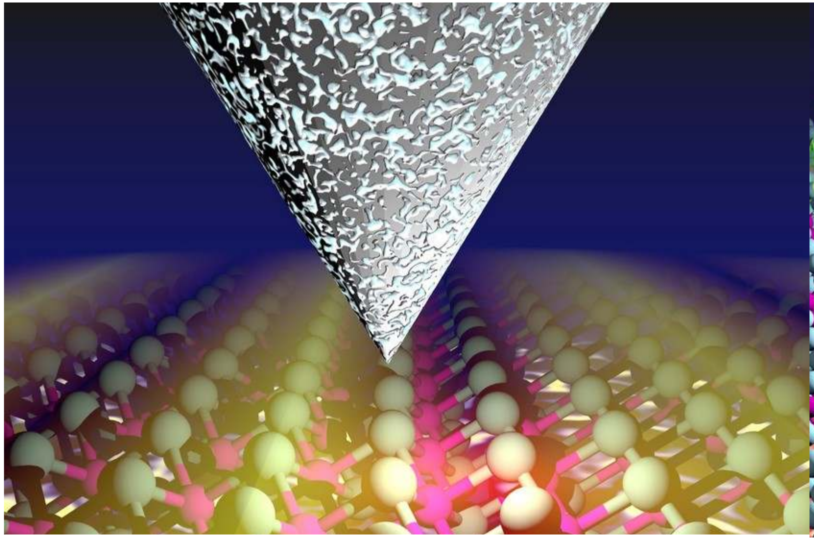
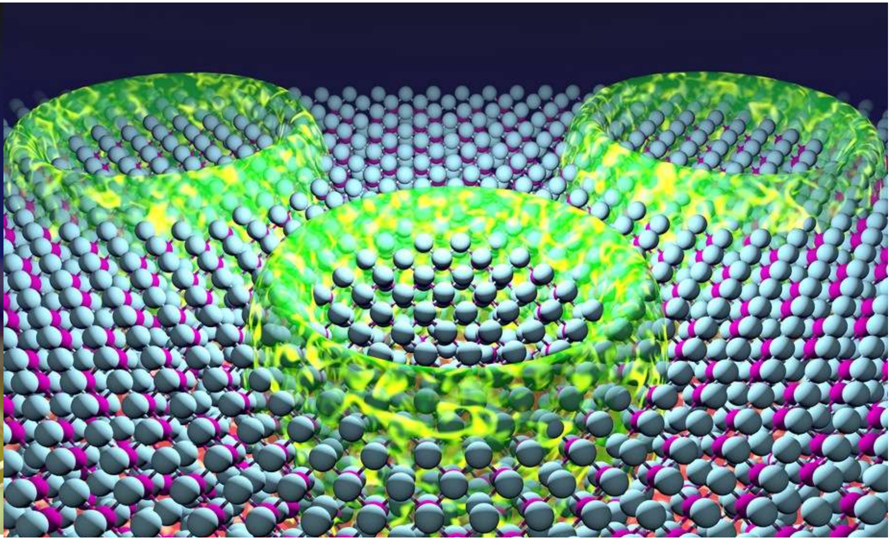
Artificial van der Waals multiferroics with twisted two-dimensional materials

Jose Lado

Department of Applied Physics, Aalto University, Finland





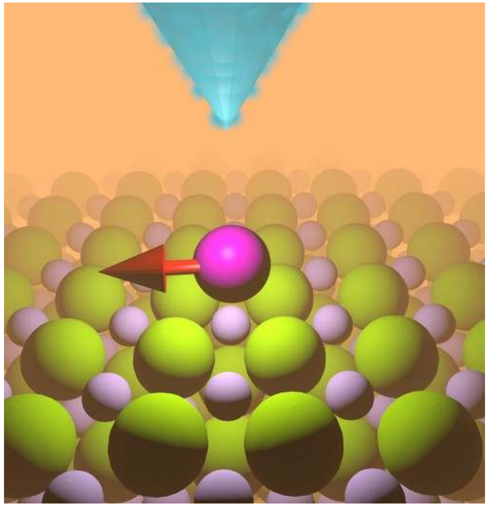
2D Materials 9 (2), 025010 (2022)
Advanced Materials, 2311342 (2024)

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

International Conference on Magnetism, ICM2024, Bologna, Italy, July 5th 2024

Building quantum matter with artificial lattices

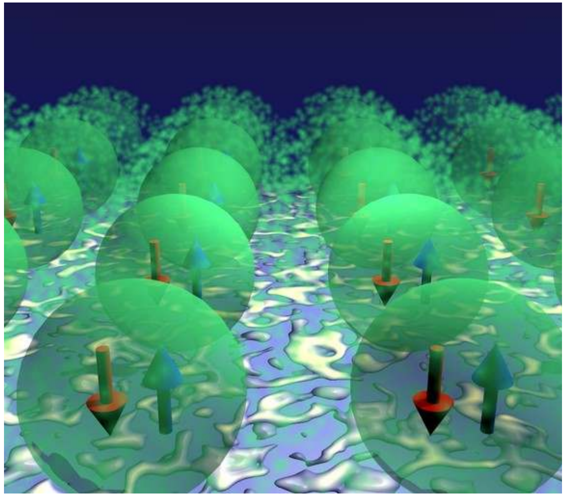
Atomic lattices



a = 0.5 nm

Nature Physics 12 (7), 656 (2016)
Nature Physics 13 (7) 668 (2017)
Science 335 (6065), 196-199 (2012)

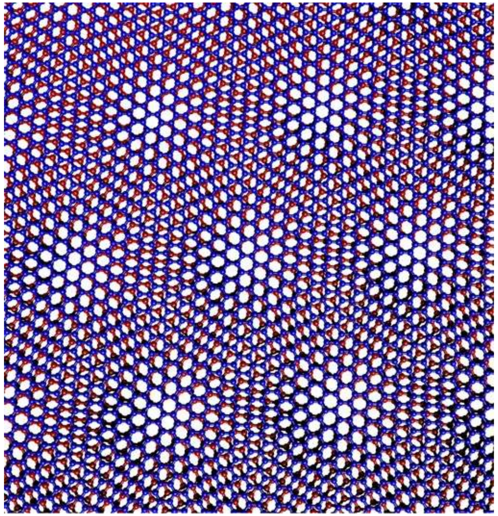
Molecular lattices



a = 3 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



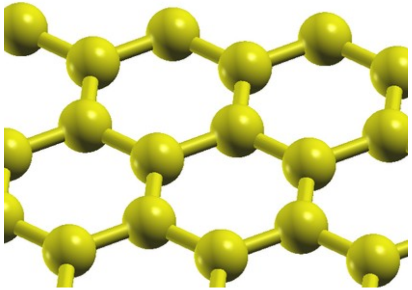
a = 20 nm

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

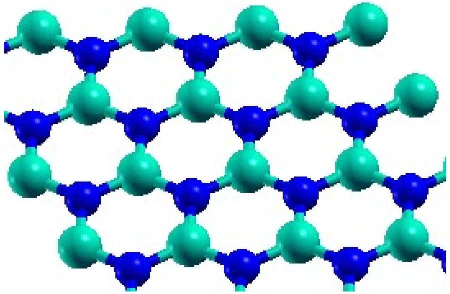
Semimetal

Graphene



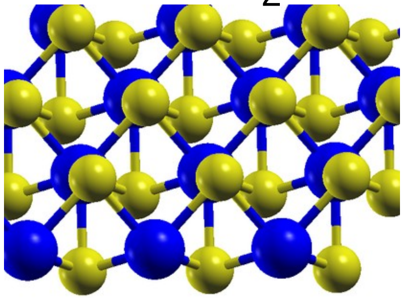
Insulator

BN



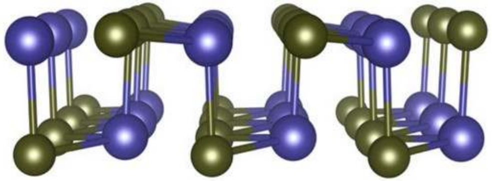
Superconductor

$NbSe_2$



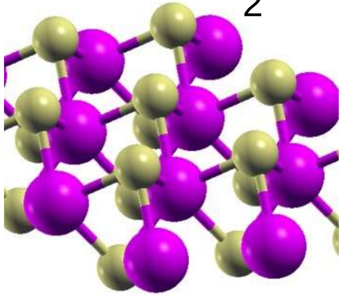
Ferroelectric

SnTe



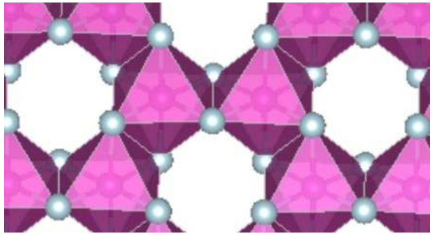
Semiconductor

WSe_2



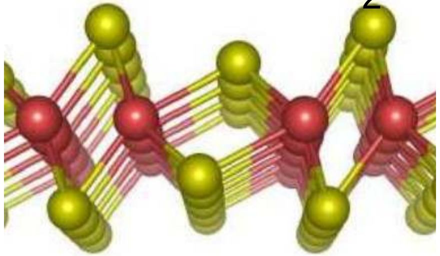
Ferromagnet

CrI_3



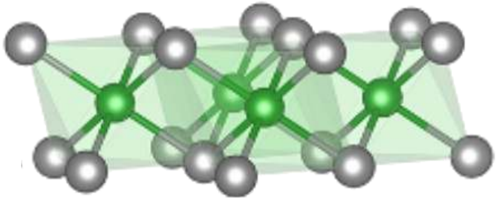
Quantum spin Hall insulator

1T’- WTe_2



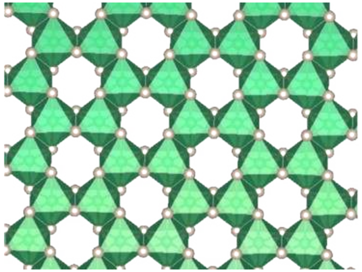
Multiferroic

NiI_2



Van der Waals magnetic materials

Ferromagnet, antiferromagnets

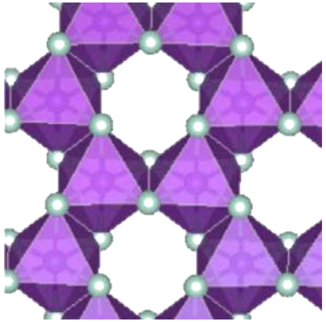


CrI_3, $CrCl_3$, $CrBr_3$

Nature 546, 270–273 (2017)

Break time-reversal

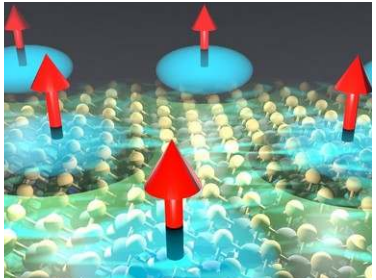
(proximal) Quantum spin-liquids



$RuCl_3$, 1T-TaS_2

Nature Physics 17, 1154–1161 (2021)

Heavy-fermion Kondo insulators

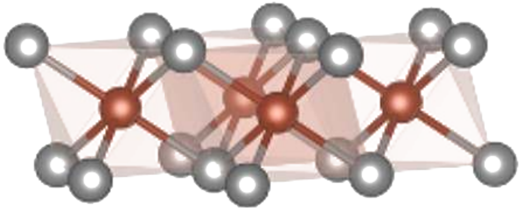


1T-TaS_2/1H-TaS_2

Nature 599, 582–586 (2021)

Do not break time-reversal

Multiferroics



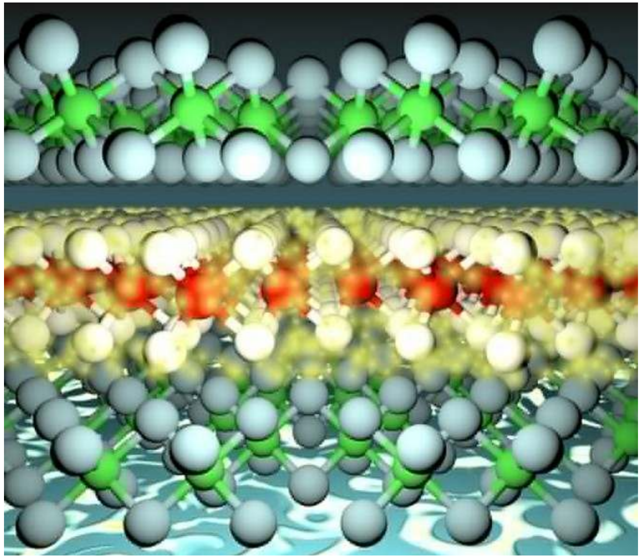
NiI_2

Nature 602, 601–605 (2022)

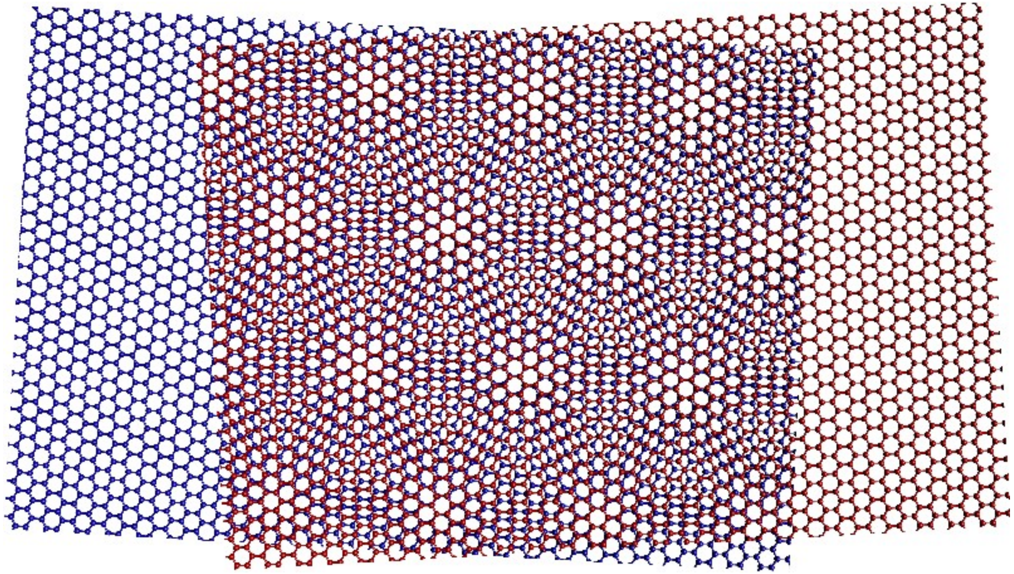
Break time-reversal and inversion symmetry

