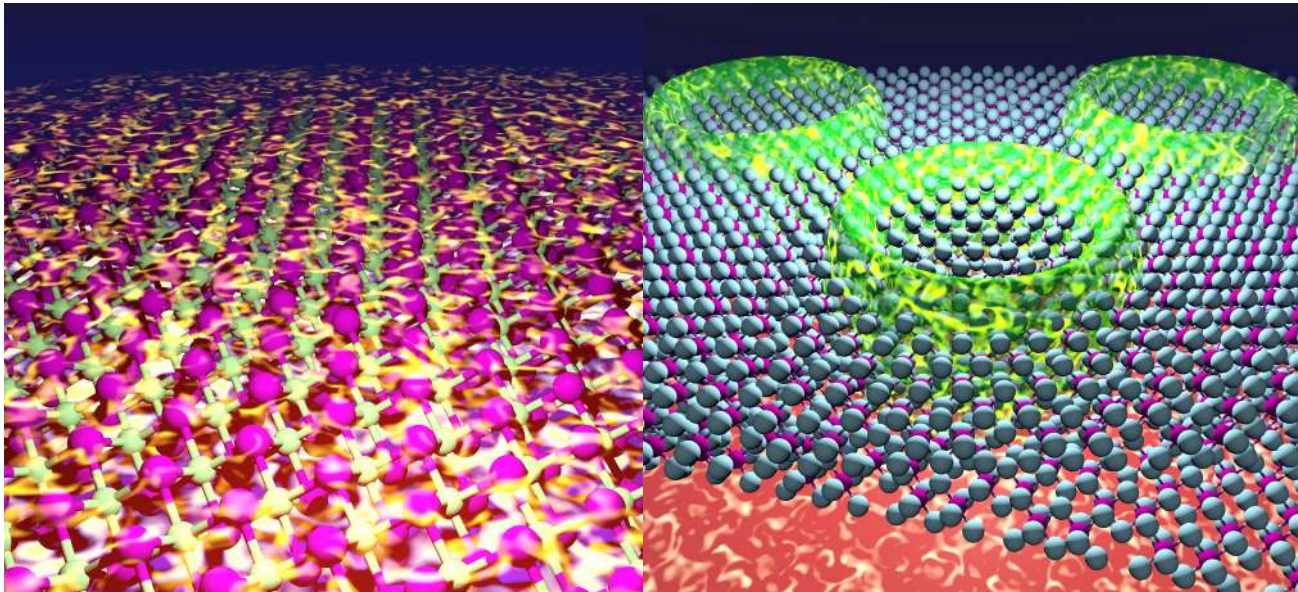


Artificial van der Waals multiferroics with twisted two-dimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland



2D Materials 9 (2), 025010 (2022)

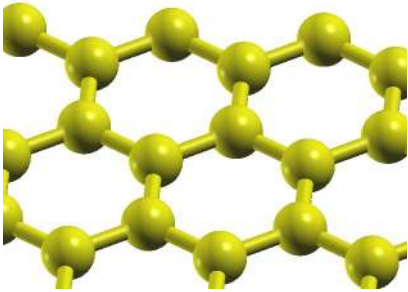
arXiv:2207.01416 (2022)

MATSUS nanoGe: Materials for Quantum Technology, Valencia, Spain, March 7th 2023

The two-dimensional materials world

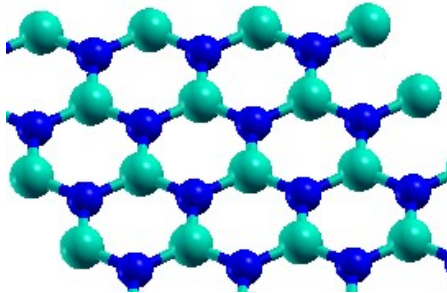
Semimetal

Graphene



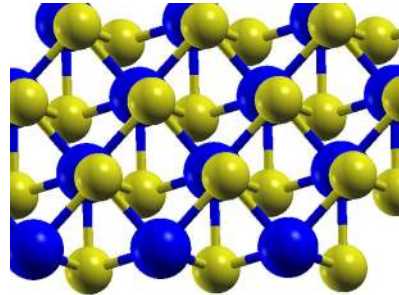
Insulator

BN



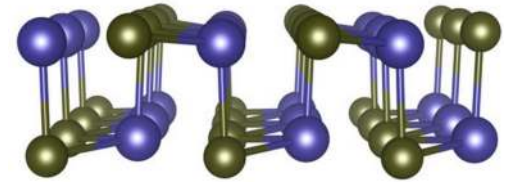
Superconductor

NbSe₂



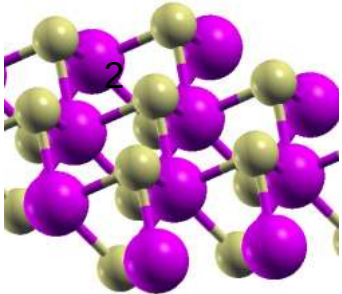
Ferroelectric

SnTe



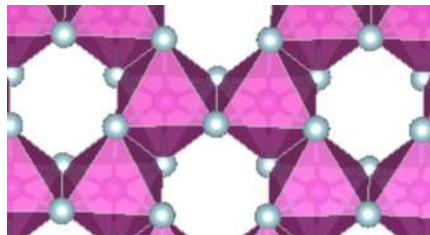
Semiconductor

WSe



Ferromagnet

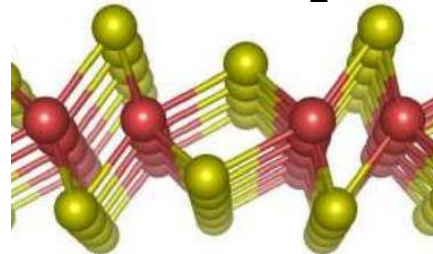
CrI₃



Quantum spin

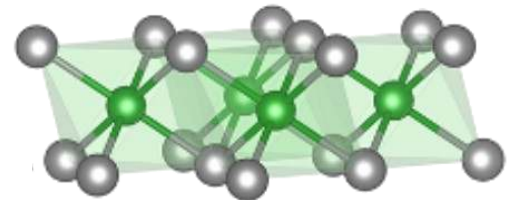
Hall insulator

WTe₂



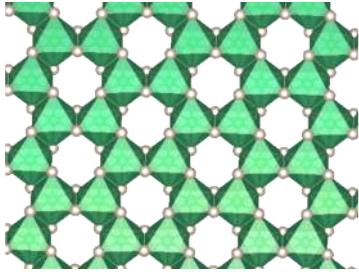
Multiferroic

NiI₂



Van der Waals magnetic materials

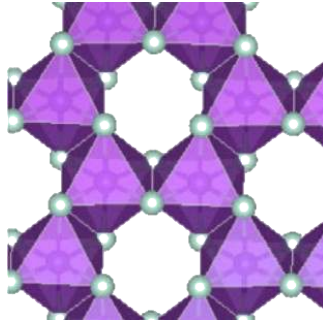
**Ferromagnet,
antiferromagnets**



CrI_3 , CrCl_3 , CrBr_3

Nature 546, 270–273 (2017)

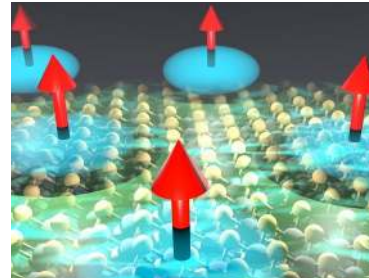
**(proximal)
Quantum
spin-liquids**



RuCl_3 , 1T-TaS₂

Nature Physics 17, 1154–1161 (2021)

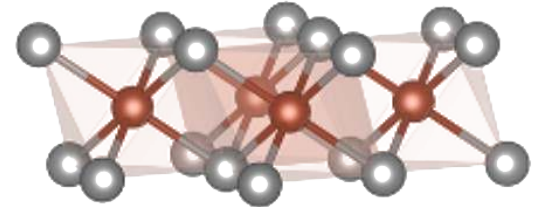
**Heavy-fermion
Kondo insulators**



1T-TaS₂/1H-TaS₂

Nature 599, 582–586 (2021)

Multiferroics



NiI_2

Nature 602, 601–605 (2022)

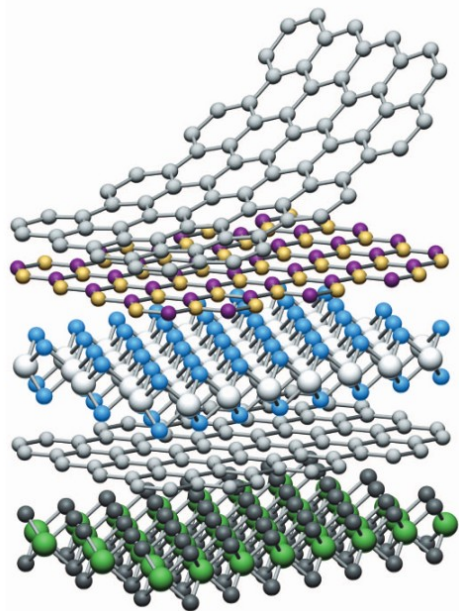
Break time-reversal

Do not break time-reversal

Break time-reversal
and inversion symmetry

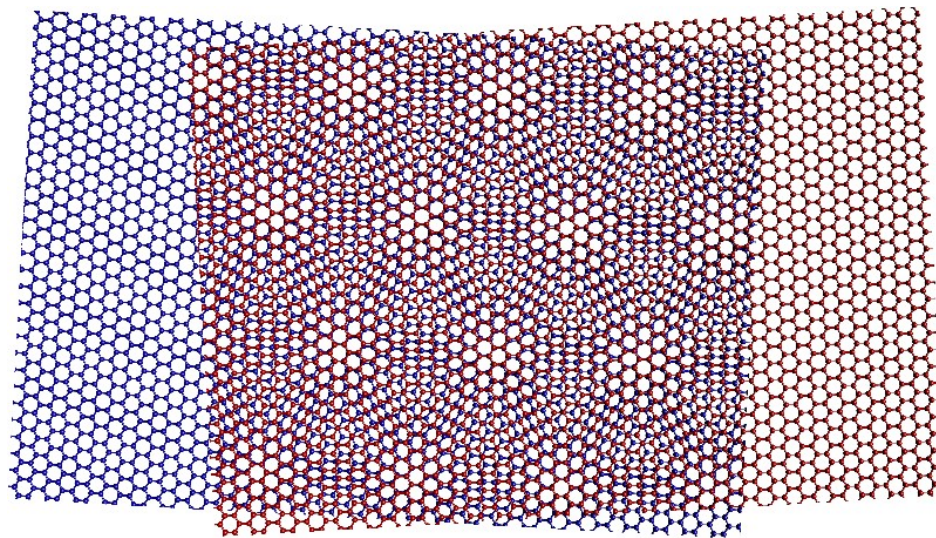
The flexibility of two-dimensional materials

They can be stacked



Nature 499, 419–425 (2013)

