Microscopic evidence of type-II multiferroicity and topological spin textures in monolayer NiI_2

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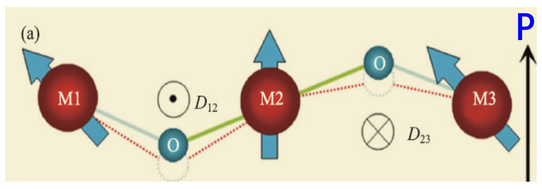
Introduction

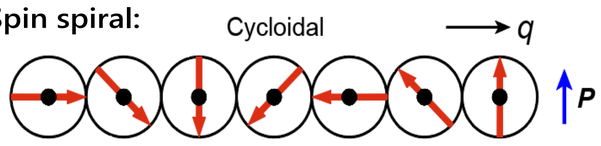
Type-II Multiferroicity



Ferroelectricity driven by noncolinear spin structure:

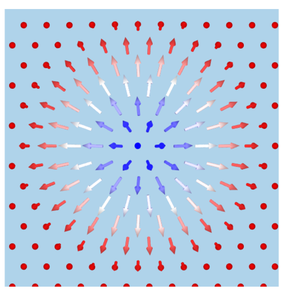
$$\boldsymbol{p} = \boldsymbol{e}_{ij} \times (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

Spin spiral: Cycloidal $\rightarrow q$



Microscopic investigation is lacking!

Topological spin textures

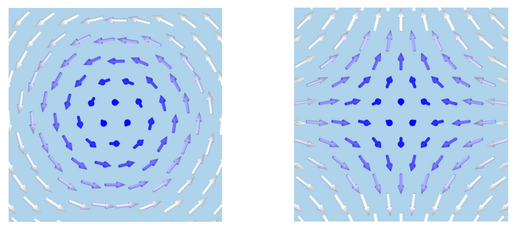


Skyrmion ($N = 1$)

$$\rho_T = \boldsymbol{n} \cdot \left(\frac{\partial \boldsymbol{n}}{\partial x} \times \frac{\partial \boldsymbol{n}}{\partial y}\right)$$

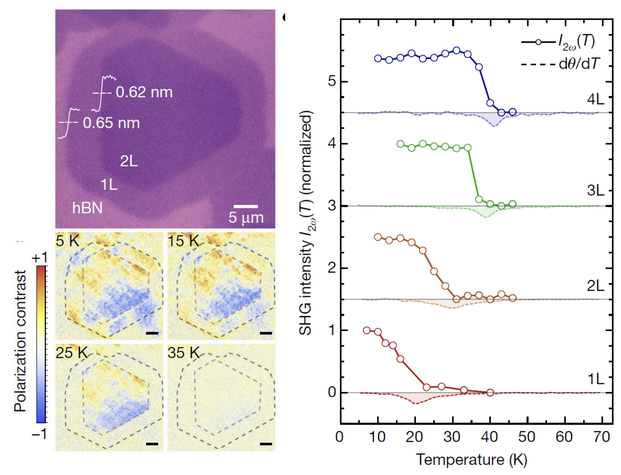
$$N = \frac{1}{4\pi} \iint \rho_T \, dx dy$$

Meron (N = 1/2) Antimeron (N = -1/2)



Can topological spin structure induce electric polarization?

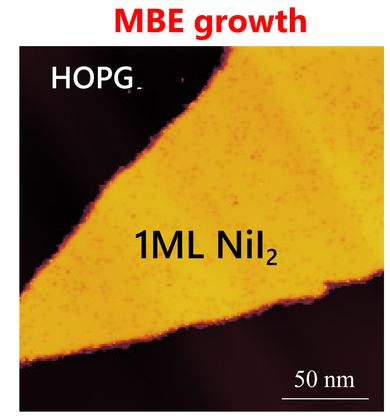
Monolayer NiI_2: a 2D type-II multiferroic ?

