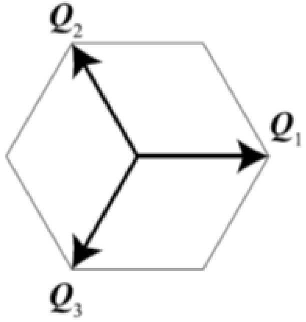
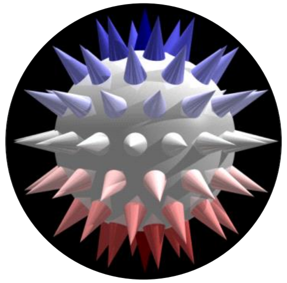
# Magnetism at the Limit: from Skyrmions to Antiferromagnets in Model-type Systems, studied with STM

Kirsten von Bergmann
University of Hamburg

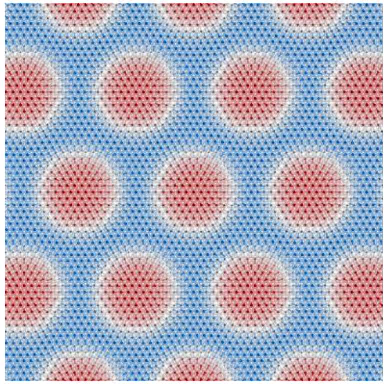
# From the skyrmion lattice to the triple-q state

**Multi-q states**



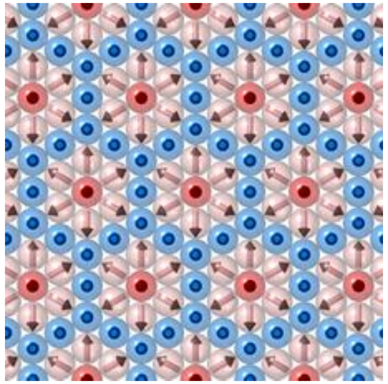
multi-q state
magnetic field induced
**Skyrmion lattice**



governed by
$J$, DMI, Zeeman

S. Mühlbauer et al., Science **323**, 915 (2009).
X.Z. Yu et al., Nature **465**, 901 (2010).
N. Romming, …KvB et al., Science **341**, 636 (2013).
A.O. Leonov, M. Mostovoy, Nat. Commun. 6, 8275 (2015).
C.D. Batista, et al., Rep. Progr. Phys. **79**, 084504 (2016).
N. D. Khanh et al., Nat. Nanotechnol. 15, 444 (2020).
Z. Wang et. al, Phys. Rev. Lett. **124**, 207201 (2020).
S. Hayami and Y. Motome, J. Phys.: Cond. Mat. **33**, 443001 (2021).

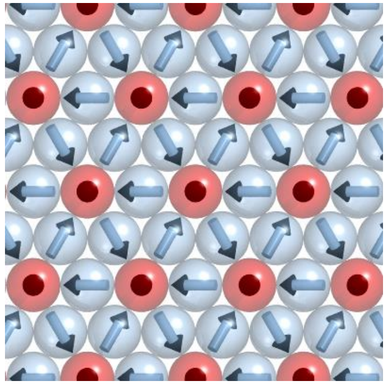
zero field HOI induced
(higher-order interactions)
**Nanoskyrmion lattice**

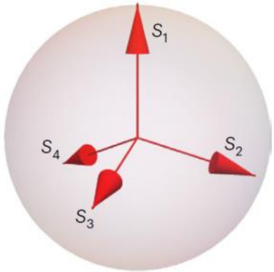


governed by
$J_{ij}$, HOI
(higher order
interactions)

S. Heinze, KvB et al. Nature Phys. **7**, 713 (2011).
KvB et al, Nano Lett. **15**, 3280 (2015).
M. Gutzeit, …, KvB, Nature Commun. **13**, 5764 (2022).

**triple-q state = 3Q**
4 atoms in the unit cell



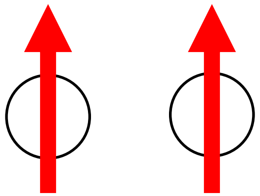



P. Kurz et al., PRL **86**, 1106 (2001).
J. Spethmann, ... KvB et al., PRL **124**, 227203 (2020).
H. Takagi et al., Nature Phys. **19**, 961 (2023).
P. Park et al., Nat. Commun. **14**, 8346 (2023).
F. Nickel, ... KvB, Phys. Rev. B **108**, L180411 (2023).

# atomic- and nano-scale spin textures

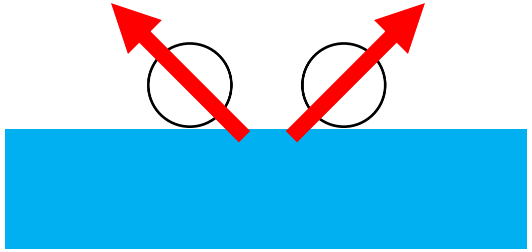
magnetic exchange interaction

$$E = -J\,(S_1 \cdot S_2)$$



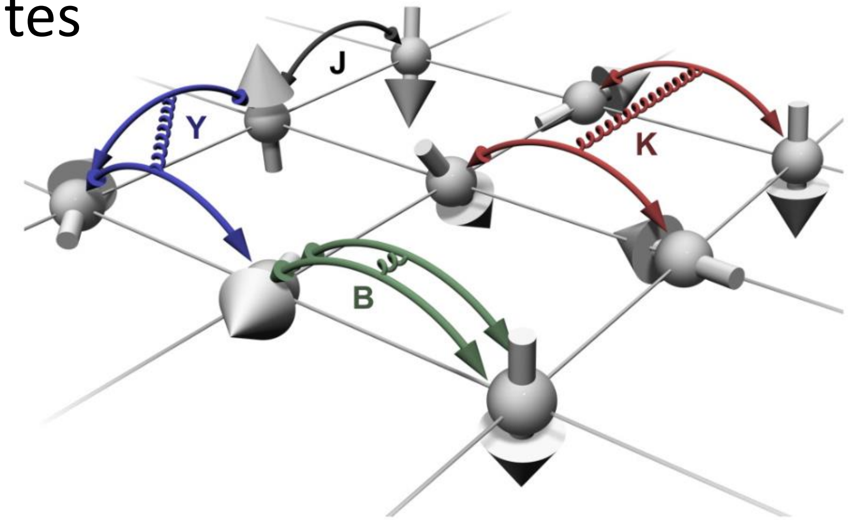
antisymmetric exchange (DMI)
Dzyaloshinskii-Moriya interaction
due to spin-orbit coupling
and broken inversion symmetry

$$E = -D\,(S_1 \times S_2)$$



driving force for
multi-q states

no requirements
for HOIs to exist




M. Hoffmann and S. Blügel, Phys. Rev. B **101**, 024418 (2020).

higher order (4-spin)
exchange interactions

$$H_{\mathrm{HO}} = -\sum_{<ij>} B\left(\boldsymbol{m}_i \boldsymbol{m}_j\right)^2$$

→ biquadratic exchange

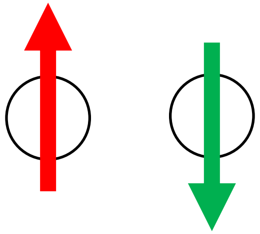
$$-2\sum_{<ijk>} Y_3\left[\left(\boldsymbol{m}_i\boldsymbol{m}_j\right)\left(\boldsymbol{m}_j\boldsymbol{m}_k\right)+\left(\boldsymbol{m}_j\boldsymbol{m}_k\right)\left(\boldsymbol{m}_k\boldsymbol{m}_i\right)+\left(\boldsymbol{m}_k\boldsymbol{m}_i\right)\left(\boldsymbol{m}_i\boldsymbol{m}_j\right)\right]$$

$$-\sum_{<ijkl>} K_4\left[\left(\boldsymbol{m}_i\boldsymbol{m}_j\right)\left(\boldsymbol{m}_k\boldsymbol{m}_l\right)+\left(\boldsymbol{m}_i\boldsymbol{m}_l\right)\left(\boldsymbol{m}_j\boldsymbol{m}_k\right)-\left(\boldsymbol{m}_i\boldsymbol{m}_k\right)\left(\boldsymbol{m}_j\boldsymbol{m}_l\right)\right]$$

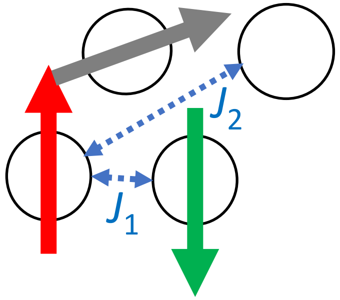
# antiferromagnetic order on a hexagonal lattice

magnetic exchange interaction

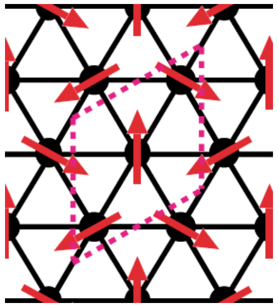
$$E = -J\,(S_1 \cdot S_2)$$



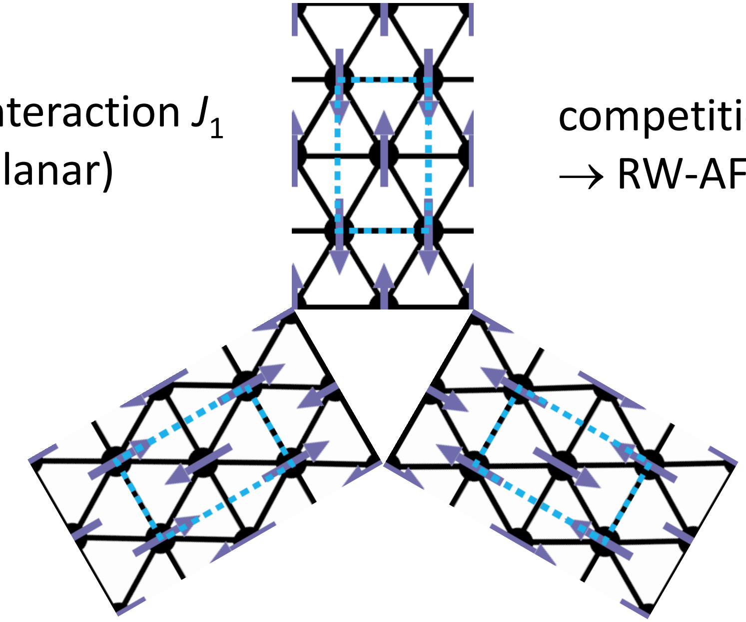
geometric frustration




afm nearest neighbor interaction $J_1$
→ Néel state (120° coplanar)

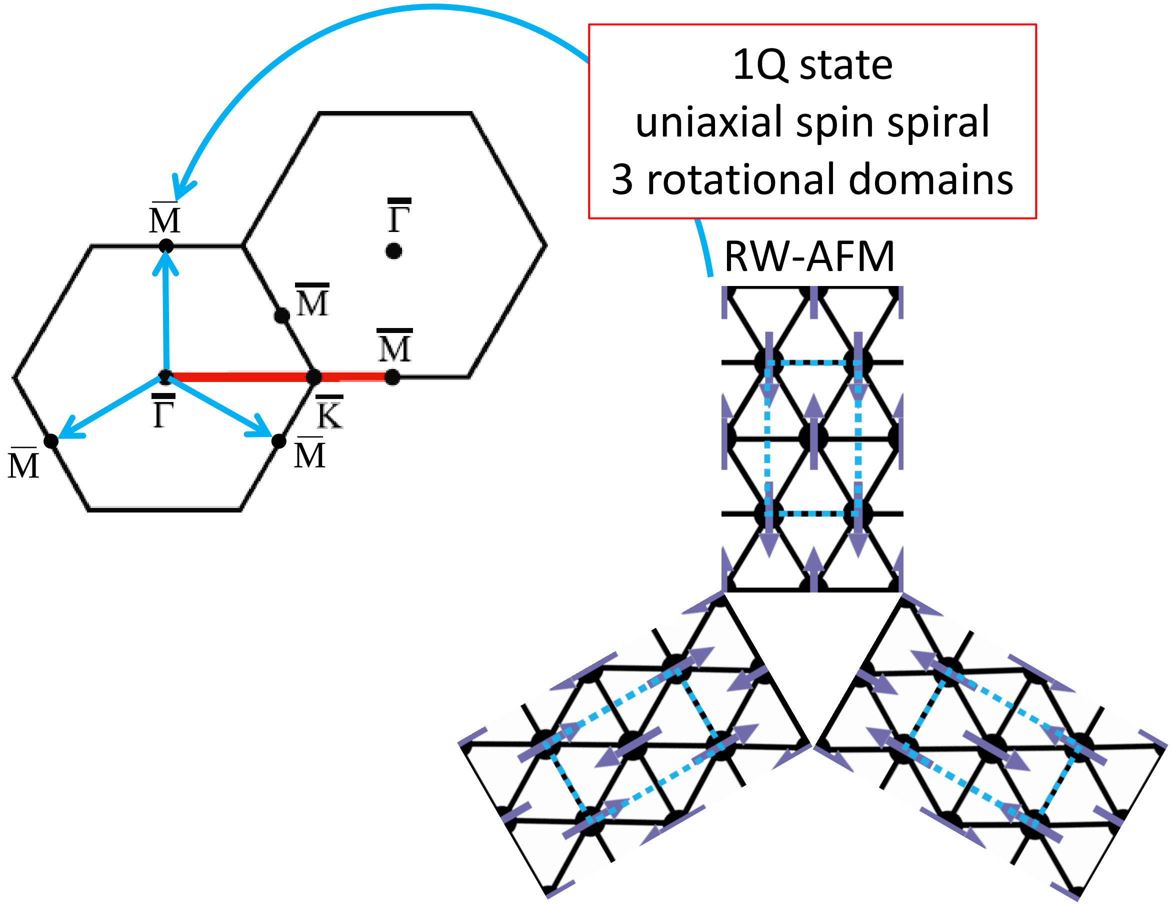


RW-AFM



competition afm $J_1$ and afm $J_2$
→ RW-AFM (180° collinear)

# antiferromagnetic order on a hexagonal lattice

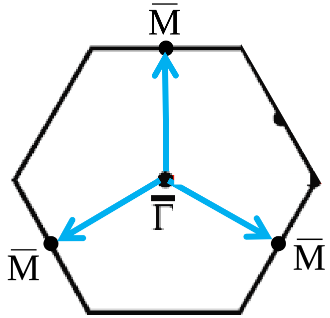


# atomic-scale afm order on a hexagonal lattice

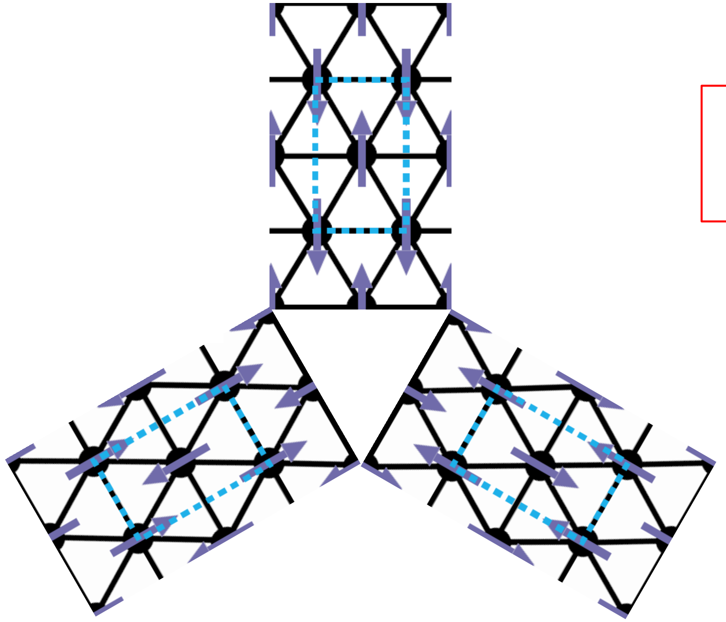
superposition

uniaxial 1Q state
3 rotational domains

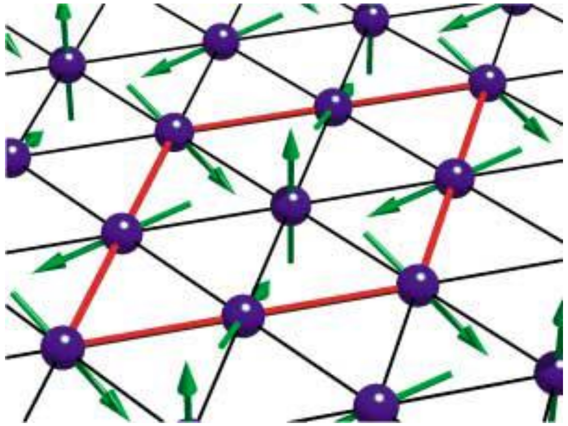
3Q state, triple-q state
3-dimensional spin structure
on 2-dimensional lattice




