Artificial van der Waals multiferroics with twisted two-dimensional materials

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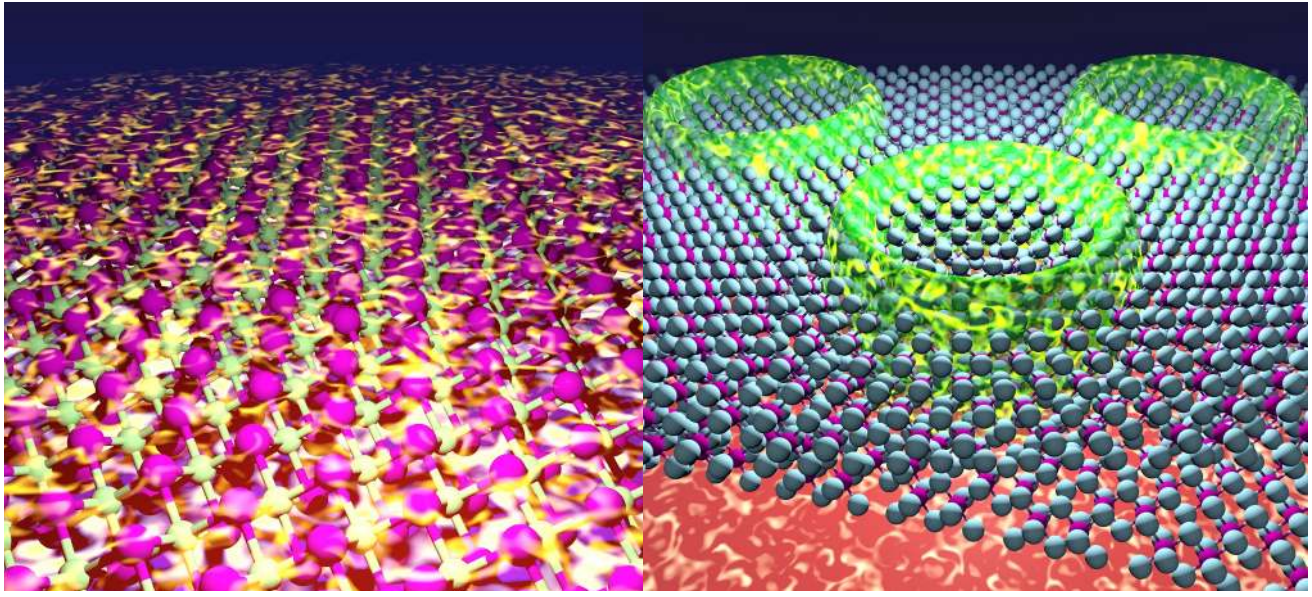


2D Materials 9 (2), 025010 (2022)

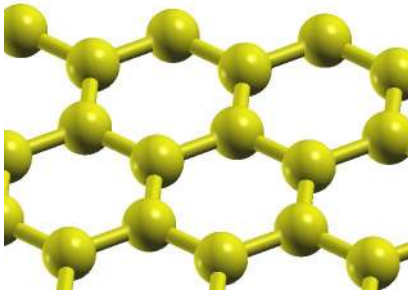
2D Materials 10 (2), 025026 (2023)

36th European Conference on Surface Science, Lodz, Poland, August 30th 2023

The two-dimensional materials world

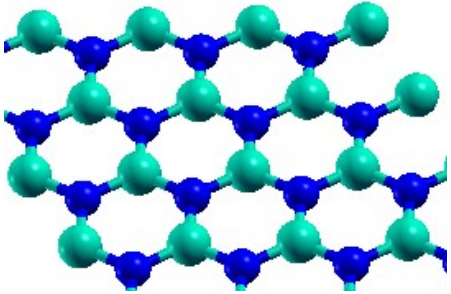
Semimetal

Graphene



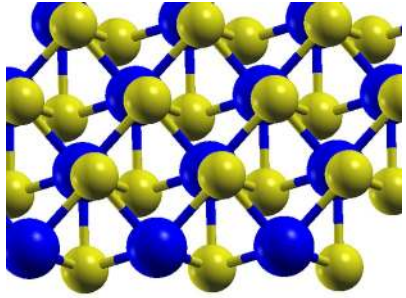
Insulator

BN



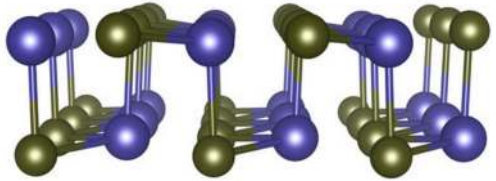
Superconductor

$NbSe_2$



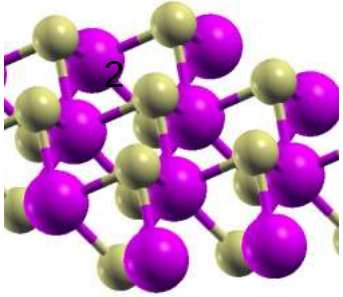
Ferroelectric

SnTe



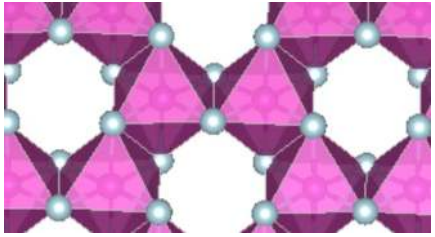
Semiconductor

WSe



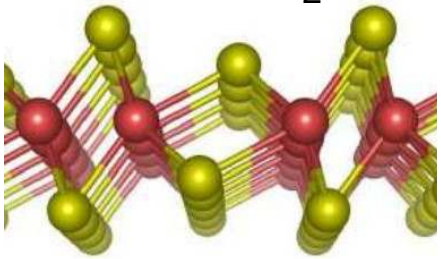
Ferromagnet

CrI_3



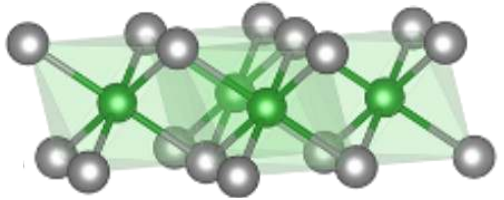
Quantum spin Hall insulator

WTe_2



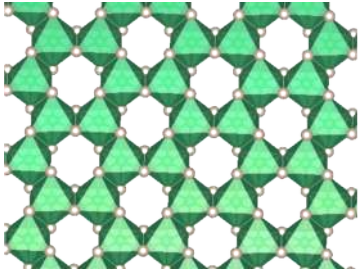
Multiferroic

NiI_2



Van der Waals magnetic materials

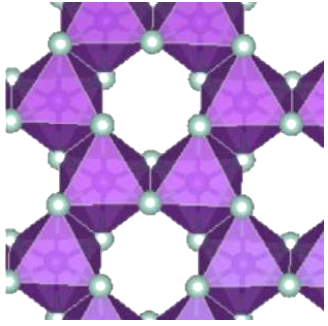
Ferromagnet, antiferromagnets



CrI_3, $CrCl_3$, $CrBr_3$

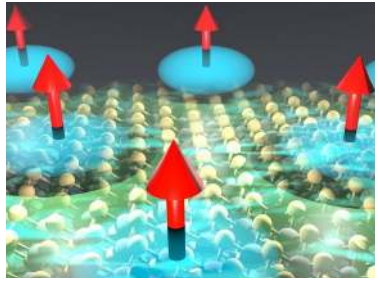
Nature 546, 270–273 (2017)

(proximal) Quantum spin-liquids



$RuCl_3$, 1T-TaS_2

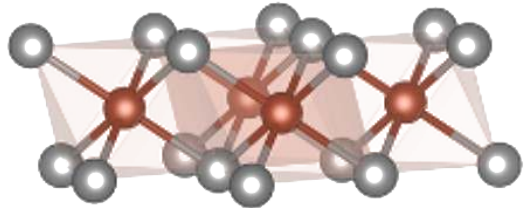
Heavy-fermion Kondo insulators



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)

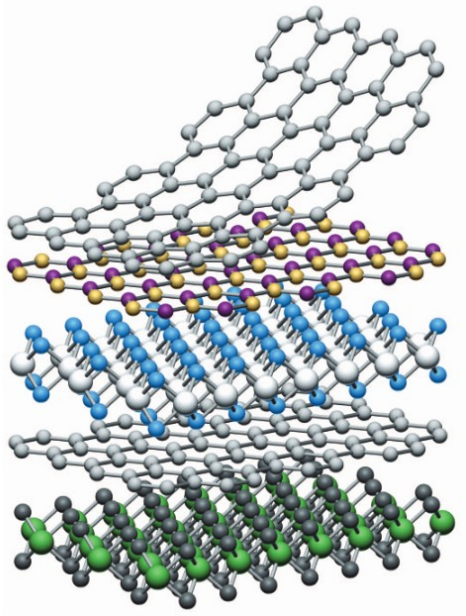
Break time-reversal

Do not break time-reversal

Break time-reversal and inversion symmetry

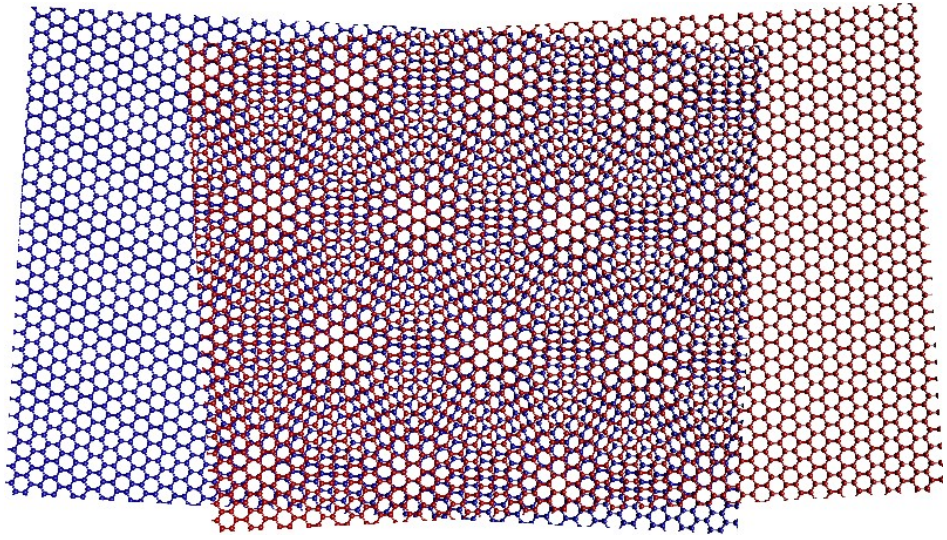
The flexibility of two-dimensional materials

They can be stacked



Nature 499, 419–425 (2013)

They can be rotated

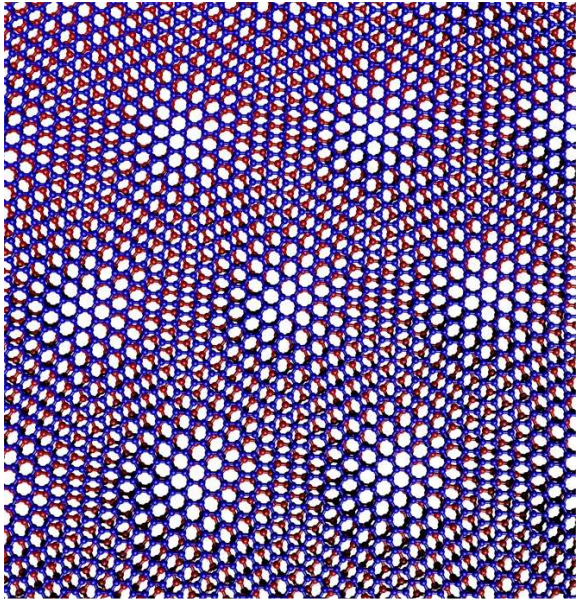


Science 361, 6403, 690-693 (2018)