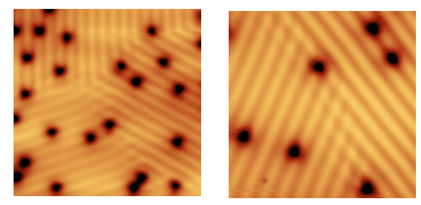


Q. Song et al., Nature 602, 601 (2022).

NiI_2 film

MBE growth

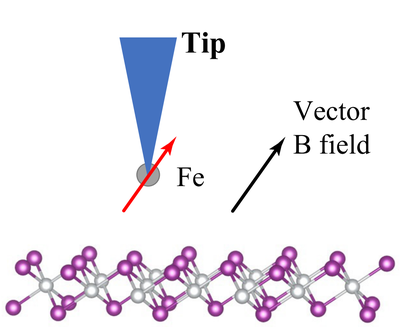


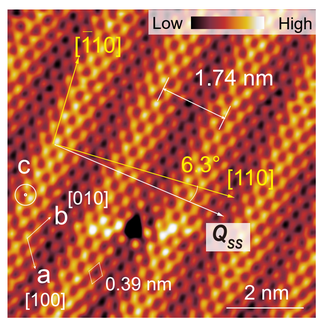


High-quality samples !

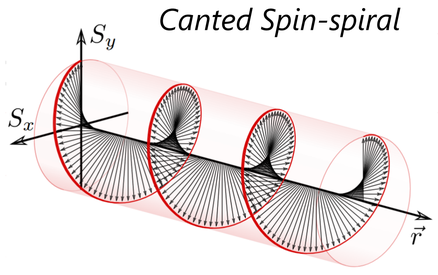
Determining the spin spiral state

Vectorial spin polarized STM: 3D spin sensitivity!

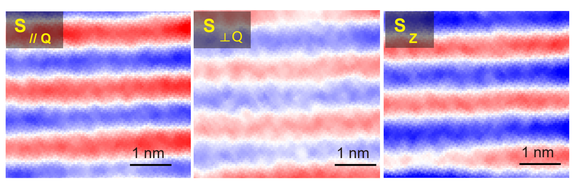


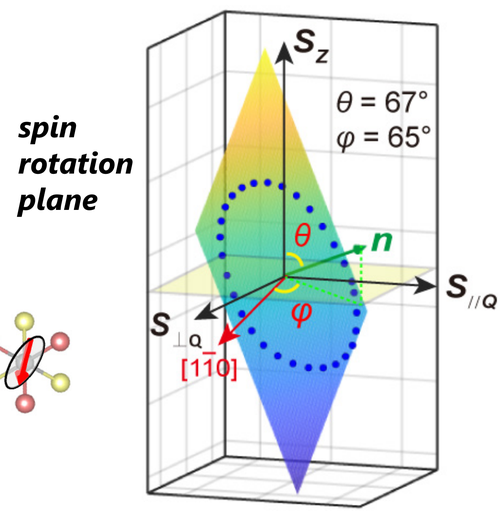


Canted Spin-spiral

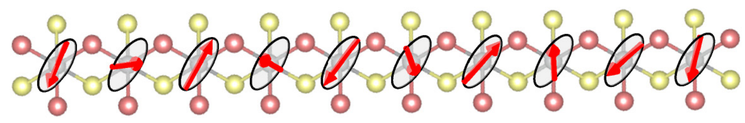


Determining 3D spin structure of spin spiral



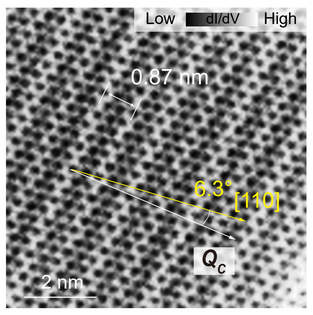


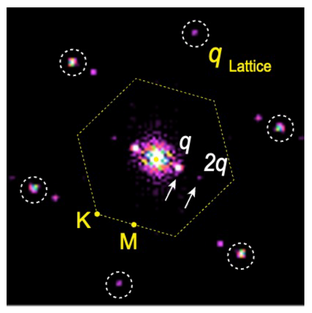
spin rotation plane



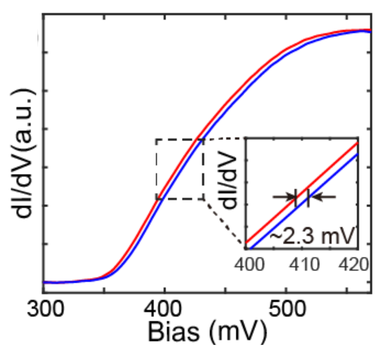
***Right-handed spin spiral with a tilted plane*!**
(Coexistence of right- and left- handed)