The flexibility of two-dimensional materials

They can be stacked



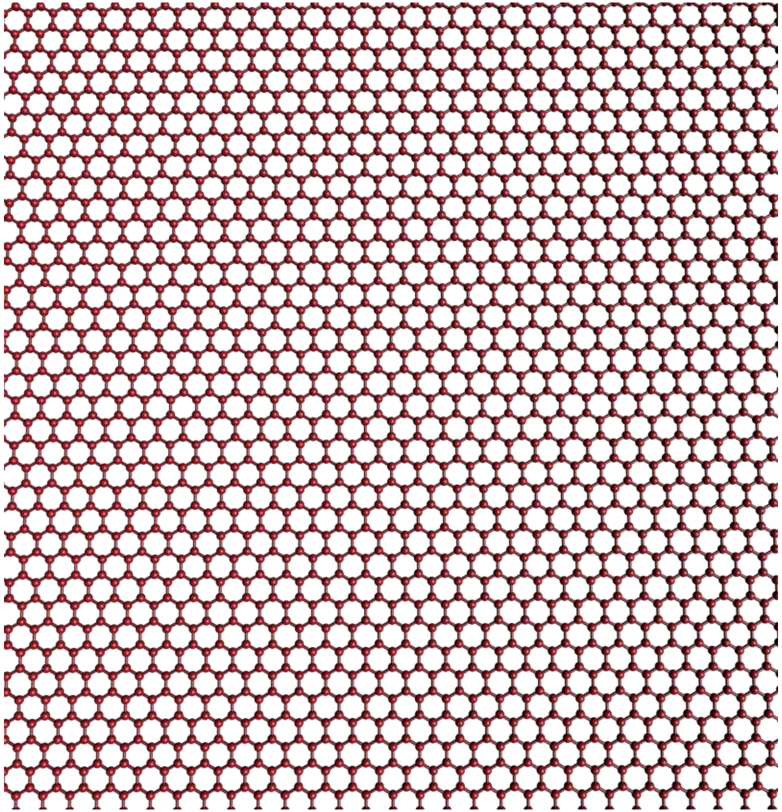
They can be rotated



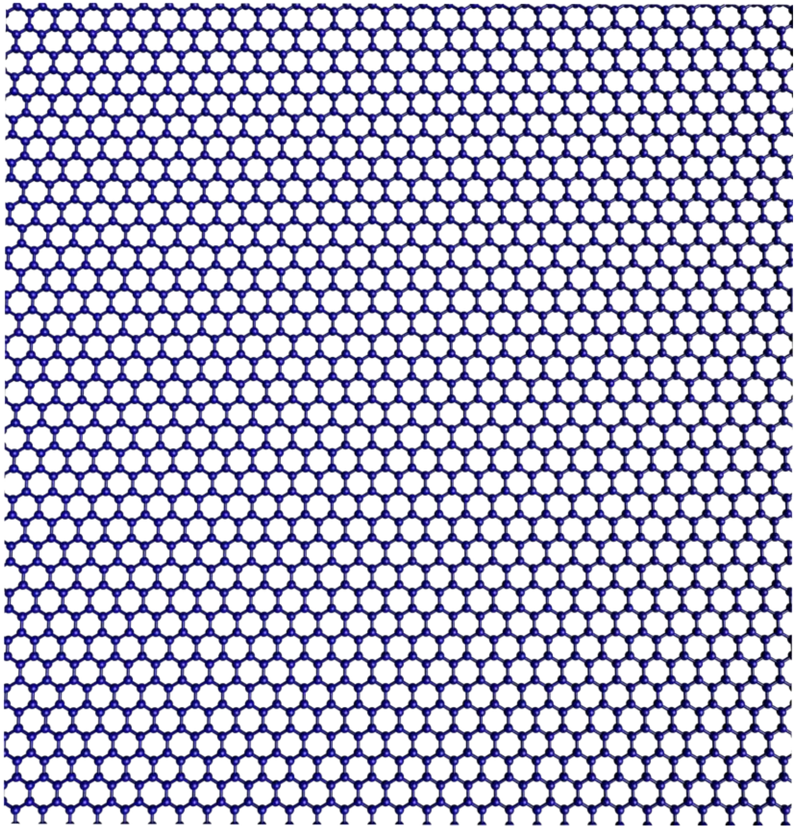
These are unique features of two-dimensional materials

Moire in twisted bilayer graphene

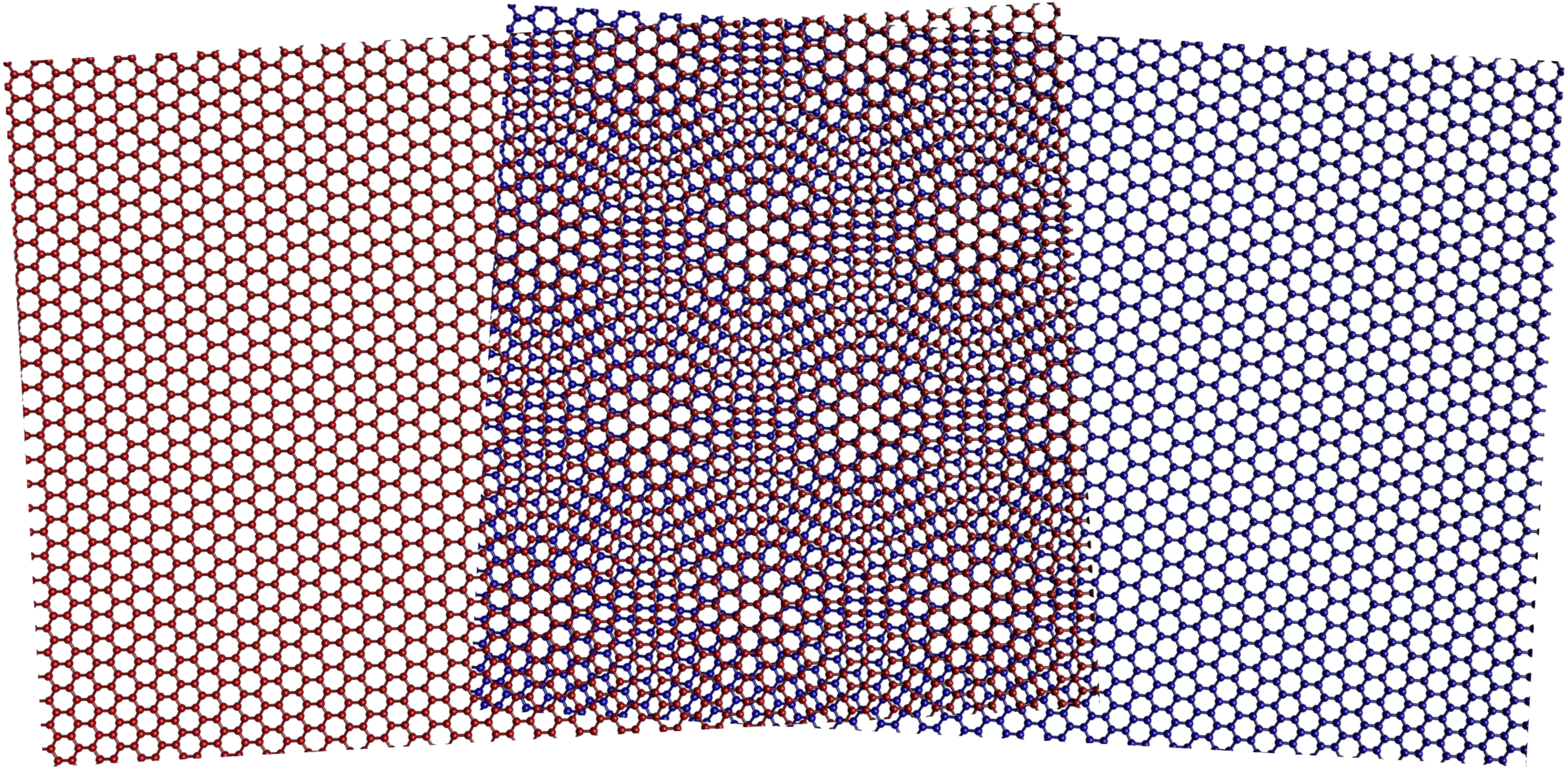
Upper graphene layer



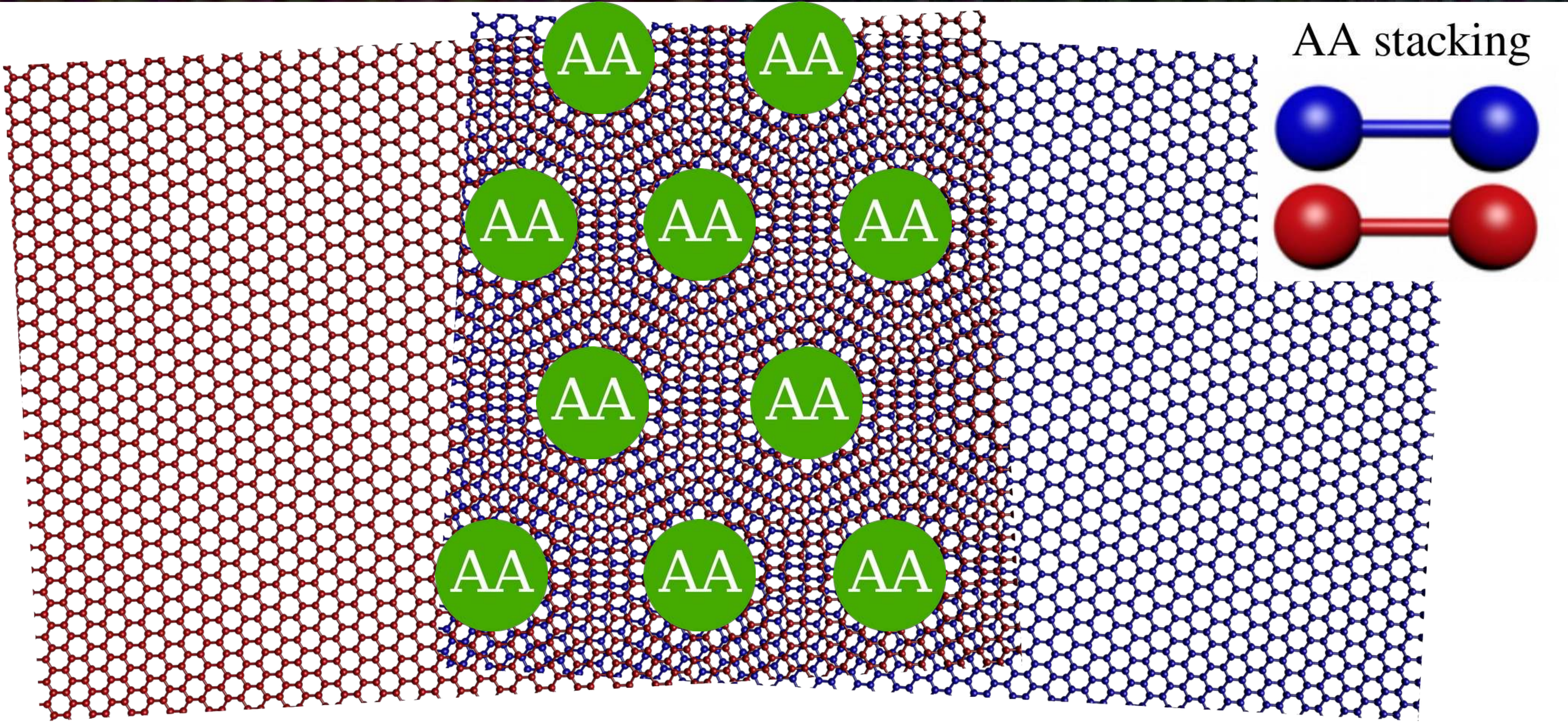
Lower graphene layer



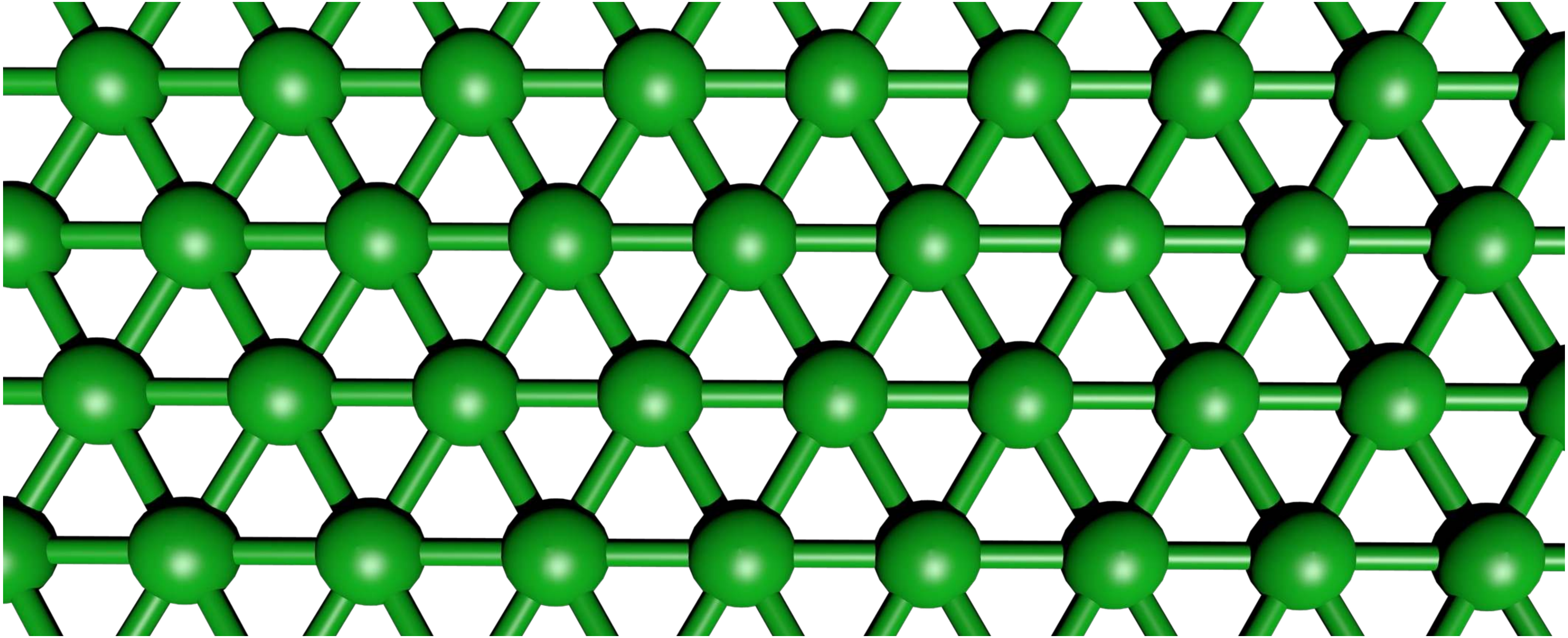
Moire in twisted bilayer graphene



Moire in twisted bilayer graphene

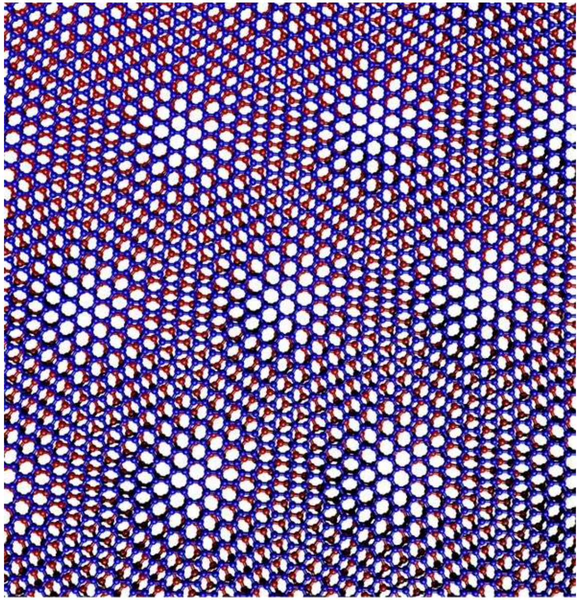


Moire in twisted bilayer graphene

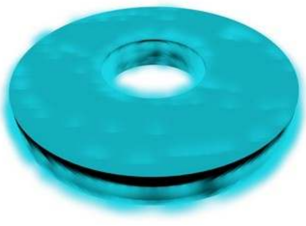


The phases of twisted bilayer graphene

Twisted bilayer graphene



Superconductivity

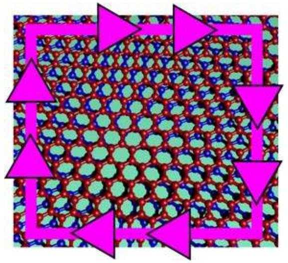


Nature 556, 43–50 (2018)

