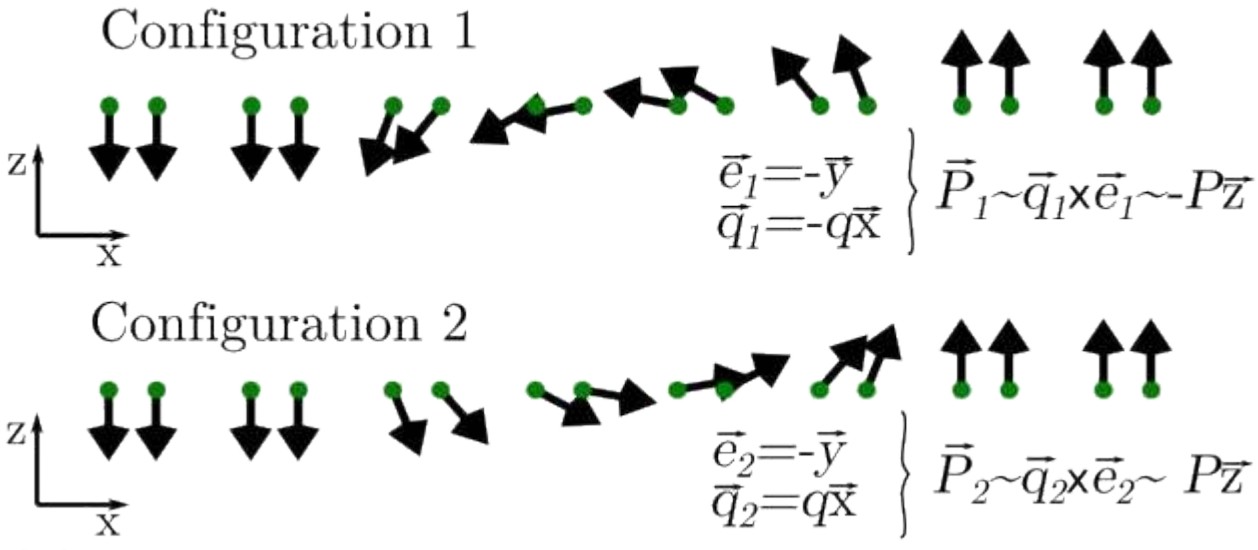


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



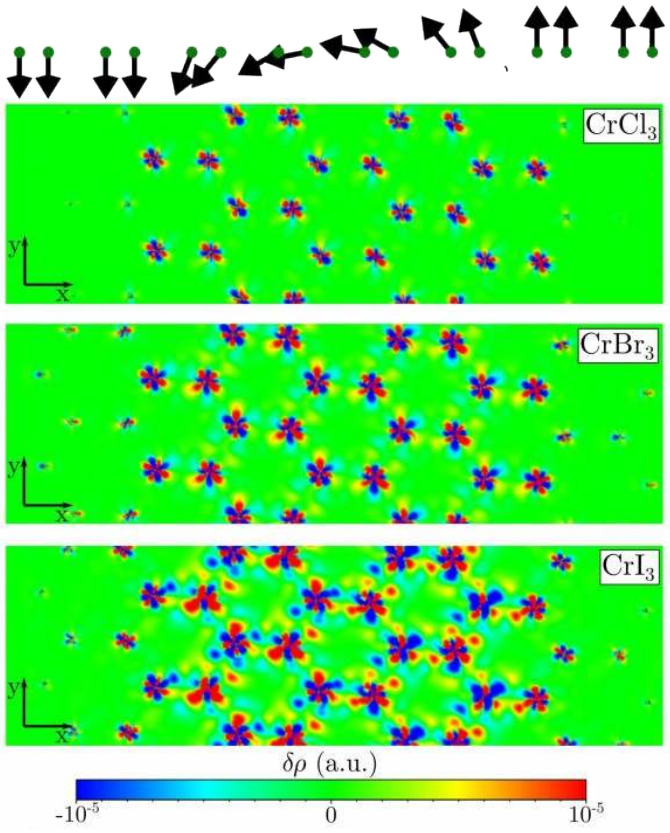
Ferroelectric electronic density

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