RW-AFM



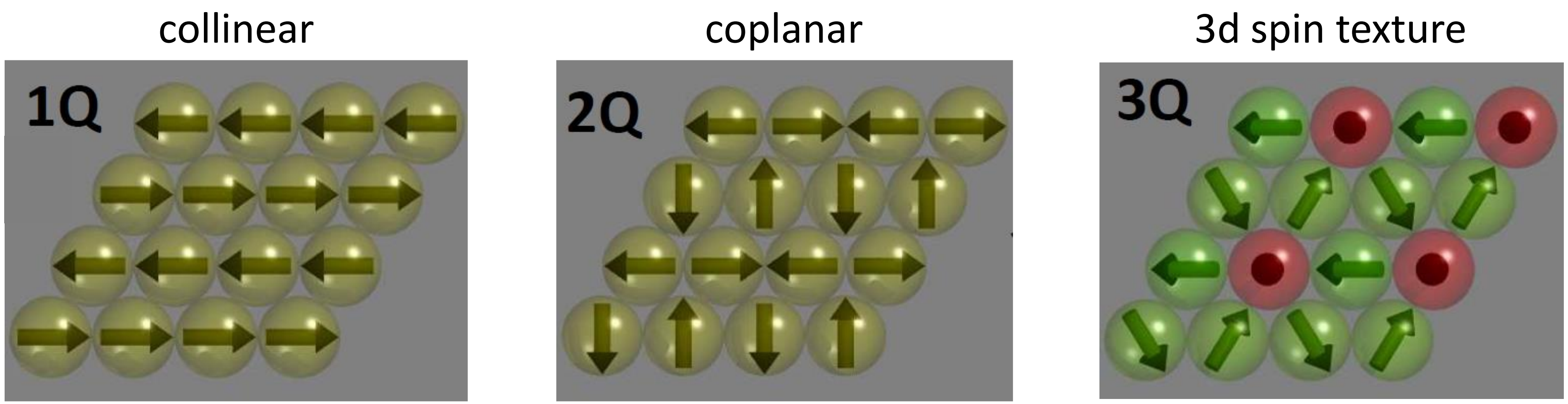
higher-order interactions
decide over 1Q vs 3Q



predicted for Mn-ML/Cu(111)

P. Kurz, et al.,
Phys. Rev. Lett. **86**, 1106 (2001).

# superposition states = multi-Q states



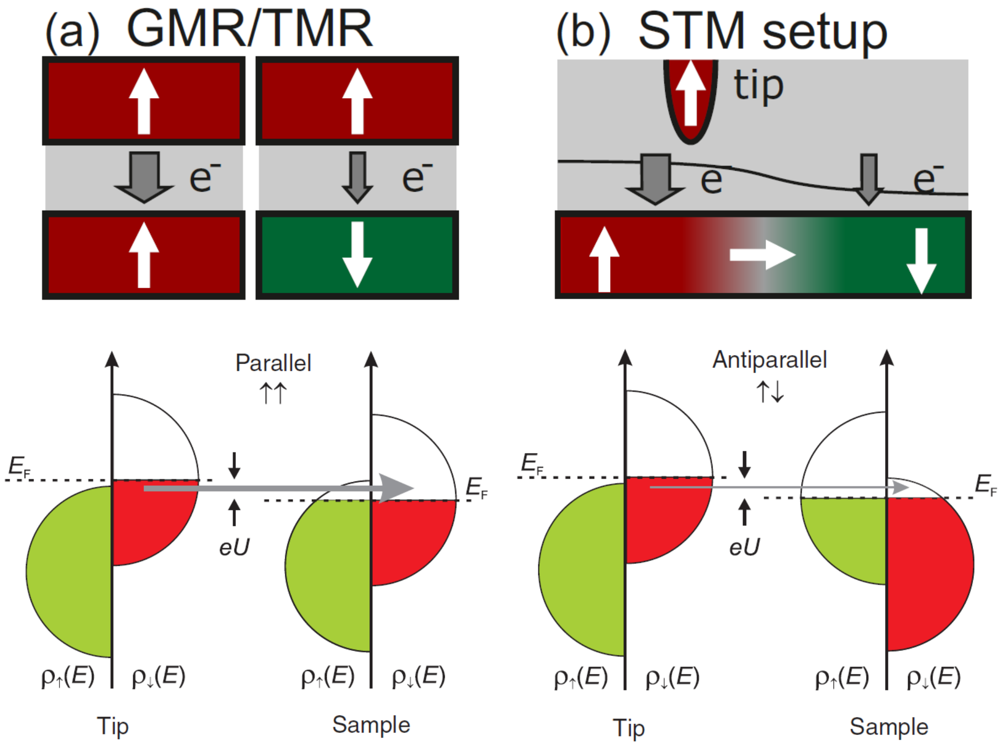

when all HOIs vanish,
these states are degenerate
and are expected to coexist

$$H_{\mathrm{HO}} = -\sum_{<ij>} B(\boldsymbol{m}_i\boldsymbol{m}_j)^2 - 2\sum_{<ijk>} Y_3[(\boldsymbol{m}_i\boldsymbol{m}_j)(\boldsymbol{m}_j\boldsymbol{m}_k) + (\boldsymbol{m}_j\boldsymbol{m}_k)(\boldsymbol{m}_k\boldsymbol{m}_i) + (\boldsymbol{m}_k\boldsymbol{m}_i)(\boldsymbol{m}_i\boldsymbol{m}_j)] - \sum_{<ijkl>} K_4[(\boldsymbol{m}_i\boldsymbol{m}_j)(\boldsymbol{m}_k\boldsymbol{m}_l) + (\boldsymbol{m}_i\boldsymbol{m}_l)(\boldsymbol{m}_j\boldsymbol{m}_k) - (\boldsymbol{m}_i\boldsymbol{m}_k)(\boldsymbol{m}_j\boldsymbol{m}_l)]$$

# spin-polarized STM


M. Bode, Rep. Prog. Phys. **66**, 523 (2003).
R. Wiesendanger, Rev. Mod. Phys. **81** 1495 (2009).
K. von Bergmann et al., J. Phys.: Condens. Matter **26**, 394002 (2014).


TMR-effect
(tunnel magnetoresistance)
in STM geometry

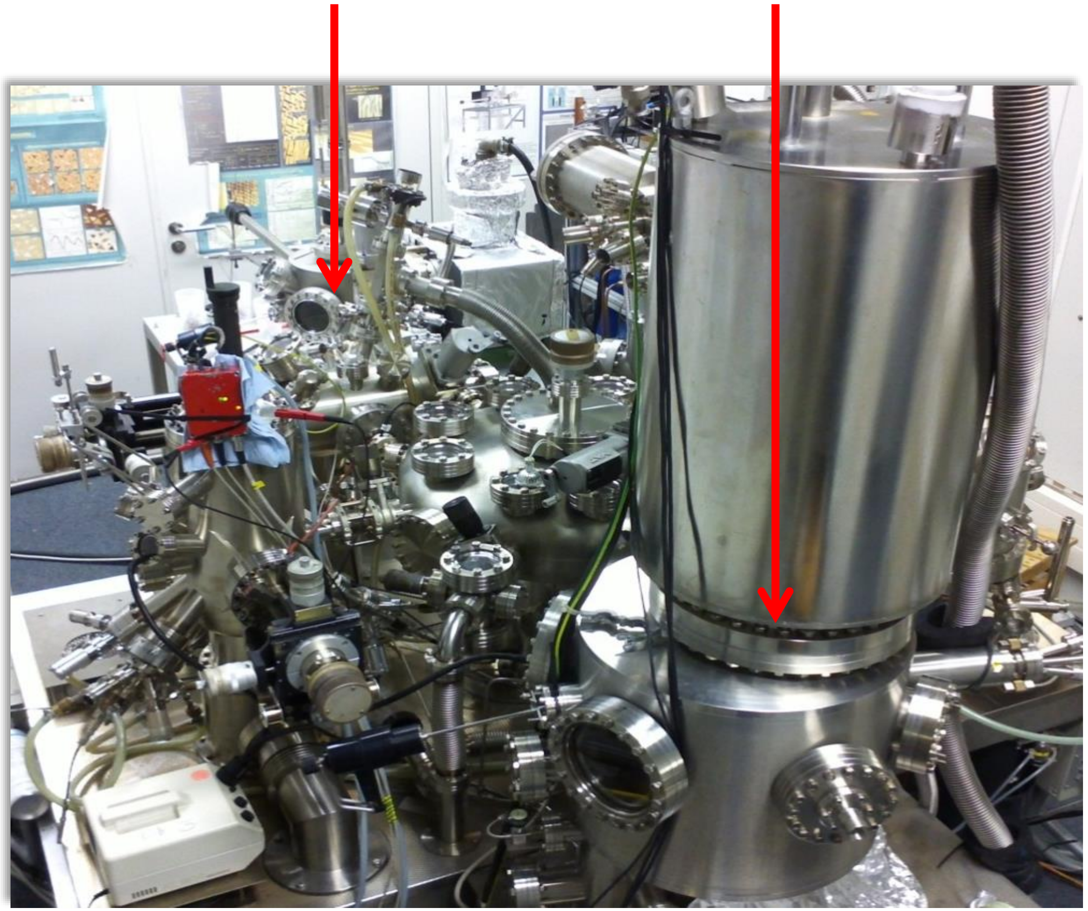



$$I_{SP}(U_0) = I_0[1 + P_s \cdot P_t \cdot \cos(\vec{M}_s, \vec{M}_t)]$$

in-situ tip and sample preparation

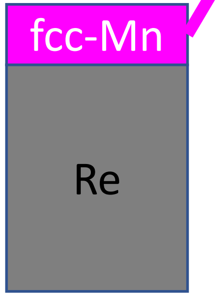
STM
$T$ = 1.3 - 4.2 K
$B_\perp$ < 9 T

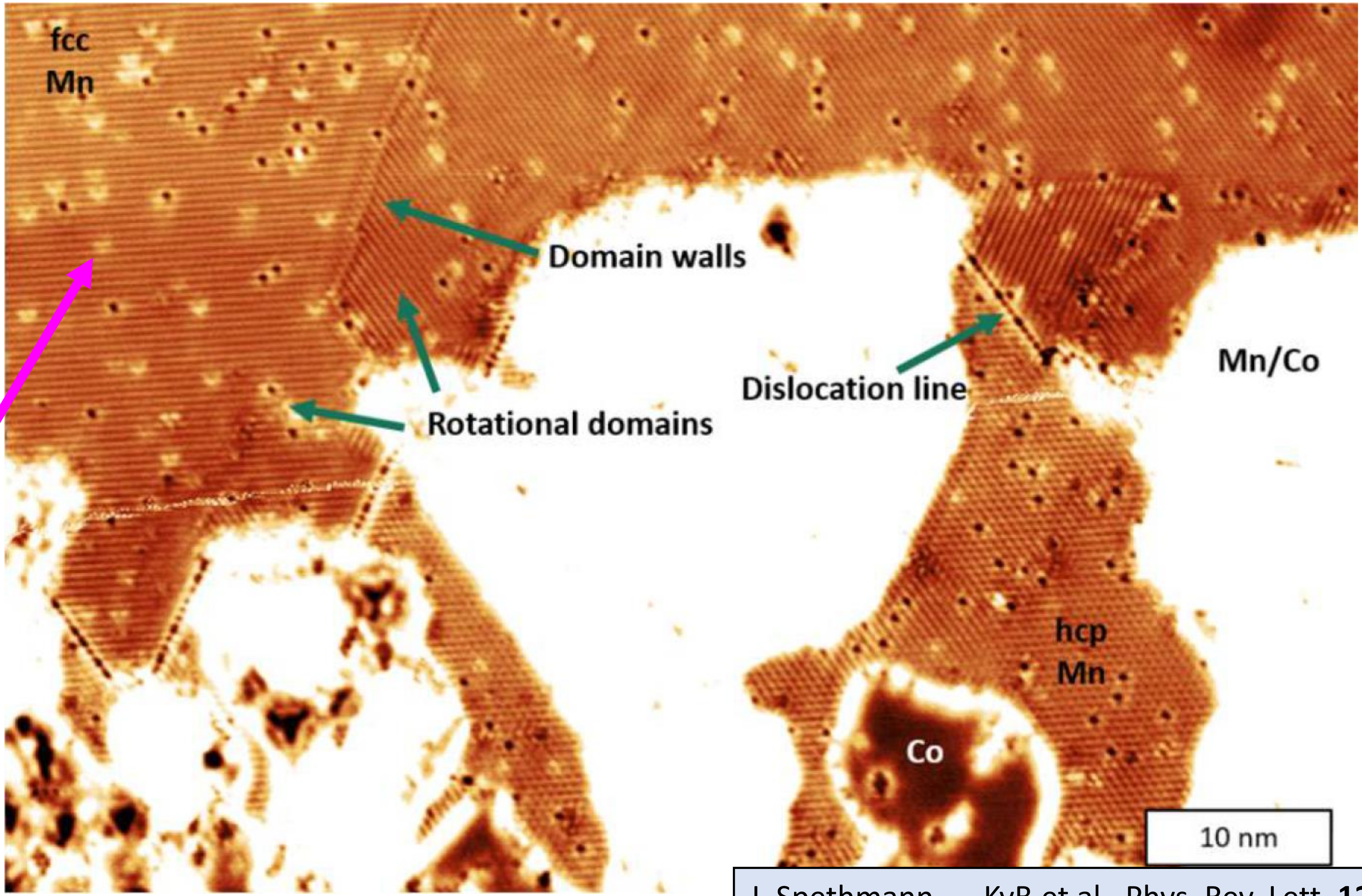
STM
$T$ = 8 - 13 K
$B_\perp$ < 2.5 T



# Mn monolayer on Re(0001)

fcc-Mn in SP-STM: stripes along the close-packed atomic rows, every other row
→ RW-AFM