These are unique features of two-dimensional materials

The flexibility of twist engineering

Twisted bilayer graphene



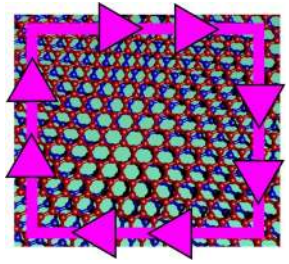
Superconductivity

Nature 556, 43–50 (2018)

Topological networks

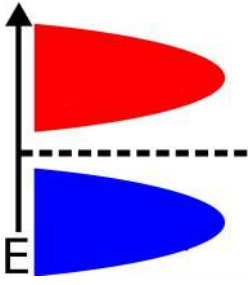
Nano Lett. 18, 11, 6725-6730 (2018)

Chern insulators



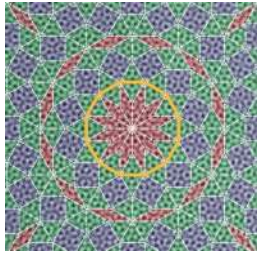
Science 365, 605-608 (2019)

Correlated insulators



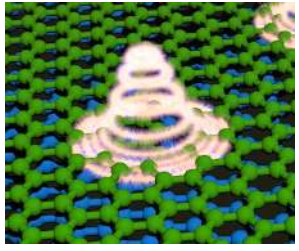
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782-786 (2018)

Fractional Chern insulators

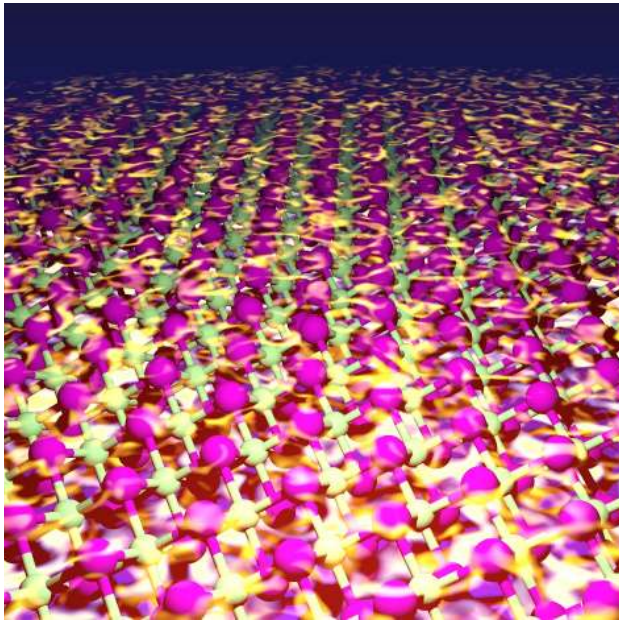


Nature 600, 439–443 (2021)

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

Two strategies for multiferroicity in van der Waals materials

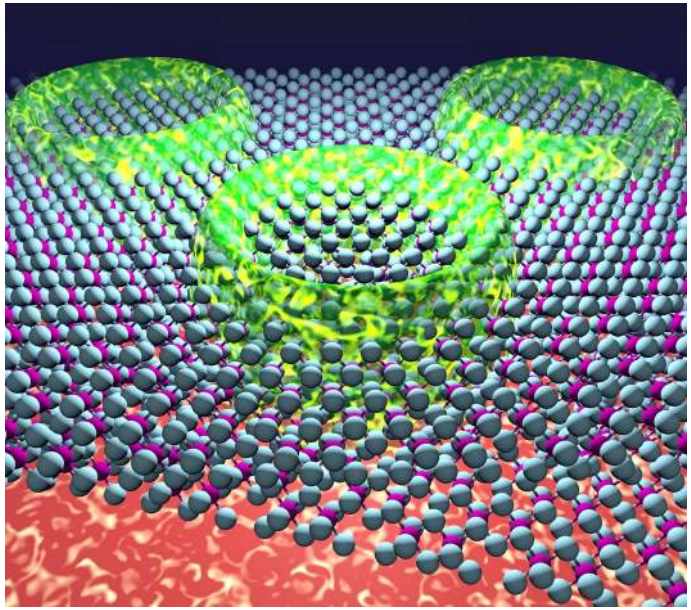
Multiferroic monolayer NiI_2



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

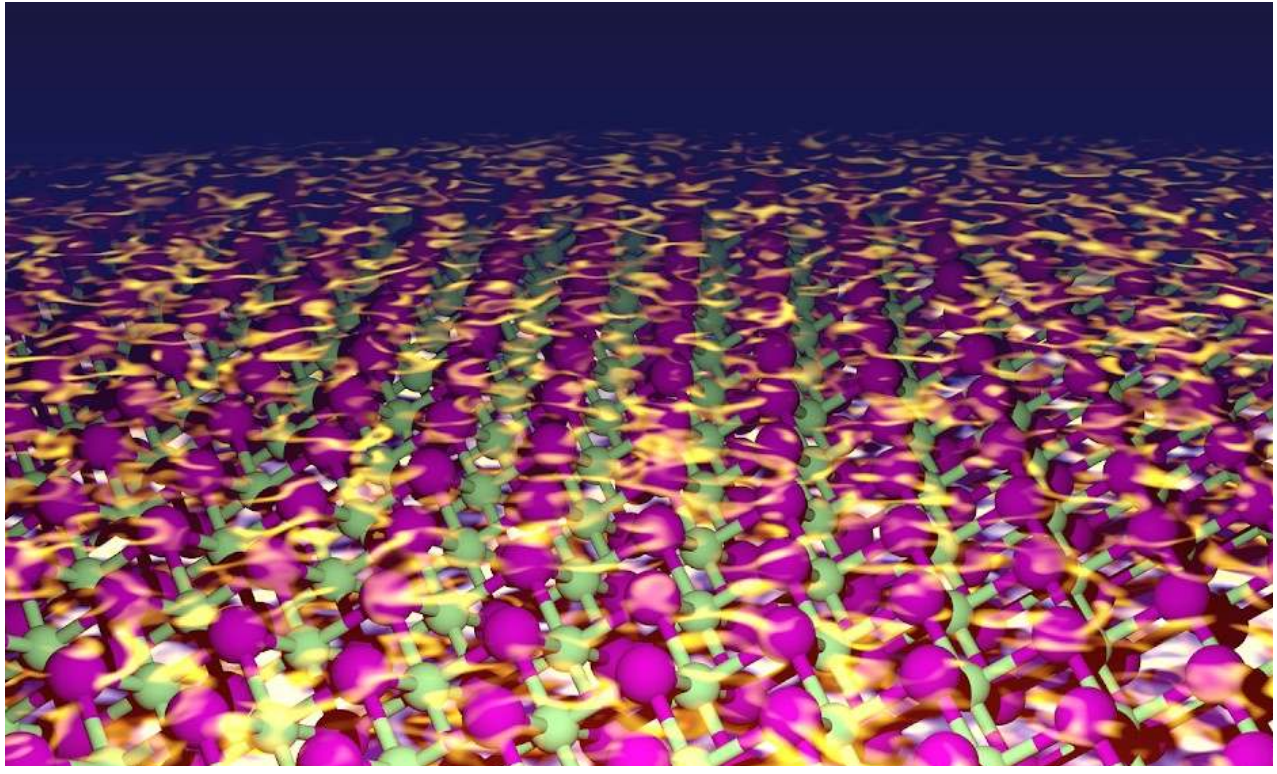
Adolfo Fumega

Artificial multiferroics via twist engineering with $CrBr_3$ bilayers



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

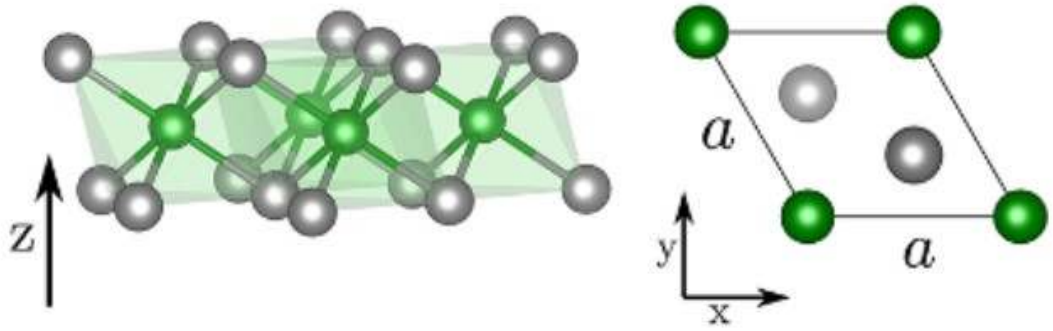
The origin of multiferroic order in NiI_2



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

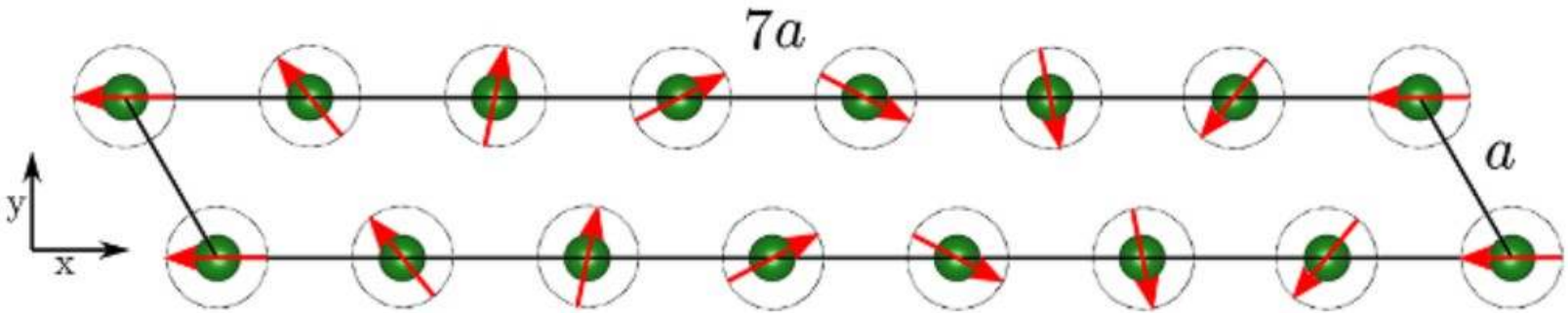
Monolayer NiI_2

Structure



Nature 602, 601–605 (2022)