They can be rotated

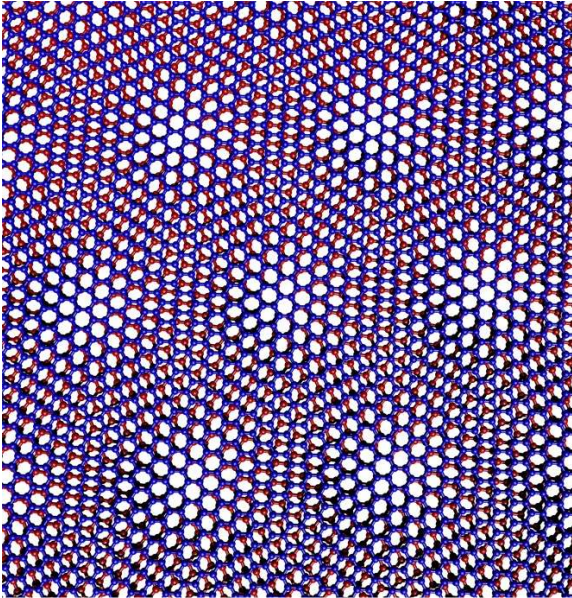


Science 361, 6403, 690-693 (2018)

These are unique features of two-dimensional materials

The flexibility of twist engineering

Twisted bilayer graphene

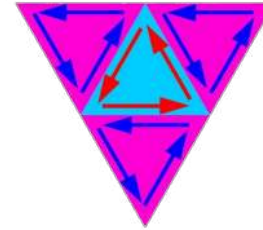


Superconductivity

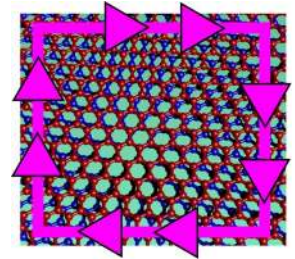


Nature 556, 43–50 (2018)

Topological networks Chern insulators

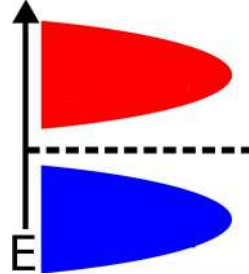


Nano Lett. 18, 11, 6725–6730 (2018)



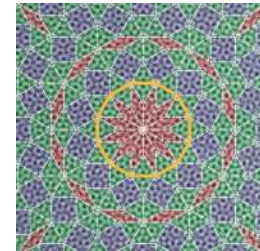
Science 365, 605–608 (2019)

Correlated insulators



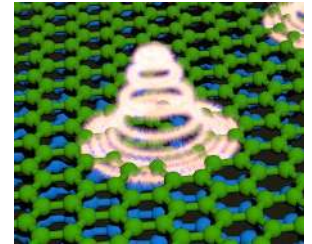
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782–786 (2018)

Fractional Chern insulators

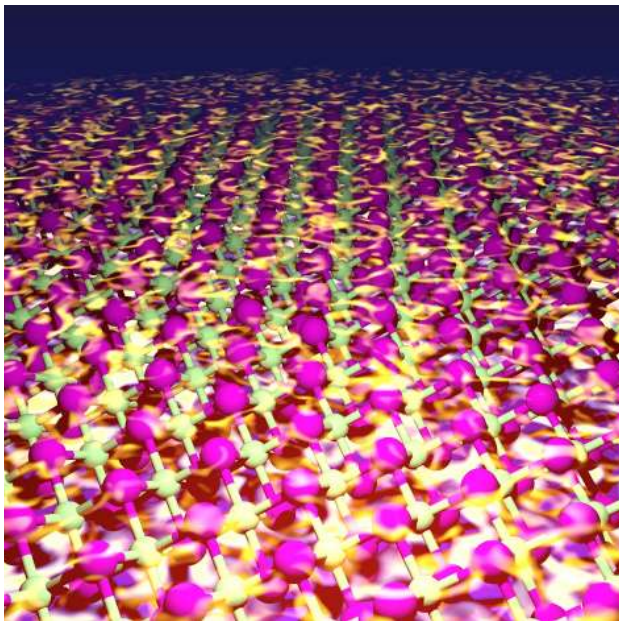


Nature 600, 439–443 (2021)

Twisted van der Waals multilayers provide a powerful platform for emergent phenomena

Two strategies for multiferroicity in van der Waals materials

Multiferroic monolayer NiI_2

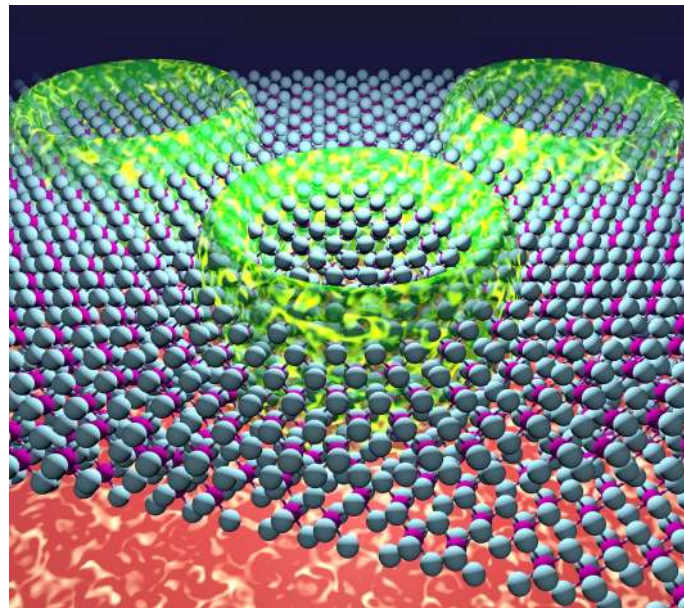


2D Materials 9 (2), 025010 (2022)

Adolfo Fumega

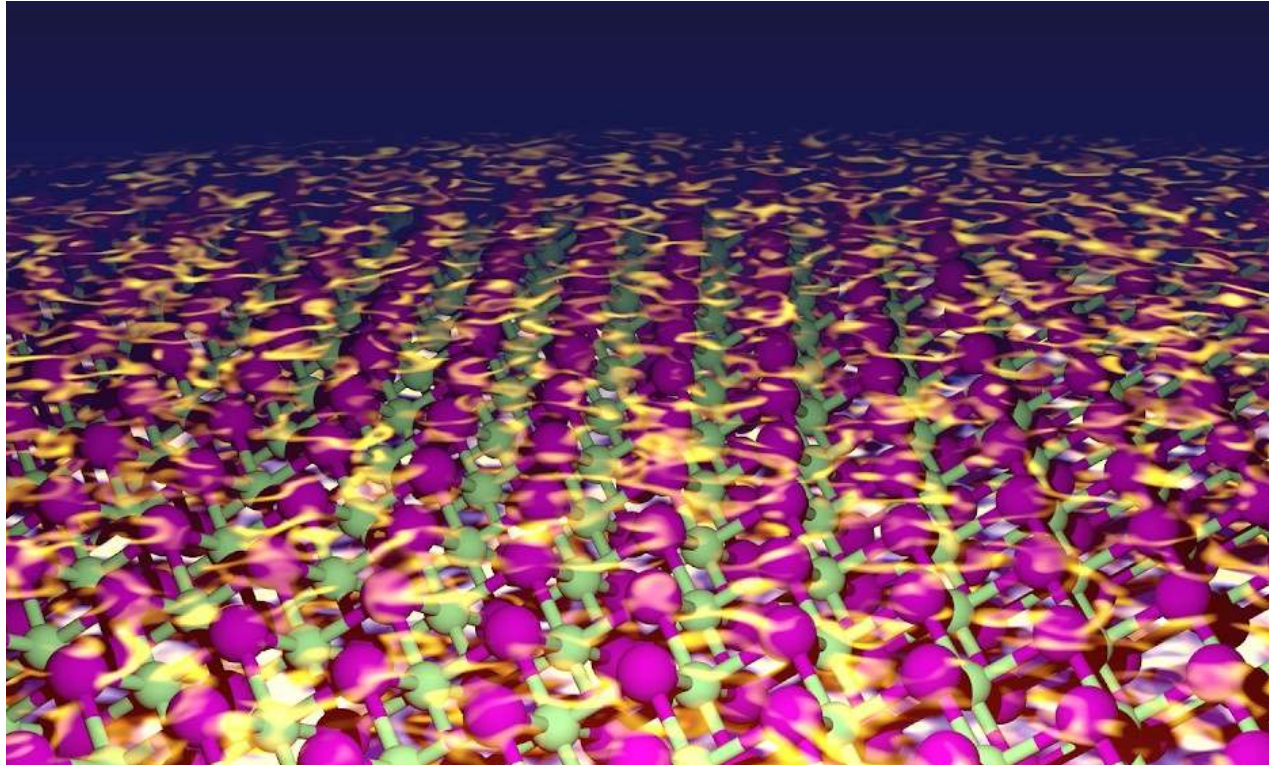


Artificial multiferroics via twist engineering with CrBr_3 bilayers



arXiv:2207.01416 (2022)

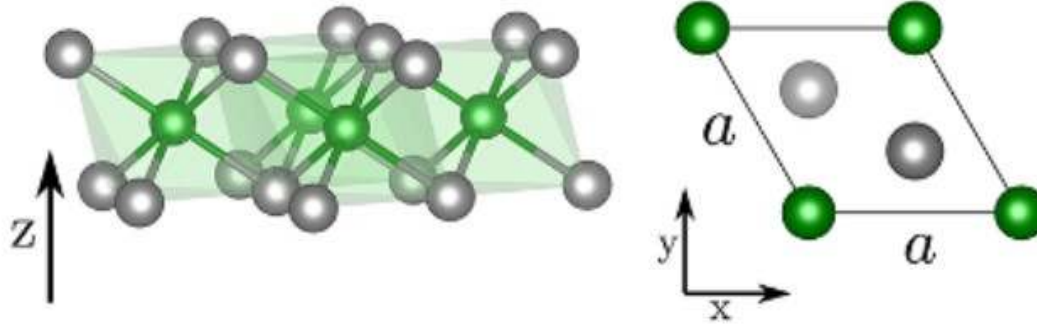
The origin of multiferroic order in NiI_2



2D Materials 9 (2), 025010 (2022)