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

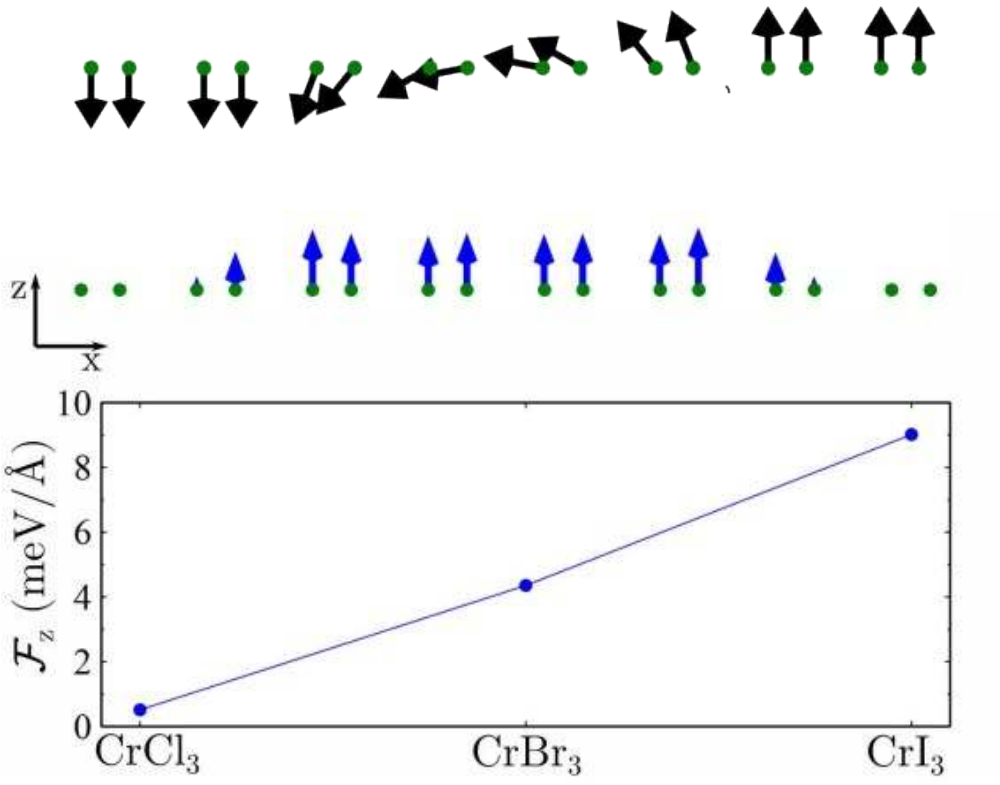
The ferroelectric force

Magnetic configuration

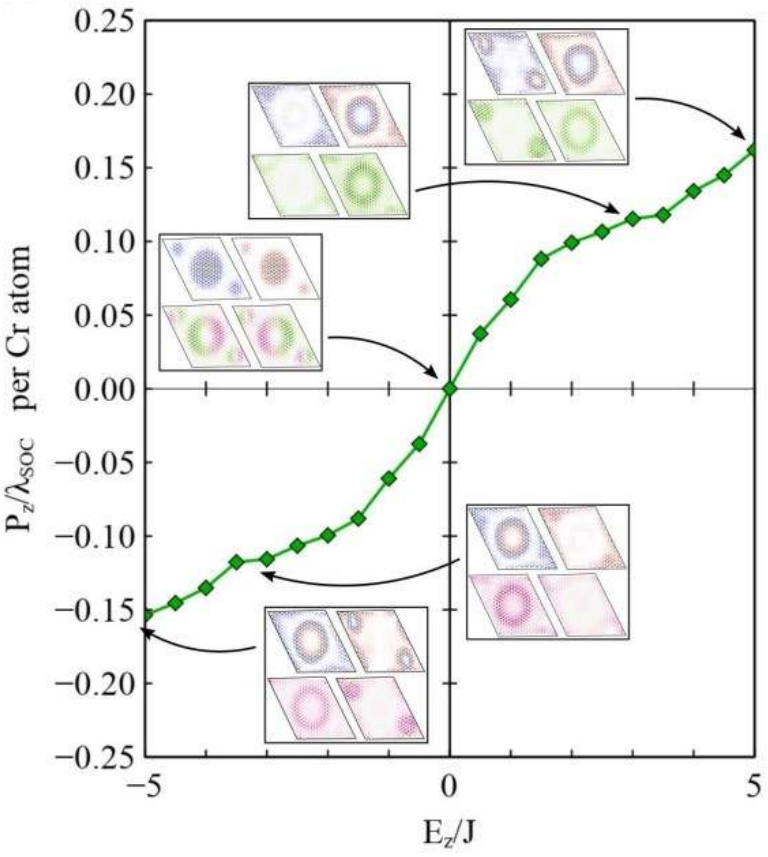
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