Topological networks

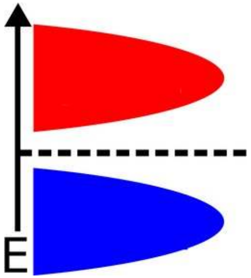
Nano Lett. 18, 11, 6725-6730 (2018)

Chern insulators



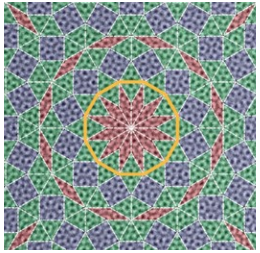
Science 365, 605-608 (2019)

Correlated insulators



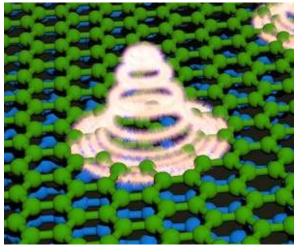
Nature 556, 80–84 (2018)

Quasicrystalline physics



Science 361, 782-786 (2018)

Proximal fractional Chern insulators

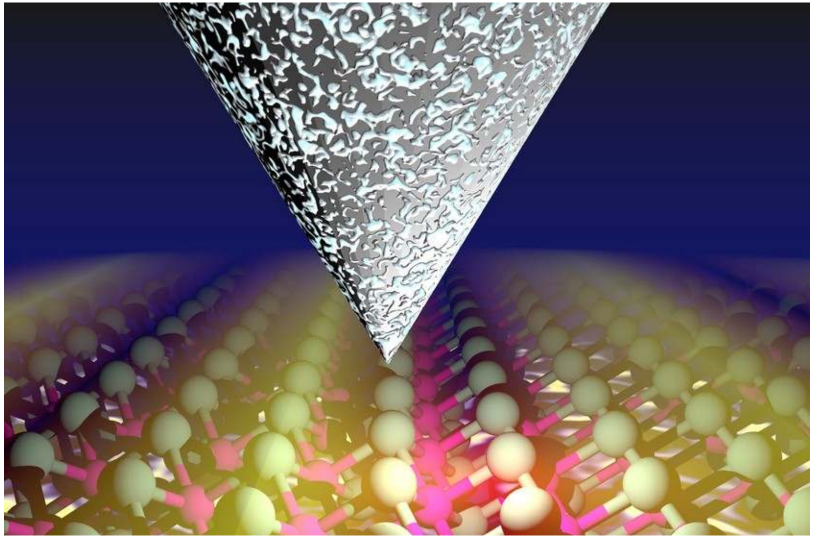


Nature 600, 439–443 (2021)

Which new phenomena could we get by twisting magnetic materials?

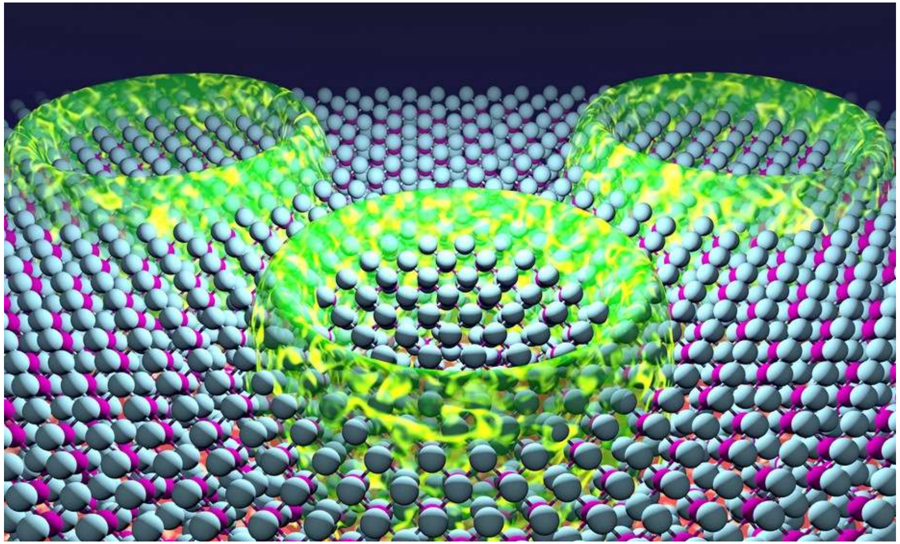
Today's plan

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)

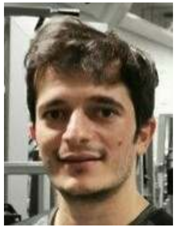
Behind the scenes

Intrinsic multiferroics, NiI_2

(theory and experiment)

V. Vaňo

M. Amini



A. Fumega

H. Gonzalez Herrero

S. Kezilebieke

P. Liljeroth

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

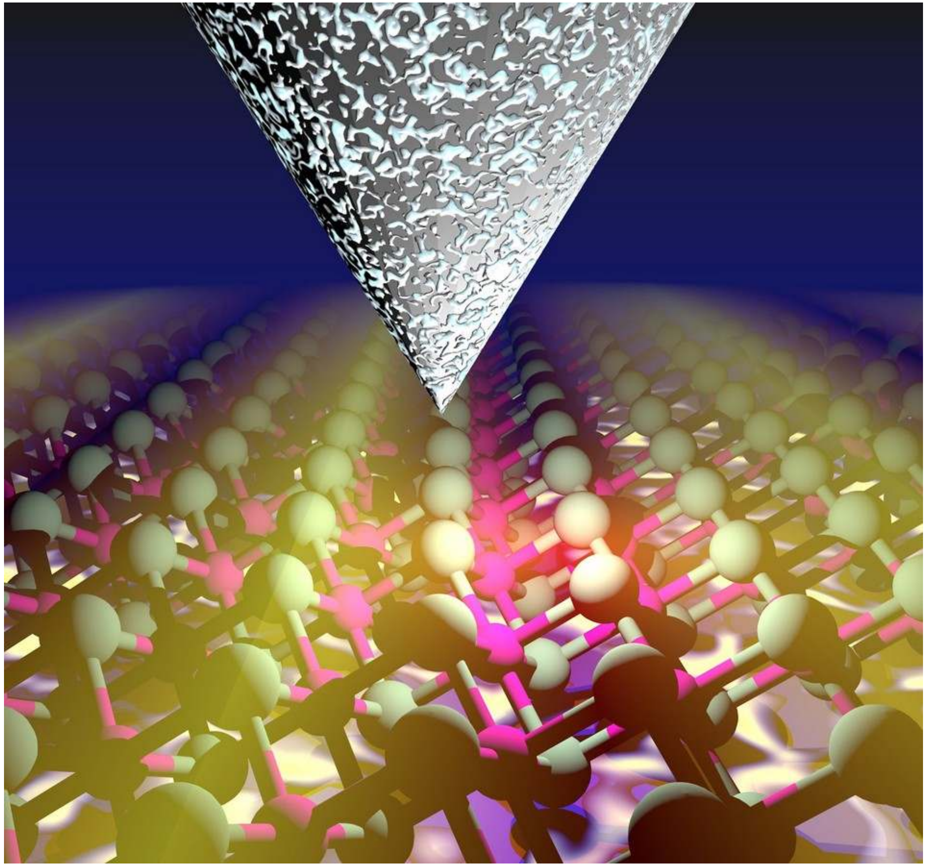
Artificial multiferroics, twisted $CrBr_3$ bilayers

(theory)

A. Fumega

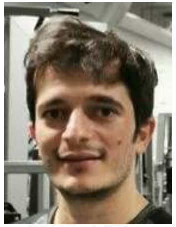
2D Materials 10 (2), 025026 (2023)

Origin and visualization of multiferroicity in NiI_2



V. Vaňo

M. Amini



A. Fumega

H. Gonzalez Herrero

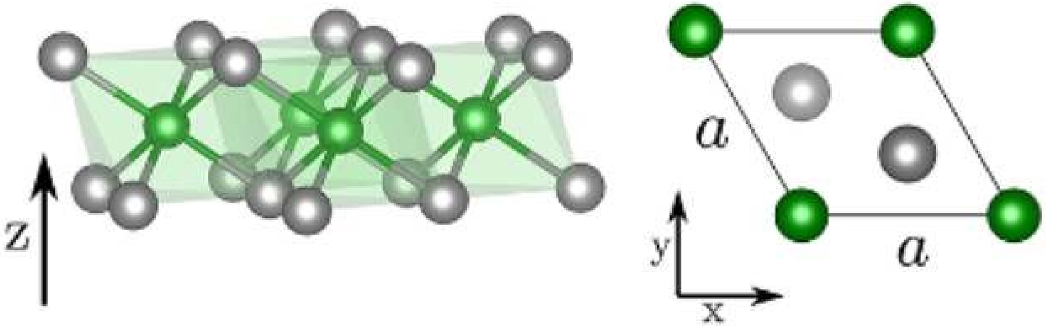
S. Kezilebieke

P. Liljeroth

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

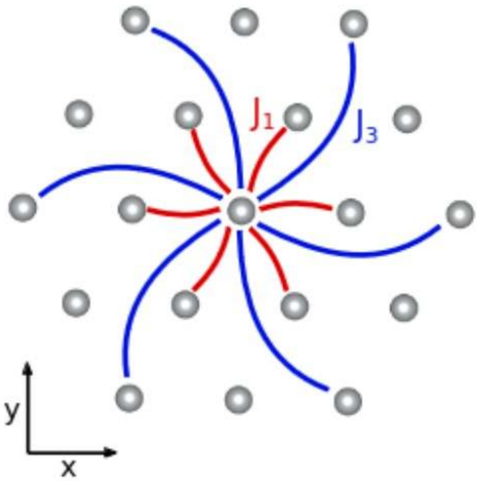
Monolayer NiI_2

Structure

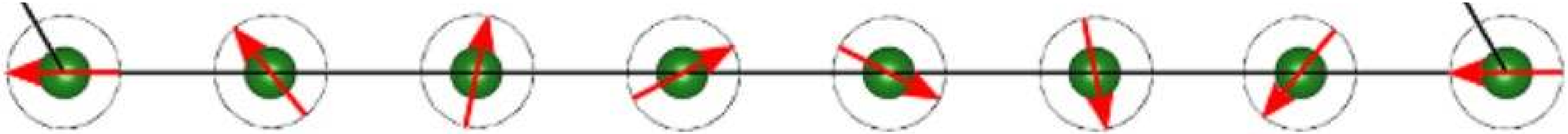


Nature 602, 601–605 (2022)

Frustrated magnetic Heisenberg model

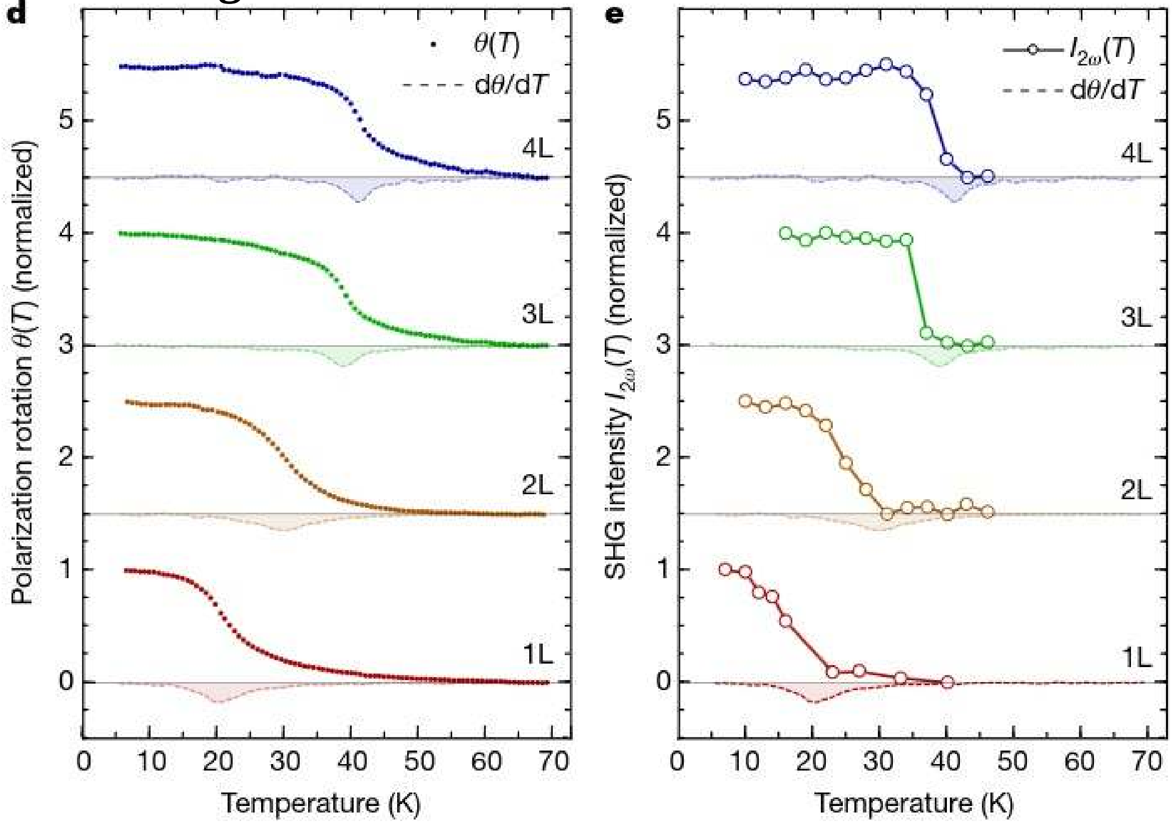


Magnetic structure



Multiferroicity in monolayer NiI_2

Magnetic order **Ferroelectric order**



Probe of magnetic order
Magneto-optic Kerr effect

Probe of ferroelectric order
Second harmonic generation

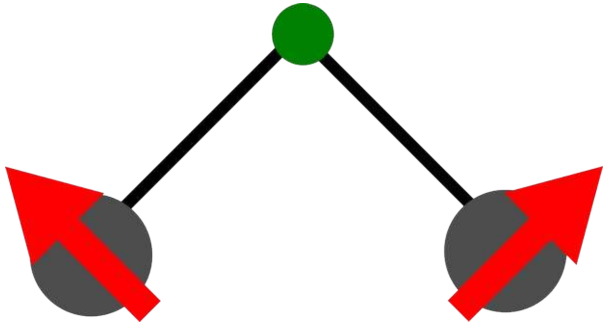
Nature 602, 601–605 (2022)