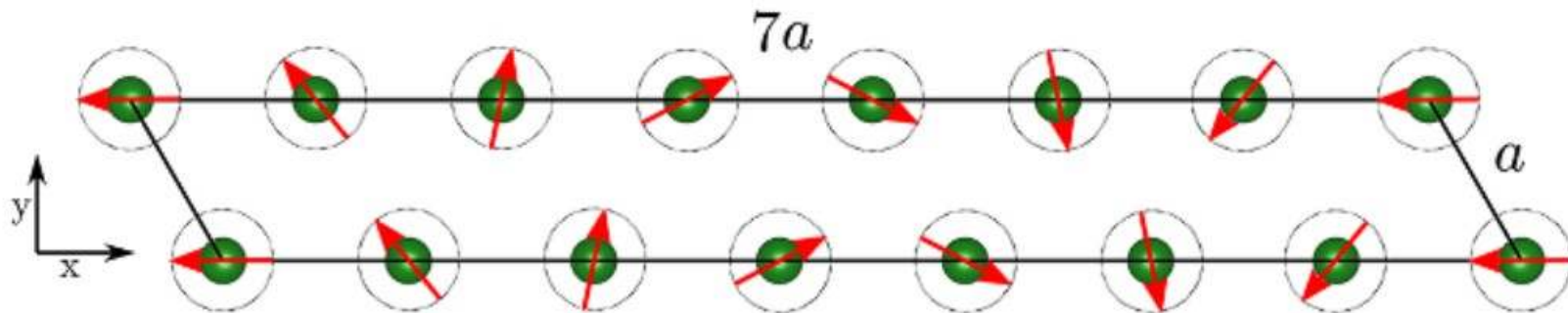
Monolayer NiI_2

Structure



Magnetic structure

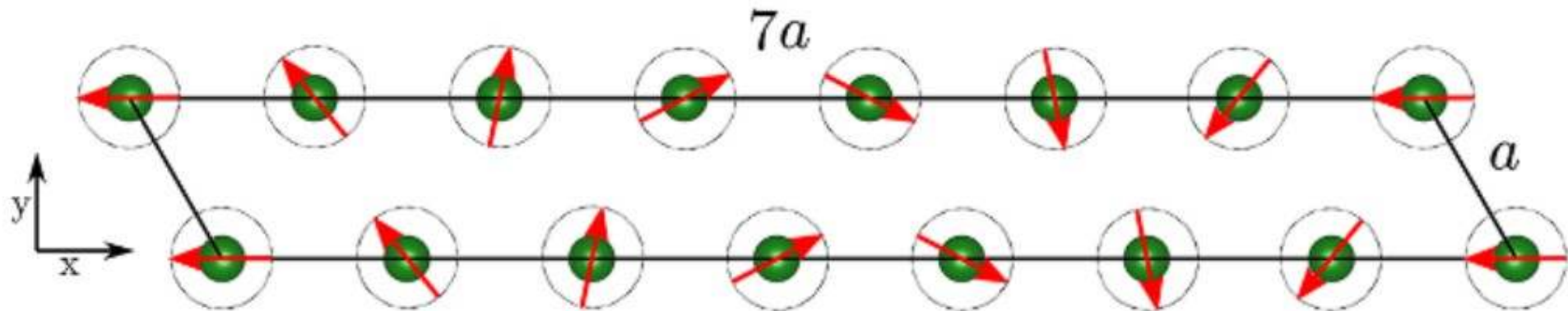
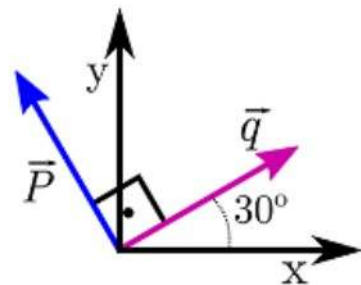
Nature 602, 601–605 (2022)



The origin of ferroelectricity in NiI_2

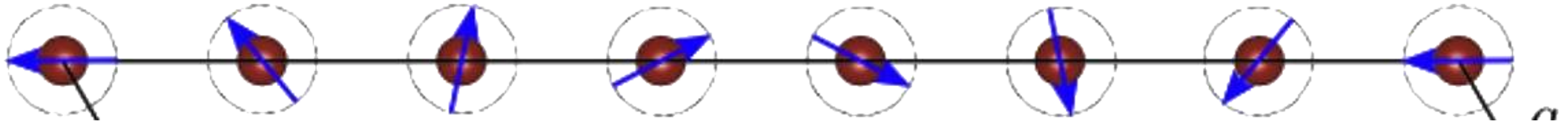
Magnetically driven ferroelectric order $\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$

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



The emergence of non-collinear order is the driving of ferroelectricity

The origin of ferroelectricity in NiI_2



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

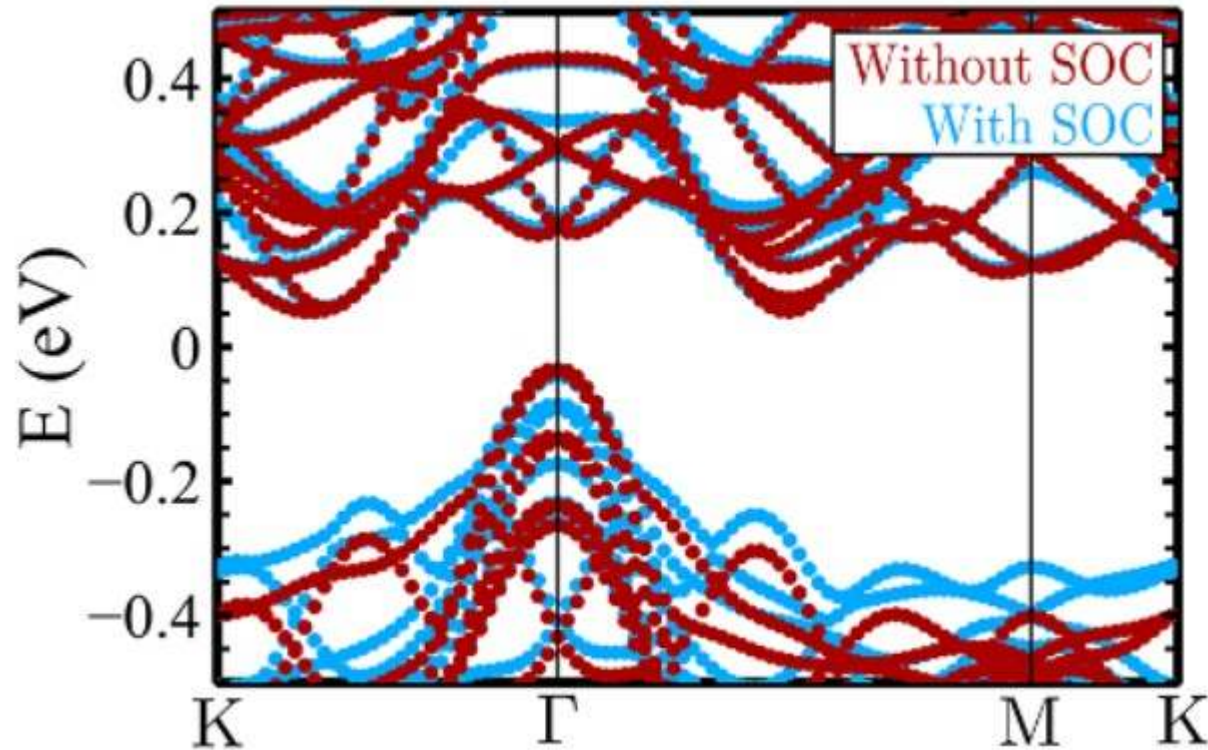
Electric polarization

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

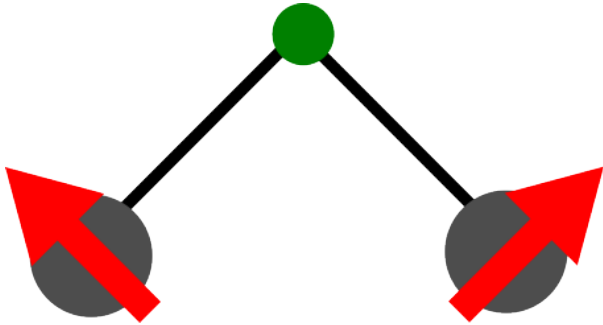
There is a major role of the halide close to the Fermi level

The impact of spin-orbit coupling in magnetic ordering

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

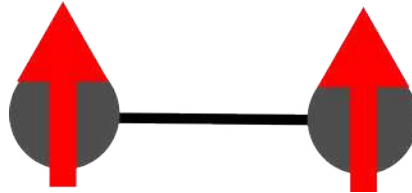
Antisymmetric exchange

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



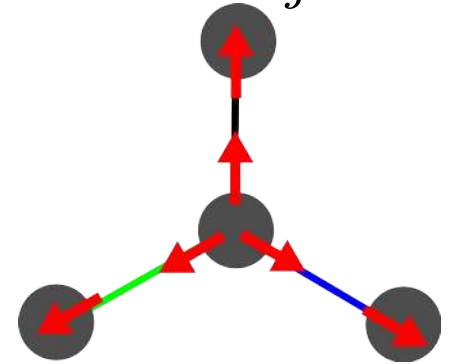
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