Electrically controllable skyrmions in twisted magnets



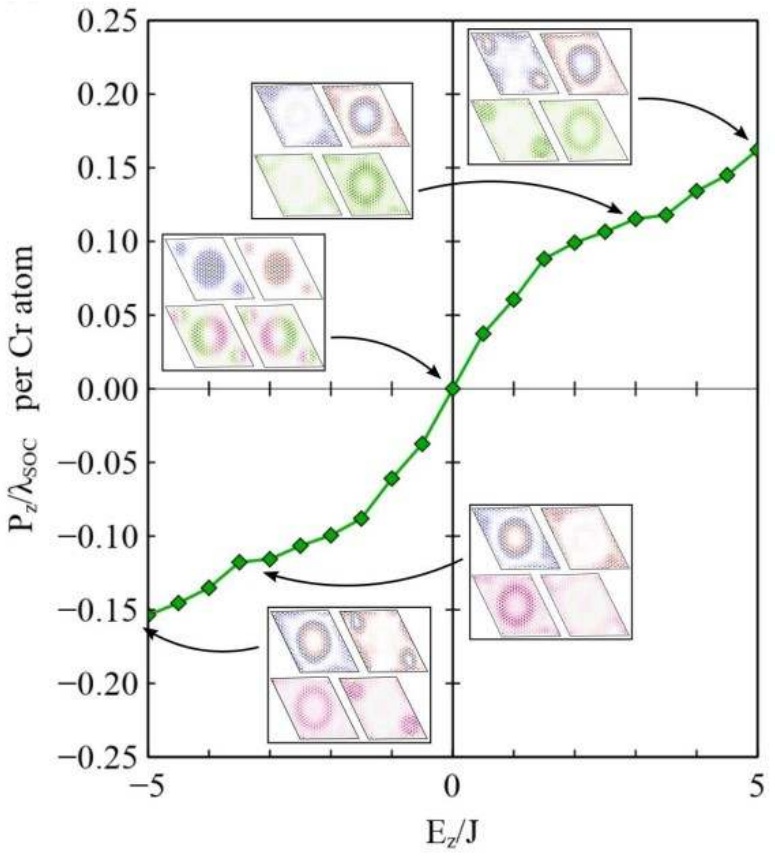
The electric polarization directly couples to an out-of plane electric field

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

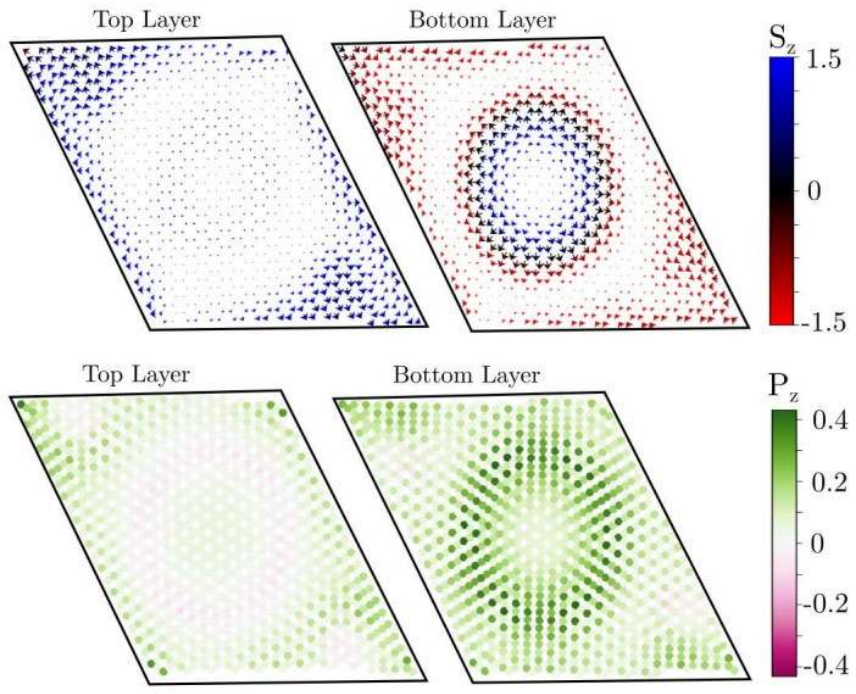
Experimental control of magnetism in twisted CrI_3