The impact of spin-orbit coupling in magnetic ordering

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

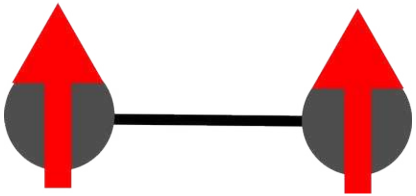
Antisymmetric exchange

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



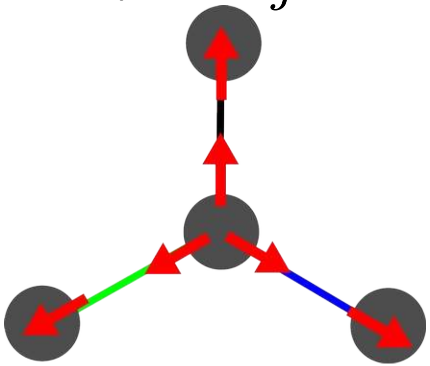
Anisotropic exchange

$$S_i^z S_j^z$$



Kitaev interaction

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



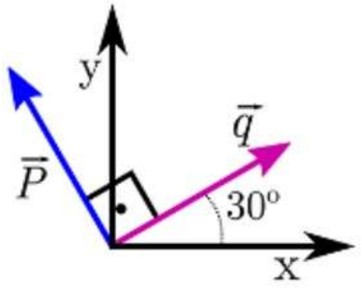
In NiI_2, the non-collinear order is promoted by isotropic exchange

In NiI_2, the ferroelectricity is created by inverse antisymmetric exchange

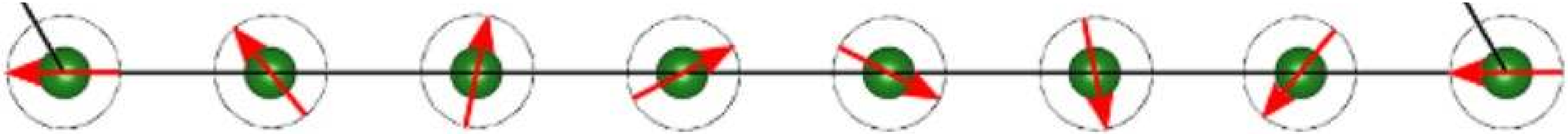
The origin of ferroelectricity in NiI_2

Magnetically driven ferroelectric order

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

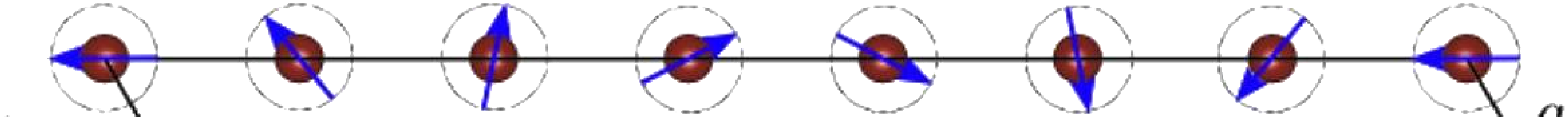


$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e}\cdot\mathbf{S})(\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}_0))}\mathbf{M}(\mathbf{r}_0)$$



The emergence of non-collinear order drives ferroelectricity

The origin of ferroelectricity in NiI_2



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

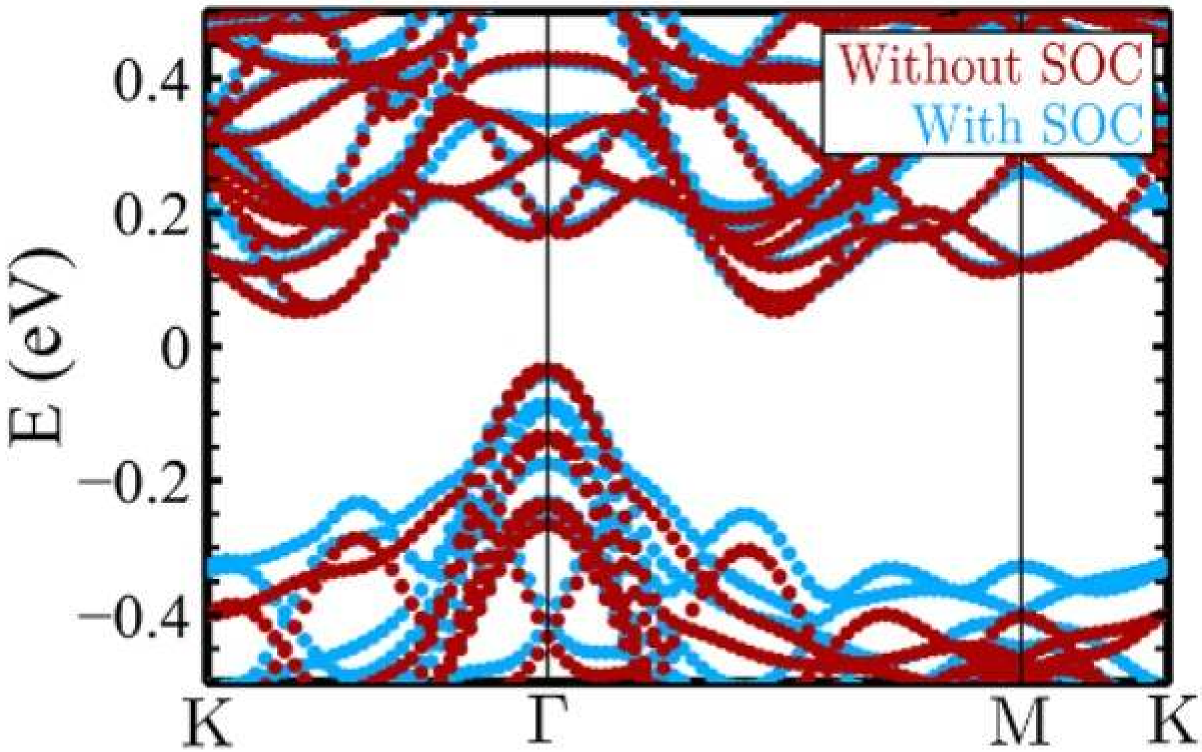
Electric polarization

SOC driven

Wavevector of the spin spiral

Spin rotation axis

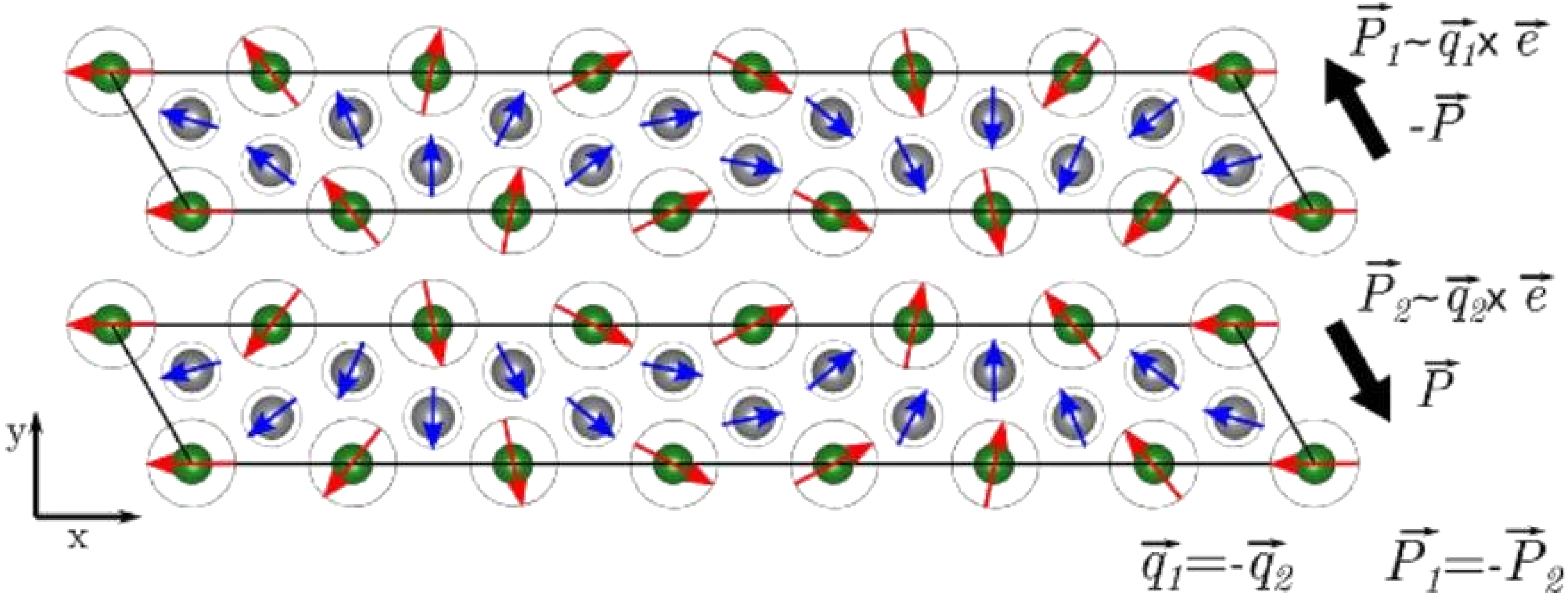
The impact of spin-orbit coupling in the electronic structure



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

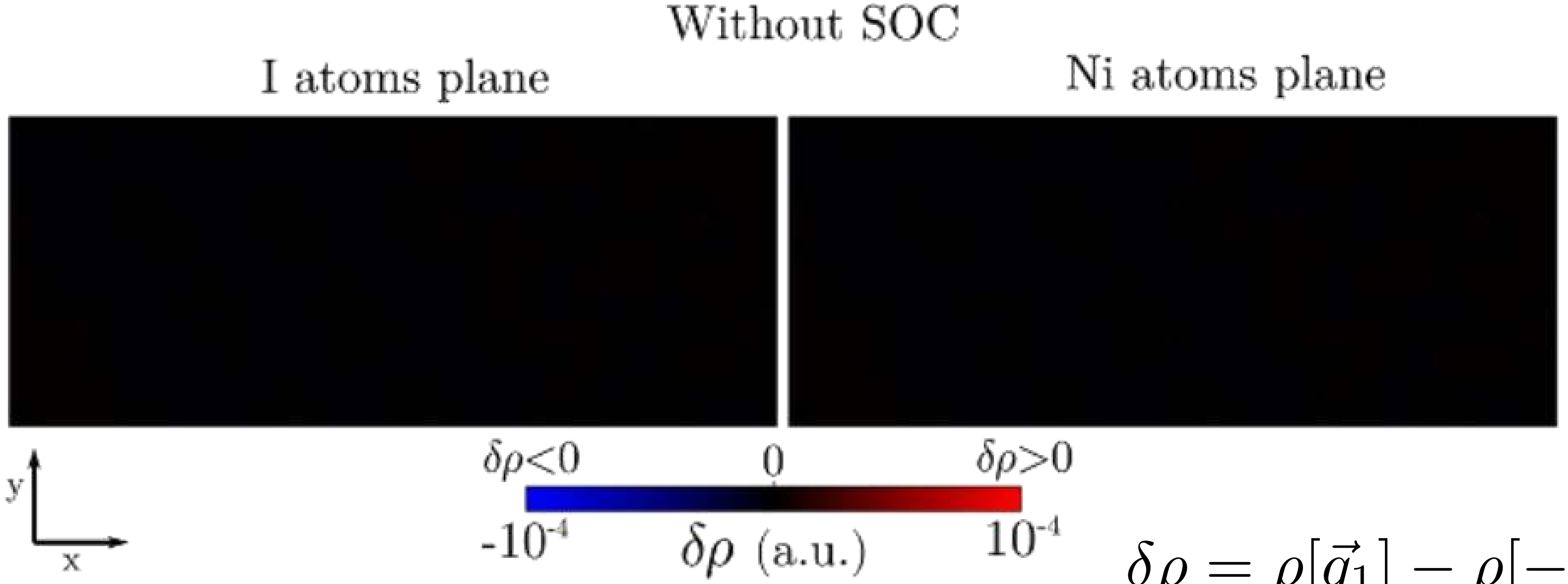
Ferroelectric correction to the electronic density



We will examine the ferroelectric density correction

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

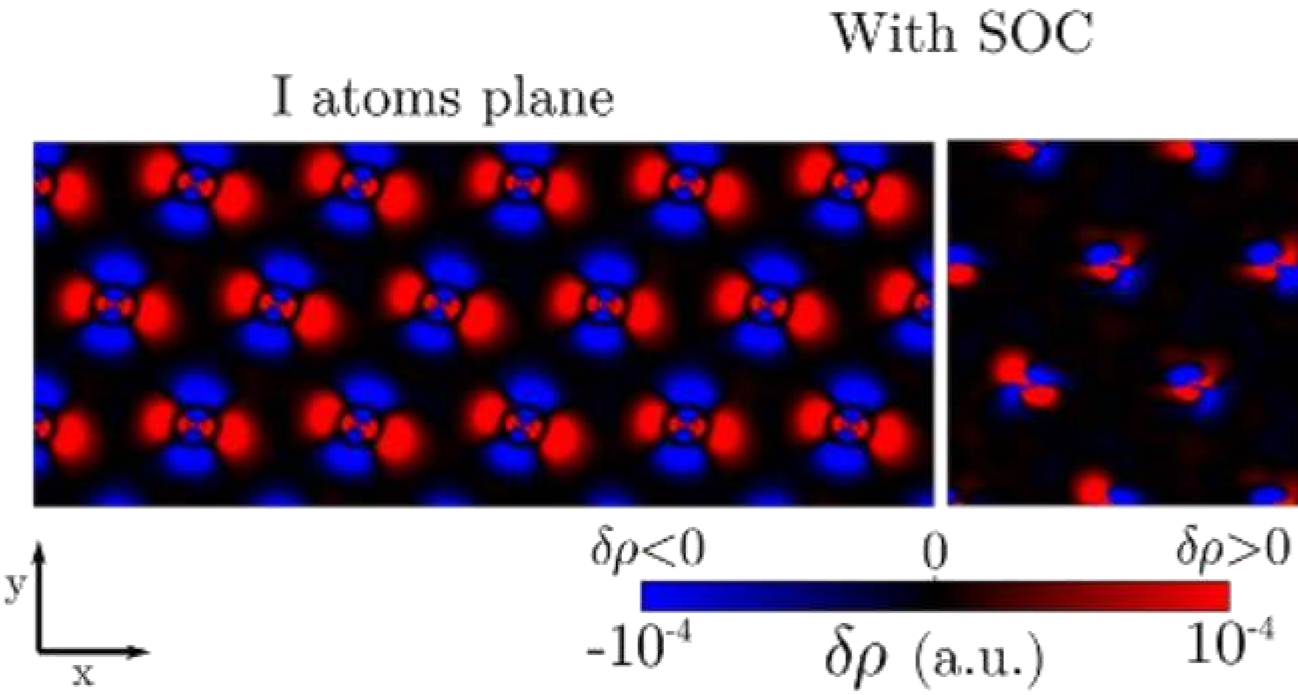
Ferroelectric correction to the electronic density