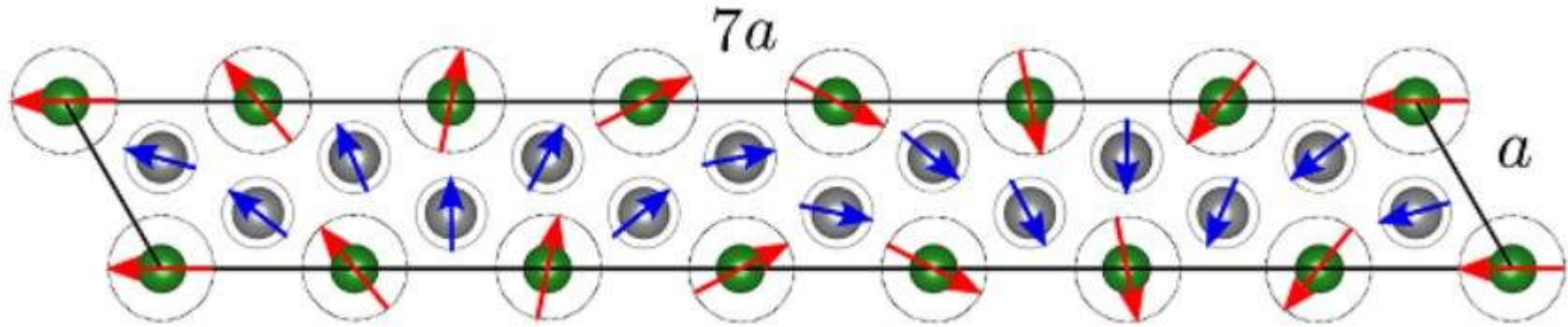
$$S_i^{\alpha(i)} S_j^{\alpha(j)}$$



In NiI₂, the non-collinear order is promoted by isotropic exchange

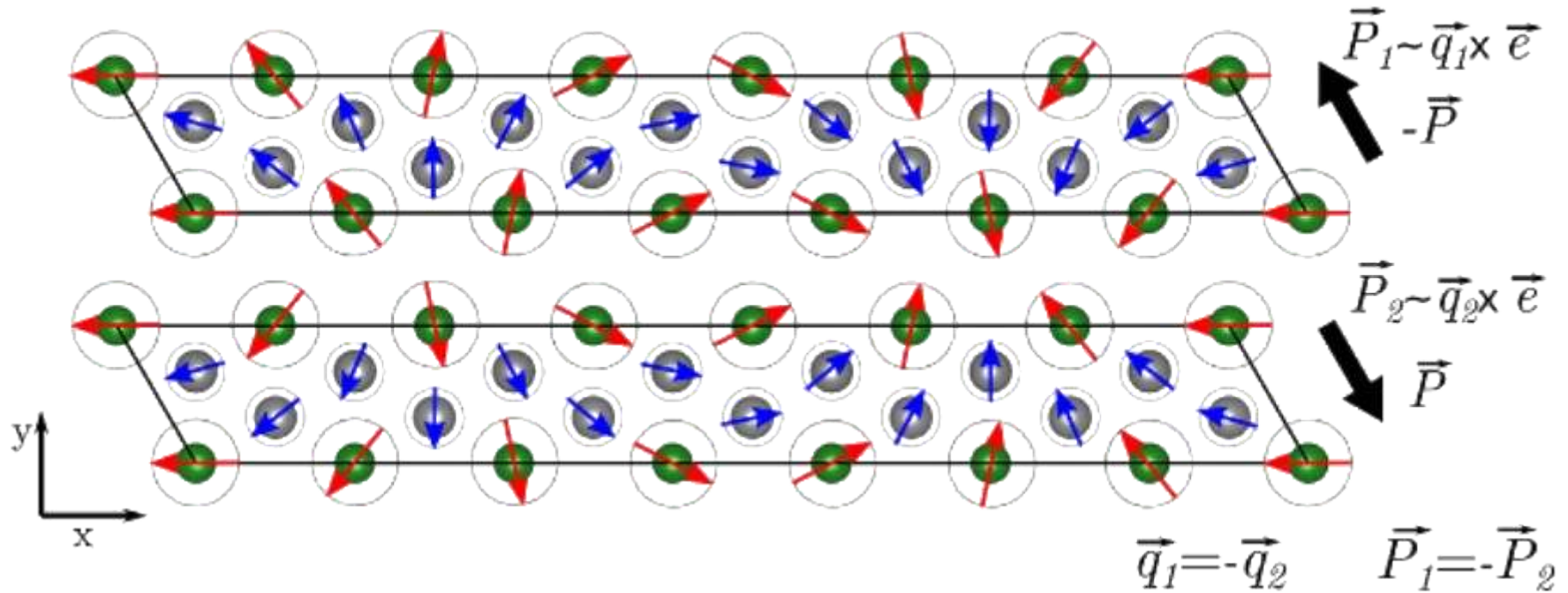
In NiI₂, the ferroelectricity is created by inverse antisymmetric exchange

The interplay between magnetism and halide electronic structure



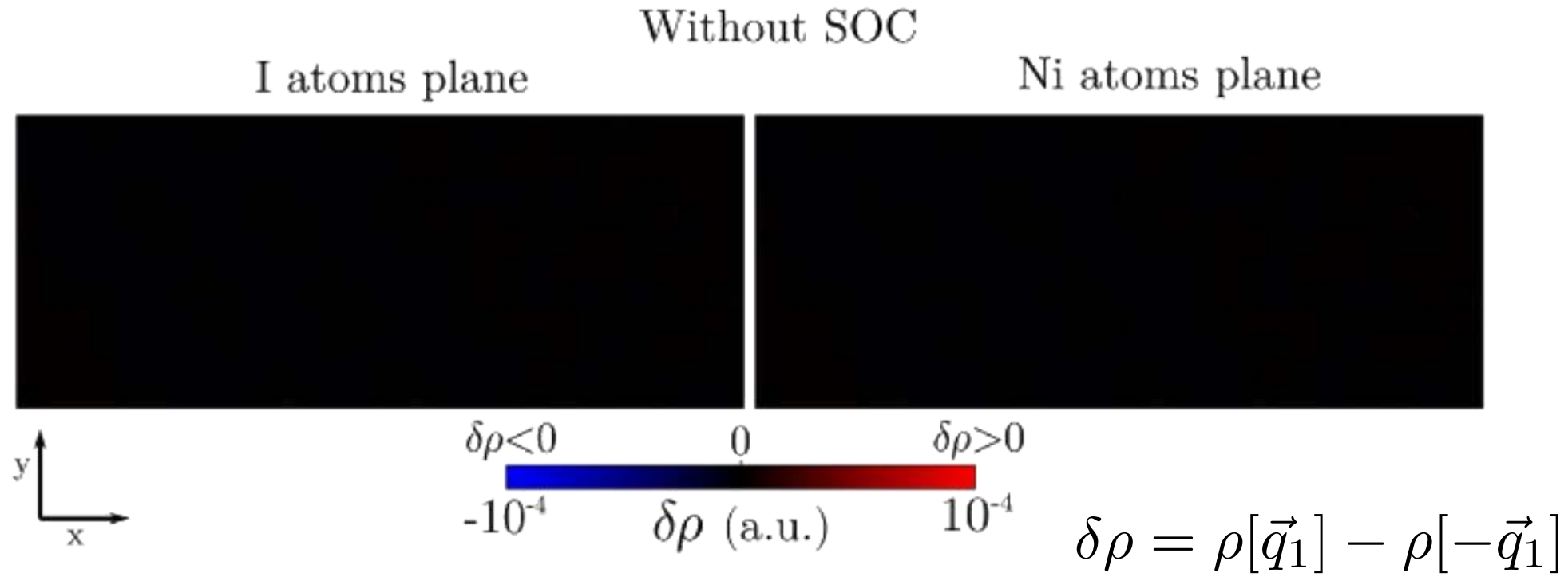
Magnetism not only appears in Ni, but a sizable value merges also in I

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



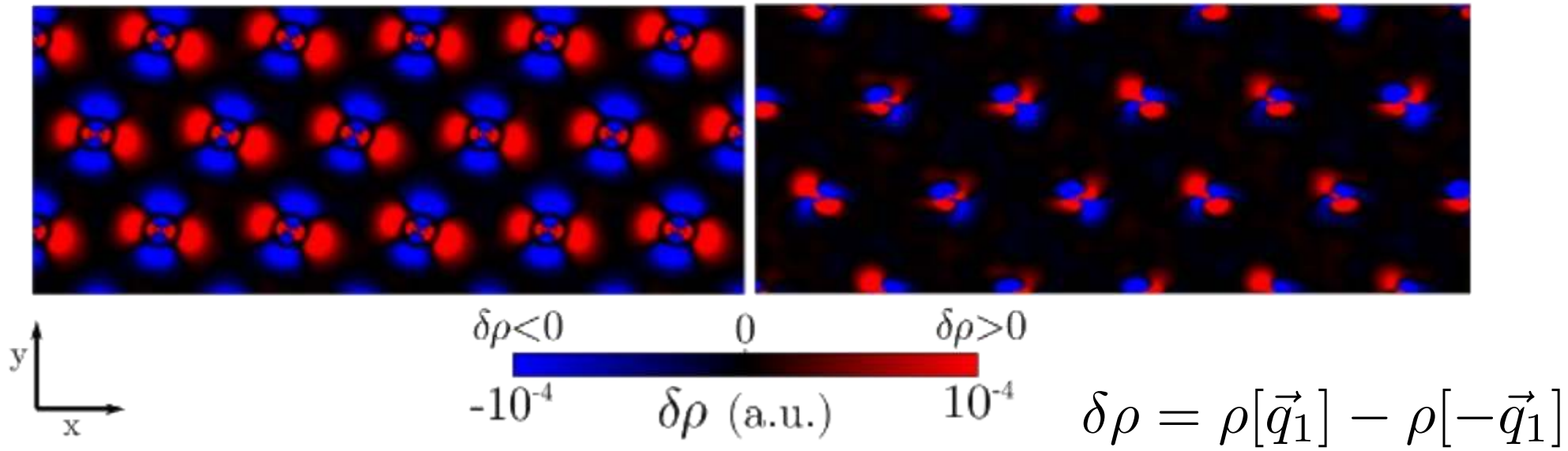
In the absence of SOC, no ferroelectric density appears