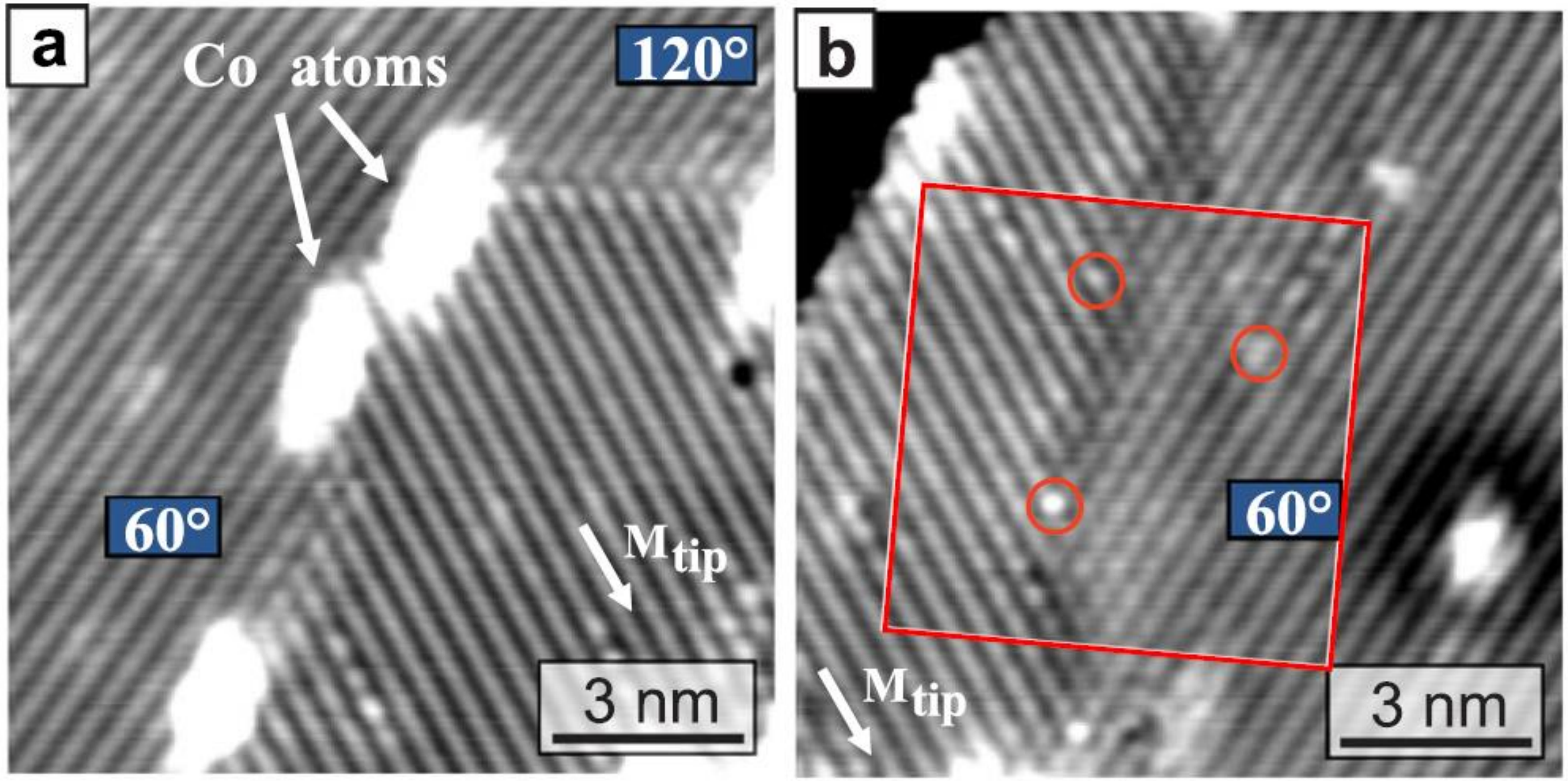


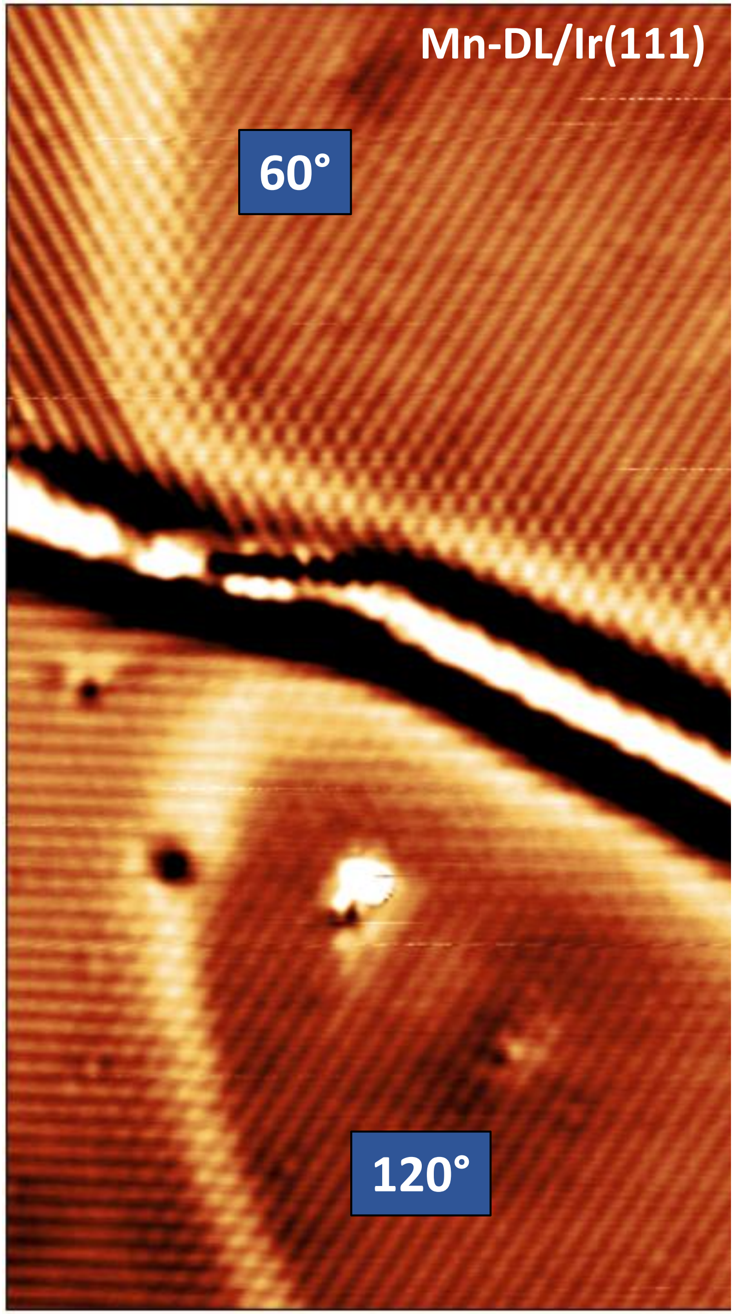

J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).
J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).

# RW-AFM domain walls (DWs)

fcc-Mn/Re(0001)



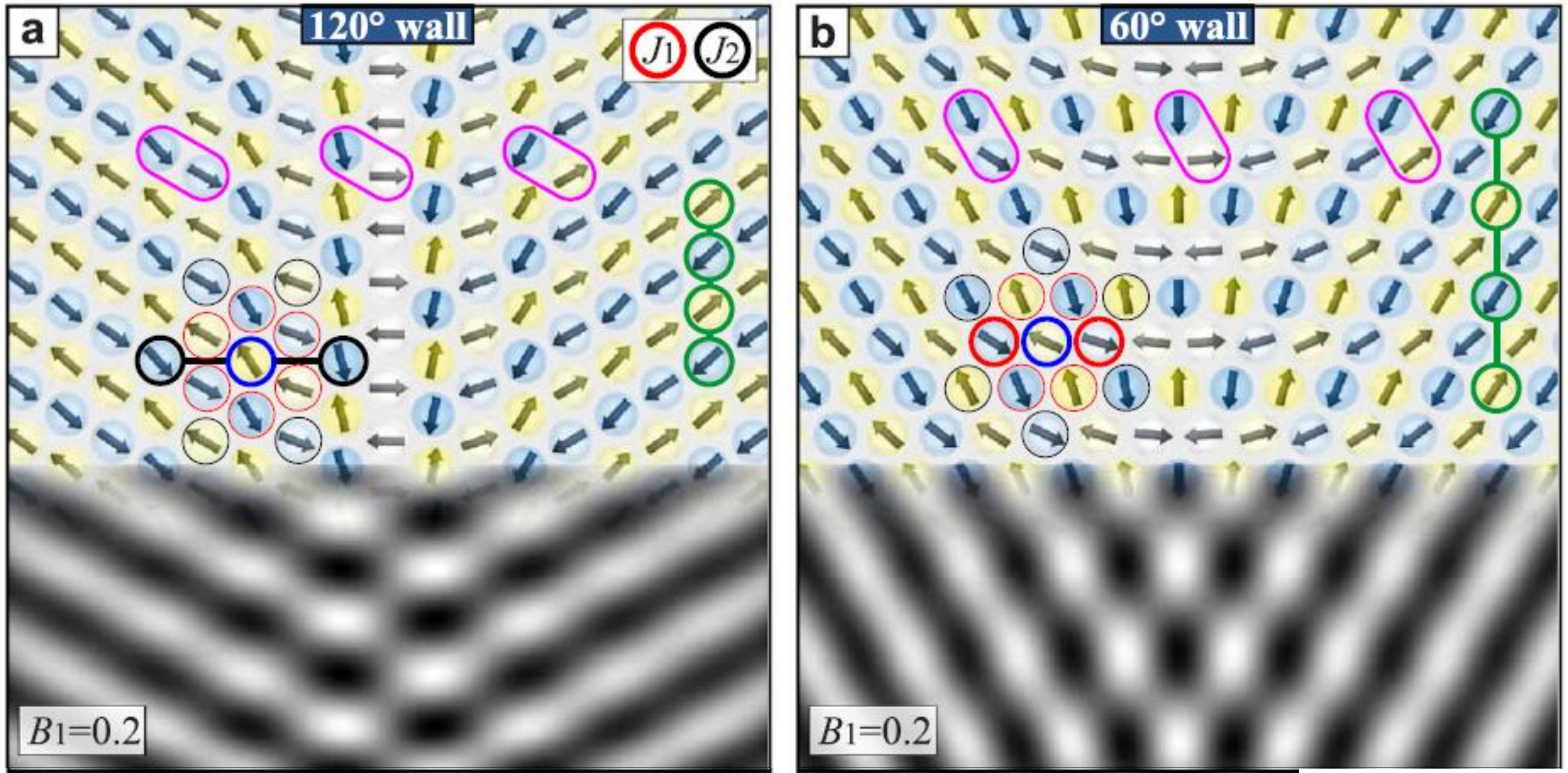

p(2×2) superstructure within the DW
→ not a coherent rotation of the afm-sublattices but a superposition of the two adjacent domains, i.e. a **2Q superposition** wall

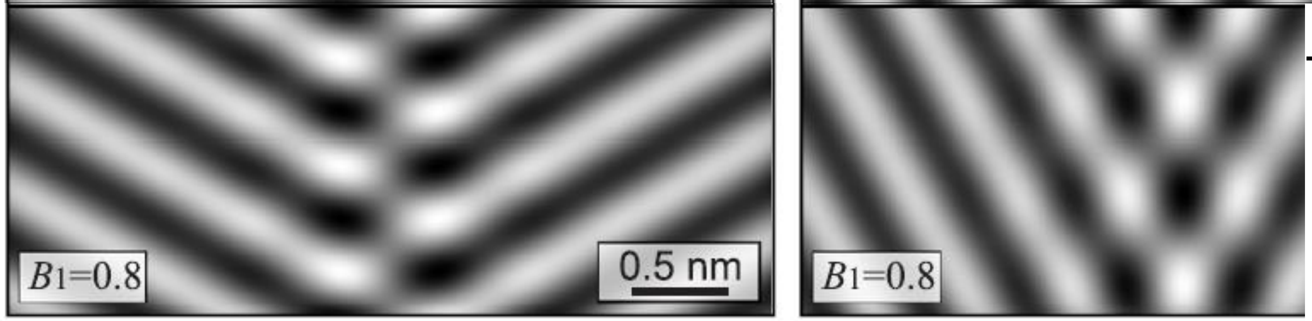





J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).

# superposition DWs




Spin dynamics simulations (MonteCrystal)

$$H = -J_1(\mathbf{S}_i \cdot \mathbf{S}_j) - J_2(\mathbf{S}_i \cdot \mathbf{S}_j) - K(S_z)^2$$
$$- J_{\mathrm{ASE}}(\mathbf{S}_i \cdot \mathbf{d}_{ij})(\mathbf{S}_j \cdot \mathbf{d}_{ij}) - B_1(\mathbf{S}_i \cdot \mathbf{S}_j)^2$$

a set of simplified DFT parameters (in meV/atom)
$J_1 = -25$, $J_2 = -5$,
$J_{\mathrm{ASE}} = +0.025$, $K = -1$, and $B_1$ as indicated.

analytical formulas for these superposition domain walls: HOIs ($B_1$) determine width and $E$

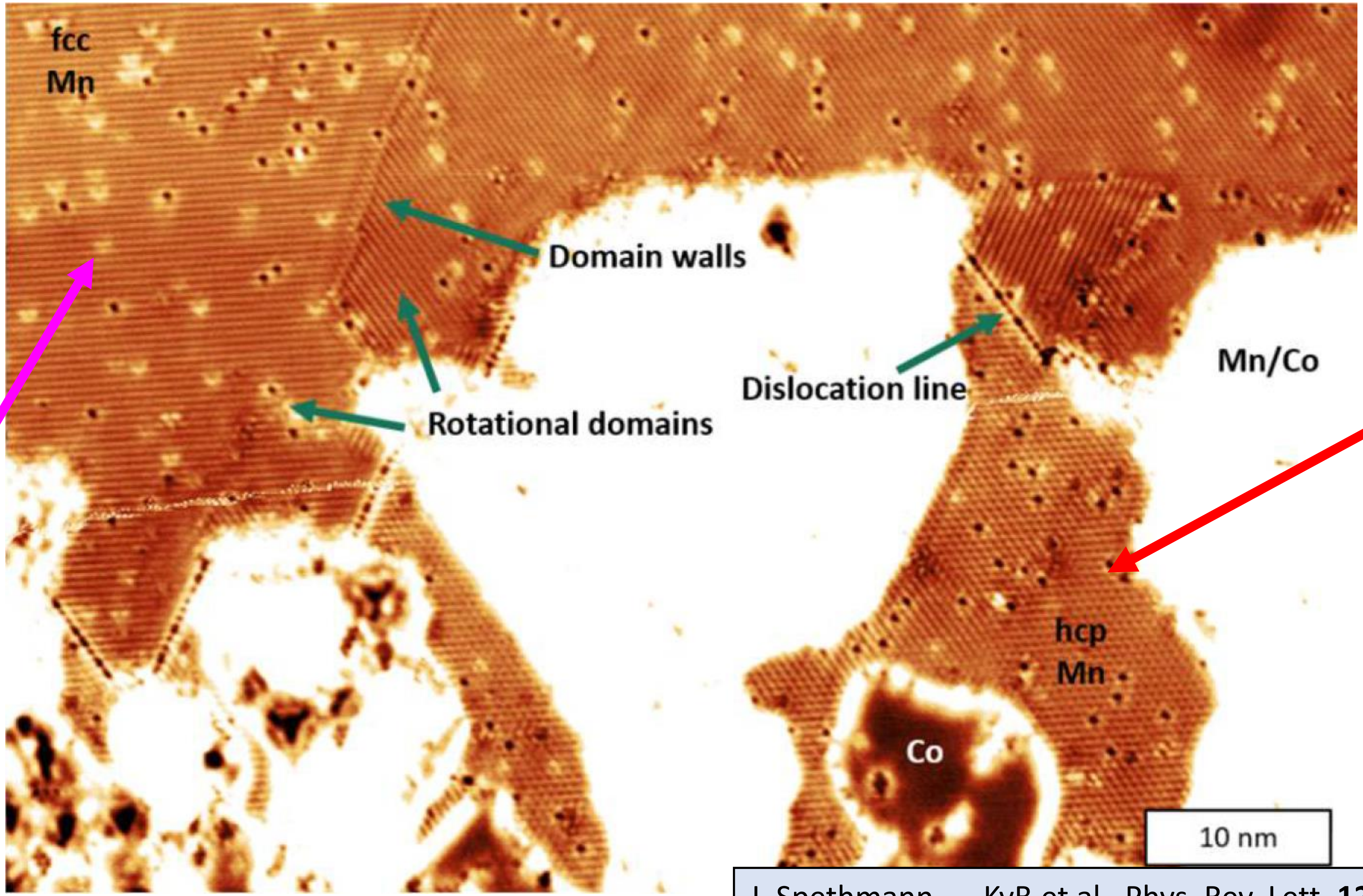
| DW type | Model | Width | Energy |
|---|---|---|---|
| 180° FM | A, K (continuum) | $2\sqrt{A/K}$ | $4\sqrt{AK}$ |
| 180° FM | $J_1 > 0$, $K > 0$ | $2a\sqrt{\frac{3}{2}J_1/K}$ | $\frac{4}{a}\sqrt{2J_1K}$ |
| 120° AFM | $J_1, J_2 < 0$, $B_1 > 0$ | $\frac{a}{2}\sqrt{3\lvert J_2\rvert/B_1}$ | $\frac{8}{a}\sqrt{\lvert J_2\rvert B_1}$ |
| 60° AFM | $J_1, J_2 < 0$, $B_1 > 0$ | $\frac{a}{2}\sqrt{\lvert J_1\rvert/B_1}$ | $\frac{8}{a}\sqrt{\frac{1}{3}\lvert J_1\rvert B_1}$ |

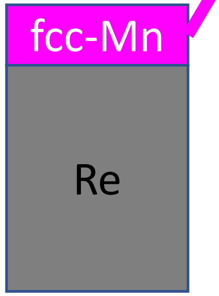


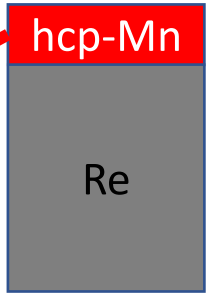
J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).