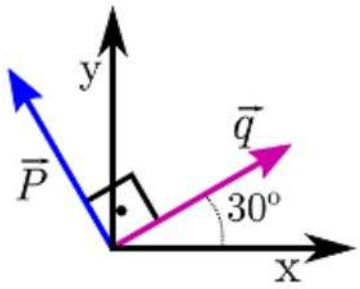
Magnetic structure



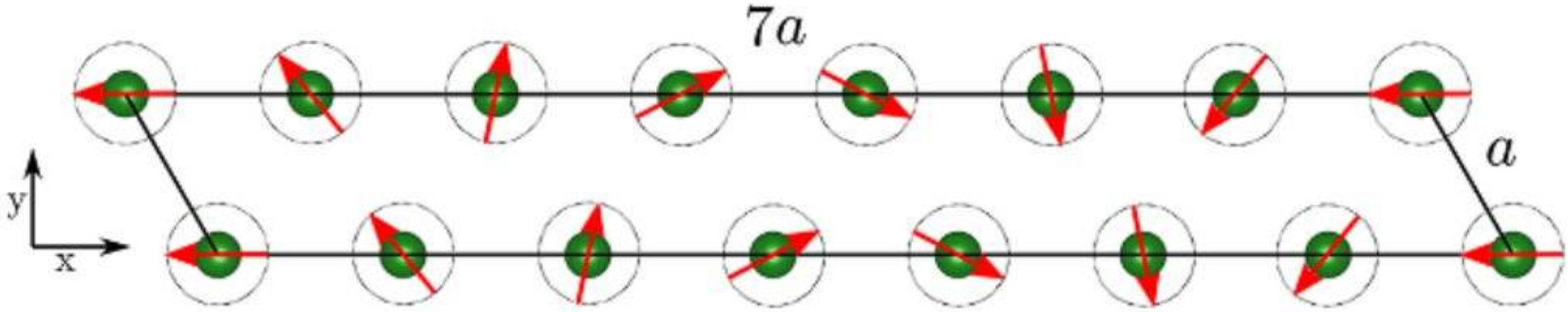
The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

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

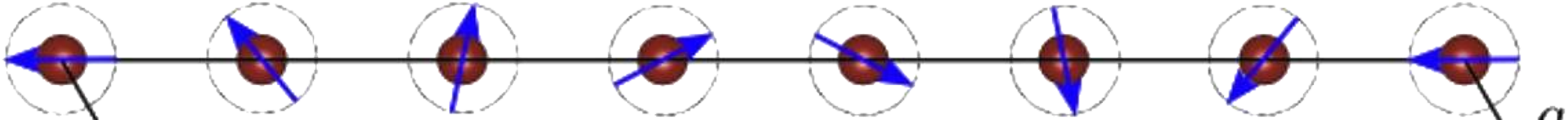


$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e}\cdot\mathbf{S})(\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}_0))}\mathbf{M}(\mathbf{r}_0)$$



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

The origin of ferroelectricity in NiI_2



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

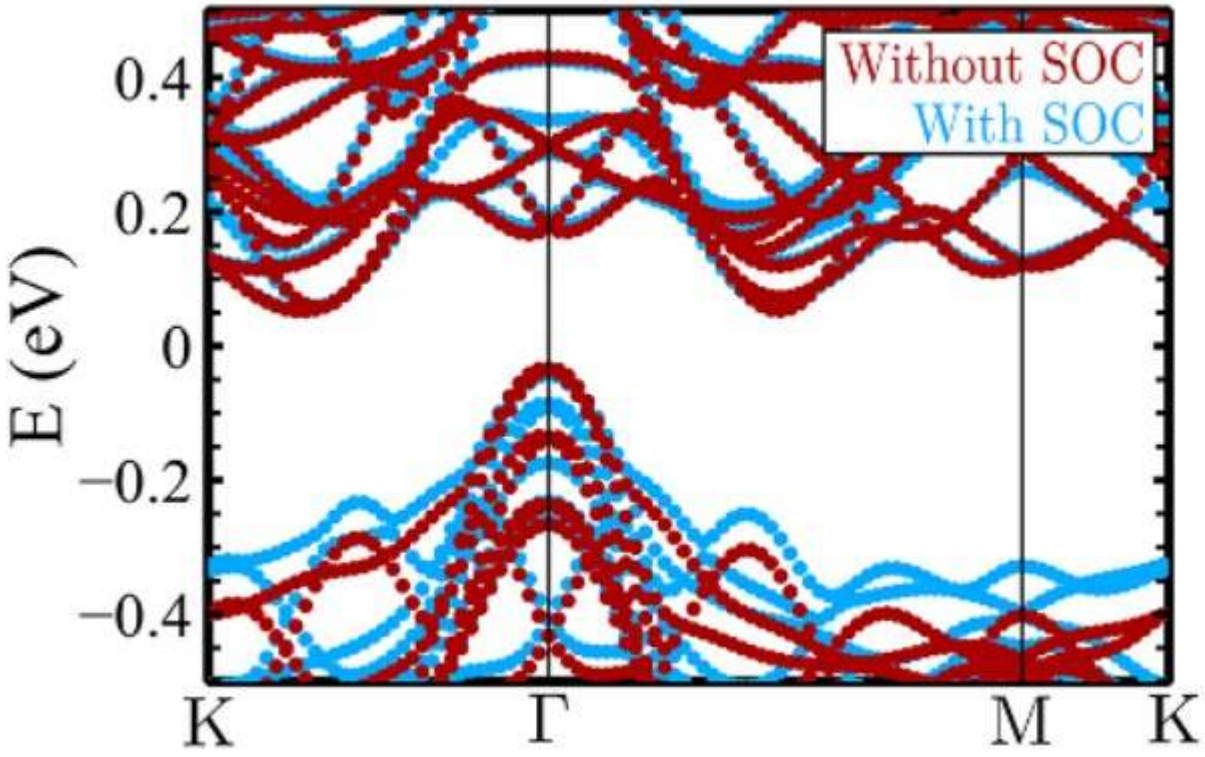
Electric polarization

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

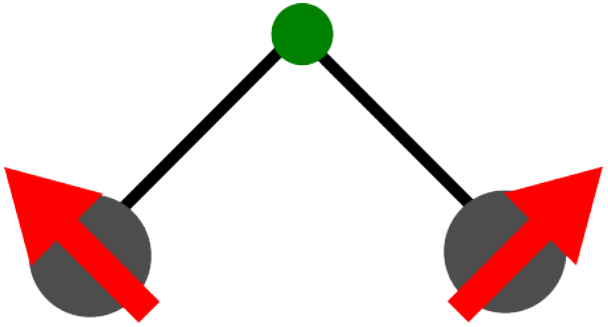
There is a major role of the halide close to the Fermi level

The impact of spin-orbit coupling in magnetic ordering

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

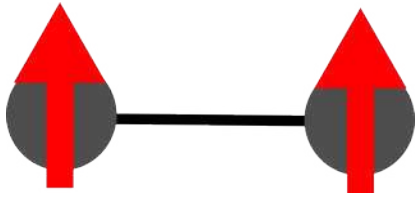
Antisymmetric exchange

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



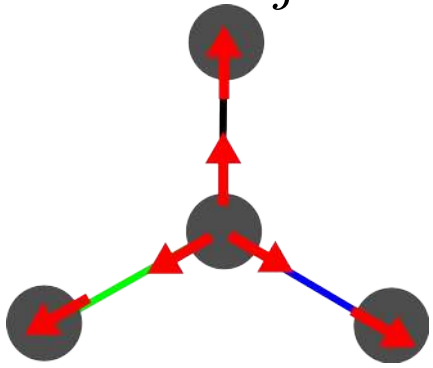
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

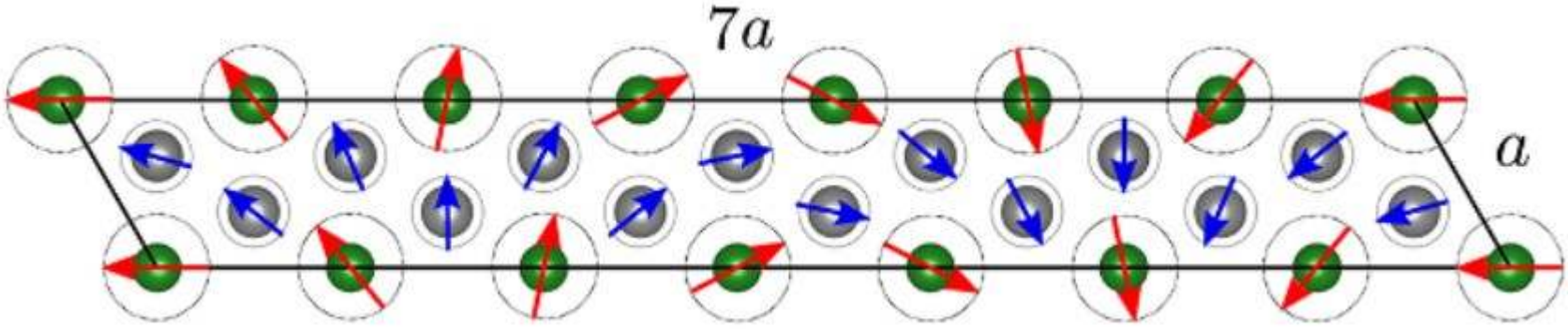
$$S_i^{\alpha(i)} S_j^{\alpha(j)}$$



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

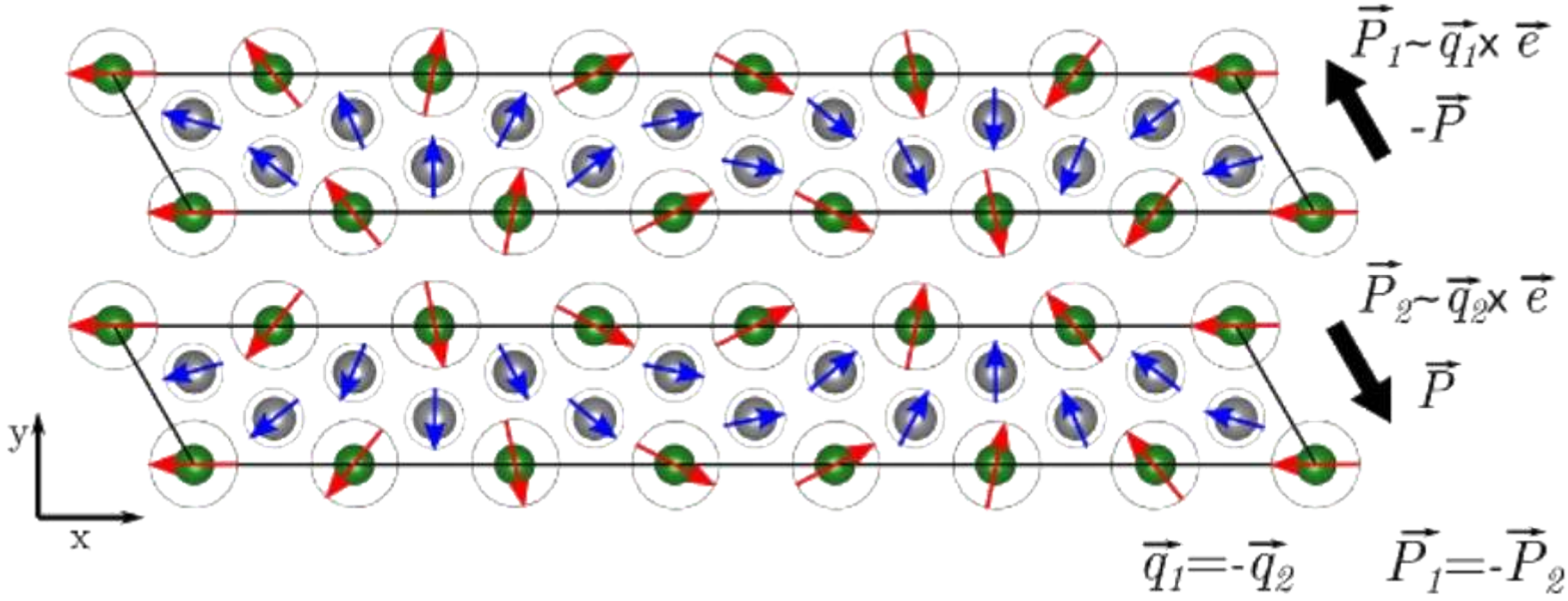
In NiI_2, the ferroelectricity is created by inverse antisymmetric exchange