Ferroelectric correction to the electronic density

With SOC

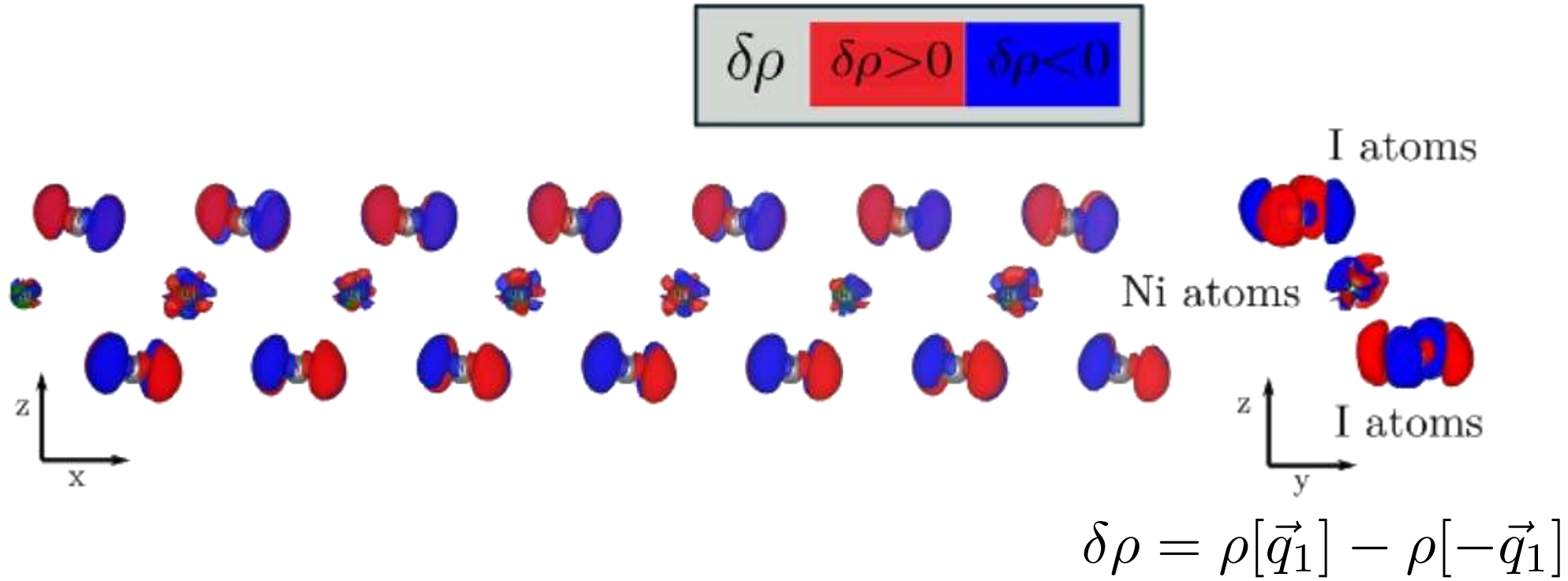
I atoms plane

Ni atoms plane



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

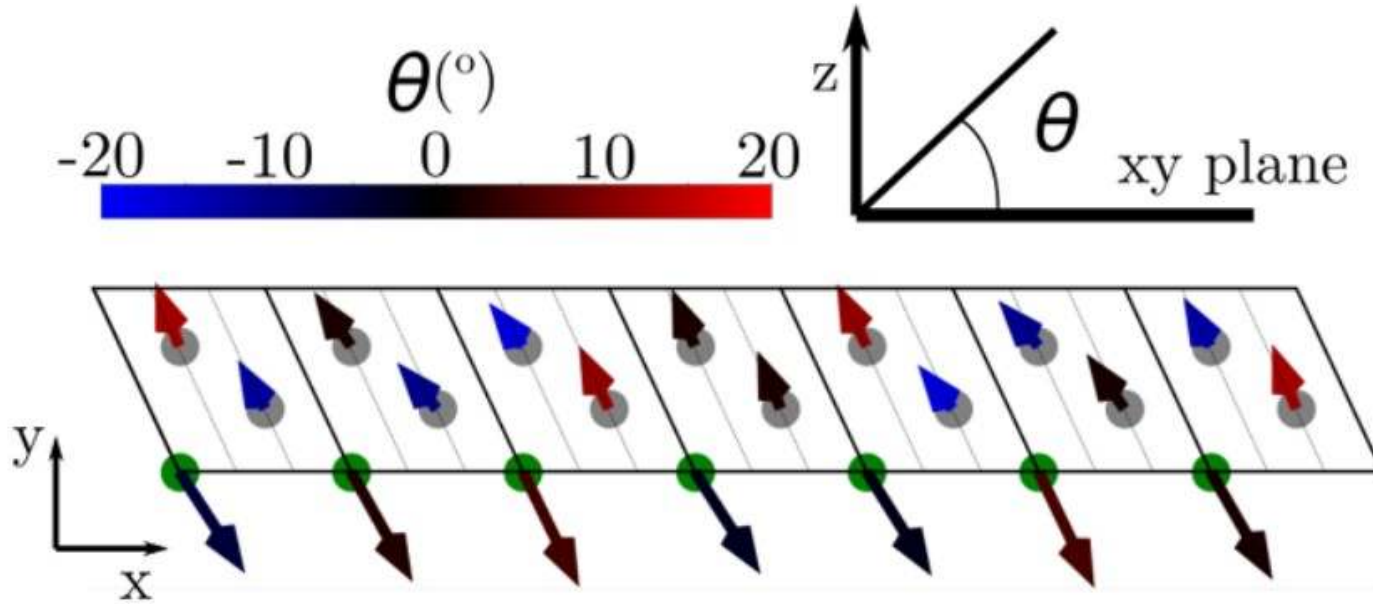
Ferroelectric correction to the electronic density



The ferroelectric density has comparable magnitudes in Ni and I

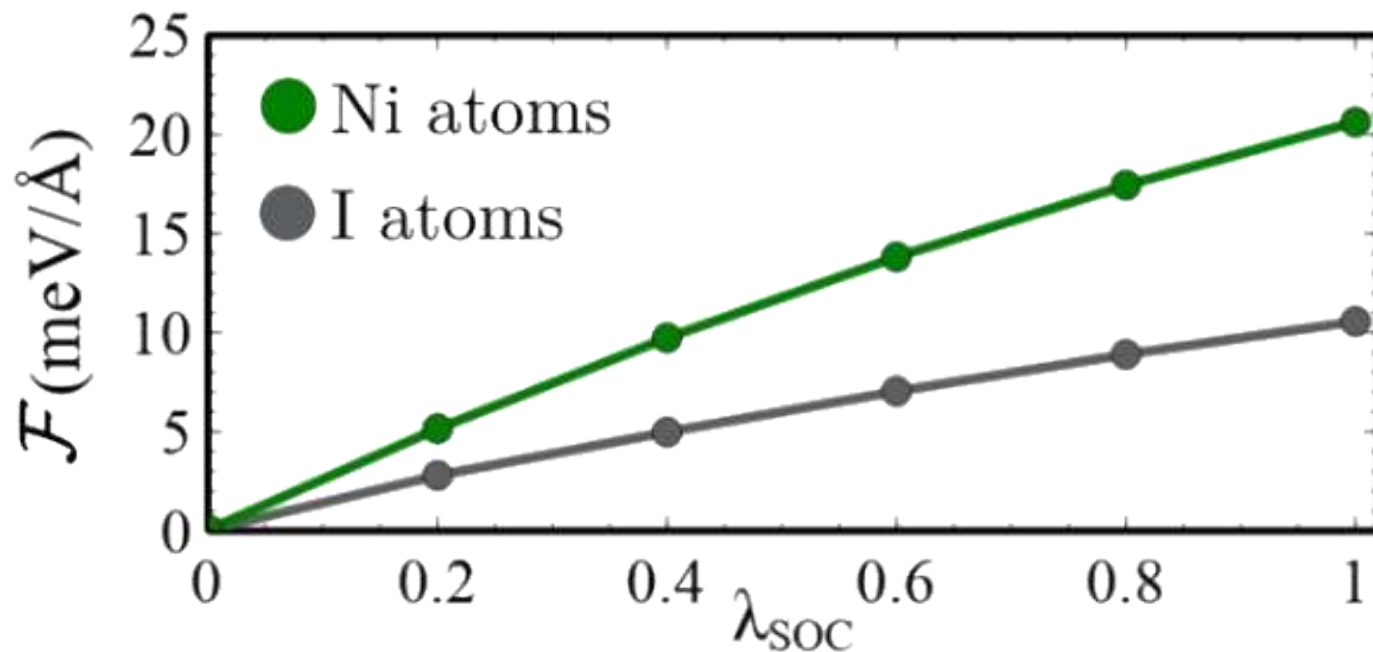
The ferroelectric force

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



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

The ferroelectric force



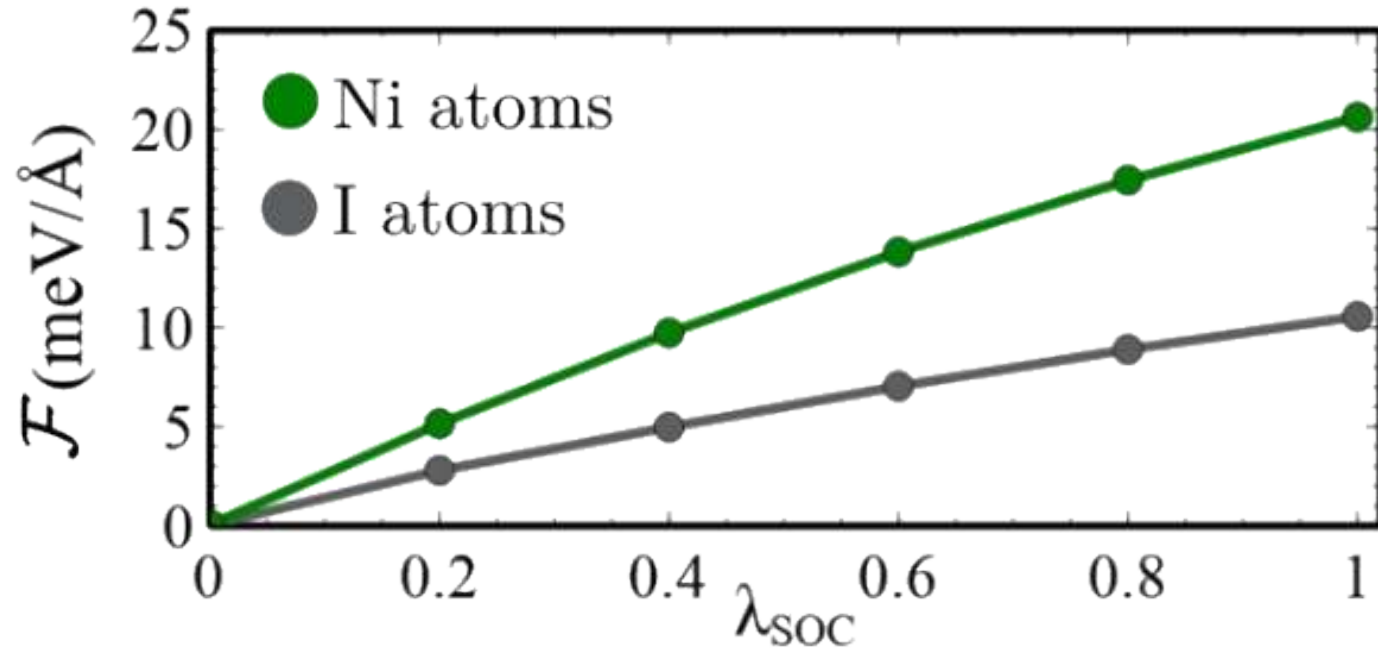
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\text{SOC}} \mathcal{H}_{\text{SOC}},$$

$$\mathcal{F}_{\text{Ni}} = \langle |\mathcal{F}_{\alpha}| \rangle_{\text{Ni}}$$

$$\mathcal{F}_{\text{I}} = \langle |\mathcal{F}_{\alpha}| \rangle_{\text{I}}$$

Both Ni and I contribute to the atomic displacement associated with ferroelectricity

The ferroelectric force



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

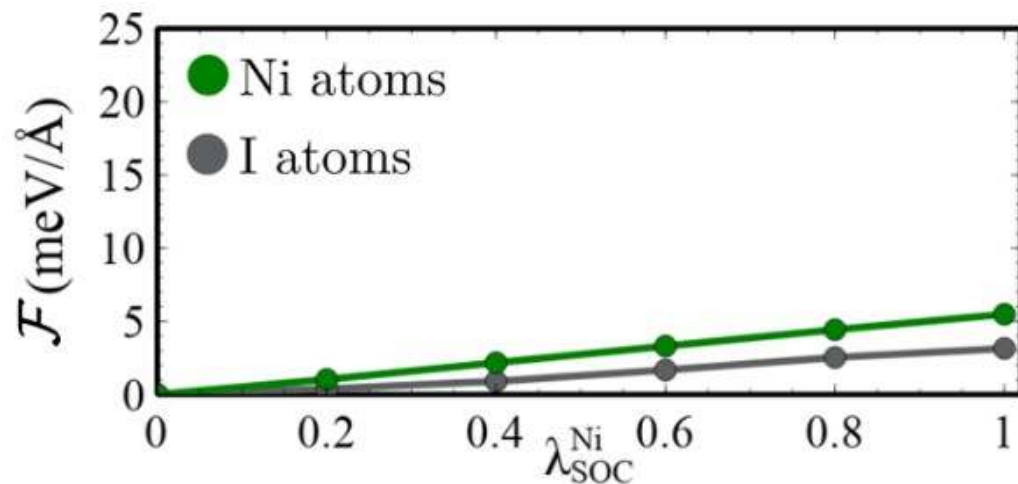
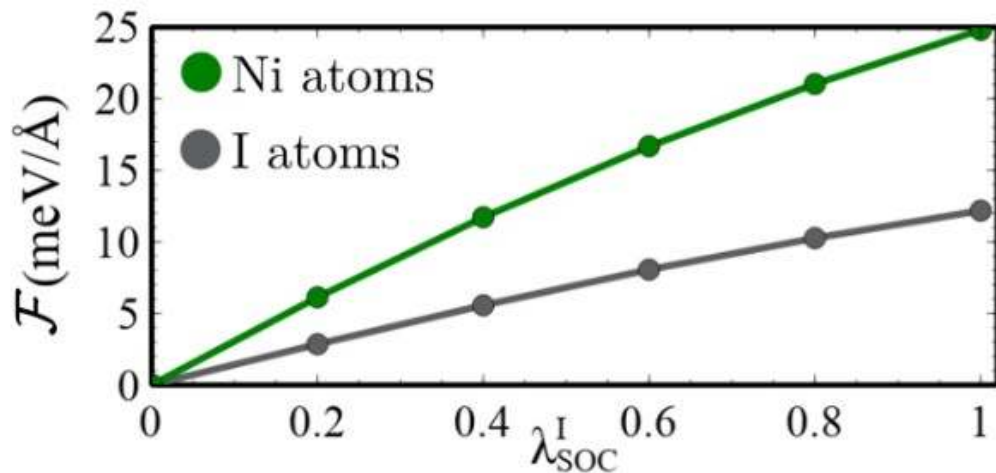
$$\mathcal{F}_{\text{Ni}} = \langle |\mathcal{F}_{\alpha}| \rangle_{\text{Ni}}$$

$$\mathcal{F}_{\text{I}} = \langle |\mathcal{F}_{\alpha}| \rangle_{\text{I}}$$

Both Ni and I contribute to the atomic displacement associated with ferroelectricity