# Mn monolayer on Re(0001)

fcc-Mn in SP-STM: stripes along the close-packed atomic rows, every other row
→ RW-AFM

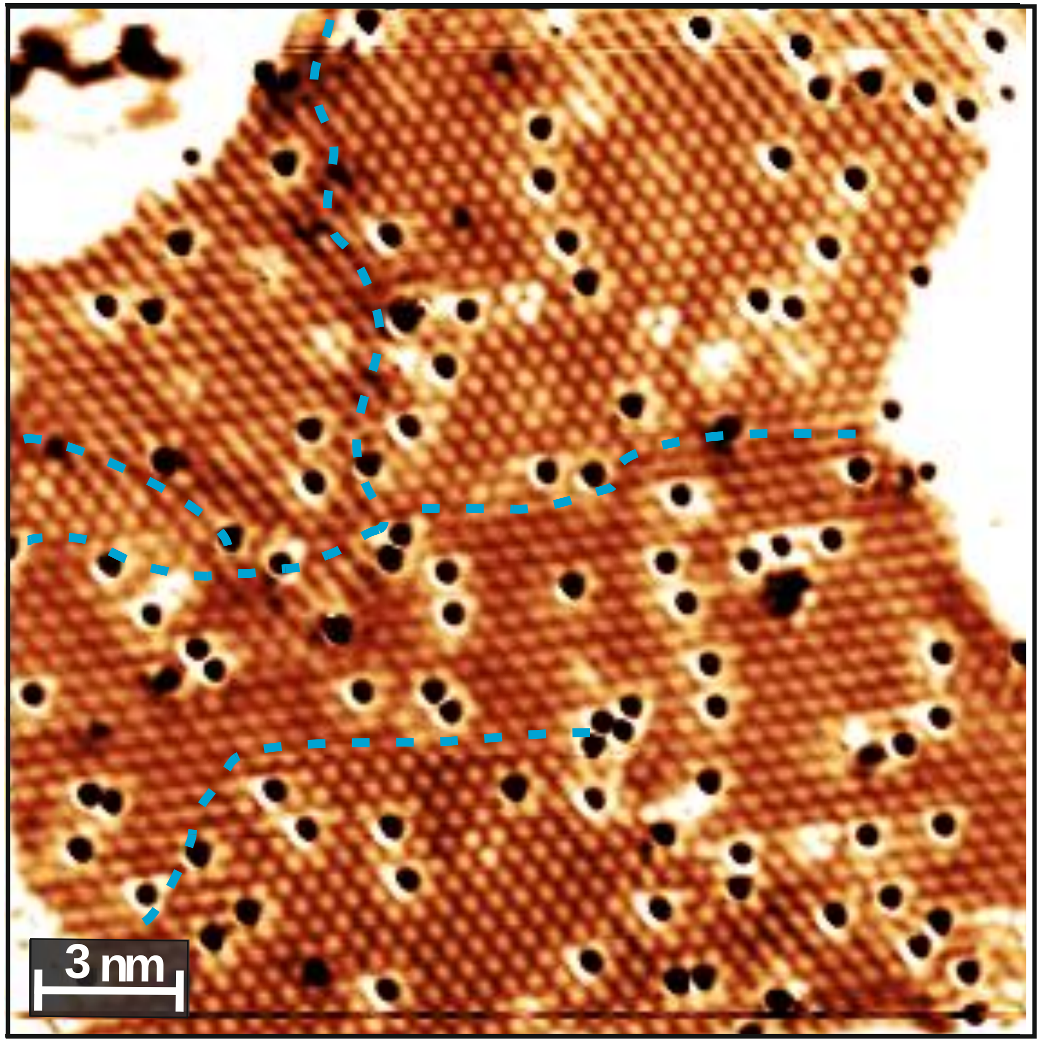


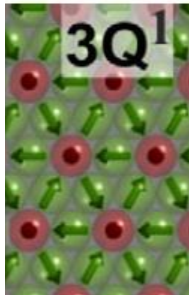




J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).
J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).

# hcp-Mn/Re(0001): the 3Q-state



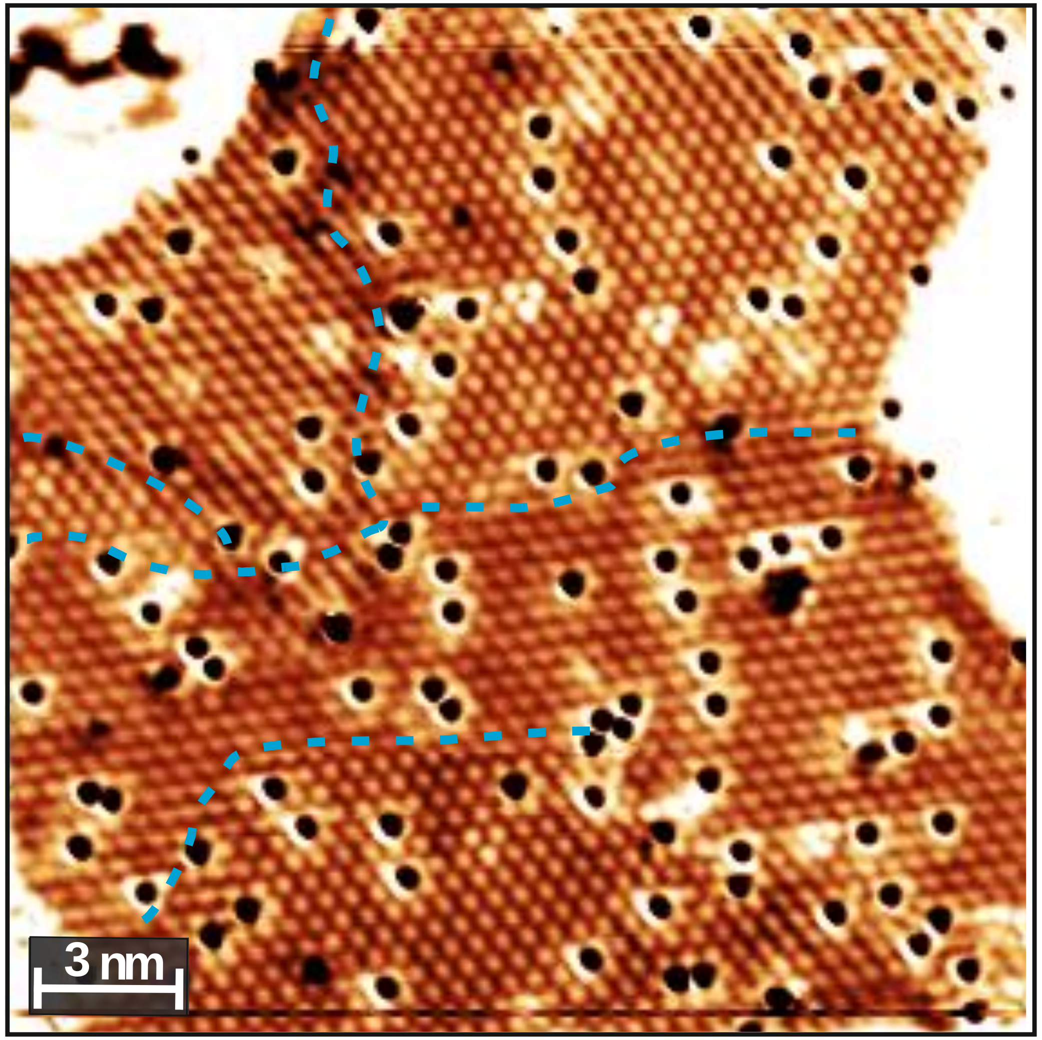

hexagonal p(2×2) pattern
→ 3Q state
with tetrahedron angles,
(equivalent to RW-AFM
superposítion state)

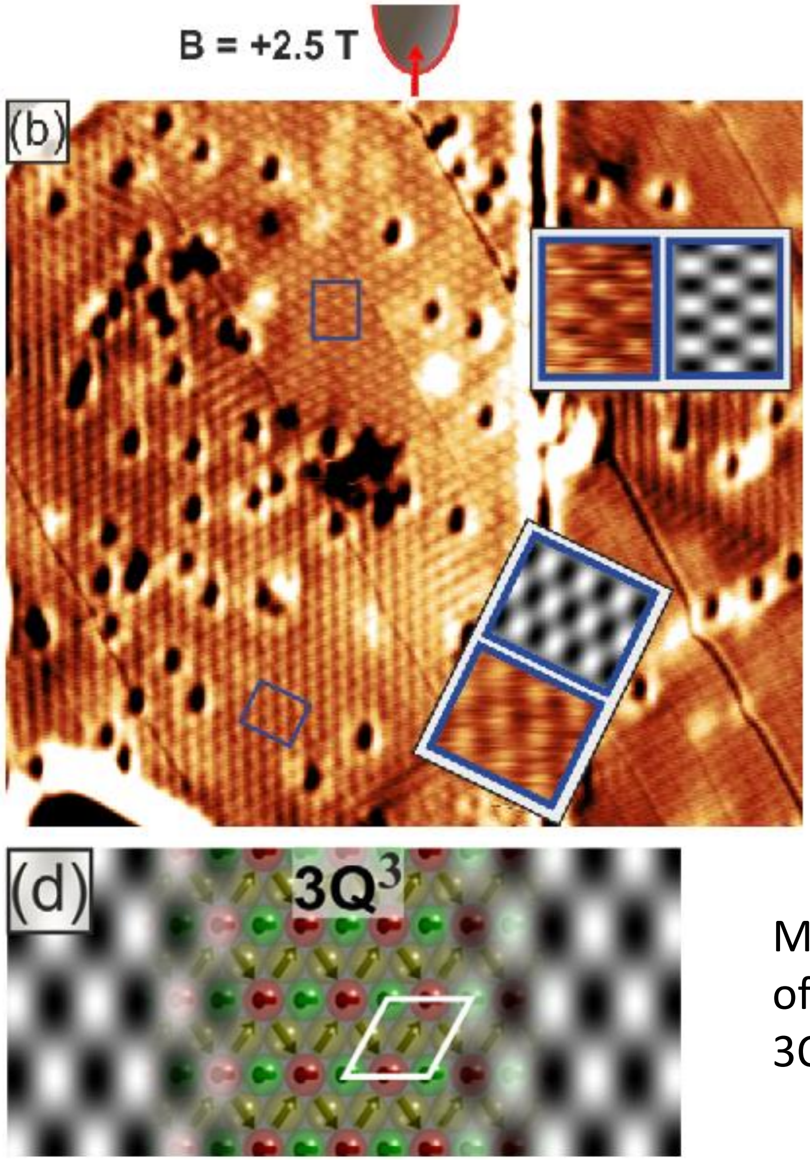
looks slightly different
in different sample areas,
occurs in 3 rotational domains



J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).

# hcp-Mn/Re(0001): the 3Q-state




Measurements with out-of-plane tips show that the $3Q^3$ is realized



J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).

# topological properties of non-coplanar magnetic states

scalar spin chirality acts like effective B field

→ topological orbital moment

→ topological Hall effect

(without the need of spin orbit coupling)

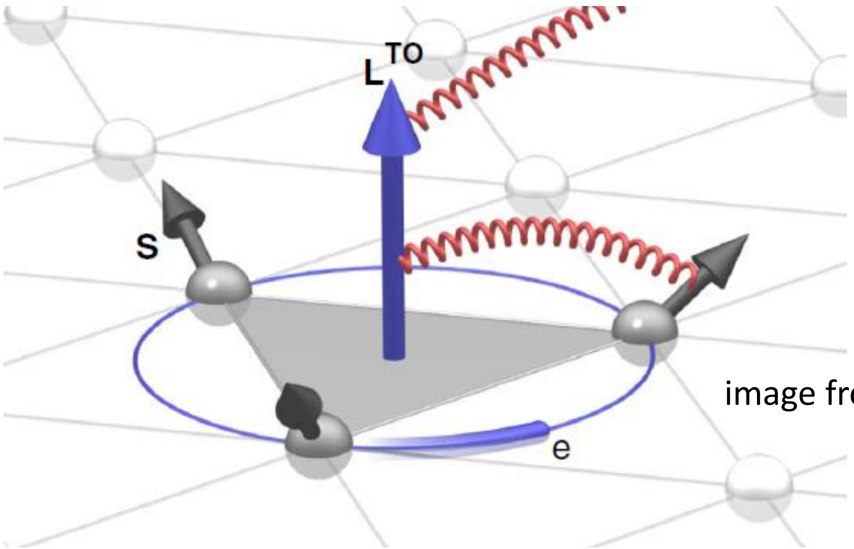



image from →

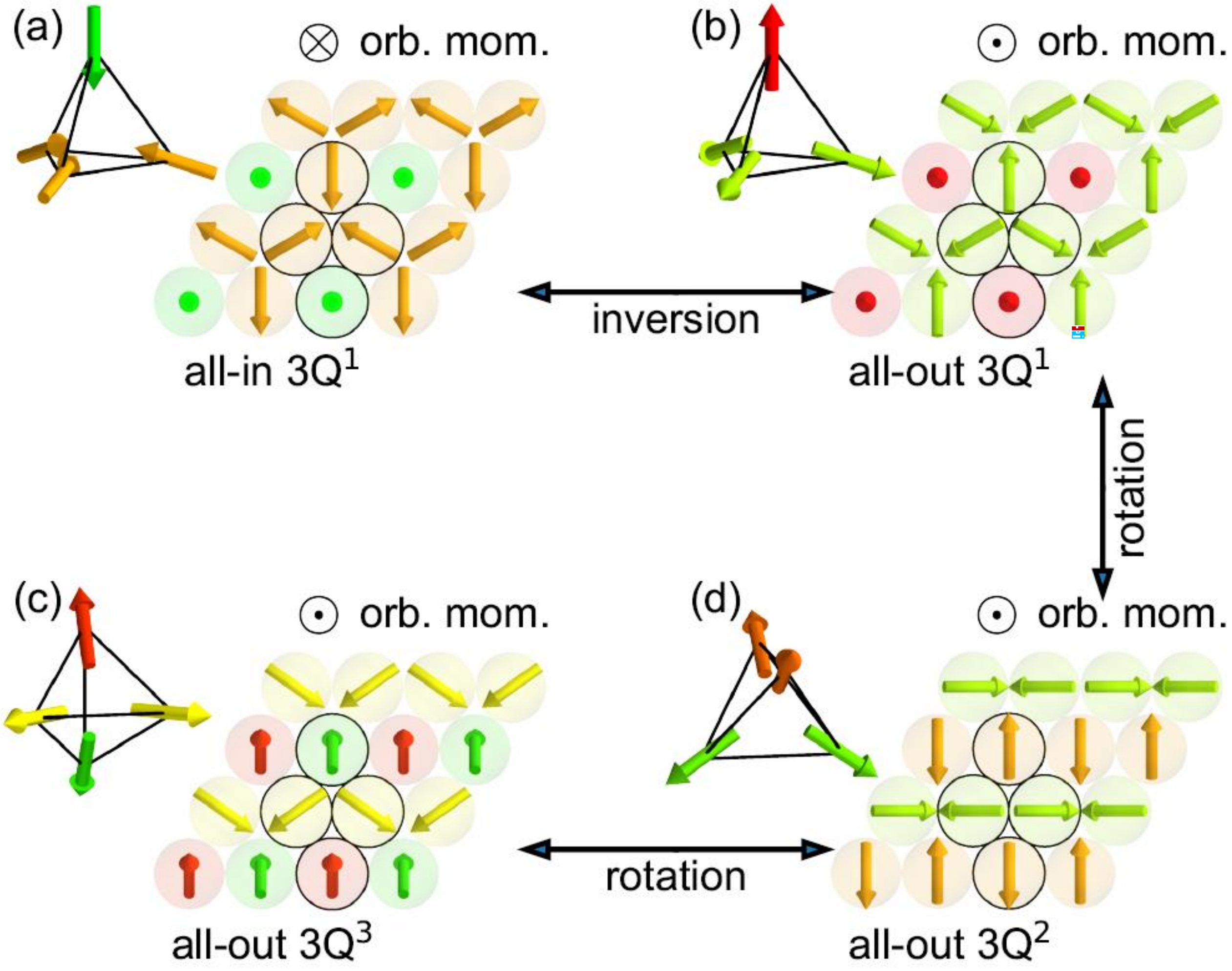
## Topological Orbital Moments (TOM)


I. Martin, C. D. Batista, PRL **101**, 156402 (2008).
M. Hoffmann et al., PRB **92**, 020401(R) (2015).
J.-P. Hanke et al, PRB **94**, 121114(R) (2016).
S. Grytsiuk et al, Nat. Commun. **11**, 511 (2020).
F. Nickel, ... KvB, PRB **108**, L180411 (2023).
F. Nickel, … KvB, et al, arXiv:2405.18088.