The interplay between magnetism and halide electronic structure



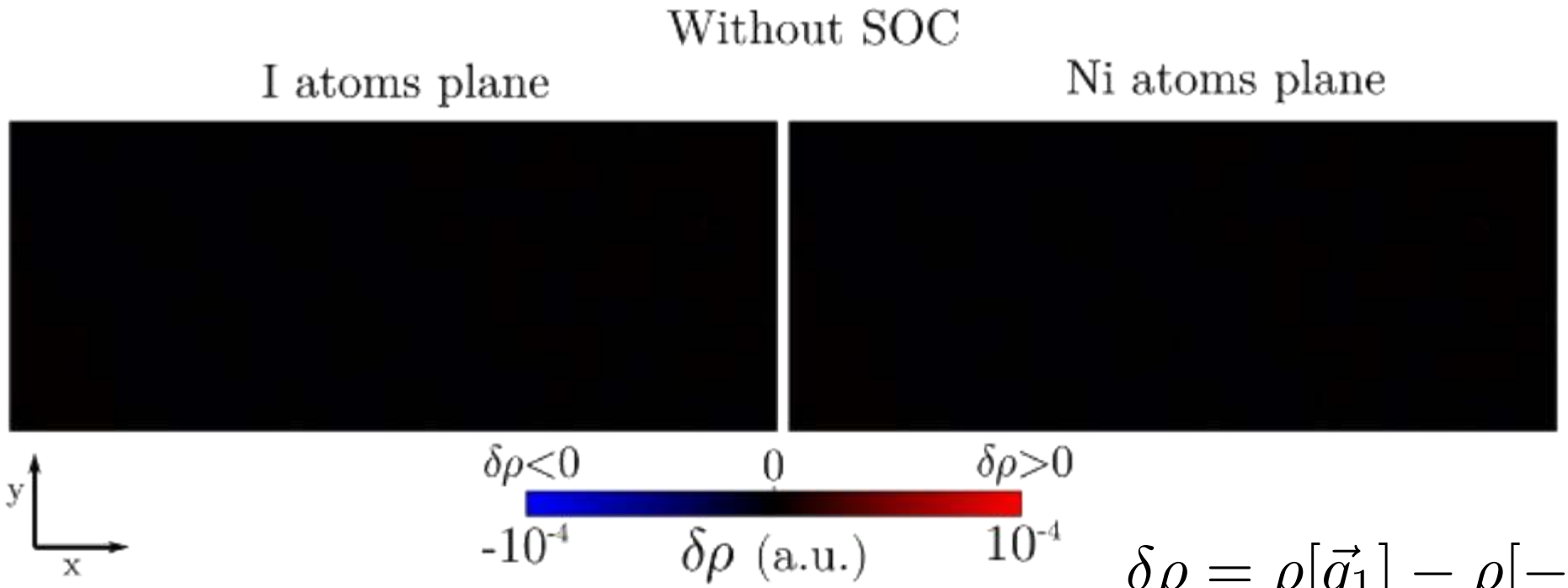
Magnetism not only appears in Ni, but a sizable value merges also in I

Ferroelectric correction to the electronic density



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

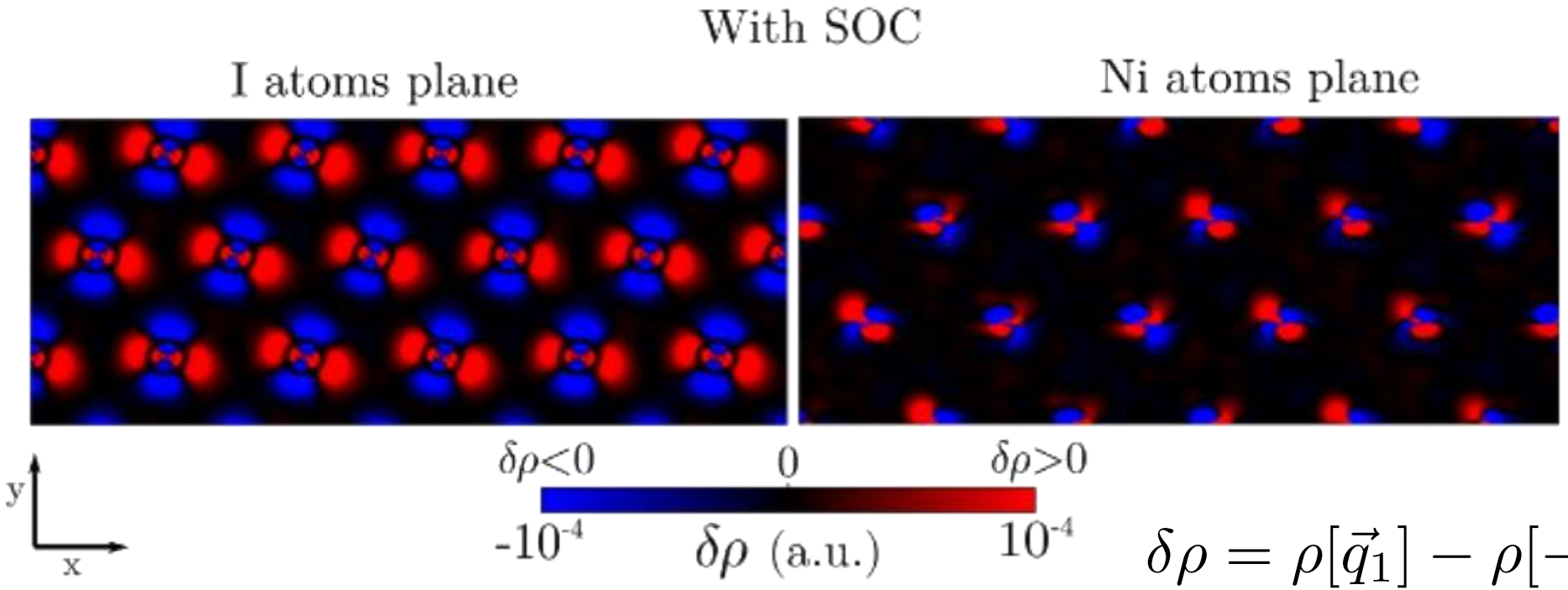
Ferroelectric correction to the electronic density



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

In the absence of SOC, no ferroelectric density appears

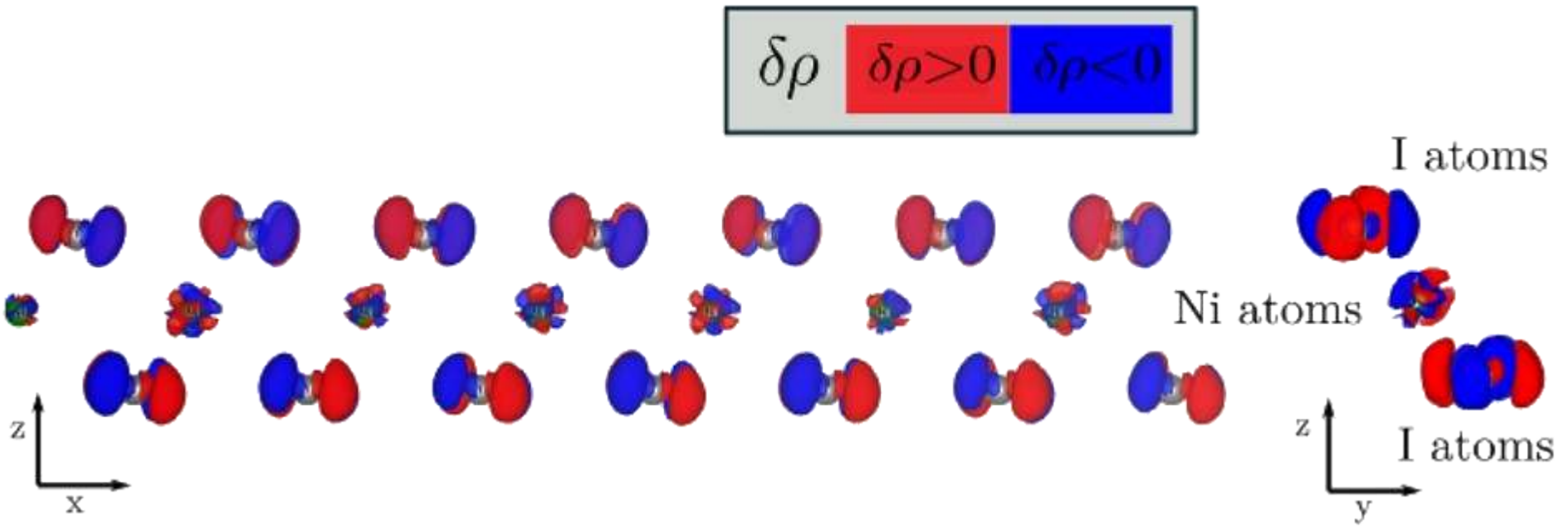
Ferroelectric correction to the electronic density