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

In the absence of SOC, no ferroelectric density appears

Ferroelectric correction to the electronic density

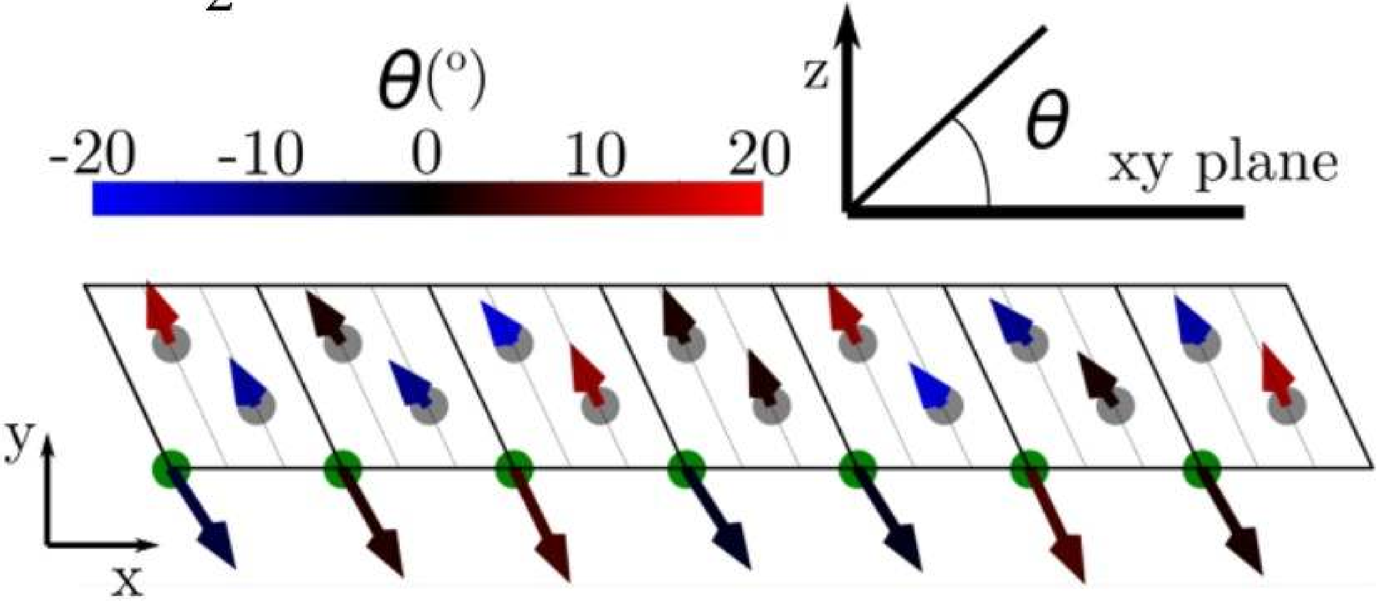


$$\delta\rho = \rho[\vec{q}_1] - \rho[-\vec{q}_1]$$

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

The ferroelectric force

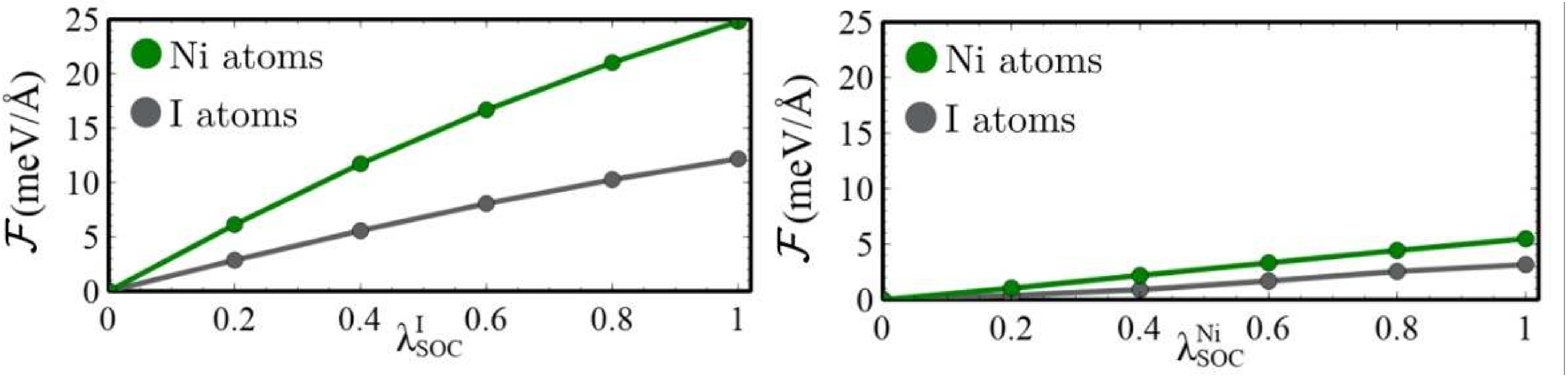
$$\mathcal{F}_\alpha = \frac{1}{2}\left(\mathbf{F}_\alpha[\mathbf{q}_1] - \mathbf{F}_\alpha[\mathbf{q}_2]\right),$$



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

The ferroelectric force

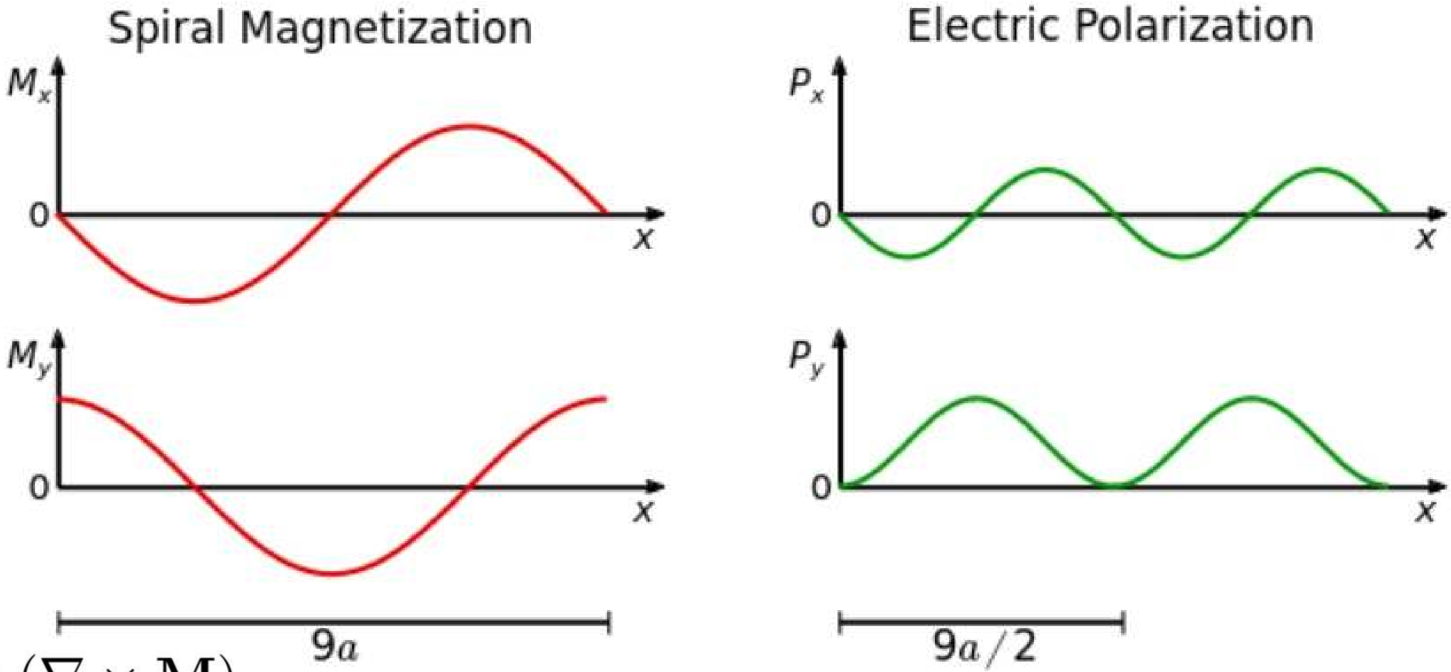
$$\mathcal{H} = \mathcal{H}_0 + \lambda_{\text{SOC}}^{\text{Ni}} \mathcal{H}_{\text{SOC}}^{\text{Ni}} + \lambda_{\text{SOC}}^{\text{I}} \mathcal{H}_{\text{SOC}}^{\text{I}},$$



The dominant contribution to the ferroelectric force comes from iodine

Modulated multiferroicity in monolayer NiI_2

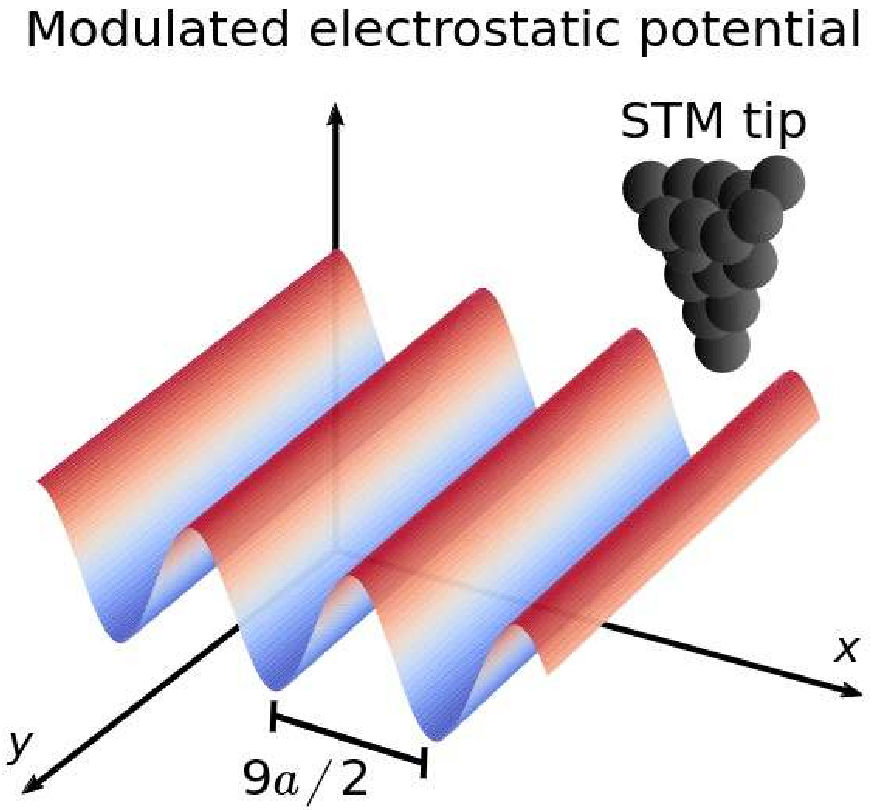
The ferroelectric order has actually a modulation in real space



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

Advanced Materials, 2311342 (2024)

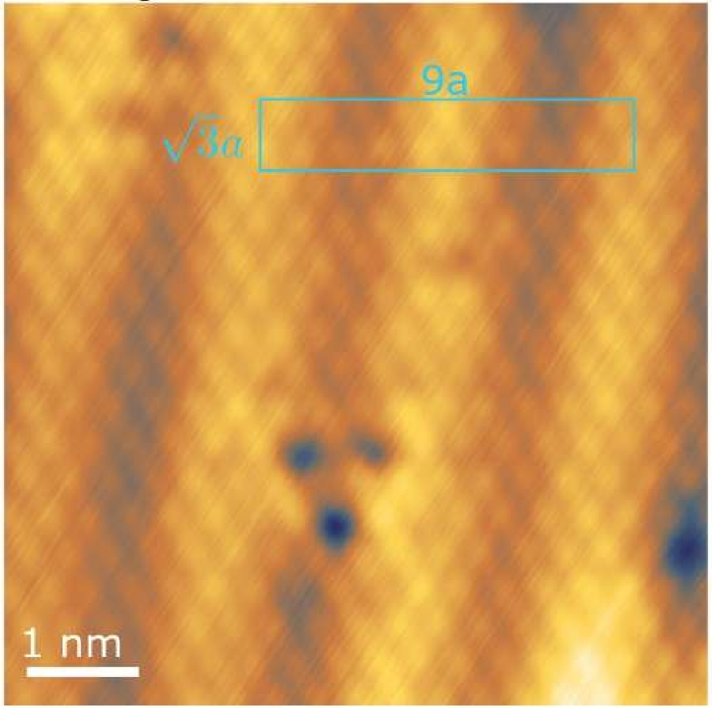
Visualizing multiferroicity in monolayer NiI_2



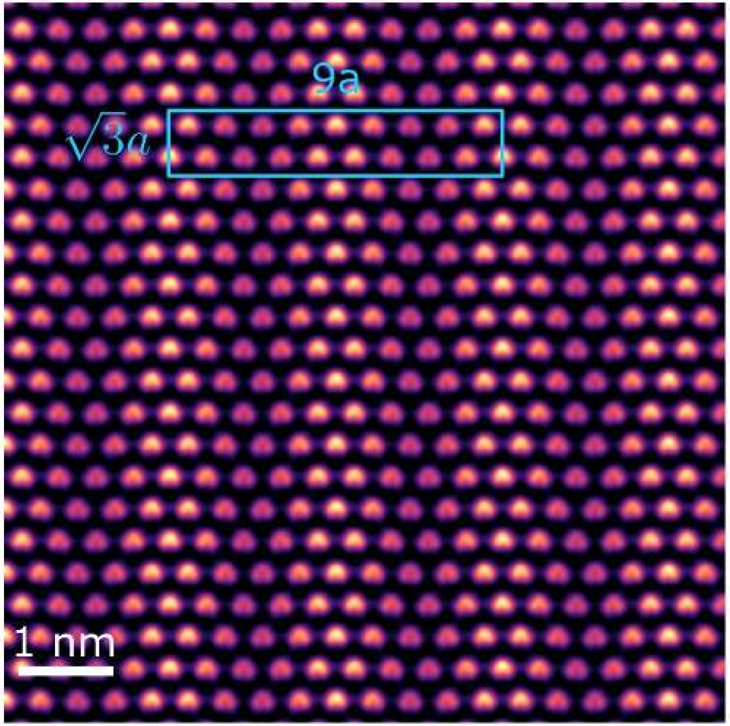
Advanced Materials, 2311342 (2024)