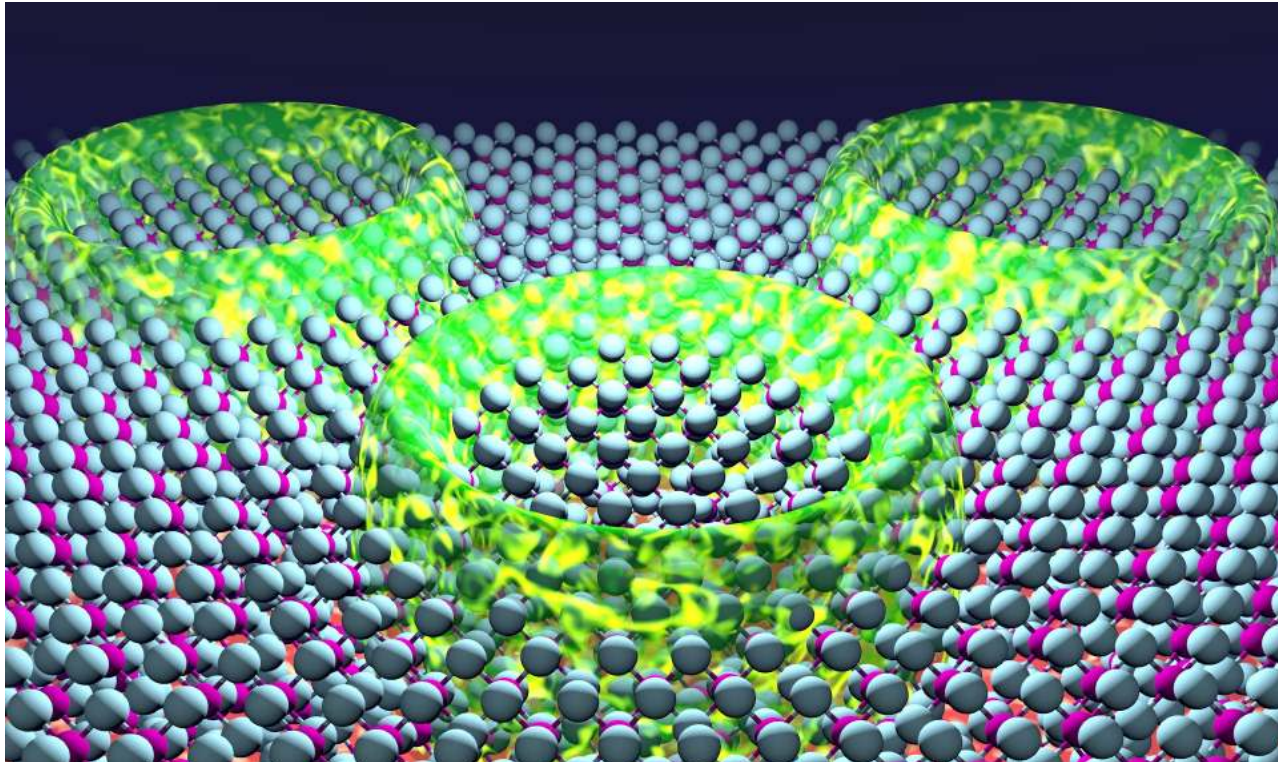
The ferroelectric force

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



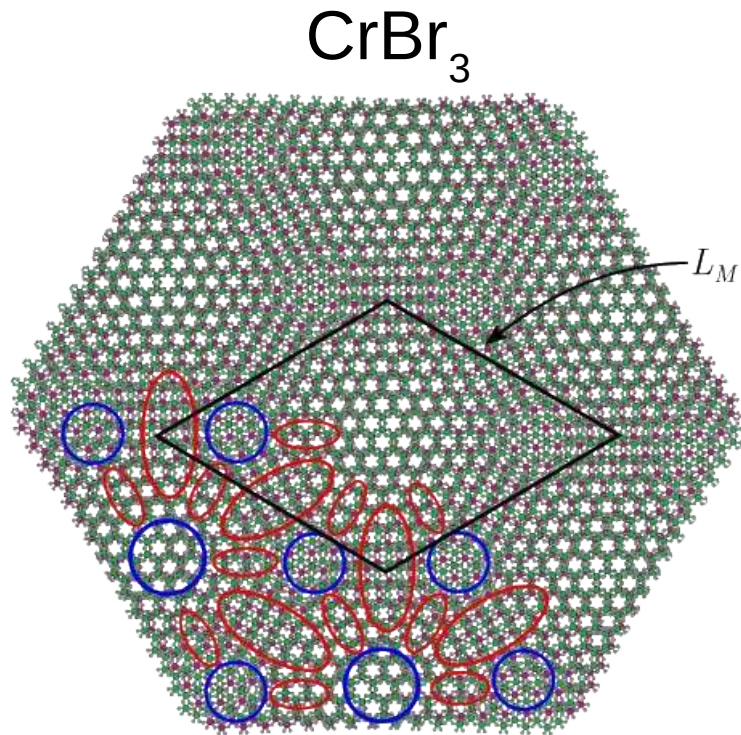
The dominant contribution to the ferroelectric force comes from I

Engineering artificial multiferroics with twisted van der Waals materials



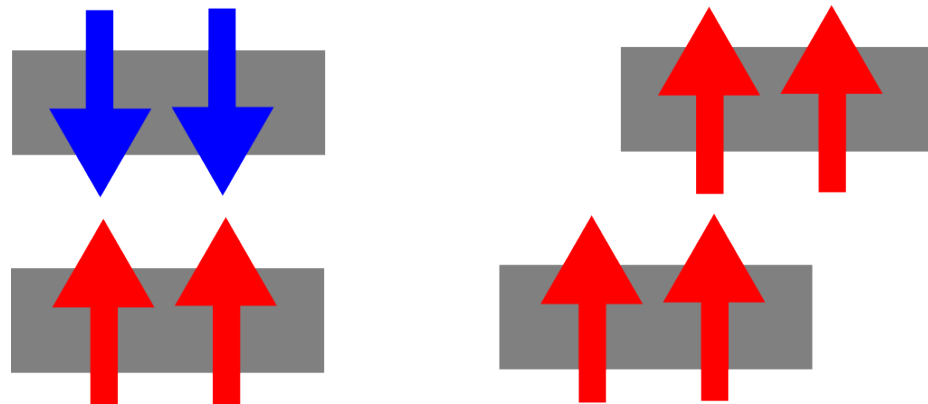
arXiv:2207.01416 (2022)

Twisted 2D trihalides



Bilayer Stacking

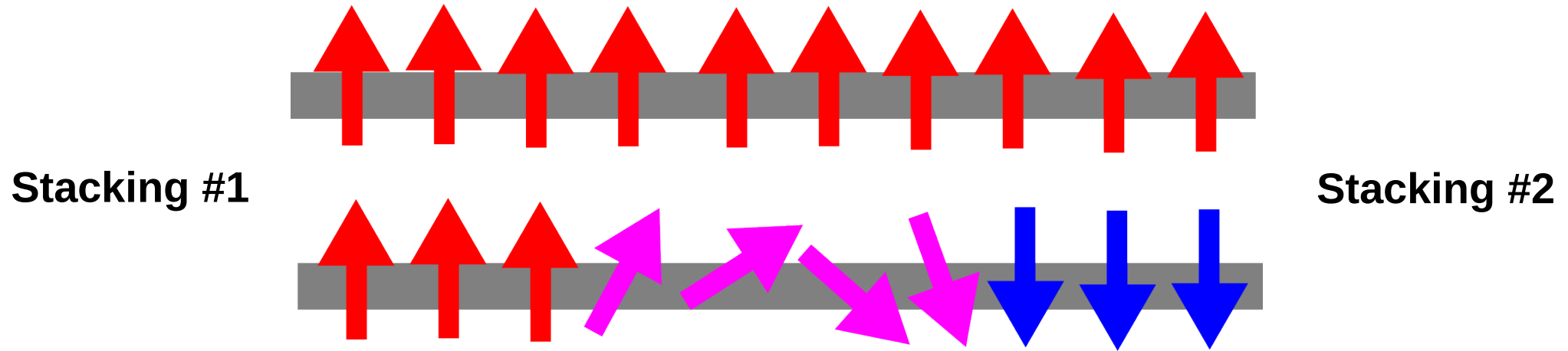
(M)	Monoclinic
(R)	Rhombohedral



The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Mixed FE/AF domains

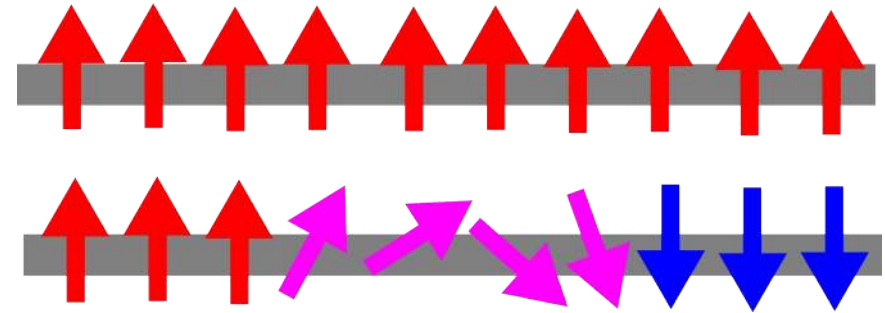
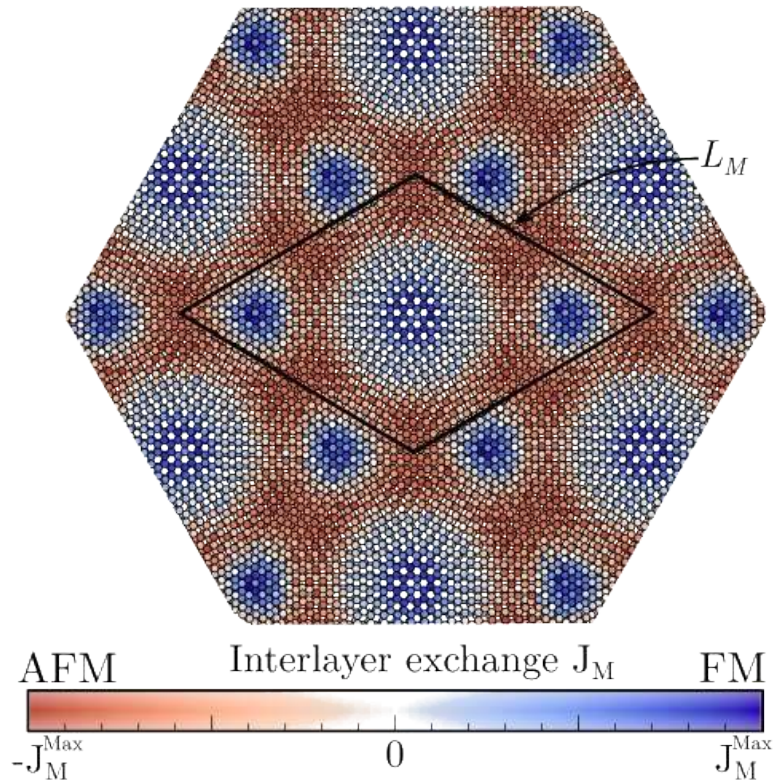
Non-collinear magnetism