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

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

Ferroelectric correction to the electronic density

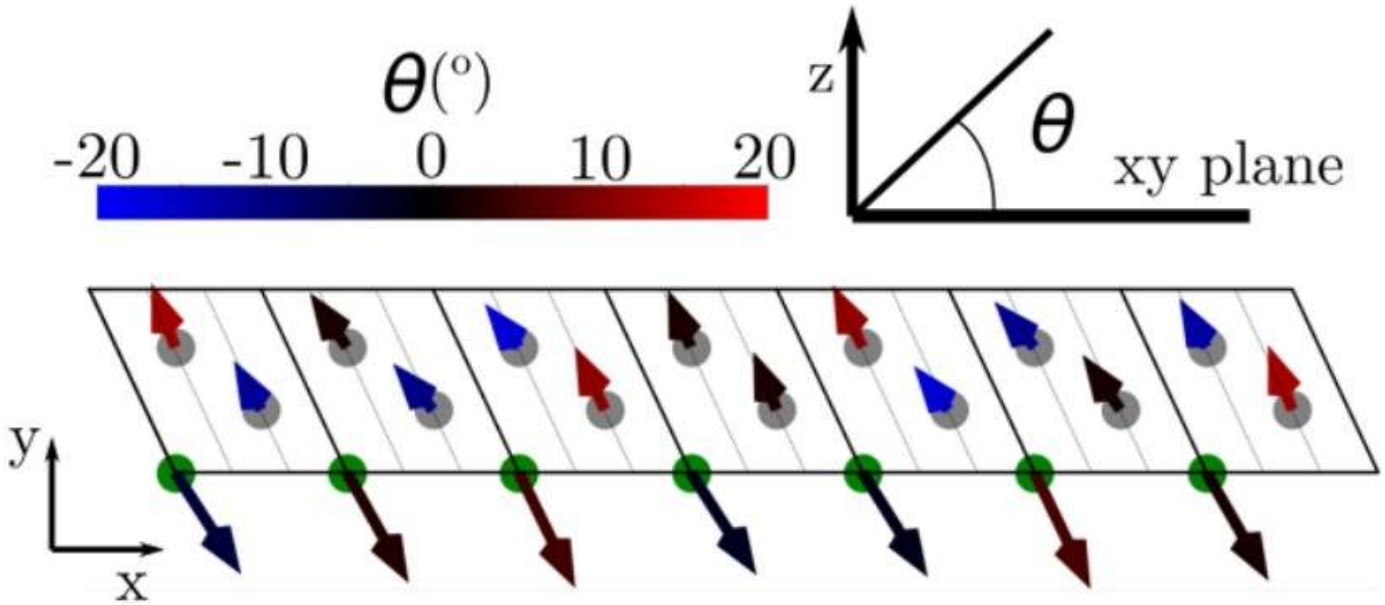


$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

The ferroelectric density has comparable magnitudes in Ni and I

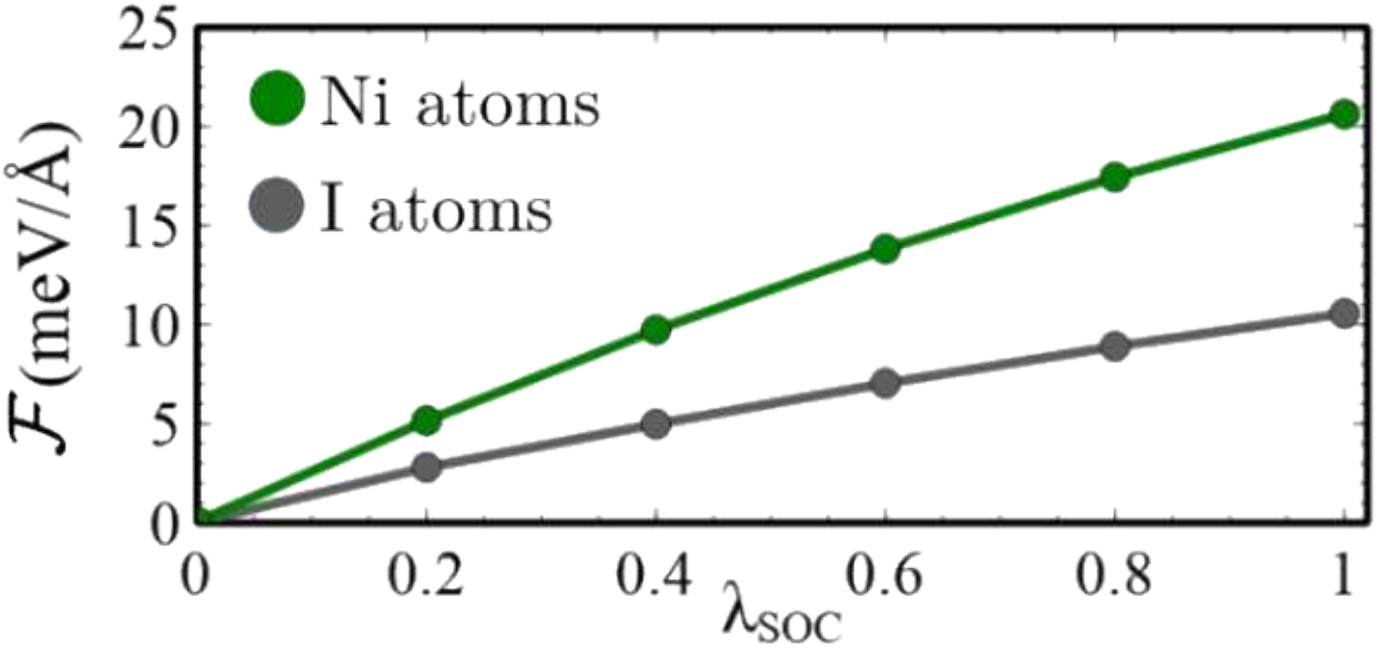
The ferroelectric force

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



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

The ferroelectric force



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

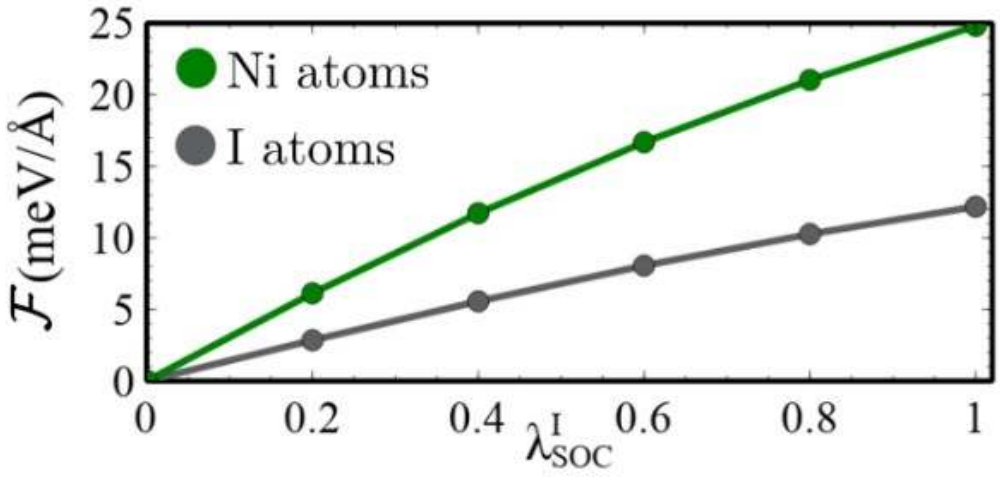
$$\mathcal{F}_{\mathrm{Ni}} = \langle |\boldsymbol{\mathcal{F}}_\alpha| \rangle_{\mathrm{Ni}}$$

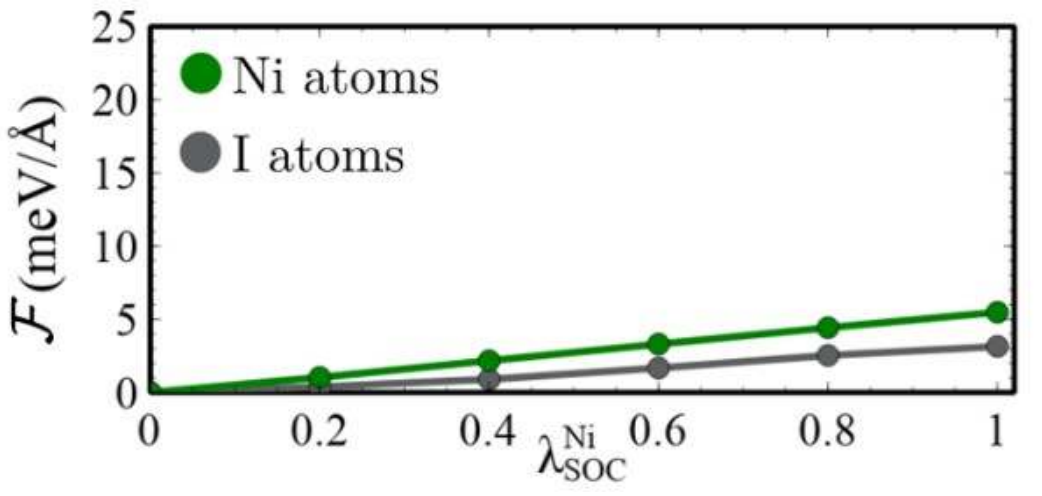
$$\mathcal{F}_{\mathrm{I}} = \langle |\boldsymbol{\mathcal{F}}_\alpha| \rangle_{\mathrm{I}}$$

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

The ferroelectric force

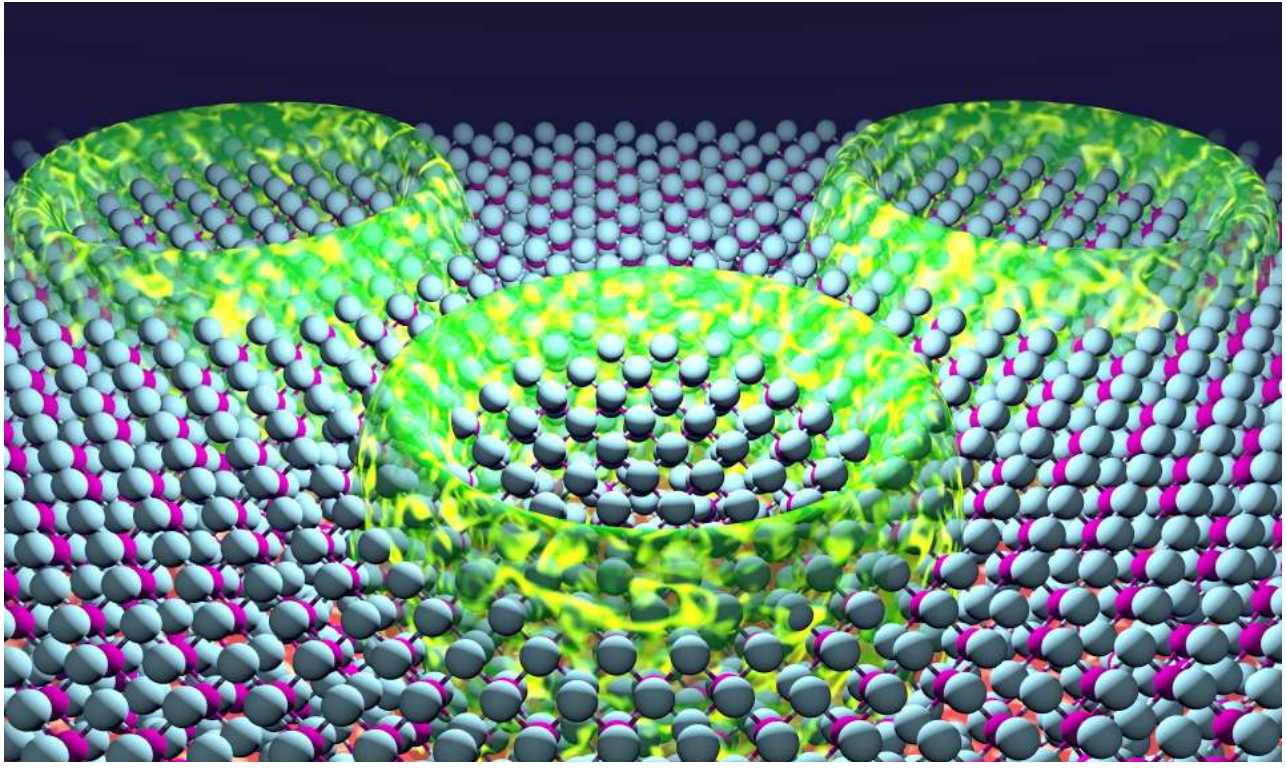
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\mathrm{SOC}}^{\mathrm{Ni}} \mathcal{H}_{\mathrm{SOC}}^{\mathrm{Ni}} + \lambda_{\mathrm{SOC}}^{\mathrm{I}} \mathcal{H}_{\mathrm{SOC}}^{\mathrm{I}},$$





The dominant contribution to the ferroelectric force comes from I

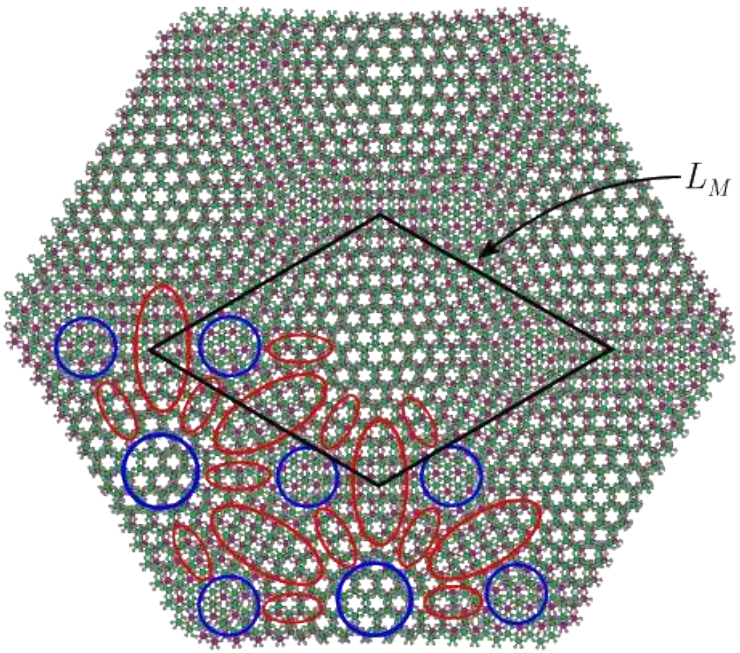
Engineering artificial multiferroics with twisted van der Waals materials