Coexistence of 2Q charge modulation



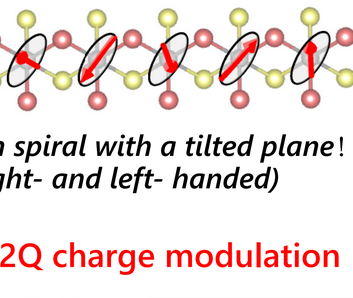


Band shift induced by 2Q



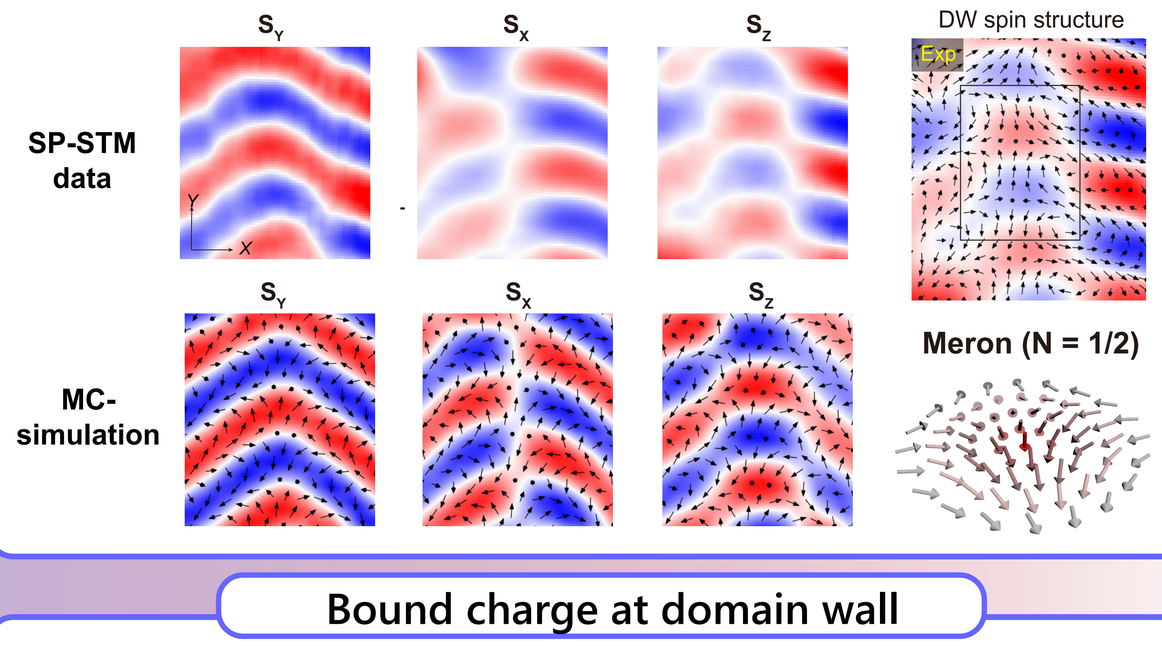
2Q modulation with band bending
(half the period of helical magnetic stripes, same wavevector direction)

Multilayer NiI_2: *Thickness dependence of spin spiral*

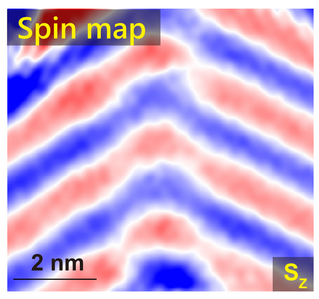


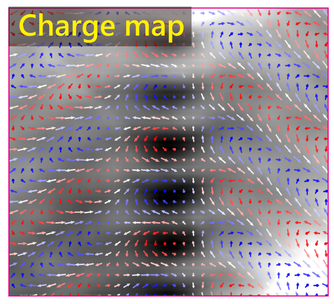
Topological spin structure at DW

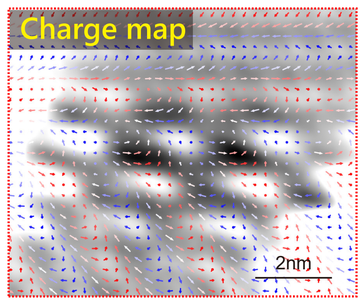
Meron/Antimeron pair is directly resolved at domain wall!