Nature Nanotechnology 17, 143–147 (2022)

arXiv:2204.01636

Twisted 2D magnetic materials

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



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

Electric
polarization

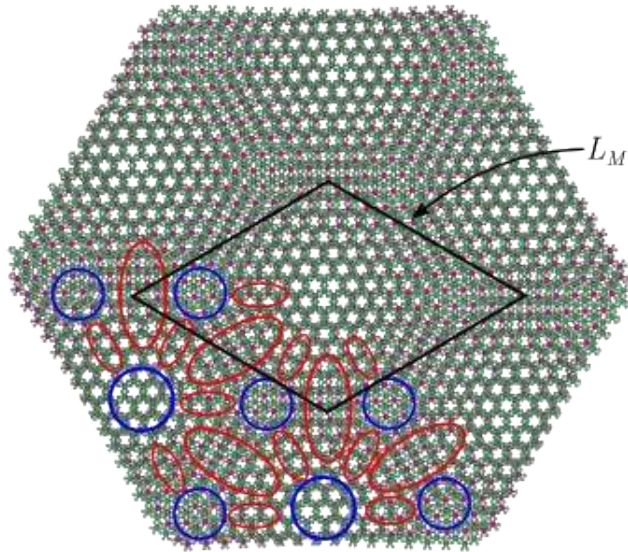
SOC driven

Spin rotation axis

Wavevector of the spin spiral

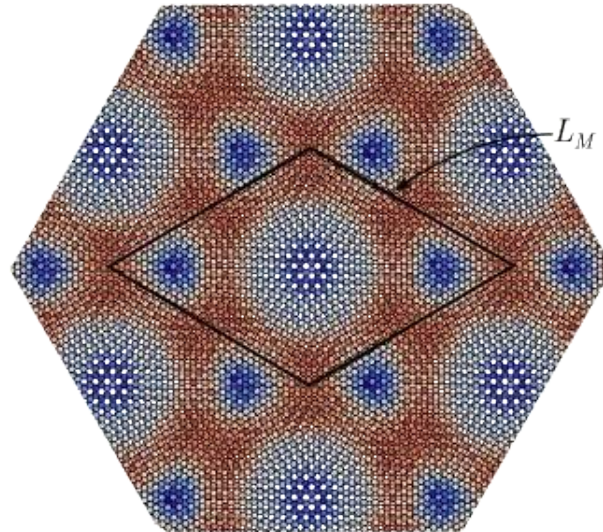
Stacking, magnetic order and electric polarization

Structure



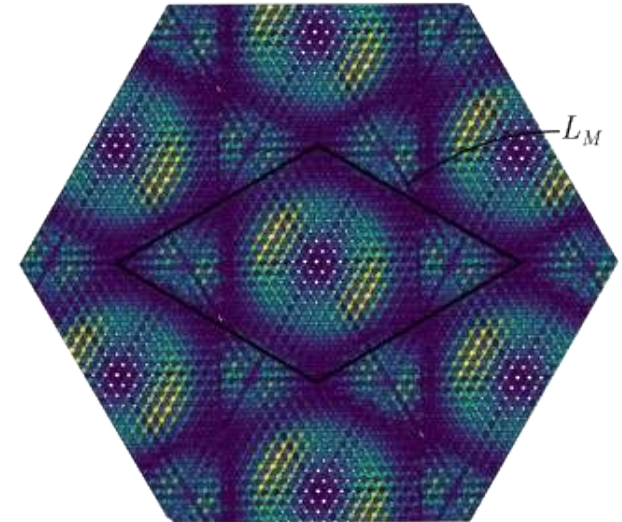
Bilayer Stacking $\left\{ \begin{array}{l} \textcircled{\text{M}} \text{ Monoclinic} \\ \textcircled{\text{R}} \text{ Rhombohedral} \end{array} \right.$

Magnetic order



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

Ferroelectric order

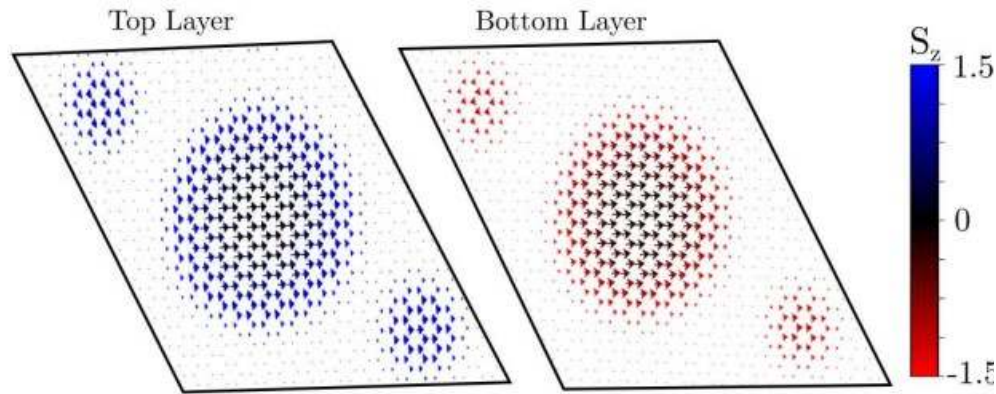


Electric Polarization
0 P^{Max}

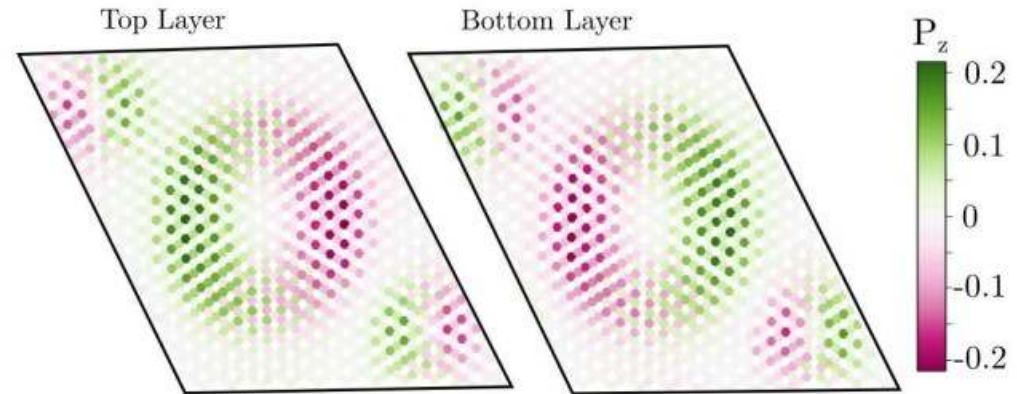
Stacking, magnetic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

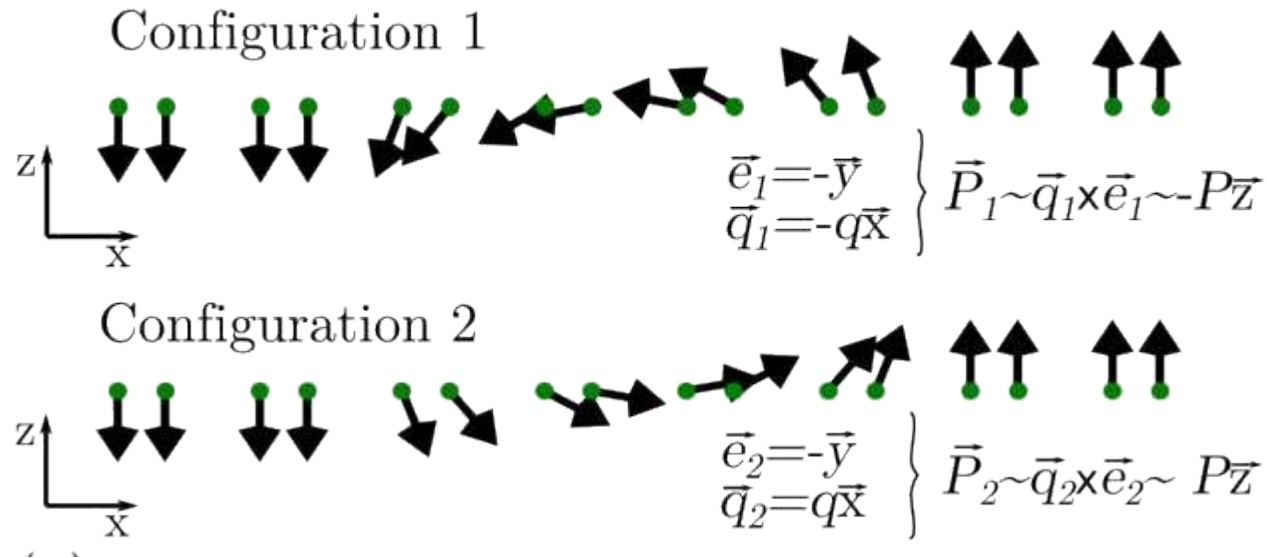


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



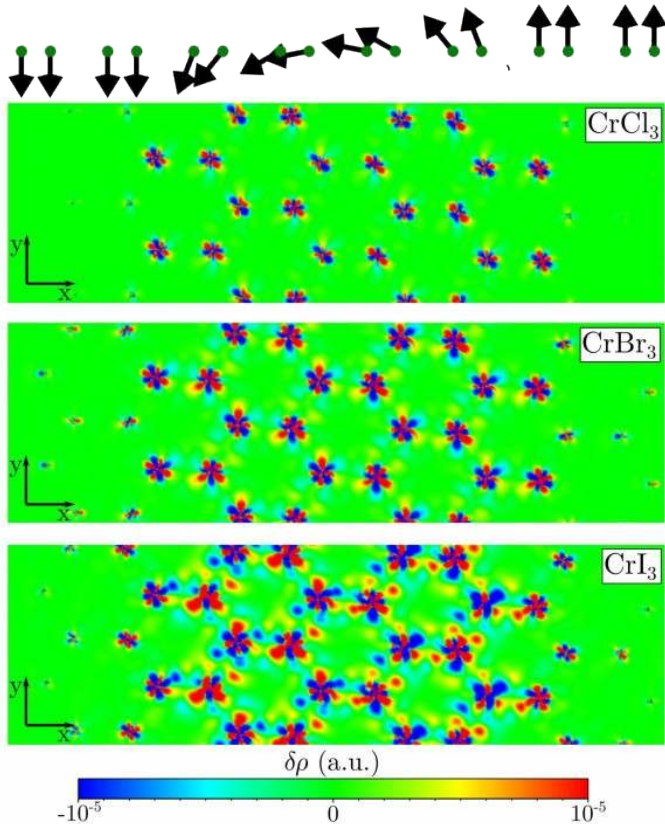
Ferroelectric electronic density

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

Ferroelectric force

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

The ferroelectric electronic correction



Ferroelectric electronic density

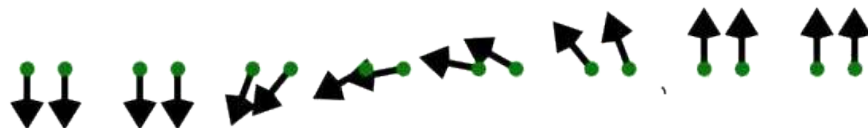
$$\delta\rho = \rho[\mathbf{q}_1] - \rho[\mathbf{q}_2]$$