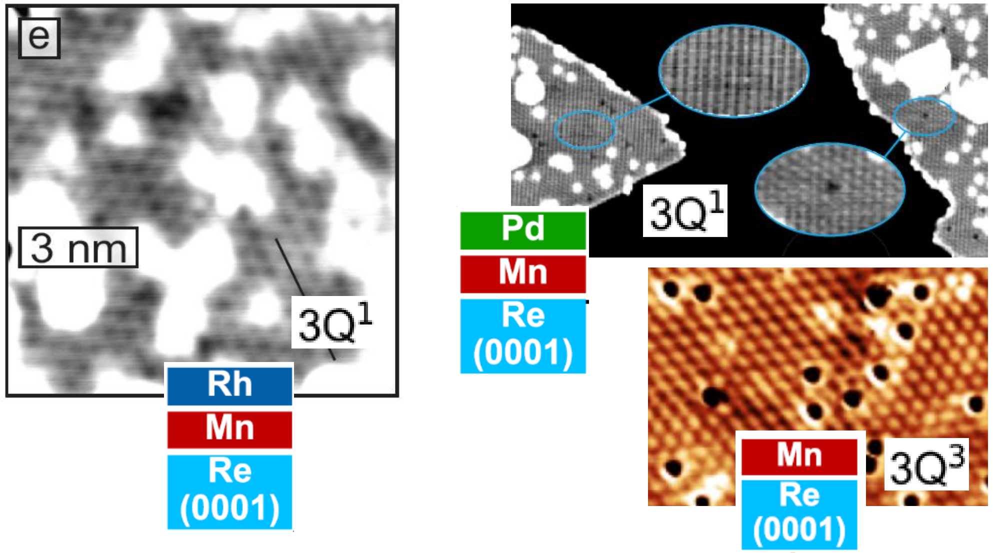

triple-q response to applied fields

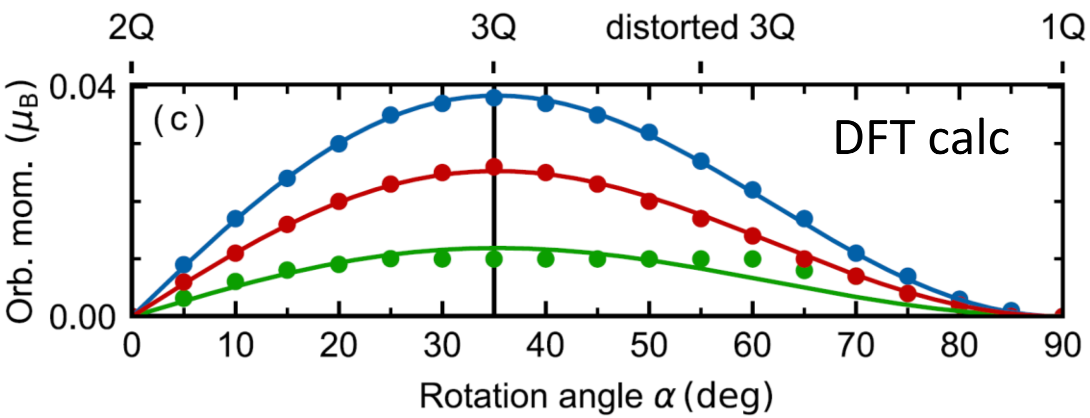

H. Takagi et al., Nature Phys. 19, 961 (2023).
P. Park et al., Nat. Commun. 14, 8346 (2023).

# anatomy of the 3Q state




3Q state can have different relative spin configurations (both net spin zero), which define the direction of the TOMs



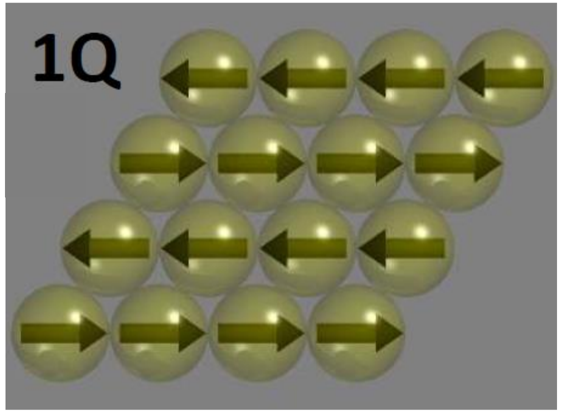





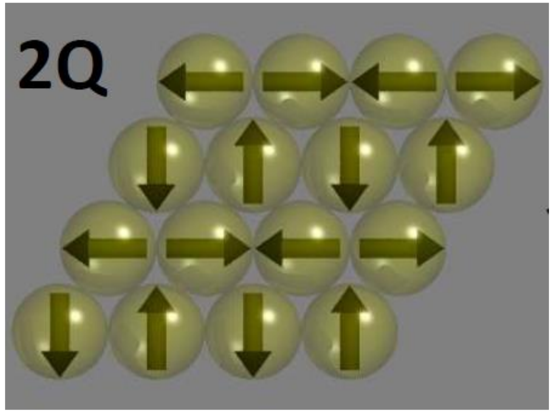

F. Nickel, ... KvB, Phys. Rev. B **108**, L180411 (2023).

# superposition states = multi-Q states

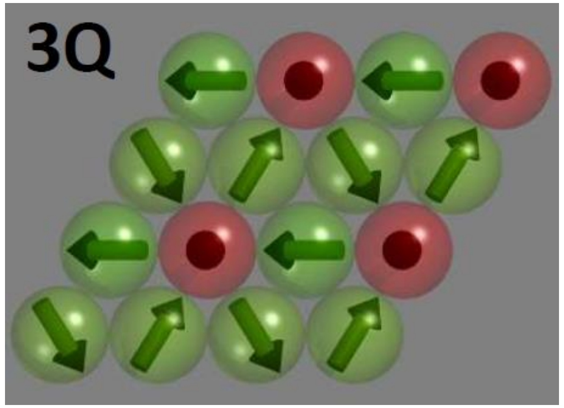
collinear




coplanar




3d spin texture

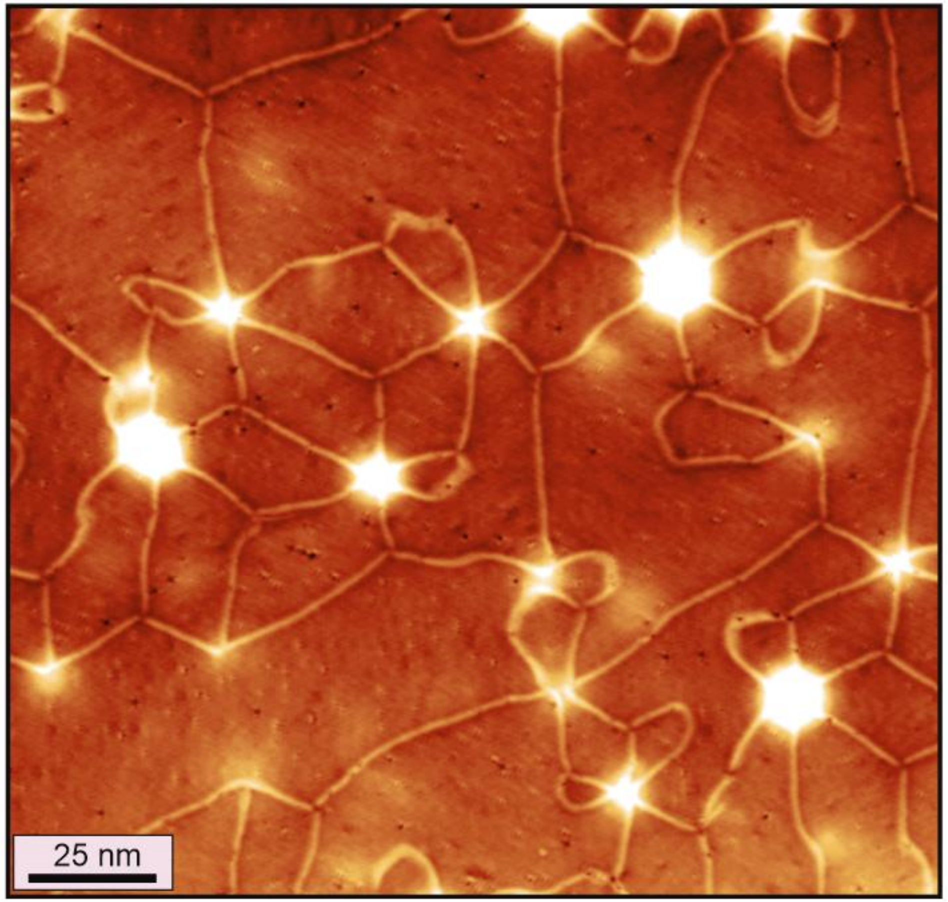


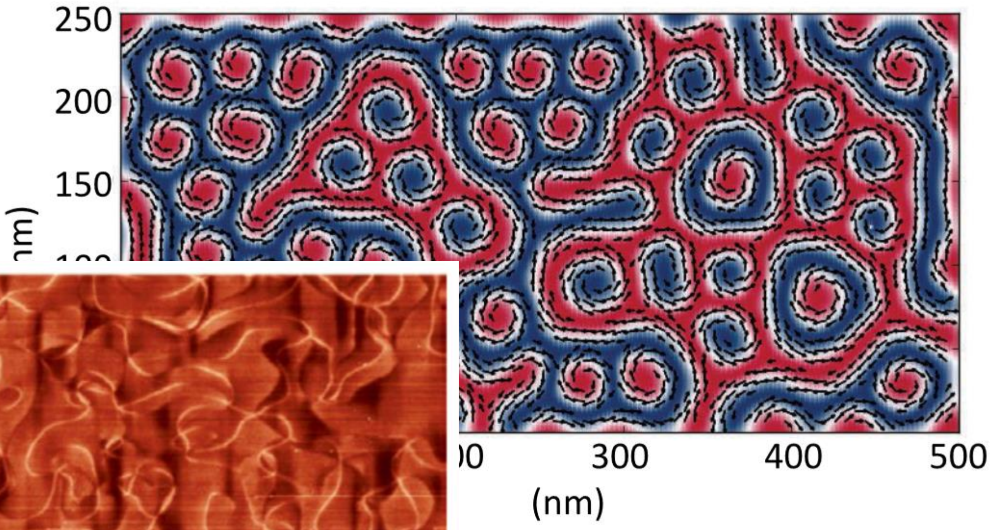

when all HOIs vanish,
these states are degenerate
and are expected to coexist

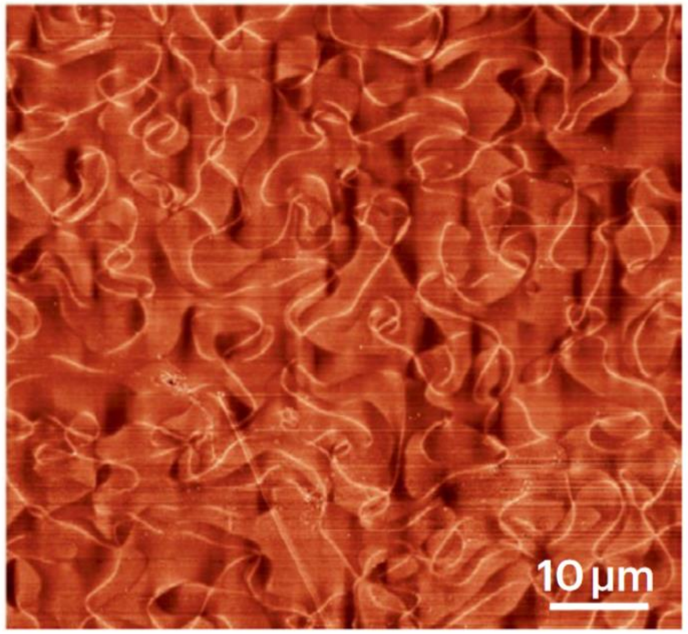
$$H_{\mathrm{HO}} = -\sum_{<ij>} B(\boldsymbol{m}_i\boldsymbol{m}_j)^2$$
$$-2\sum_{<ijk>} Y_3[(\boldsymbol{m}_i\boldsymbol{m}_j)(\boldsymbol{m}_j\boldsymbol{m}_k) + (\boldsymbol{m}_j\boldsymbol{m}_k)(\boldsymbol{m}_k\boldsymbol{m}_i) + (\boldsymbol{m}_k\boldsymbol{m}_i)(\boldsymbol{m}_i\boldsymbol{m}_j)]$$
$$-\sum_{<ijkl>} K_4[(\boldsymbol{m}_i\boldsymbol{m}_j)(\boldsymbol{m}_k\boldsymbol{m}_l) + (\boldsymbol{m}_i\boldsymbol{m}_l)(\boldsymbol{m}_j\boldsymbol{m}_k) - (\boldsymbol{m}_i\boldsymbol{m}_k)(\boldsymbol{m}_j\boldsymbol{m}_l)]$$

# Domain Wall Network

Mn double layer on Ir(111)




D. Prychynenko et al, Phys. Rev. Appl. **9**, 014034 (2018).
K. Everschor-Sitte et al, Nat. Rev. Phys. **6**, 455 (2024).