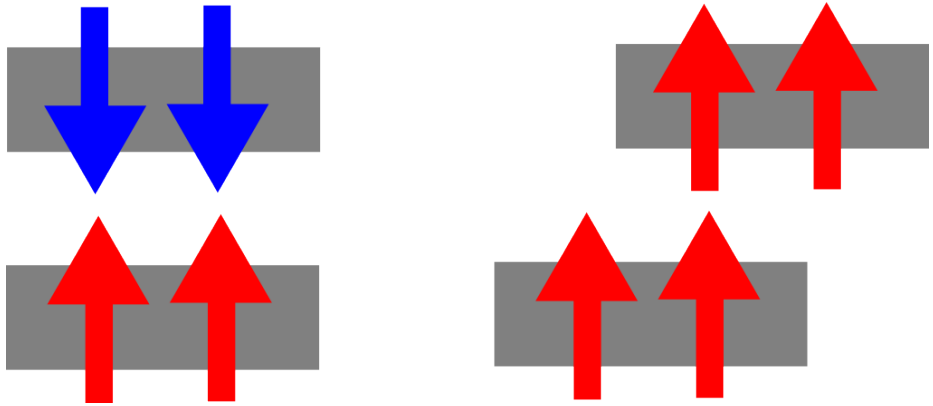


2D Materials 10 025026 (2023)

Twisted 2D trihalides

$CrBr_3$

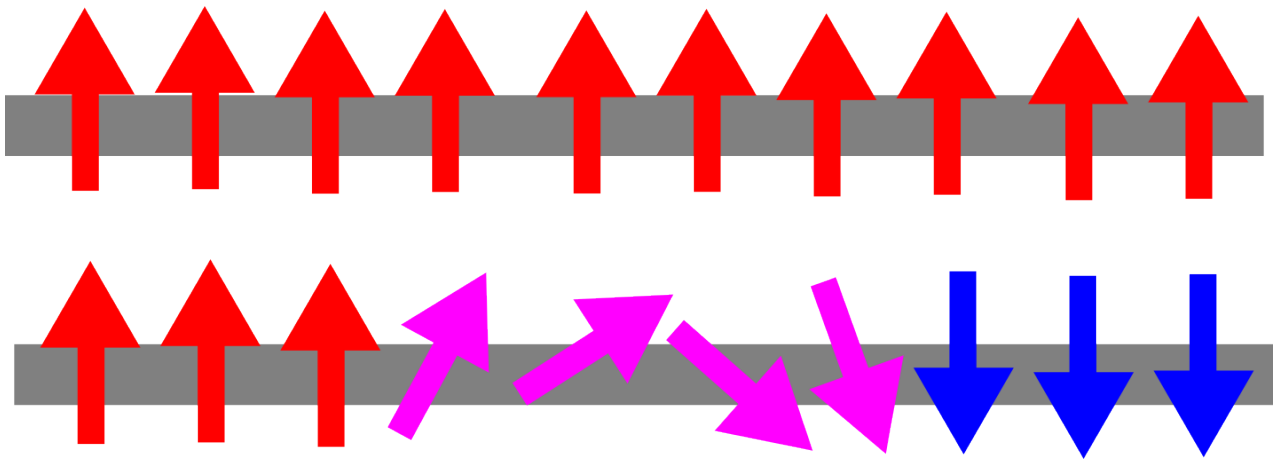




The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Stacking #1

Stacking #2

Mixed FE/AF domains

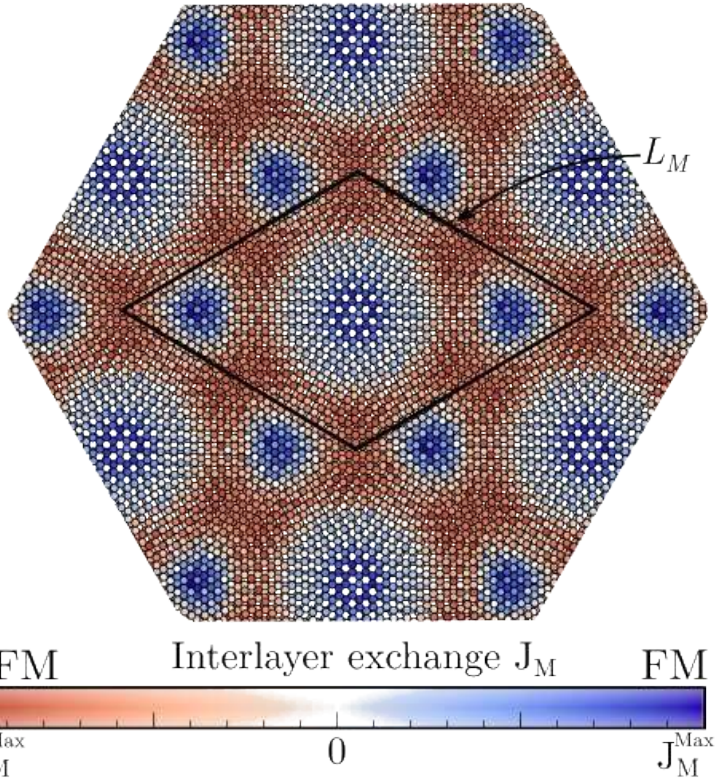
Nature Nanotechnology 17, 143–147 (2022)

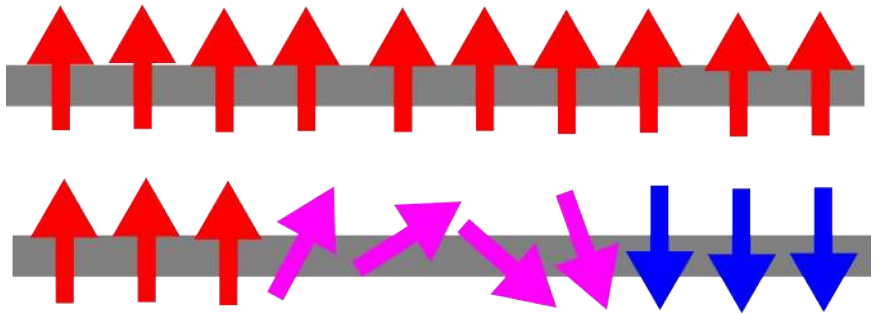
Non-collinear magnetism

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

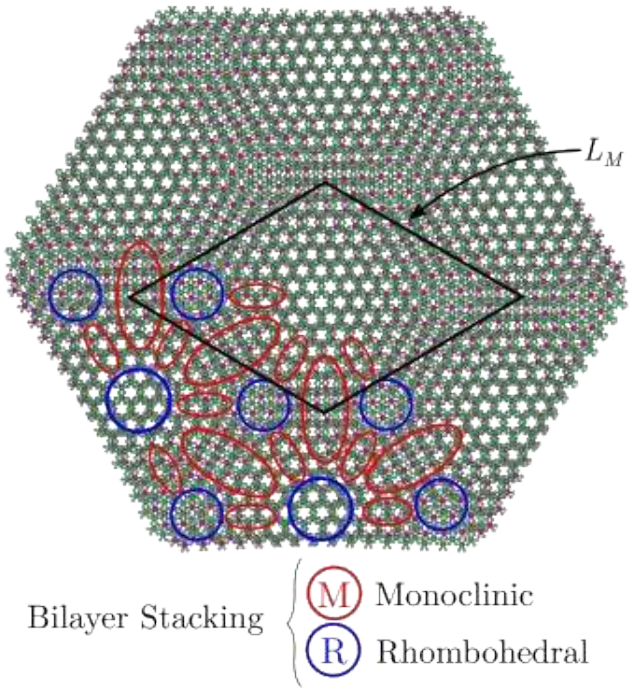
SOC driven

Wavevector of the spin spiral

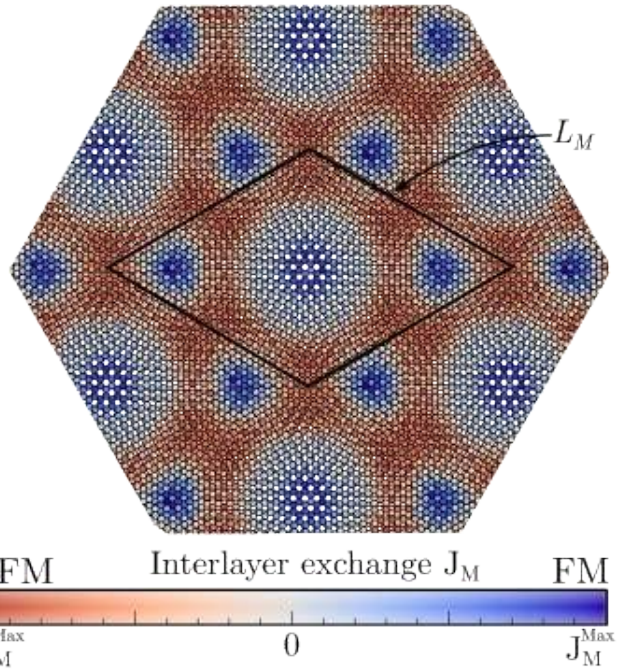
Spin rotation axis

Stacking, magnetic order and electric polarization

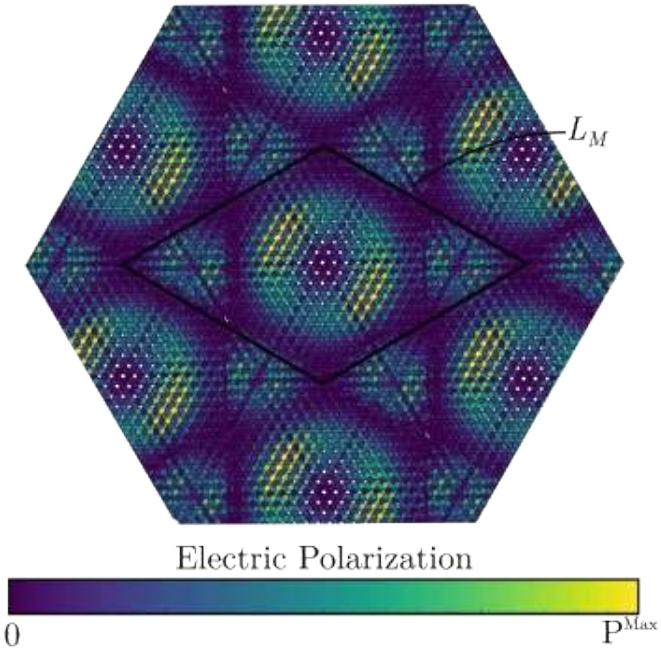
Structure



Magnetic order



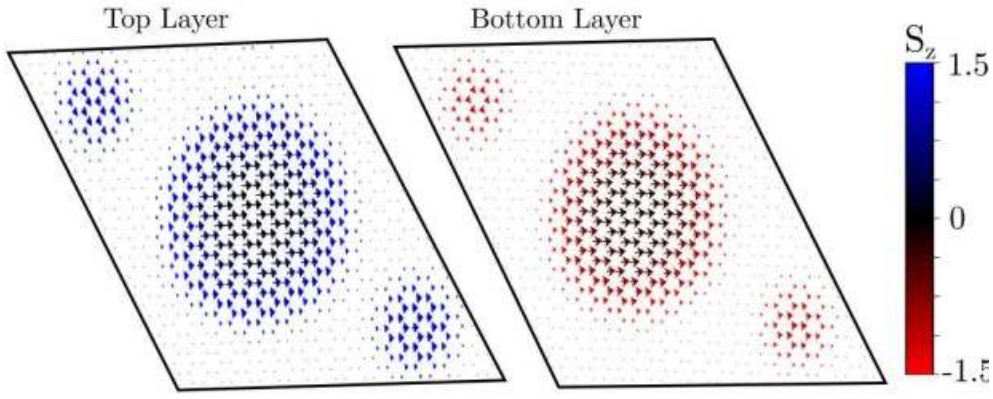
Ferroelectric order



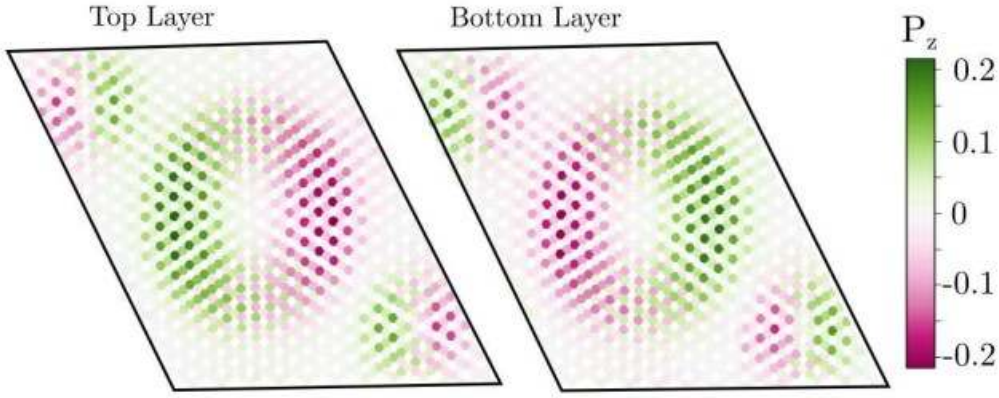
Stacking, magntic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