Visualizing multiferroicity in monolayer NiI_2

Experiment



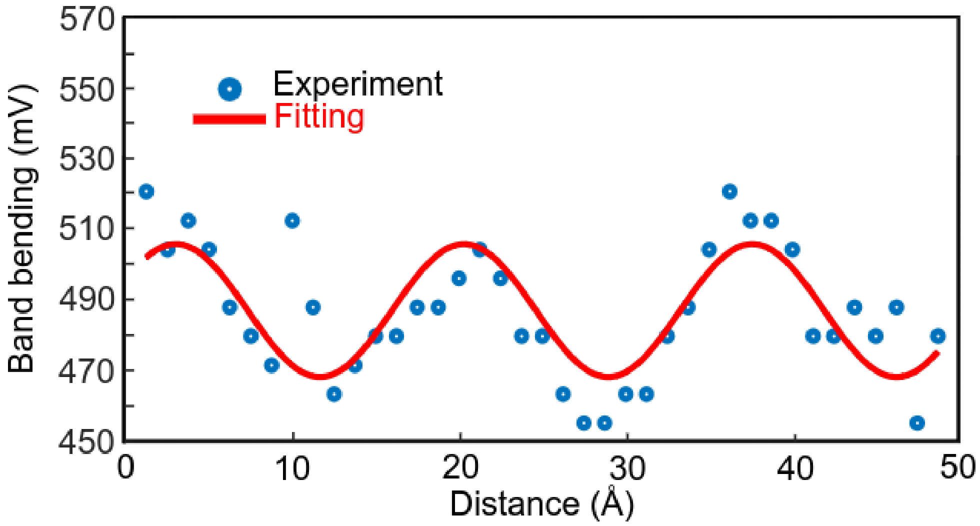
Theory



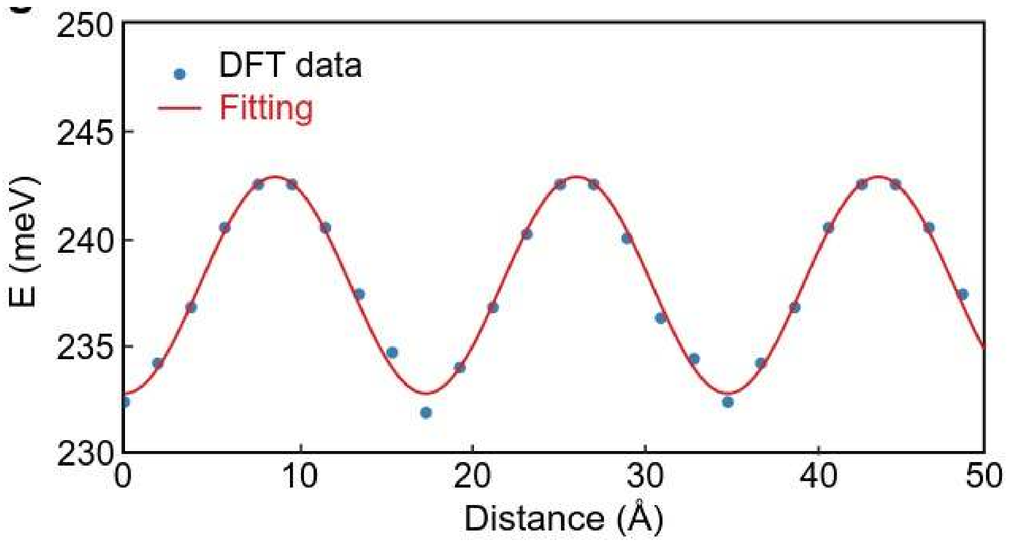
$$\Lambda \mathrm{q}(-\sin(2\mathbf{q}\cdot\mathbf{r}), \sin^2(\mathbf{q}\cdot\mathbf{r}), 0)$$

Ferroelectricity from modulated band offset

Band offset (experiment)



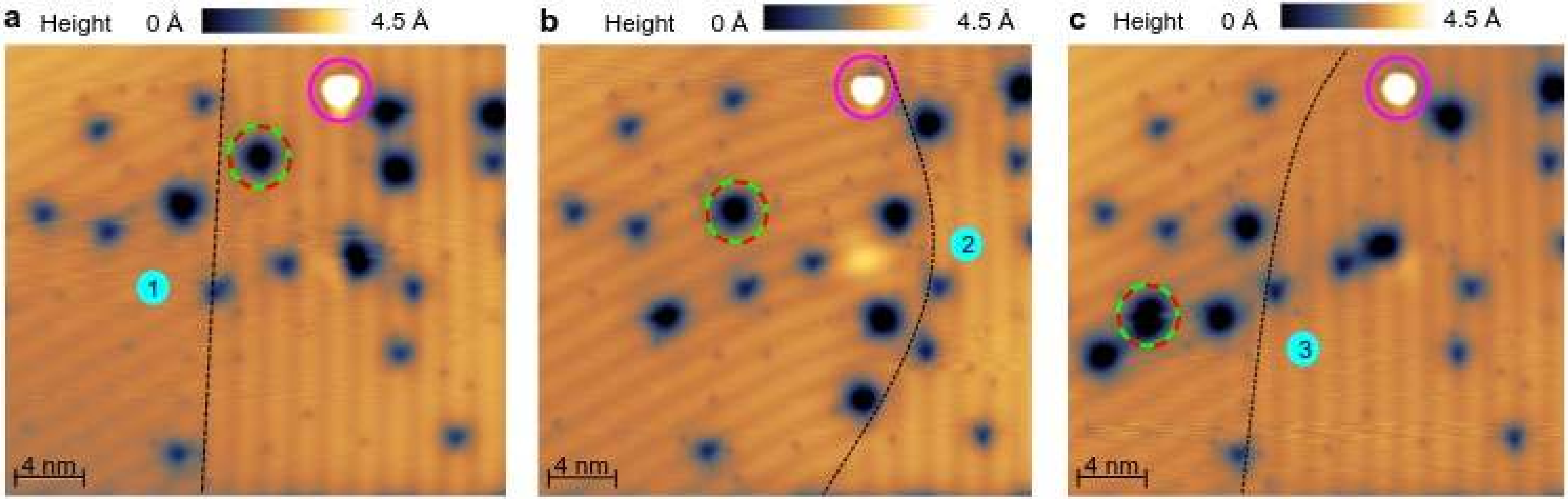
Band offset (theory)



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

Advanced Materials, 2311342 (2024)

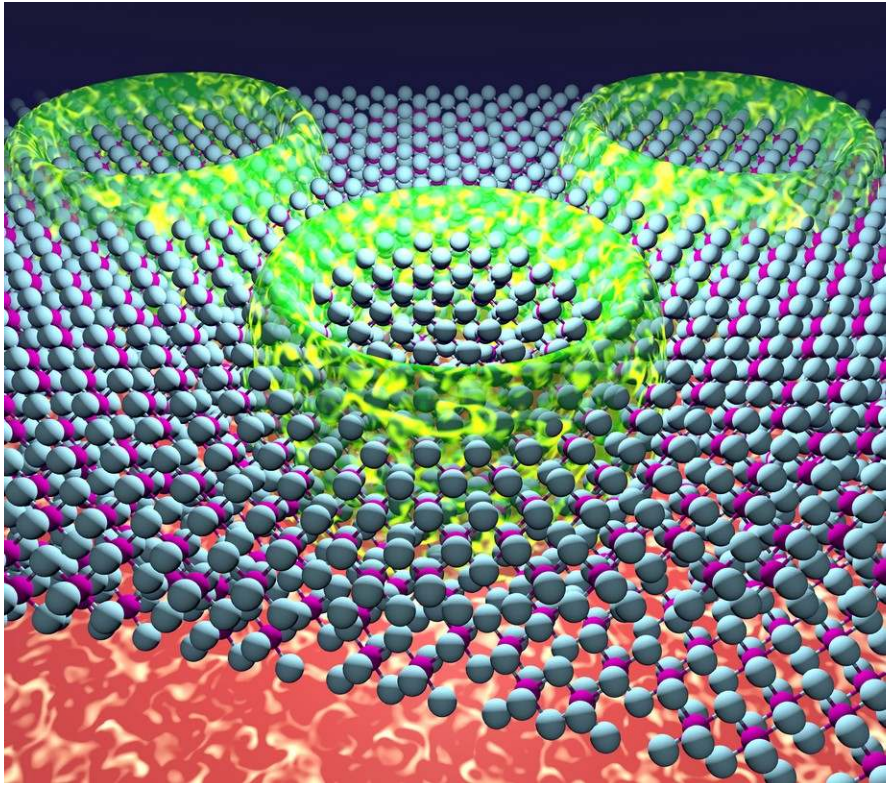
Controlling multiferroic domains with an STM tip



An STM bias allows to control the multiferroic domains

Advanced Materials, 2311342 (2024)

Engineering artificial multiferroics with twisted van der Waals materials

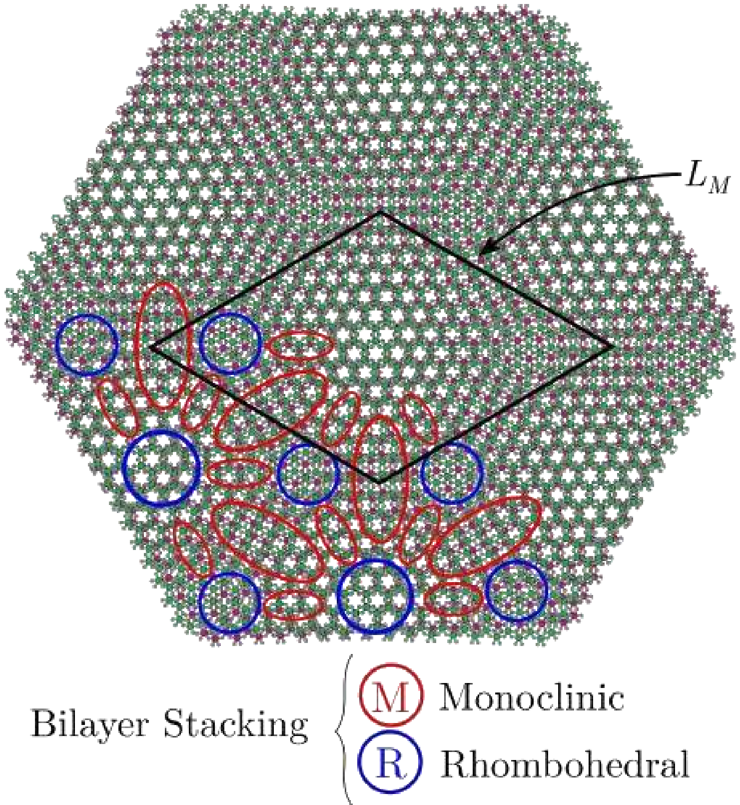


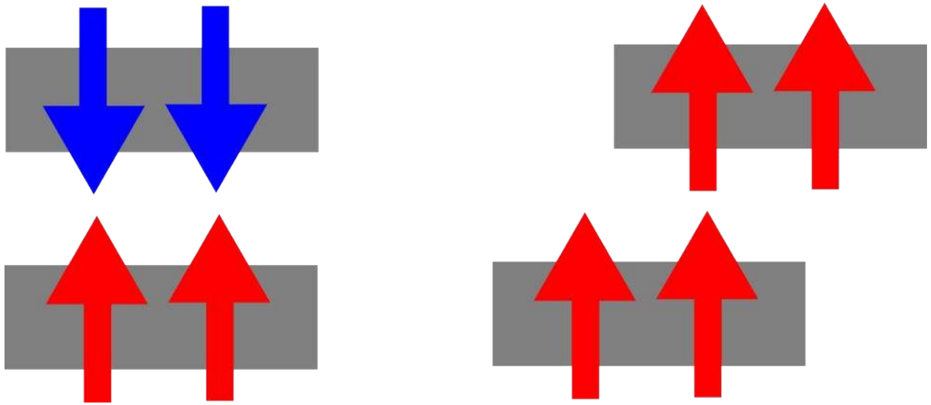
A. Fumega

2D Materials 10 025026 (2023)

Twisted 2D trihalides

$CrBr_3$



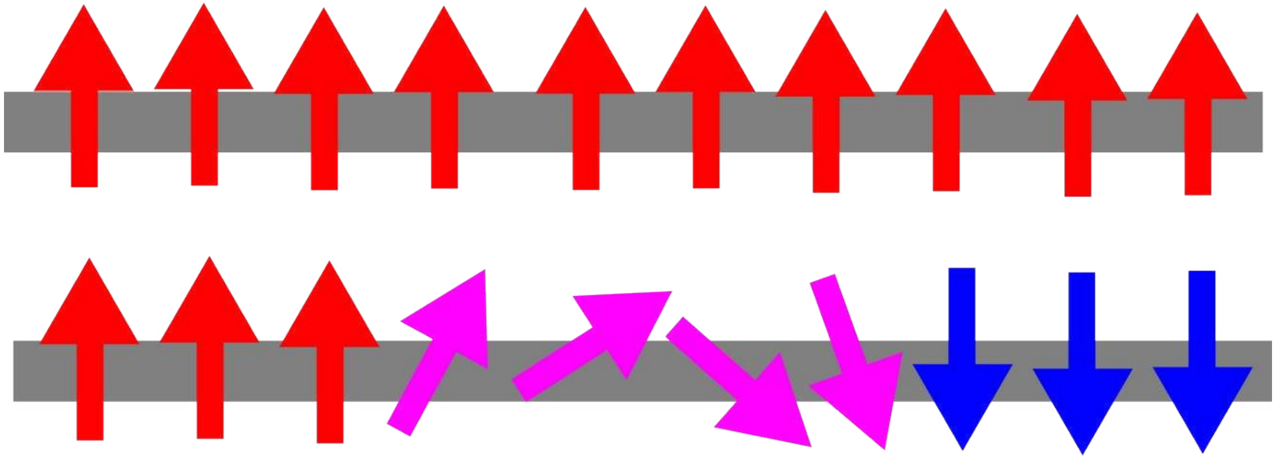


The local stacking determines the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls