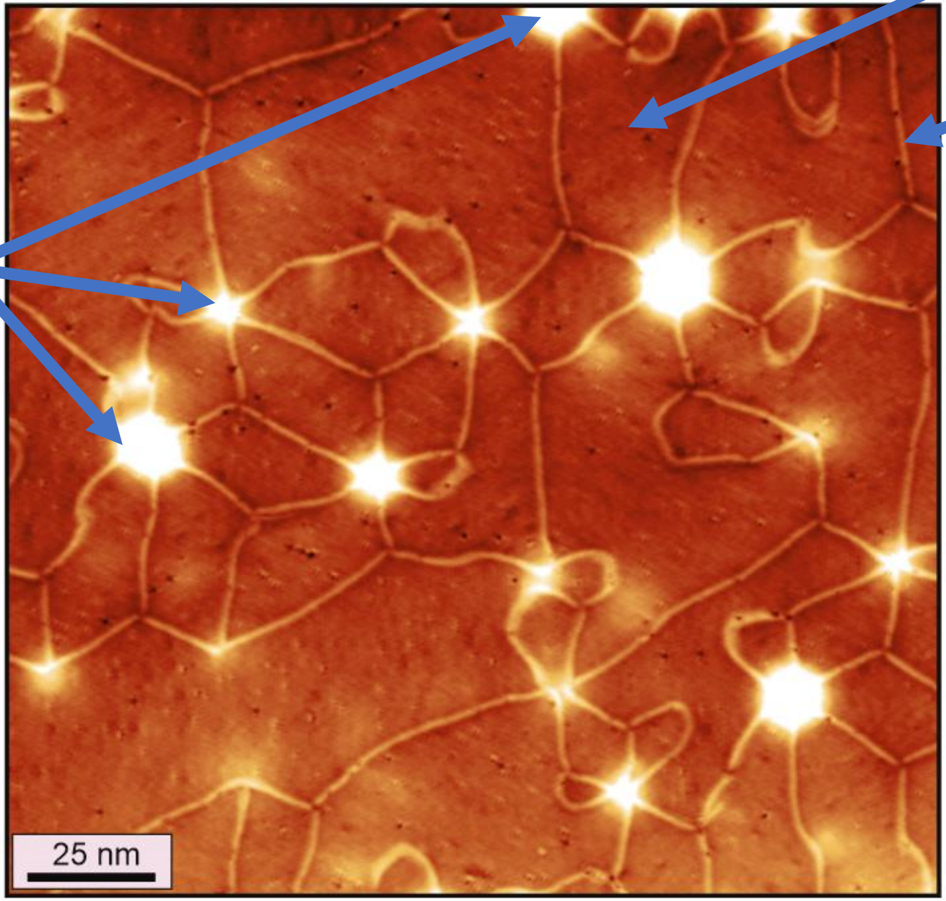
# Domain Wall Network

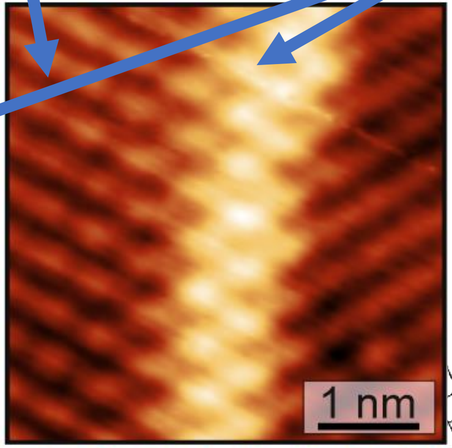
Mn double layer on Ir(111)

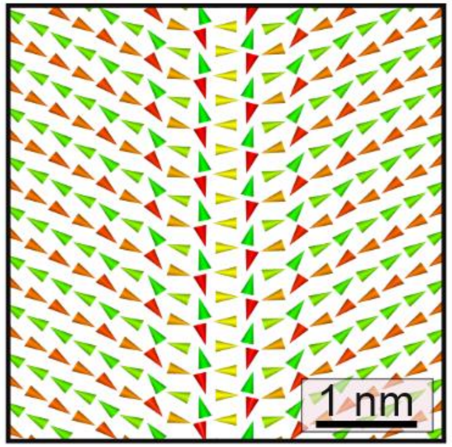
1Q ground state

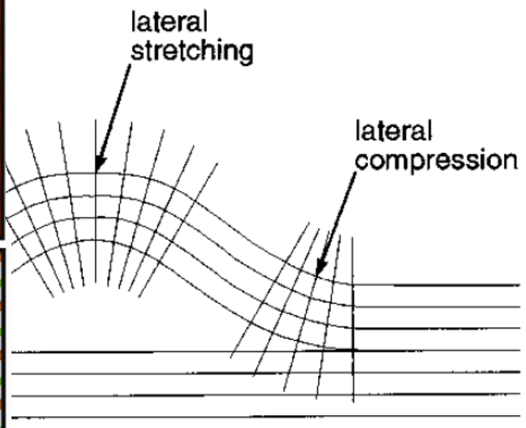
2Q domain wall
strictly 120°-type

Ar bubbles



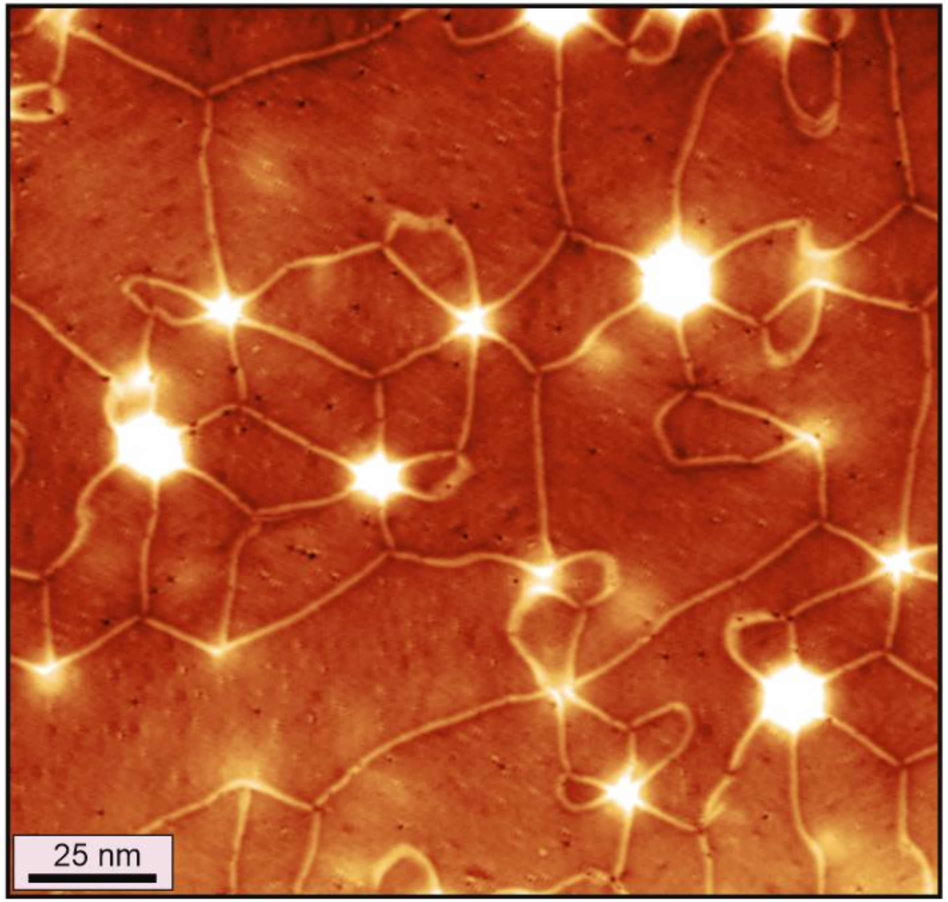




Science **280**, 717 (1998).

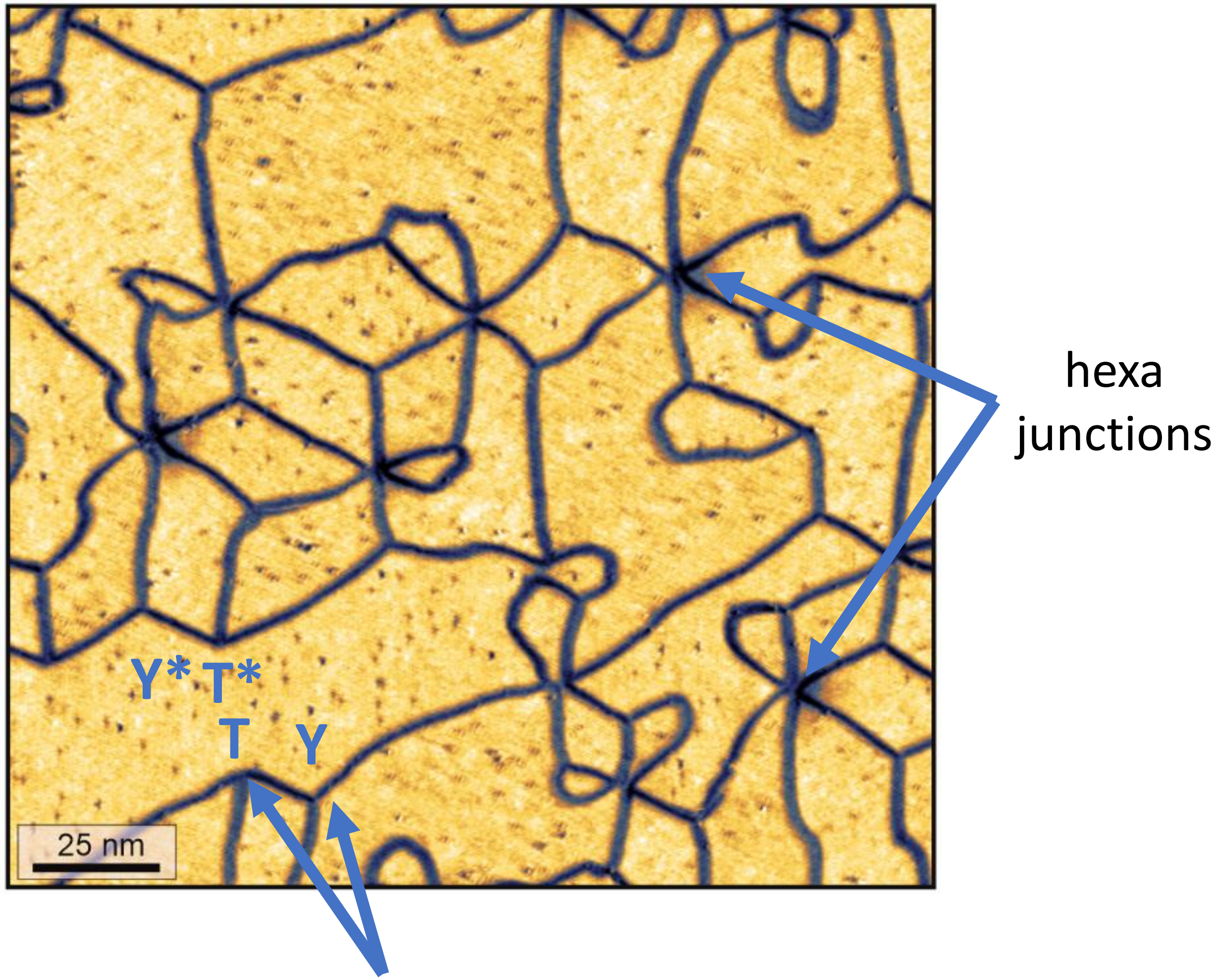
# Domain Wall Network

Mn double layer on Ir(111)

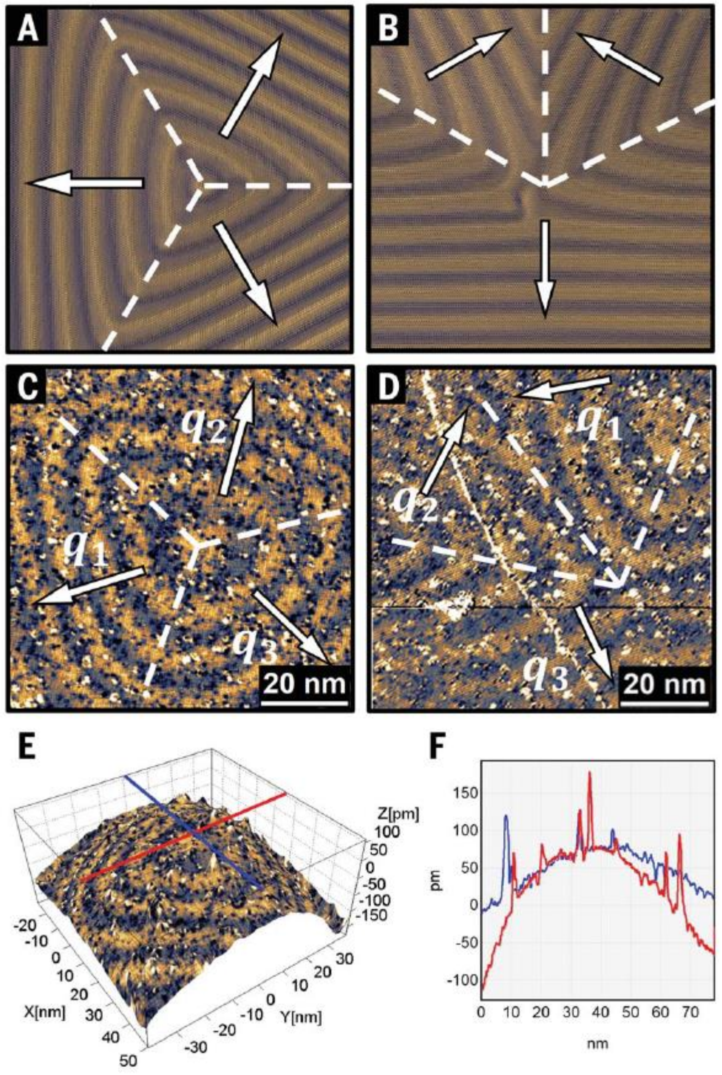
Constant-current STM

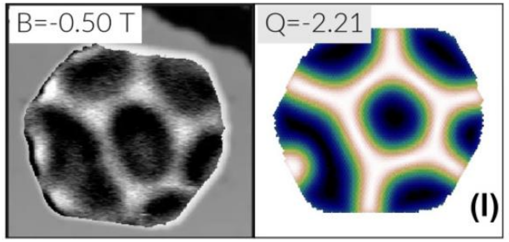


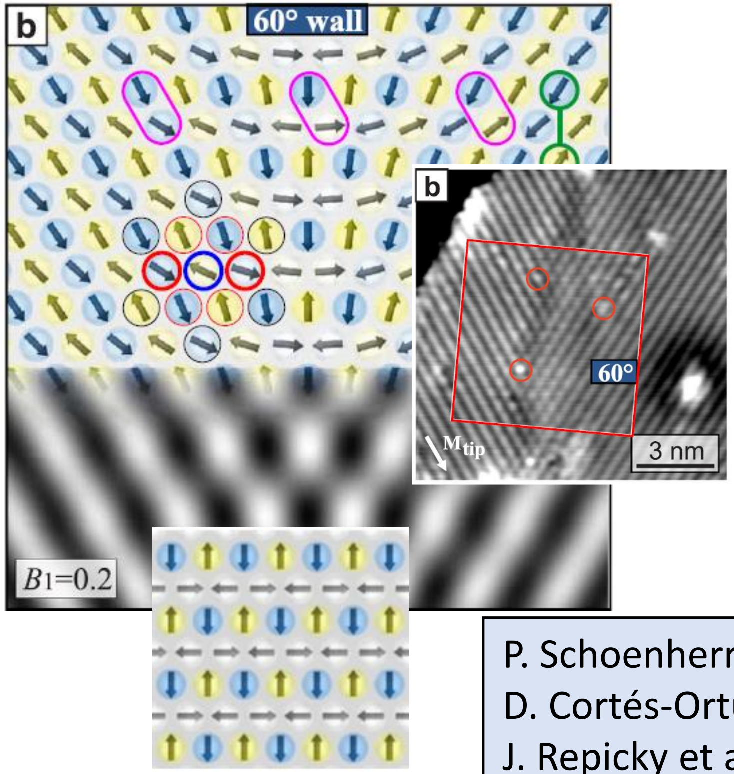

Differential conductance

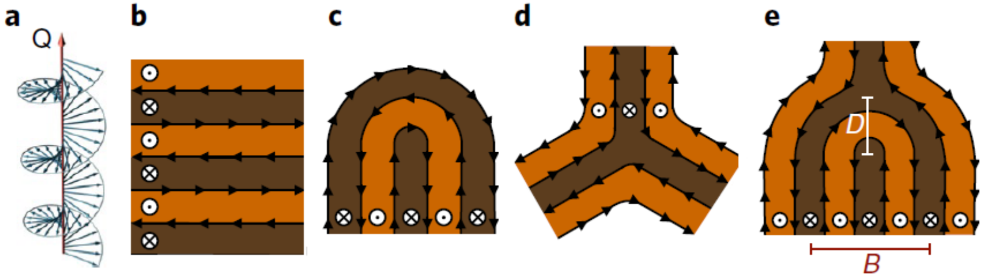


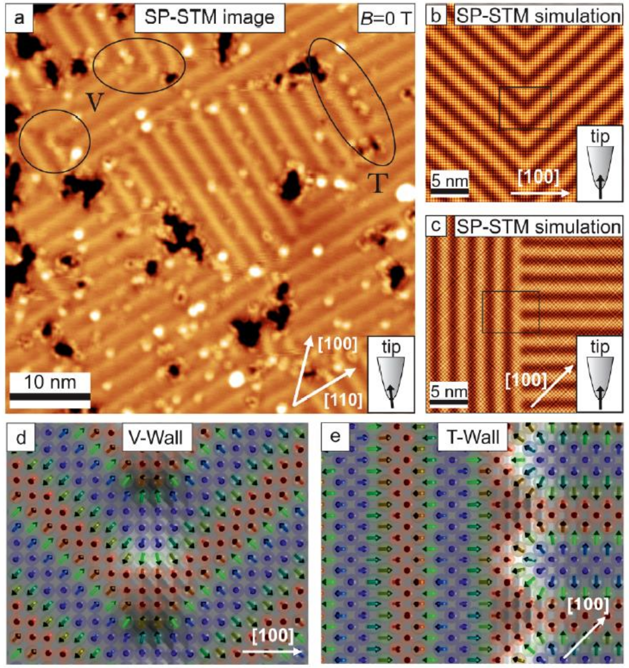
# Domain walls and junctions



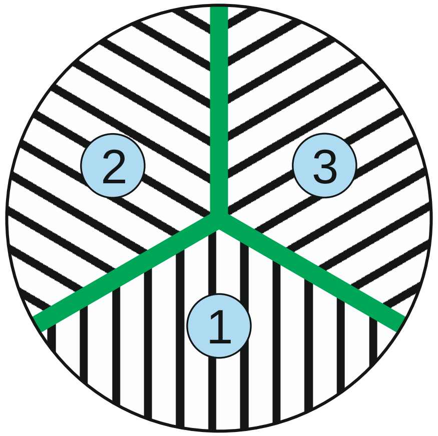



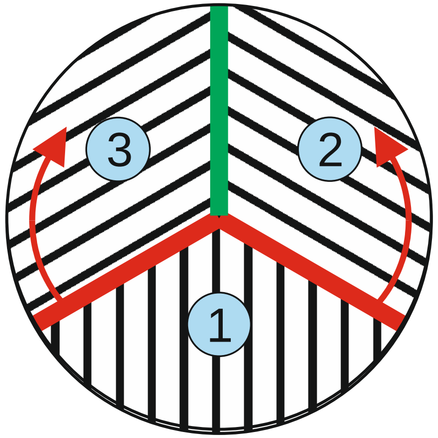



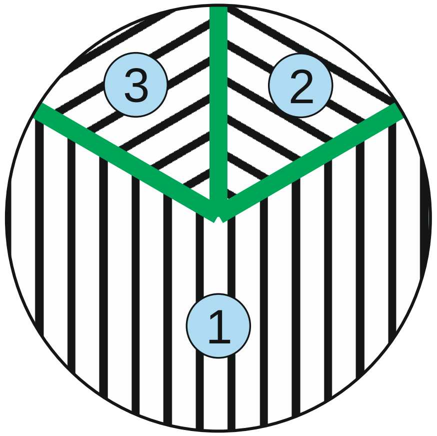

P. Schoenherr et al, Nat. Phys. **14**, 465 (2018).
D. Cortés-Ortuño, …, KvB, Phys. Rev. B **99**, 214408 (2019).
J. Repicky et al, Science **374**, 1484 (2021).
J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).
R. Brüning, …, KvB et al, Phys. Rev. B **105**, L241401 (2022).
A. Finco et al, Phys. Rev. Lett. **128**, 187201 (2022).