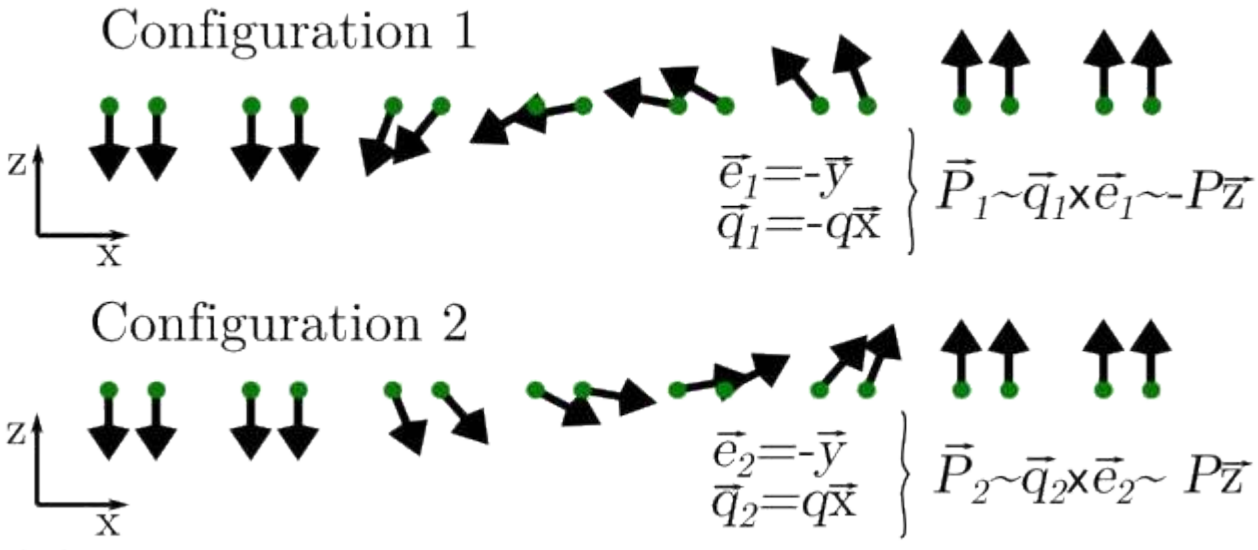


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



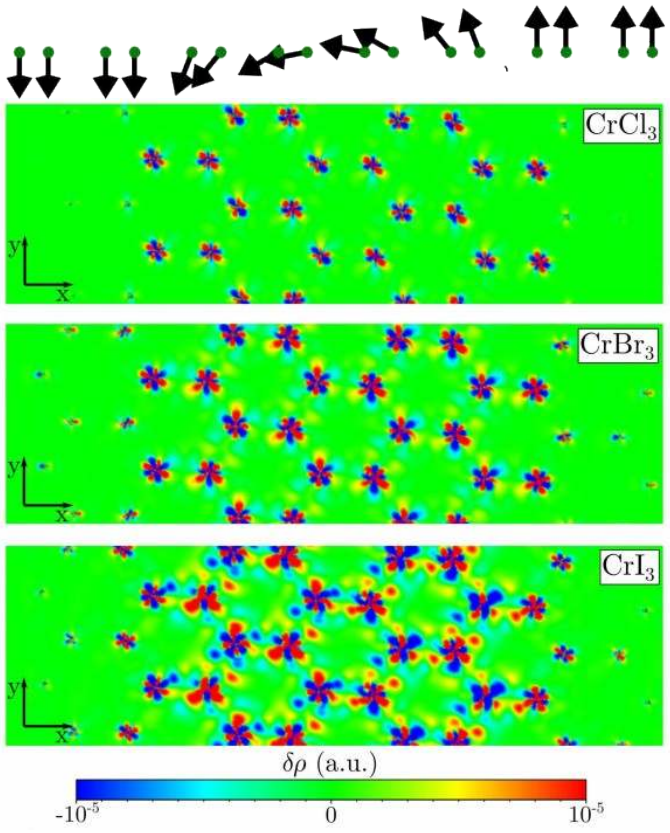
Ferroelectric electronic density

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

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

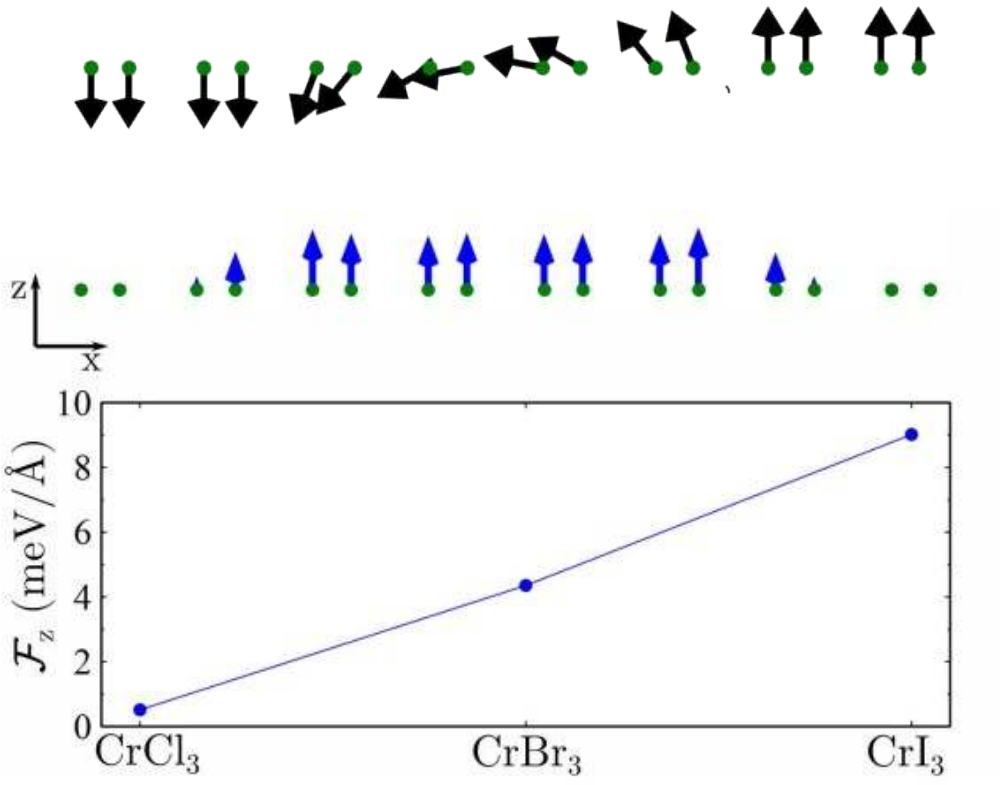
The ferroelectric force

Magnetic configuration

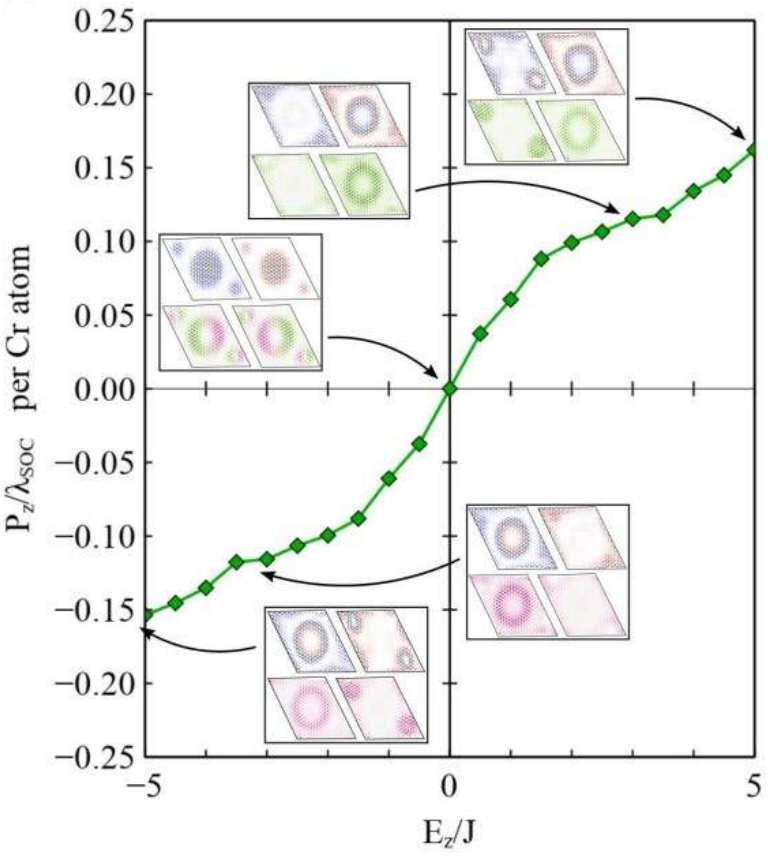
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



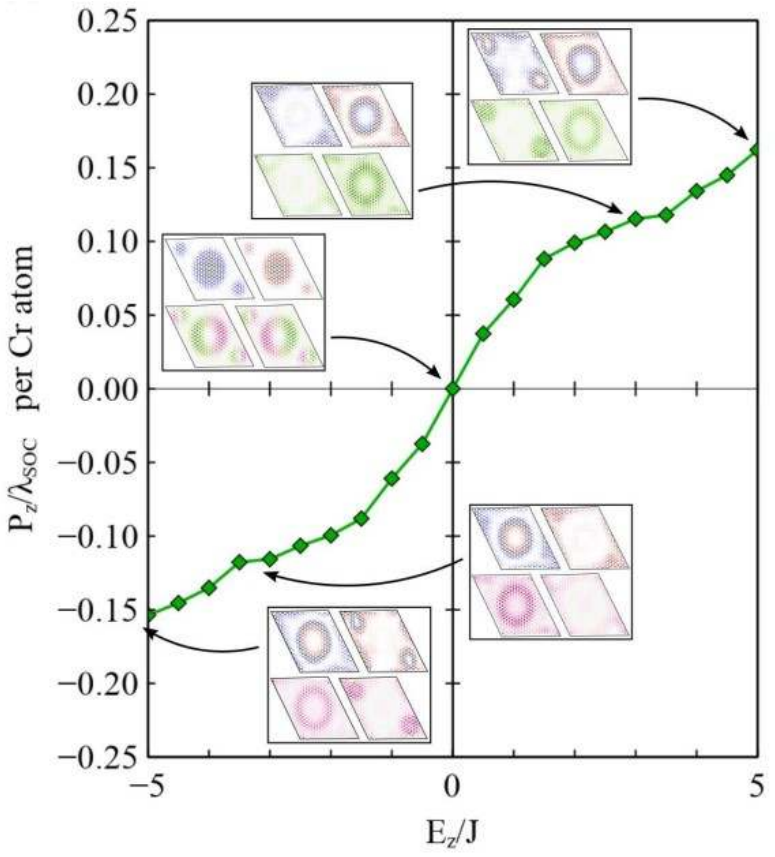
Electrically controllable skyrmions in twisted magnets