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

It becomes stronger the heavier the halide

The ferroelectric force

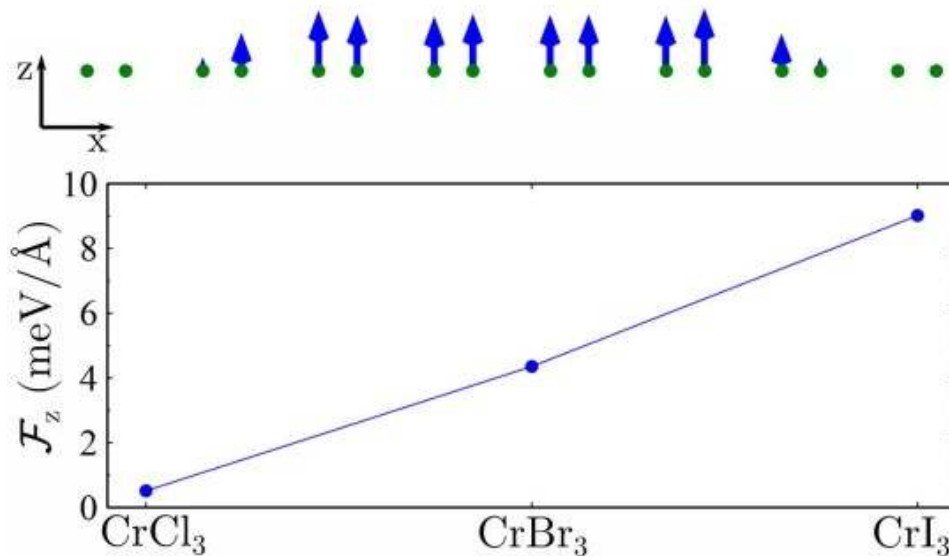
Magnetic configuration



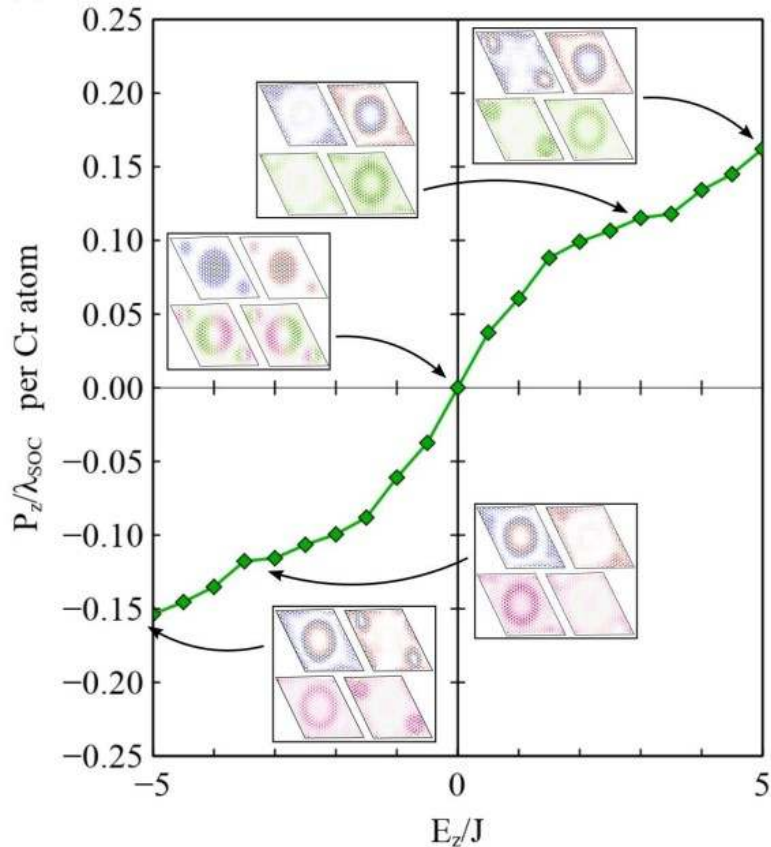
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



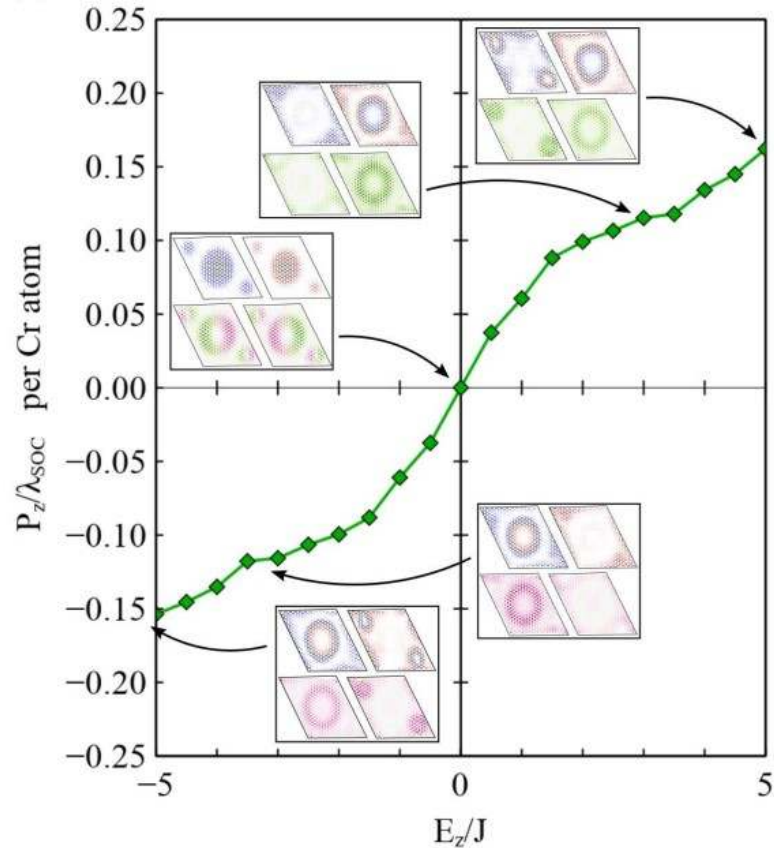
Electrically controllable skyrmions in twisted magnets



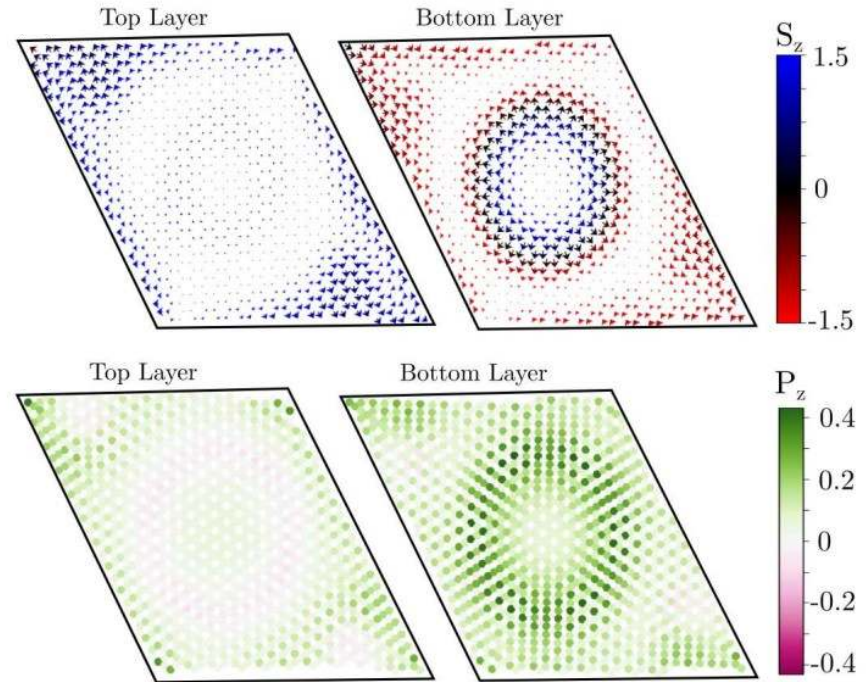
The electric polarization directly couples to an out-of plane electric field

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

Electrically controllable skyrmions in twisted magnets

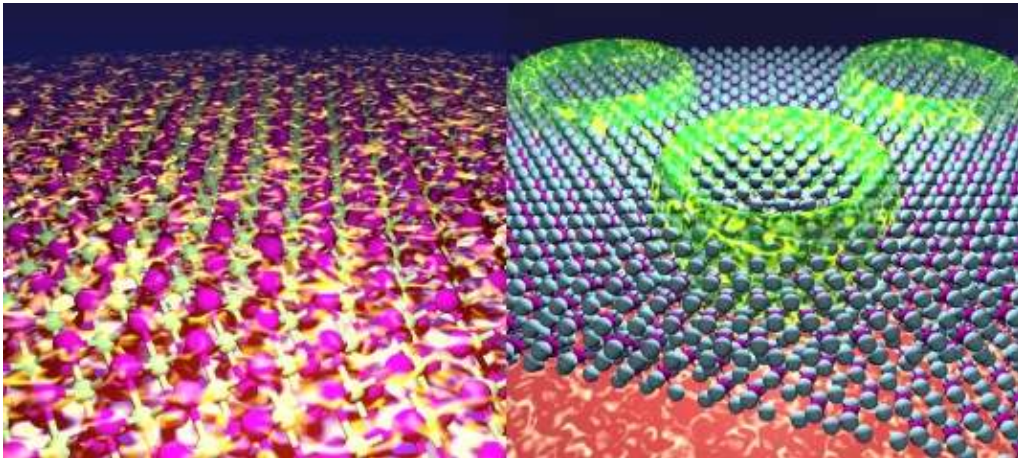


Skyrmions at finite interlayer bias



Take home

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



2D Materials 9 (2), 025010 (2022)
arXiv:2207.01416 (2022)

Thank you

Funding from