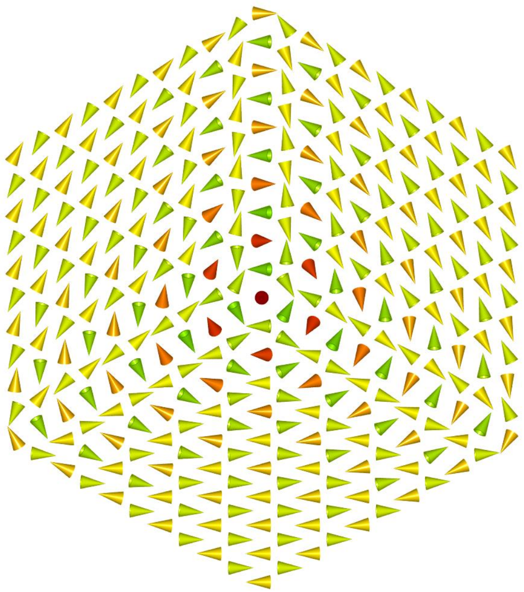
# Triple junctions

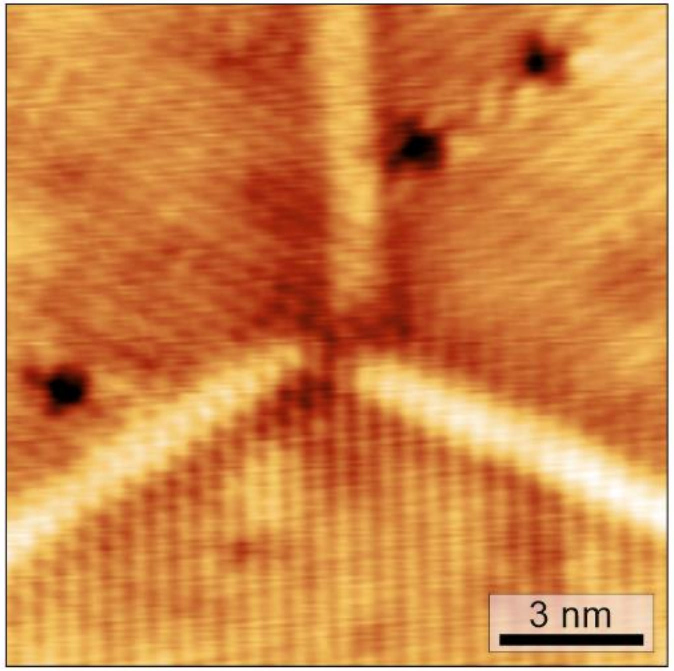
we have non-coplanar order at the triple junctions

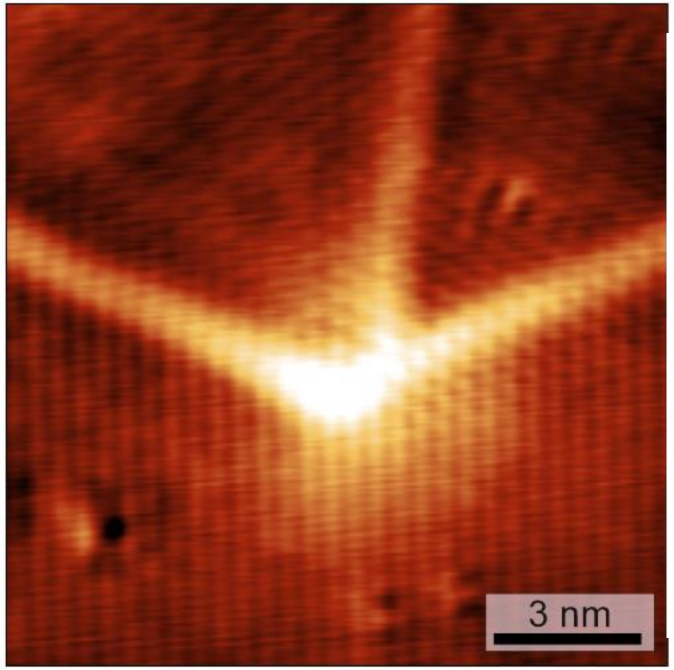


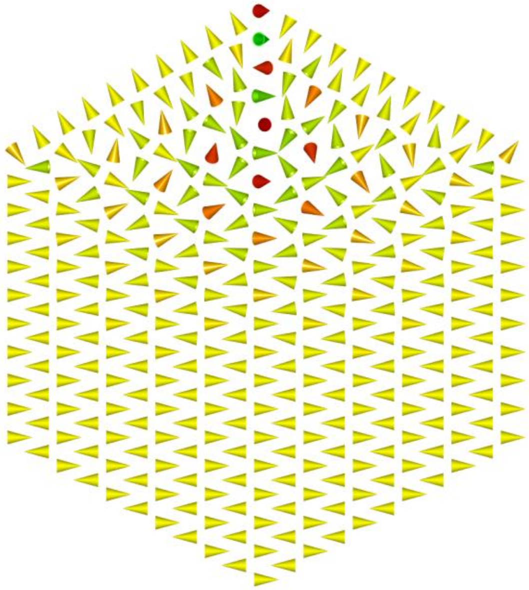



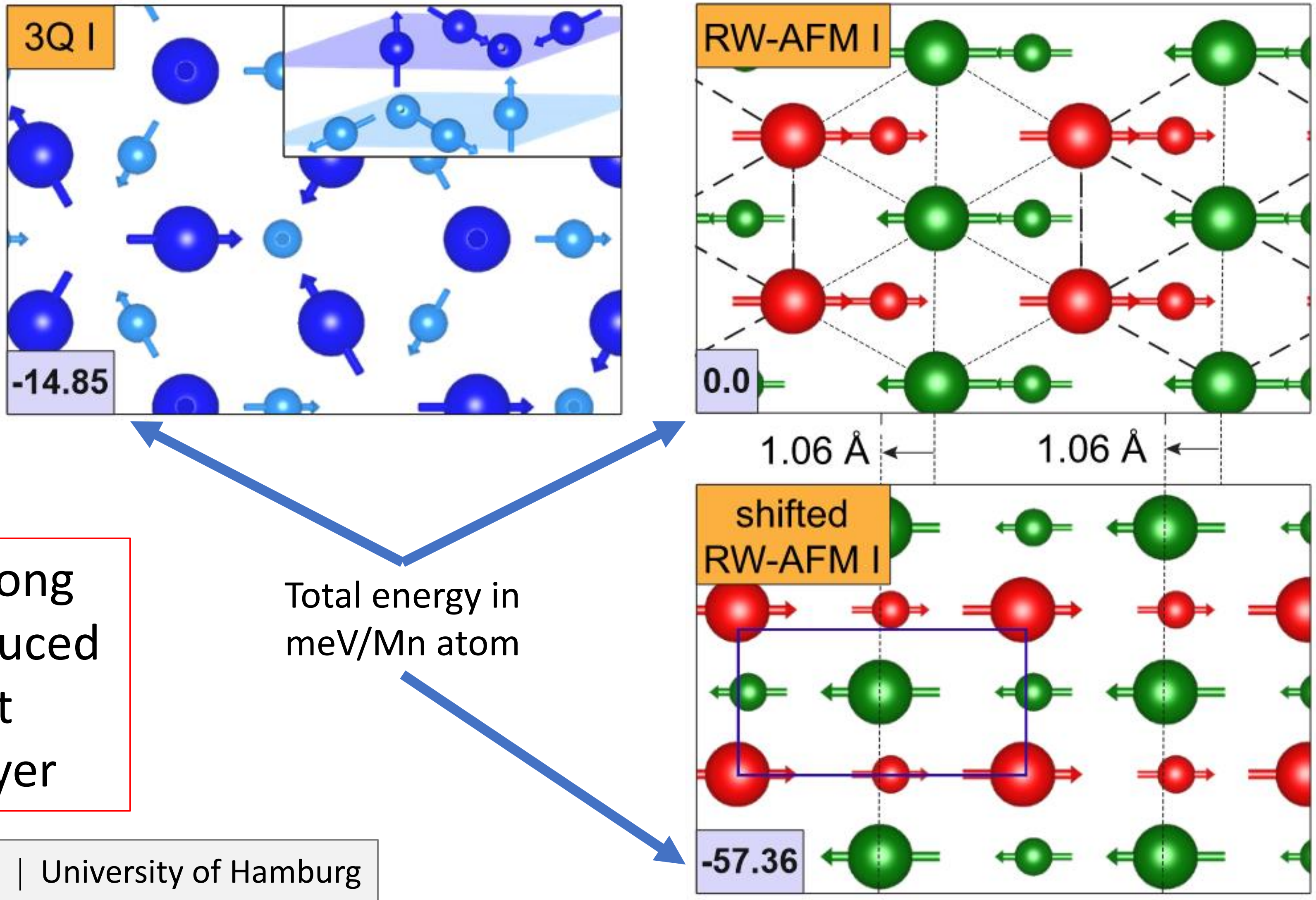


# DFT calculations of the magnetic states

Mn double layer on Ir(111)

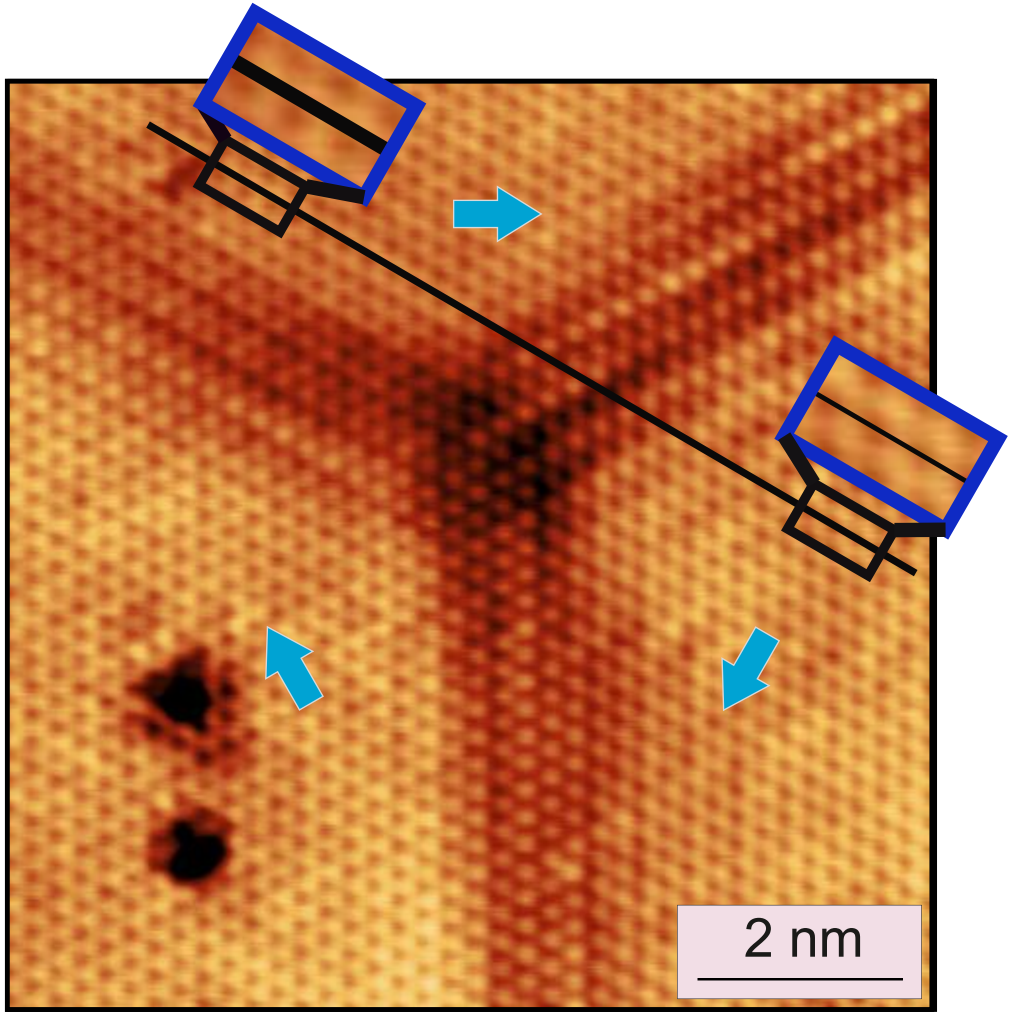


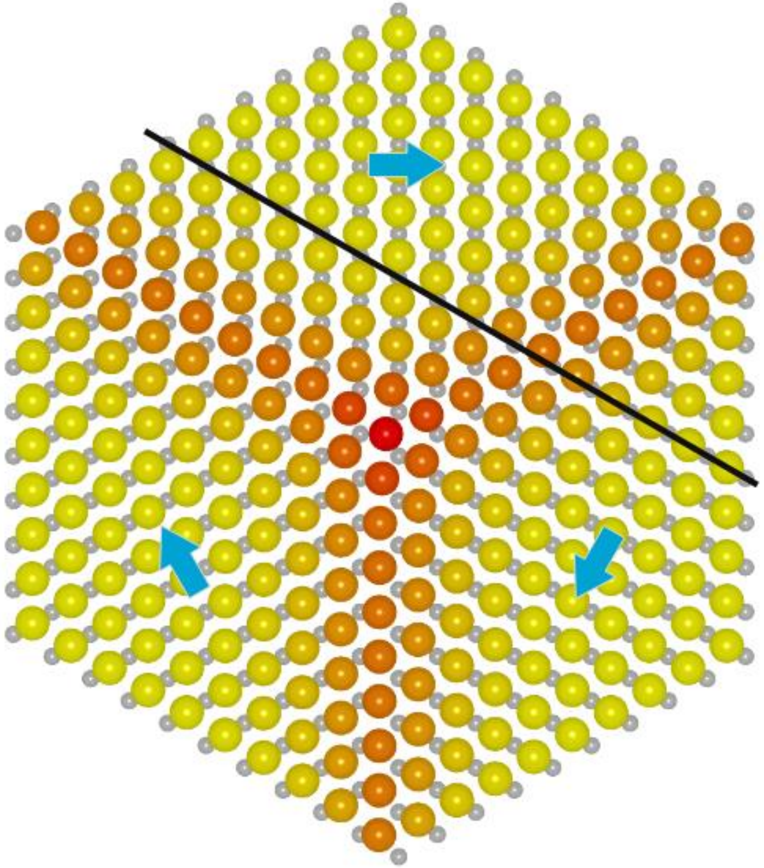

we have a strong magnetism-induced lateral shift of the top layer

# Lateral shift of the top layer

Mn double layer on Ir(111)