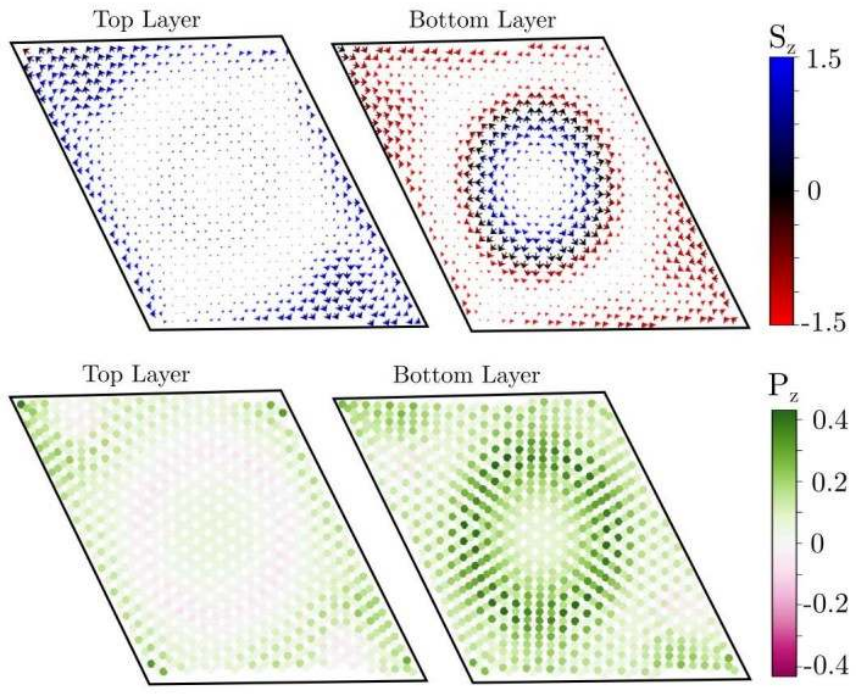
The electric polarization directly couples to an out-of plane electric field

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

Electrically controllable skyrmions in twisted magnets

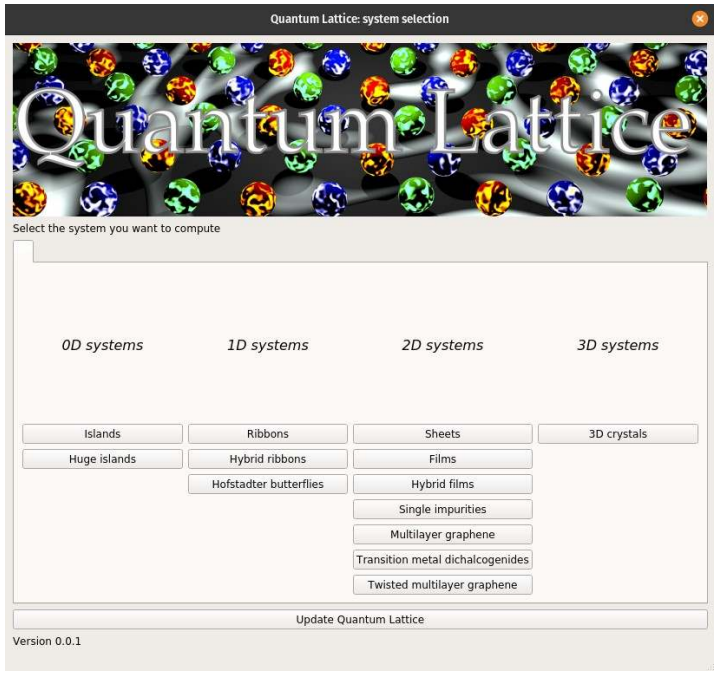


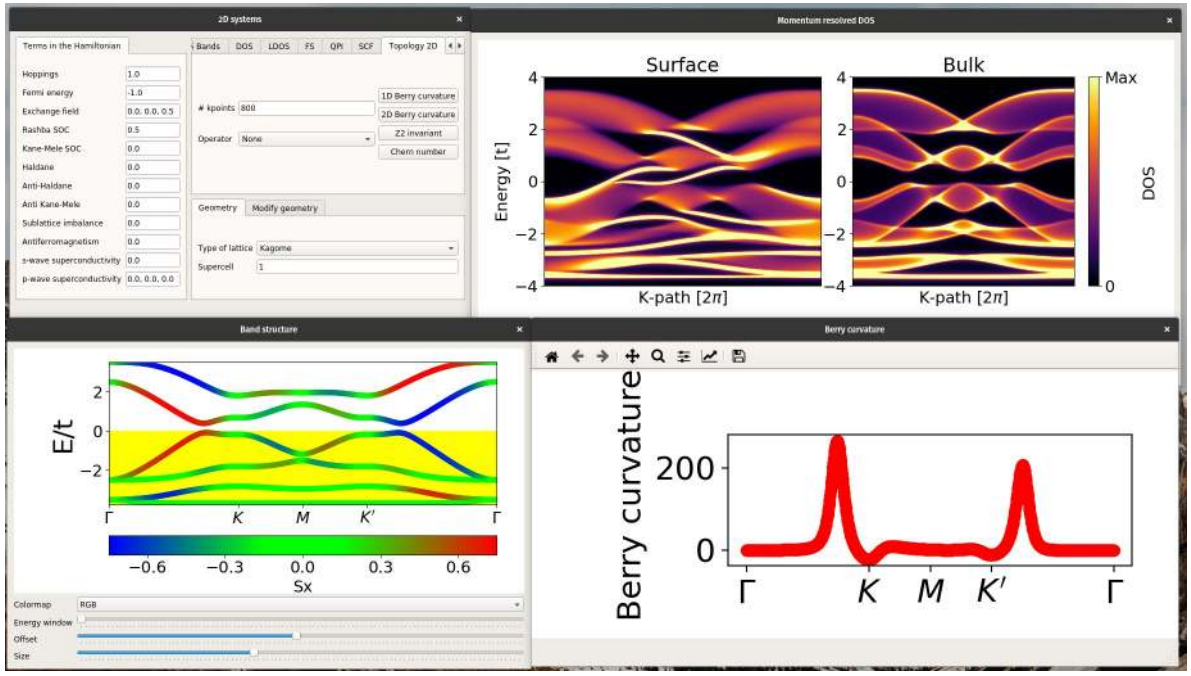
Skyrmions at finite interlayer bias



Open-source software for magnetic and electronic properties

Quantum Lattice: A user interface to compute electronic properties

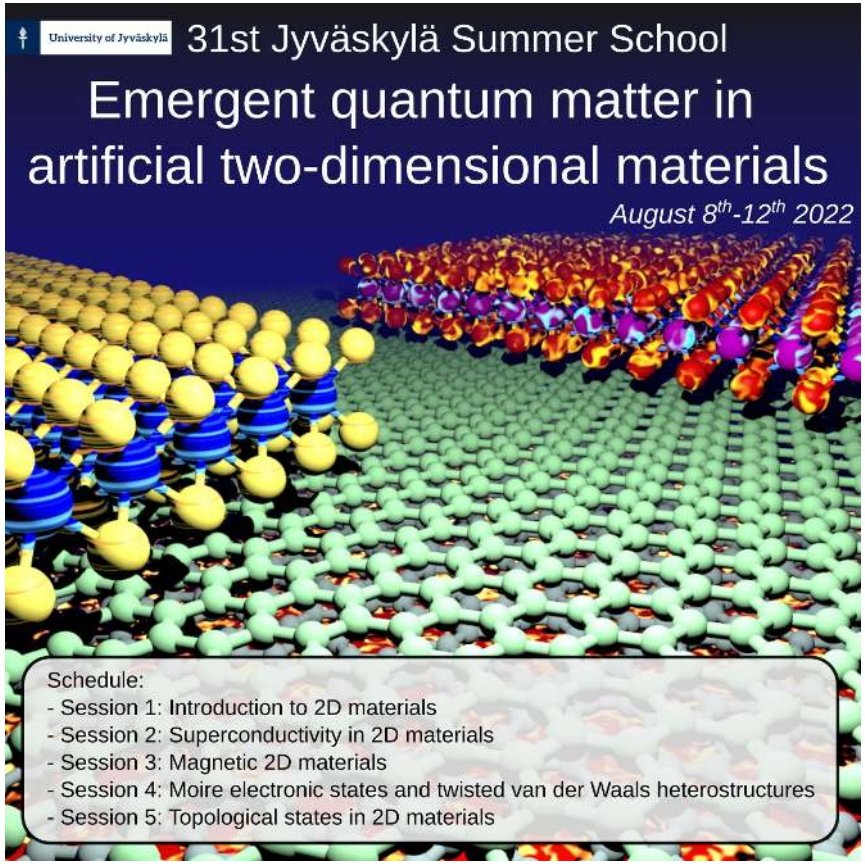




Quantum Lattice: open source interactive interface for tight binding modeling

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

Quantum matter in van der Waals materials school



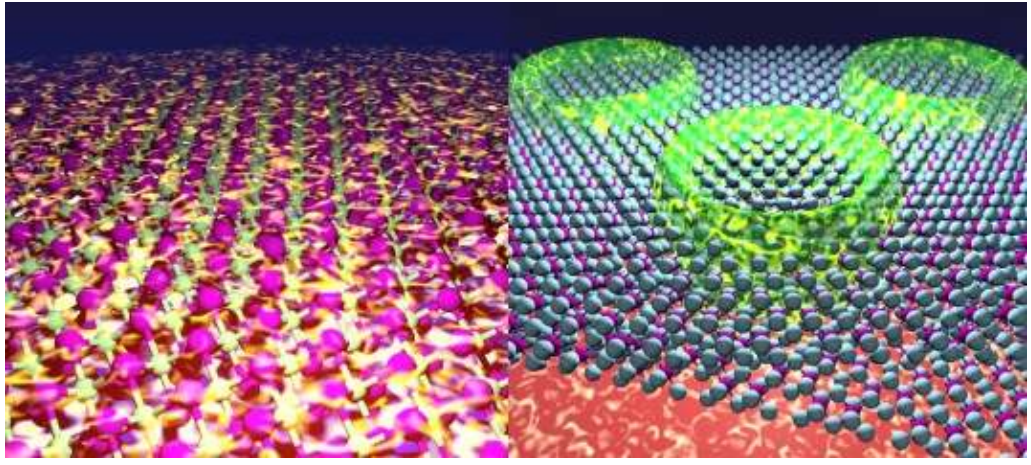
Recordings available at

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 materials provide a platform to realize tunable multiferroic order



2D Materials 9 (2), 025010 (2022)
2D Materials 10 (2), 025026 (2023)

Thank you

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JANE AND AATOS ERKKO FOUNDATION

SUOMEN AKATEMIA