Stacking #1



Stacking #2

Mixed FE/AF domains

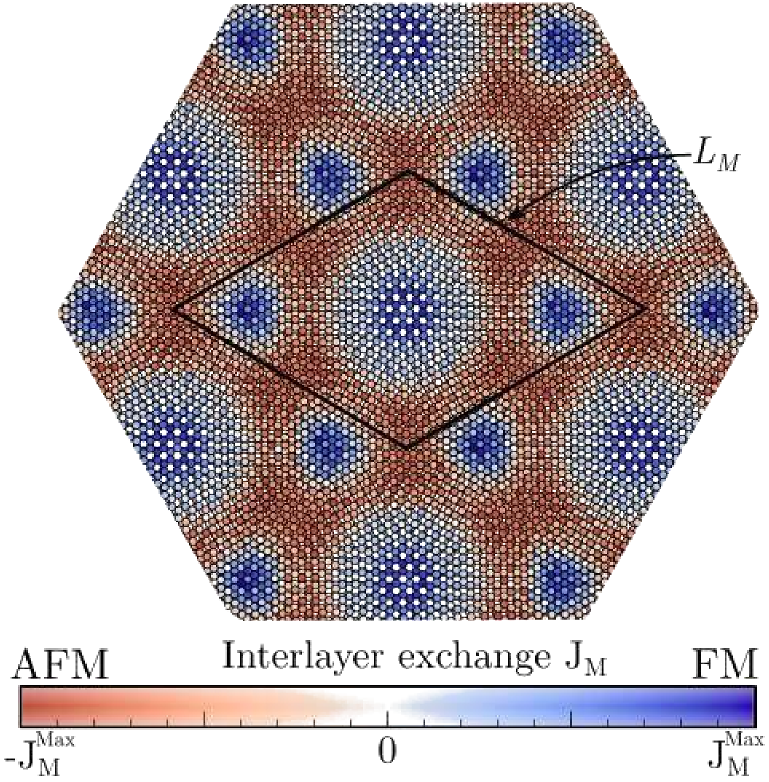
Nature Nanotechnology 17, 143–147 (2022)

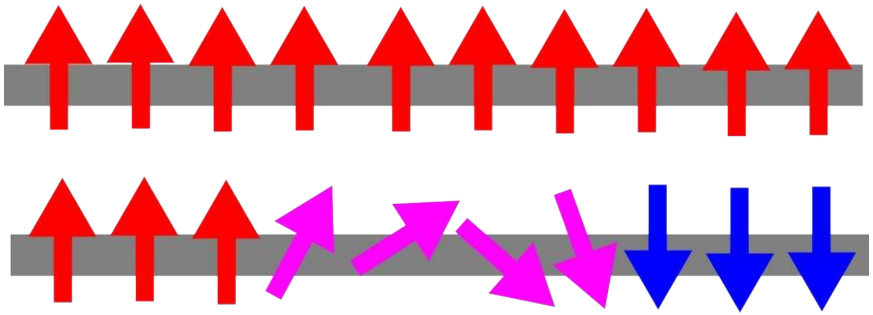
Non-collinear magnetism

Nature Physics 19, 1150–1155 (2023)

Twisted 2D magnetic materials

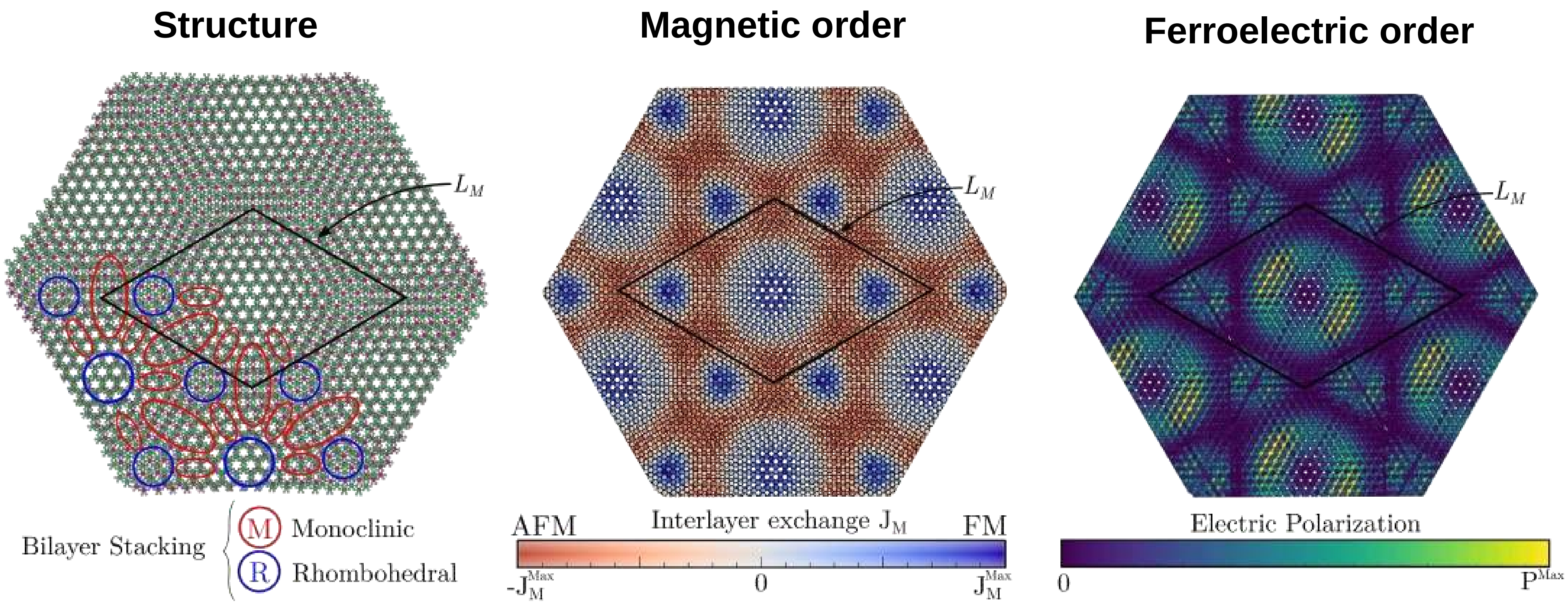
Non-collinear magnetism and multiferroic order appear due to the moire





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

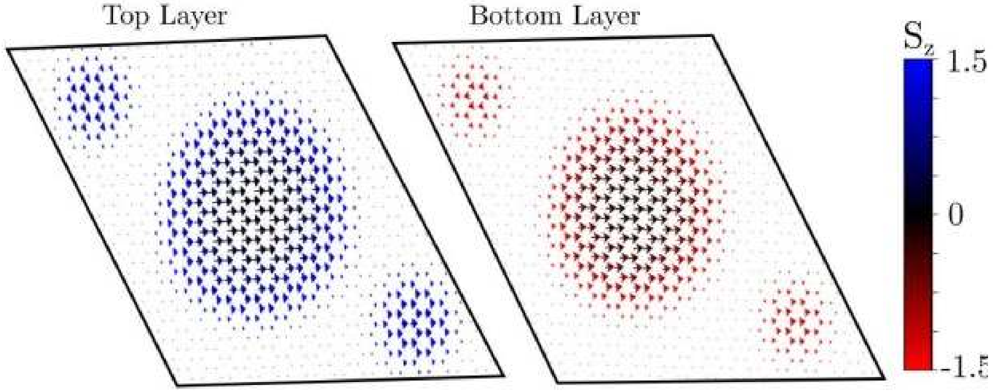
Stacking, magnetic order and electric polarization



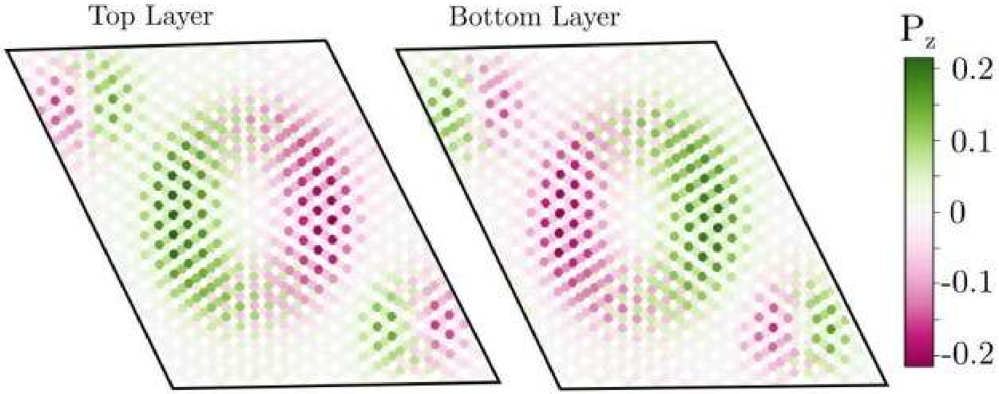
Stacking, magntic structure and ferroelectric order are correlated