Nature Elec 6, 434–442 (2023)

Electrically controllable skyrmions in twisted magnets

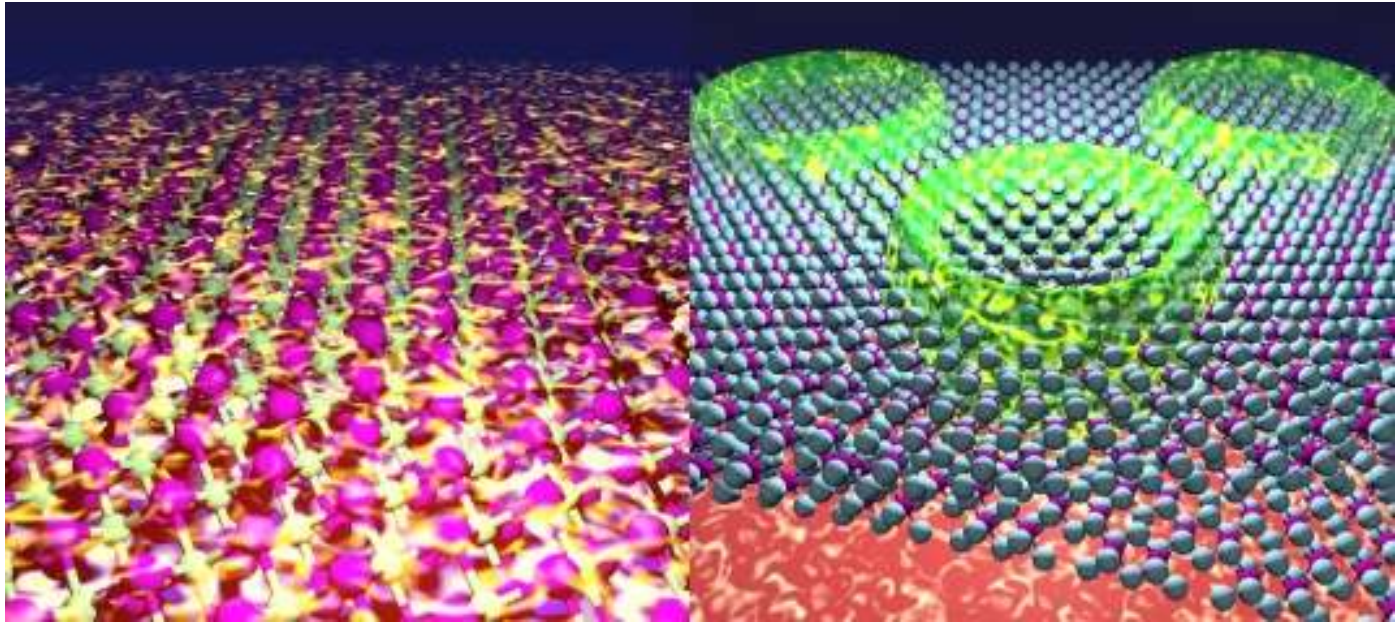


Skyrmions at finite interlayer bias



Take home

Van der Waals materials provide a platform to realize tunable multiferroic order



Two open questions in van der Waals multiferroics

- Can we drive frustrated monolayers to a multiferroic regime ($RuCl_3$, 1T-$TaSe_2$, $FeCl_3$)
- Other multiferroics from twisting beyond trihalides