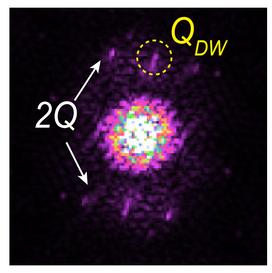


Bound charge at domain wall

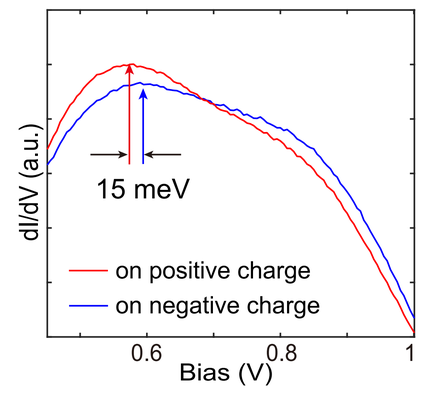








Band shift at DW



Bound charge is evidenced by band shift at domain walls!

Signature of ferroelectricity!

Modeling/Simulations

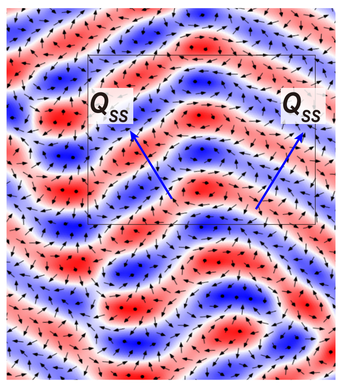
gKNB model

$$H = \sum_{\langle i,j \rangle_1} \left[J_1 \boldsymbol{S}_i \cdot \boldsymbol{S}_j + K S_i^{\gamma} S_j^{\gamma} + B(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)^2\right] + \sum_{\langle i,j \rangle_3} J_3 \boldsymbol{S}_i \cdot \boldsymbol{S}_j$$

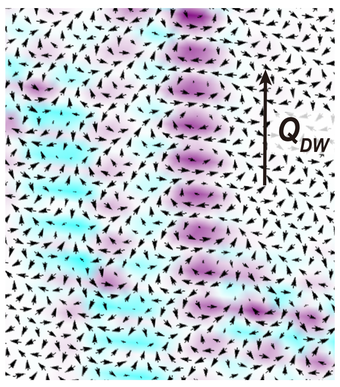
Kitaev K Biquadratic B

$$\boldsymbol{P} = \sum_{\langle ij \rangle_n}^{n=1,3} M_{ij}^n (\boldsymbol{S}_i \times \boldsymbol{S}_j)$$

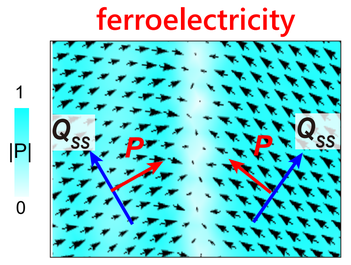
Monte Carlo simulation of spin structure

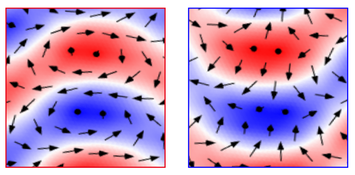


Bound charge distribution



ferroelectricity

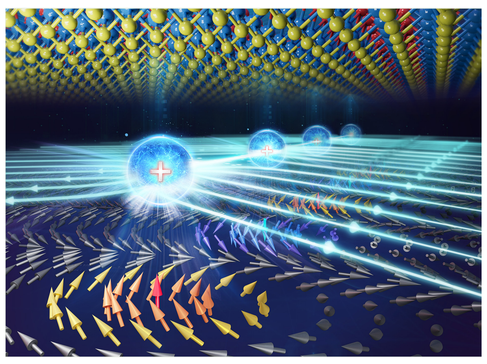




- *The spin model of NiI2 and GKNB theory well reproduced the observed spin/charge feature, further support the spin-driven multiferoricity!*

Summary

- *Spin structure of 1ML NiI_2*
  - *Spin spiral with canted rotation plane and topological Meron/Antimeron pair are directly determined by SP-STM*
- *Microscopic evidence of type-II multiferroicity.*
  - *Bound charge is associated with topological Meron/Antimeron pairs, providing microscopic evidence for Type-II multiferroicity.*



Ref: H. Wang et al., *Microscopic Evidence of Spin-Driven Multiferroicity and Topological Spin Textures in Monolayer NiI_2*, Phys. Rev. Lett. 136, 026402 (2026).