Magnetization and ferroelectricity

Magnetization

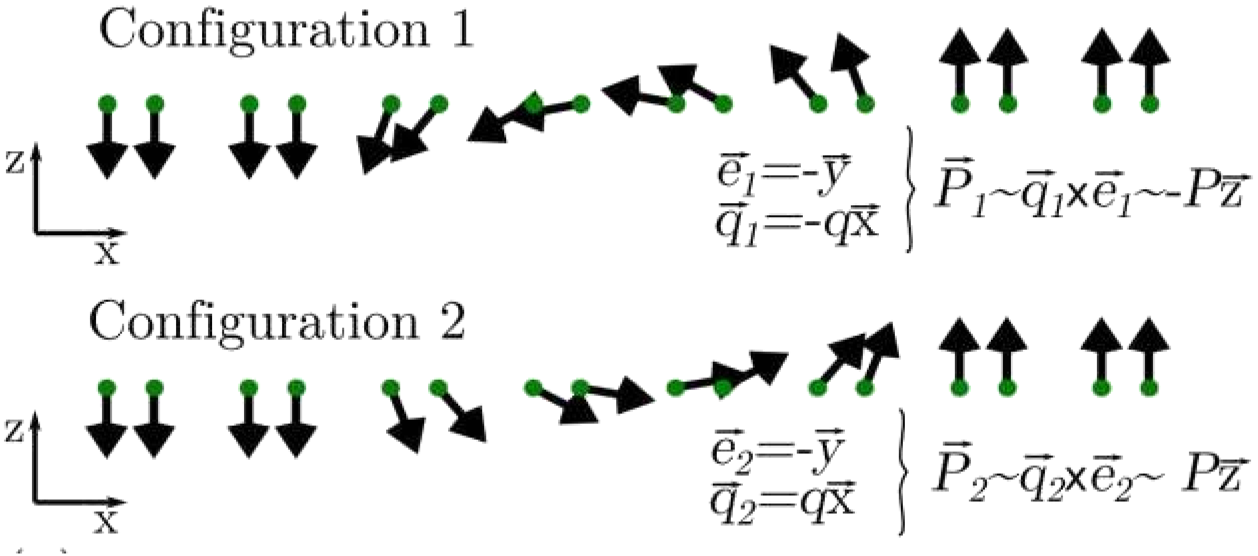


Ferroelectric order



The ferroelectric order appear at the domain walls between magnetic domains

Tracking the emergence of ferroelectricity



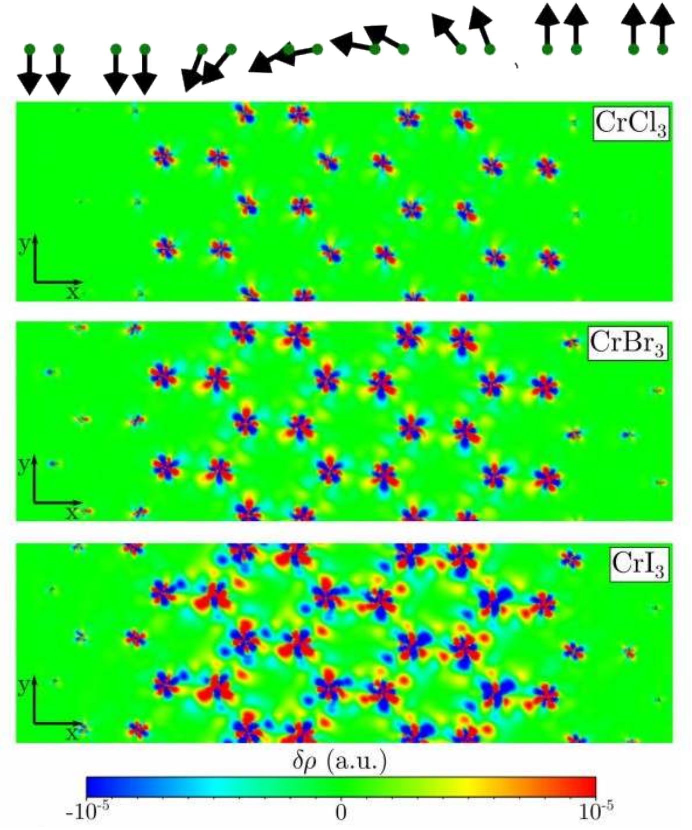
Ferroelectric electronic density

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

Ferroelectric force

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

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

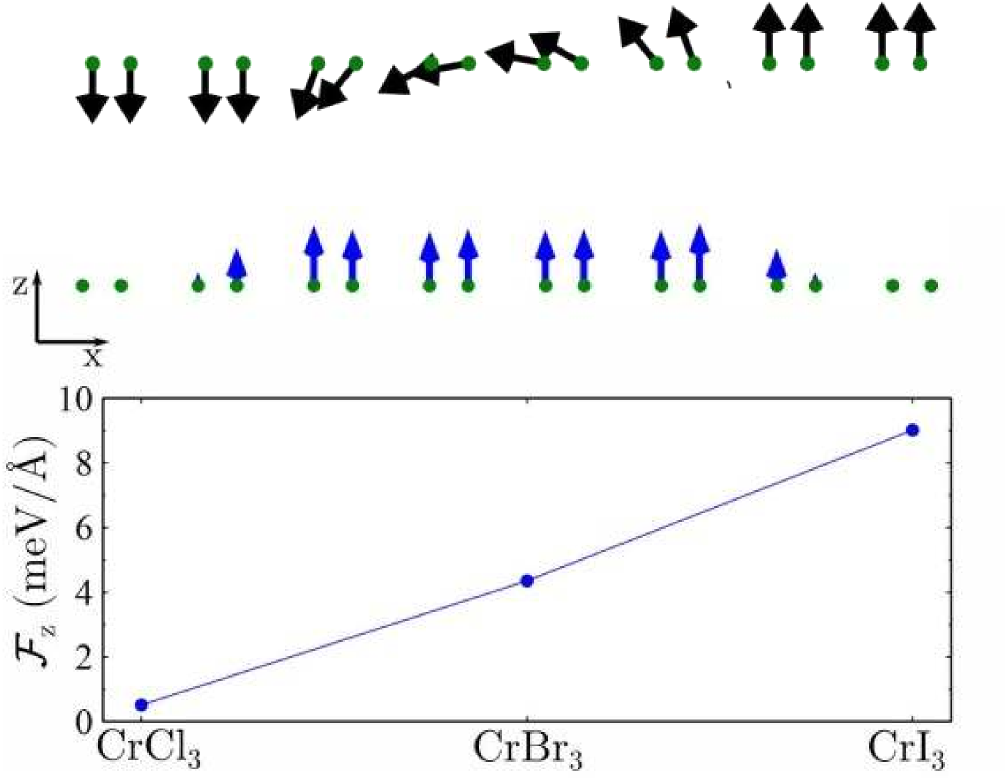
The ferroelectric force

Magnetic configuration

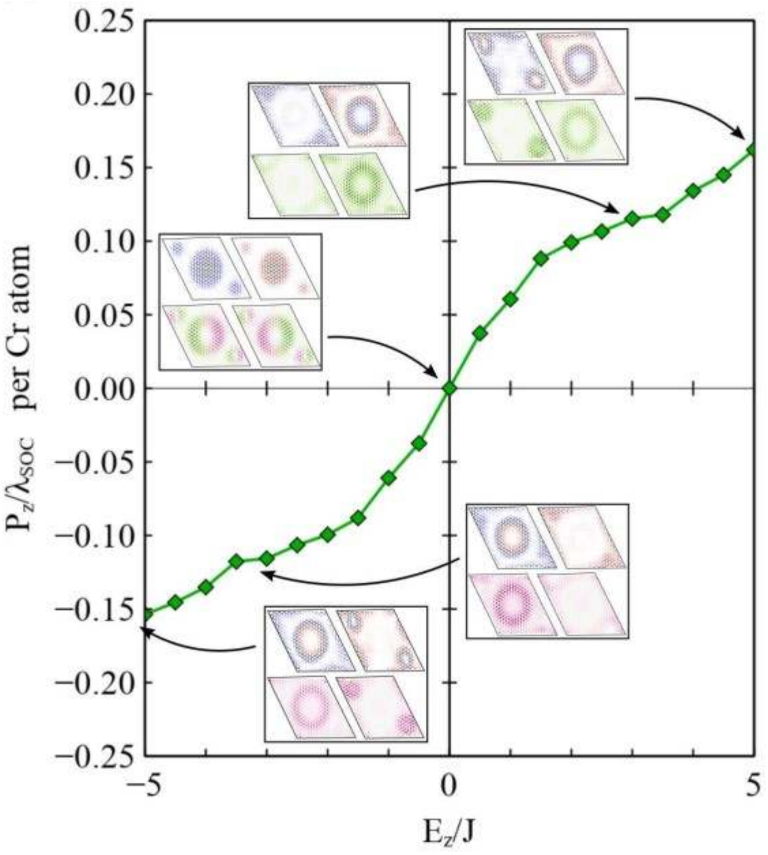
Ferroelectric force

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

The ferroelectric force becomes
Stronger for heavier halides



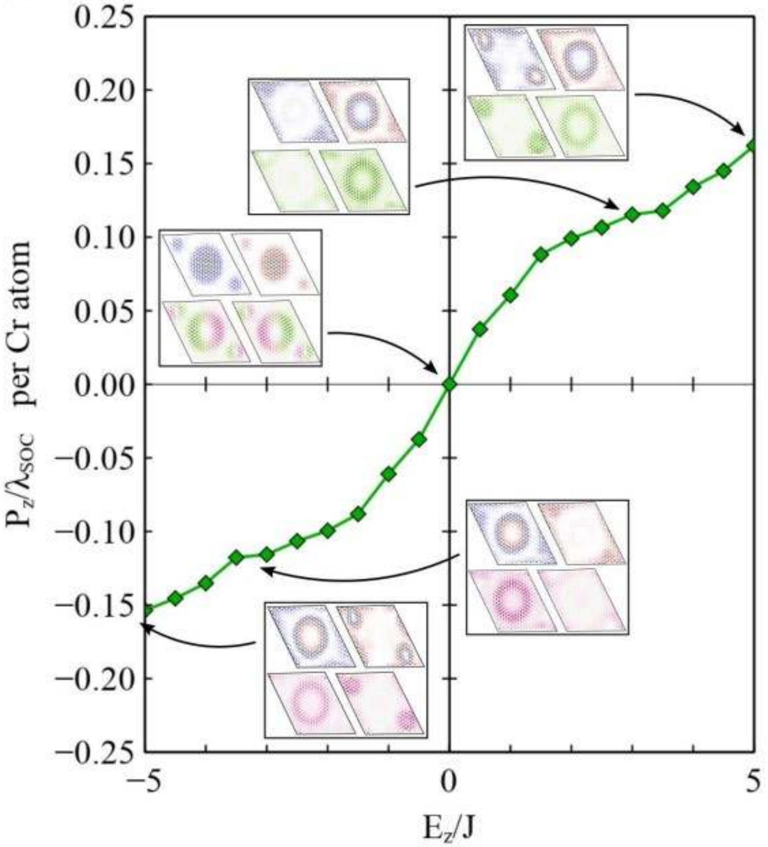
Electrically controllable skyrmions in twisted magnets



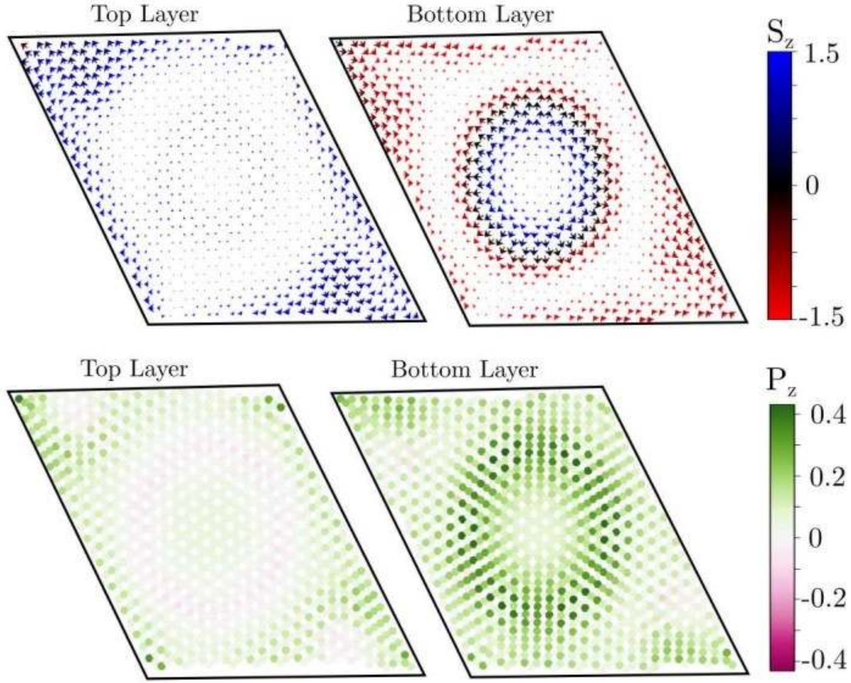
The electric polarization directly couples to an out-of plane electric field

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

Electrically controllable skyrmions in twisted magnets

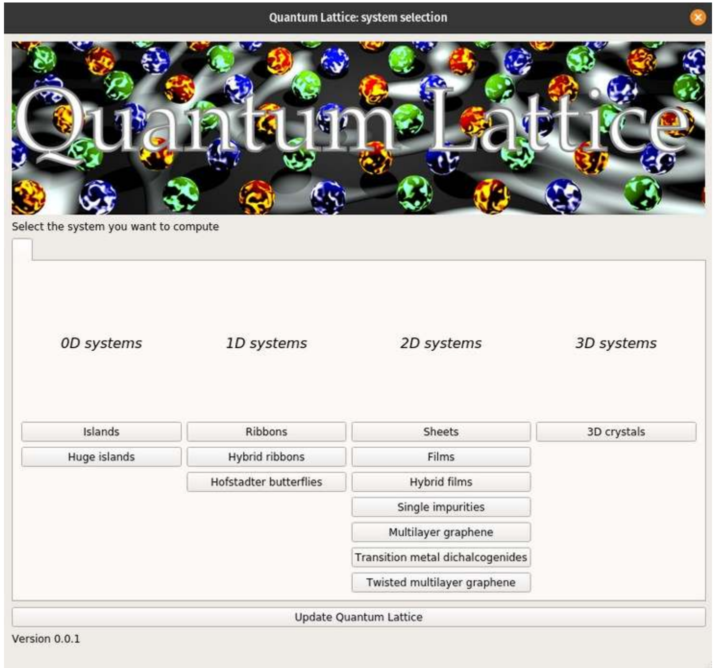


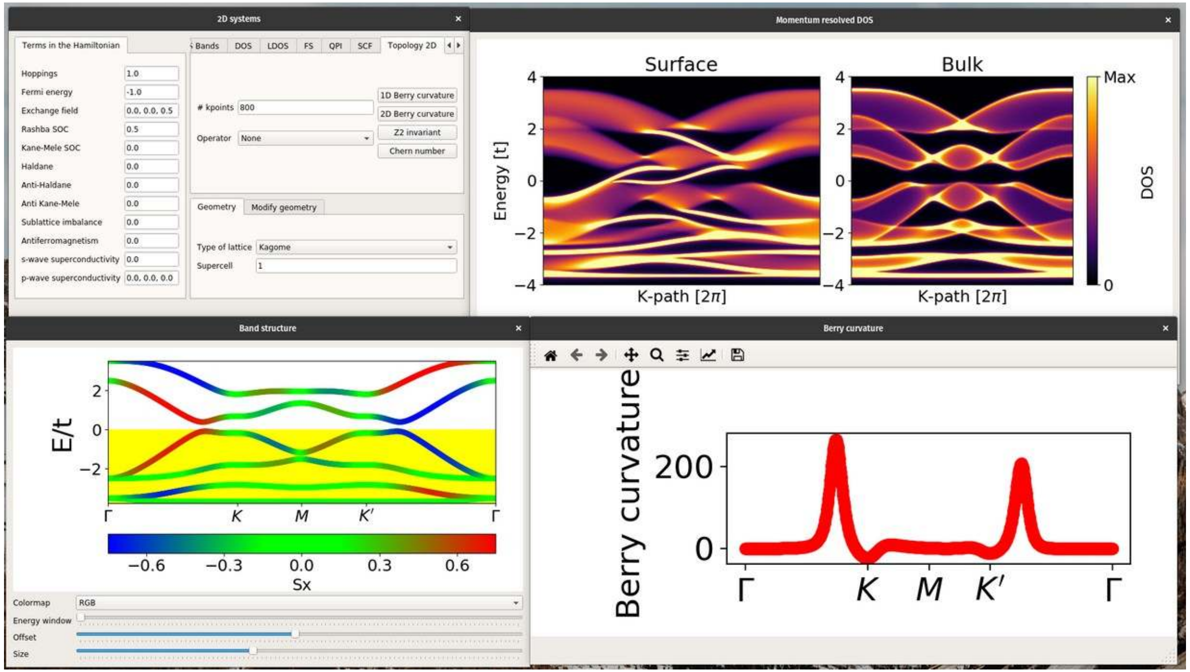
Skyrmions at finite interlayer bias



Open-source software for van der Waals materials

Quantum Lattice: A user interface to compute electronic properties

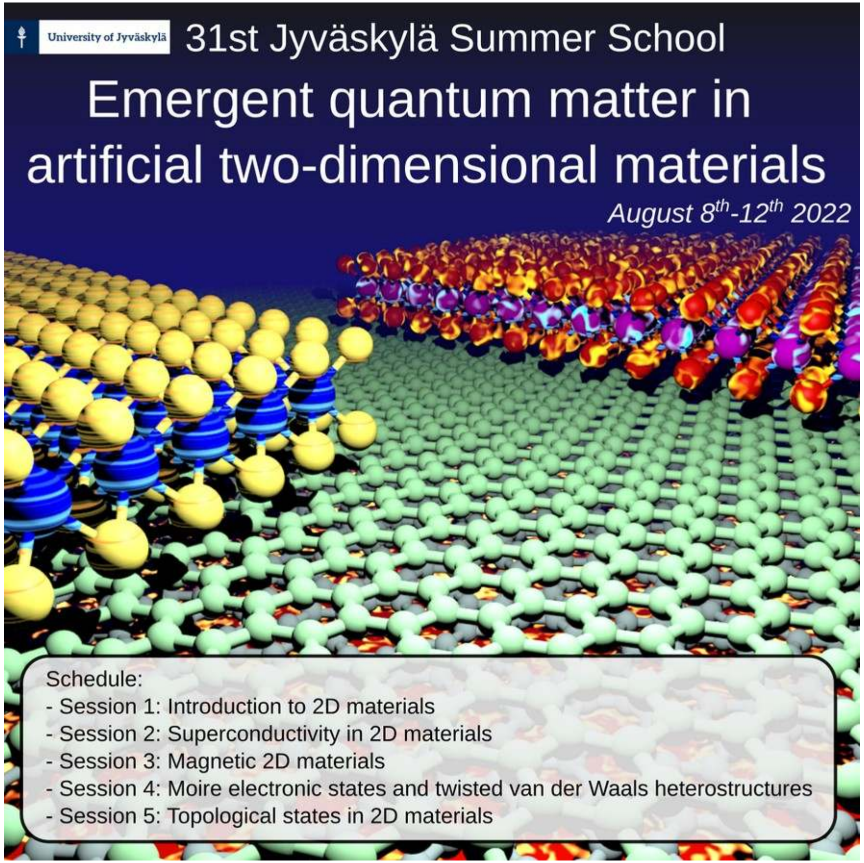




Quantum Lattice: open source interactive interface for tight binding modeling

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

Quantum matter in van der Waals materials school



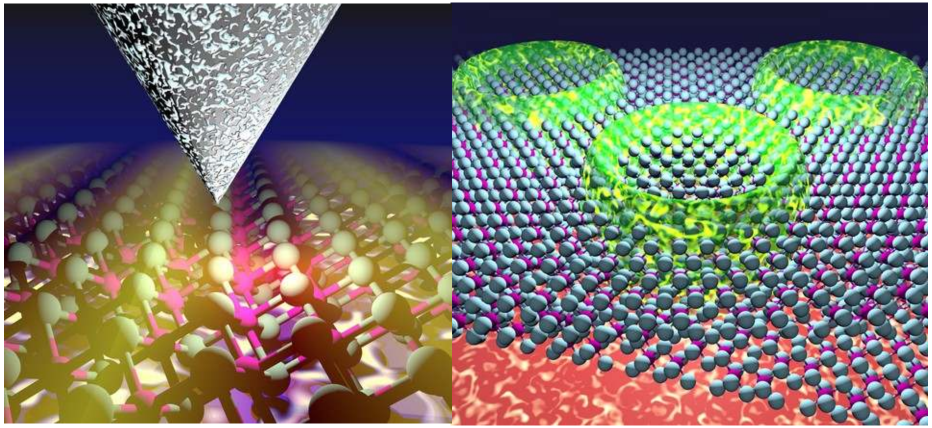
Recordings available at

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 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

J&AE JANE AND AATOS ERKKO FOUNDATION

SUOMEN AKATEMIA

FQF FINNISH QUANTUM FLAGSHIP