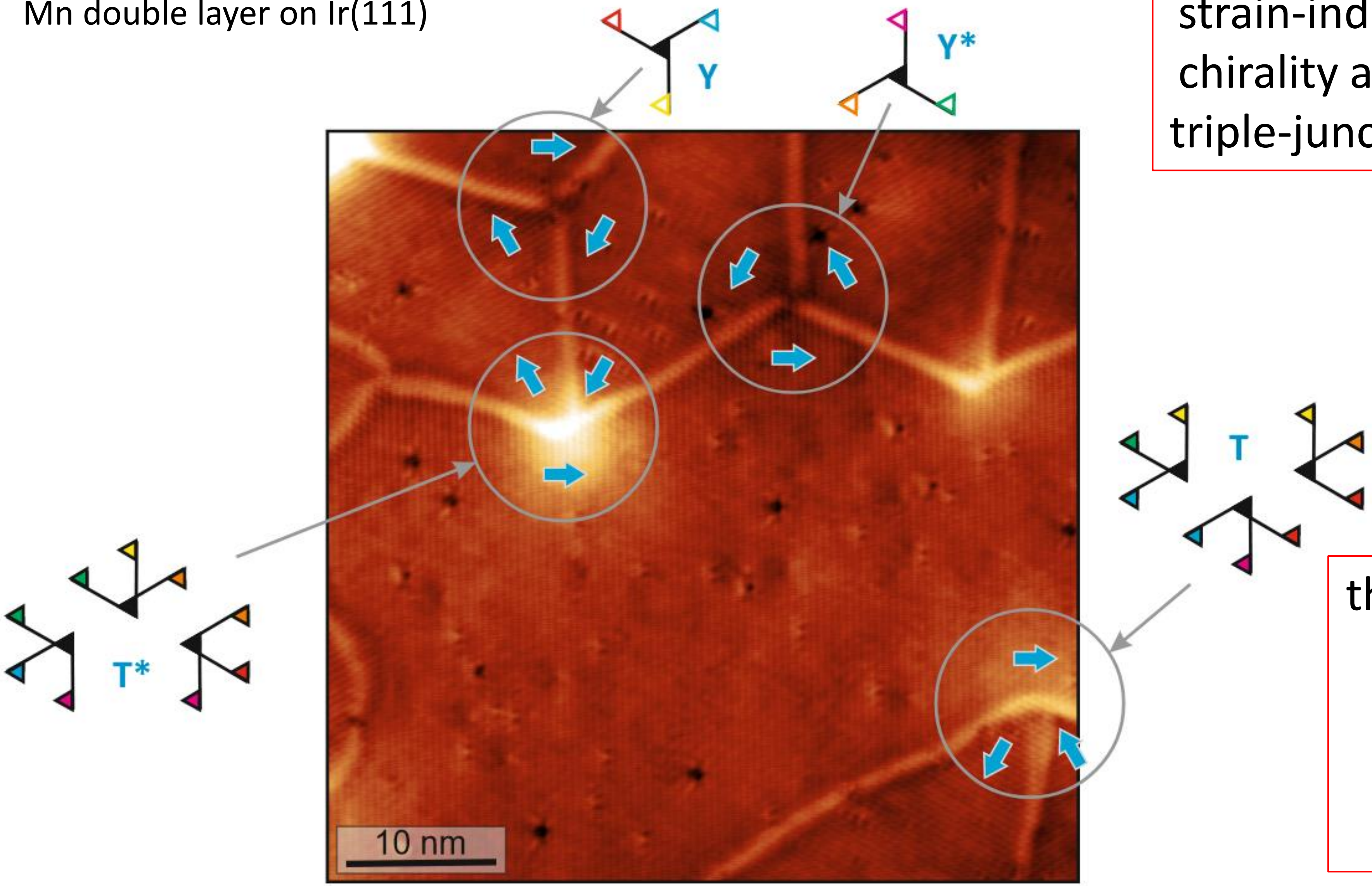
there are local strain fields at domain walls and junctions



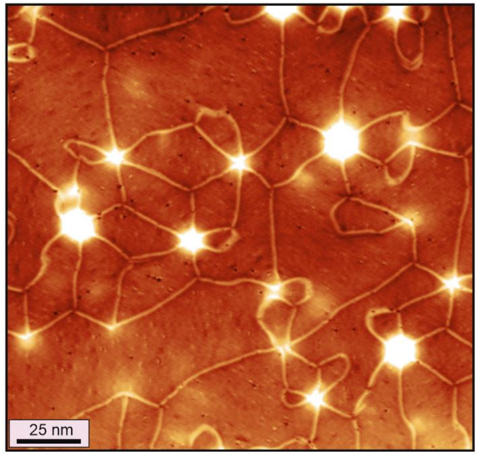



# Domain Wall Network

Mn double layer on Ir(111)



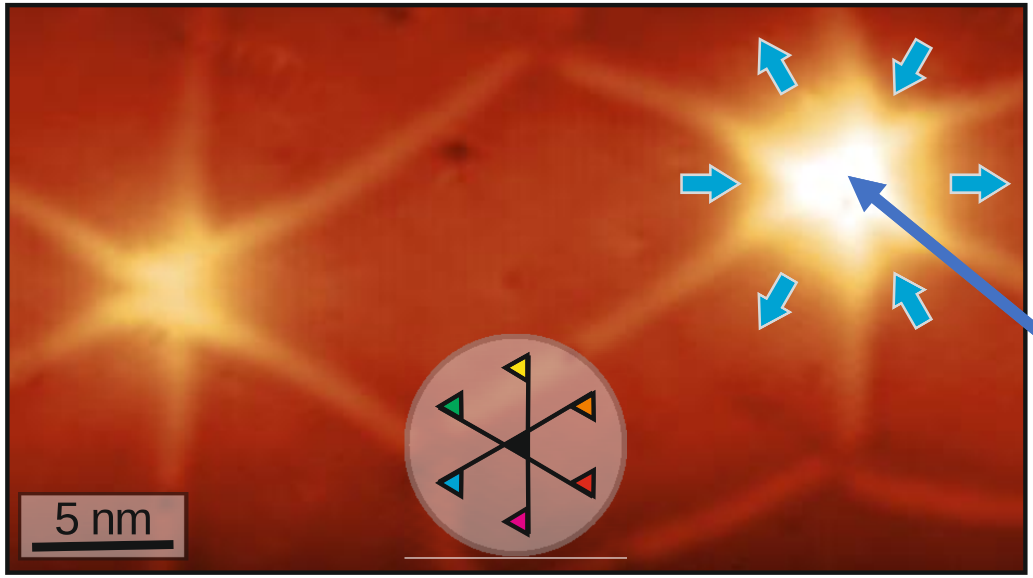

there is a strain-induced chirality at the triple-junctions



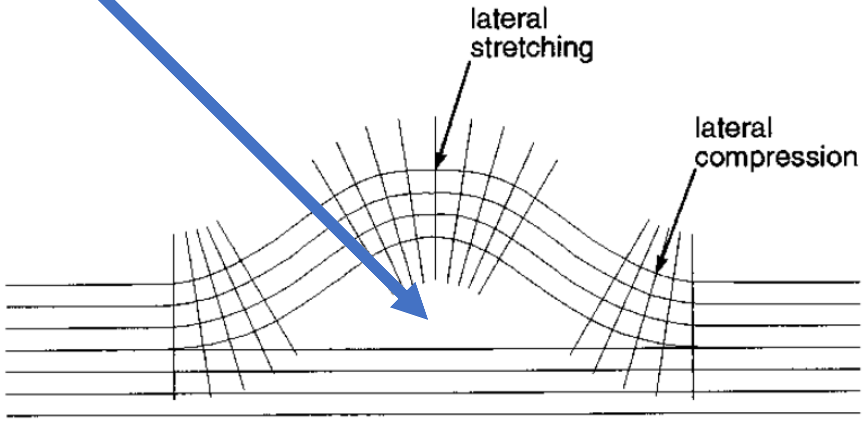

there are very simple rules which connections can form the network (connect same color, always pair $Y^{(*)}$ and $T^{(*)}$)

# Hexa-junctions

Mn double layer on Ir(111)



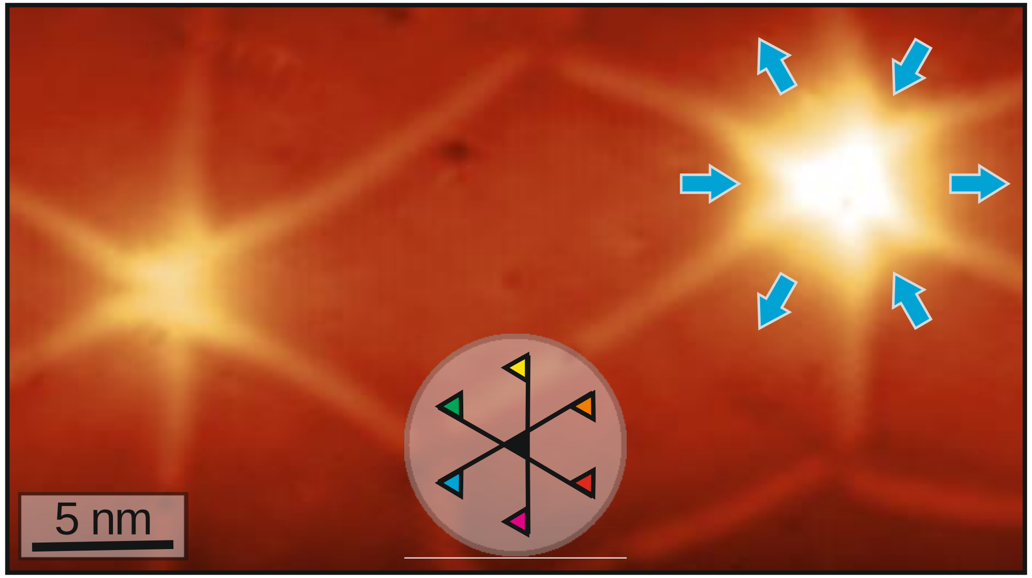

Ar bubble



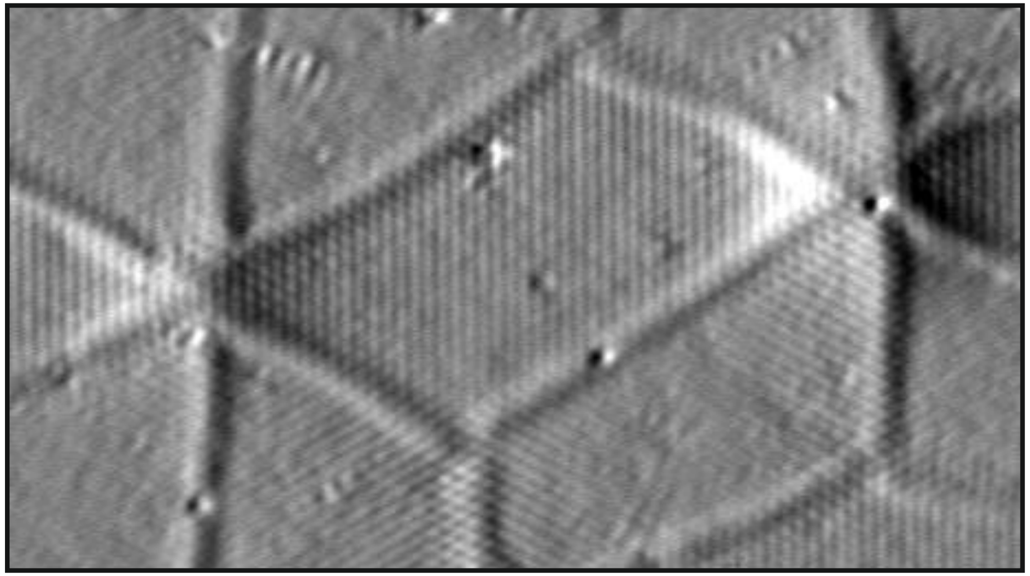

M. Gsell et al, Science **280**, 717 (1998).

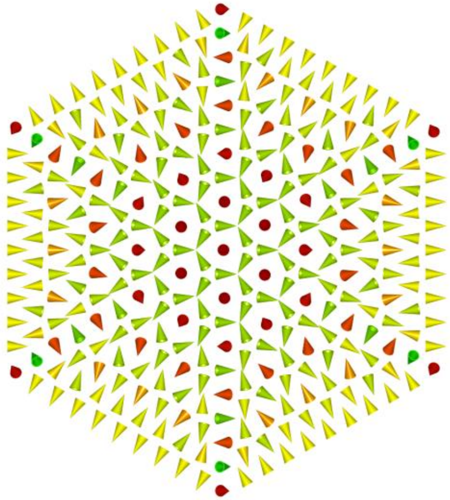
# Hexa-junctions

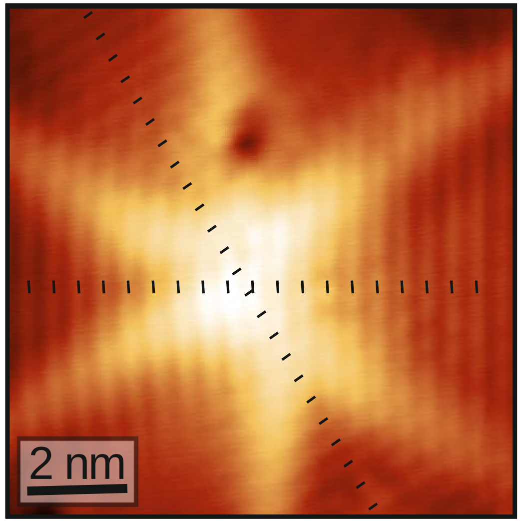
Mn double layer on Ir(111)




current channel

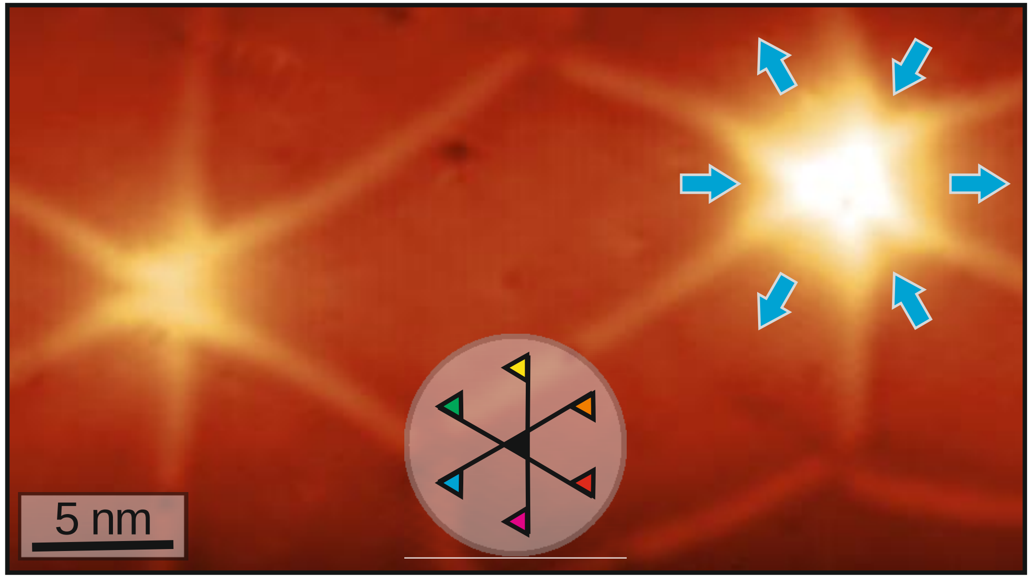


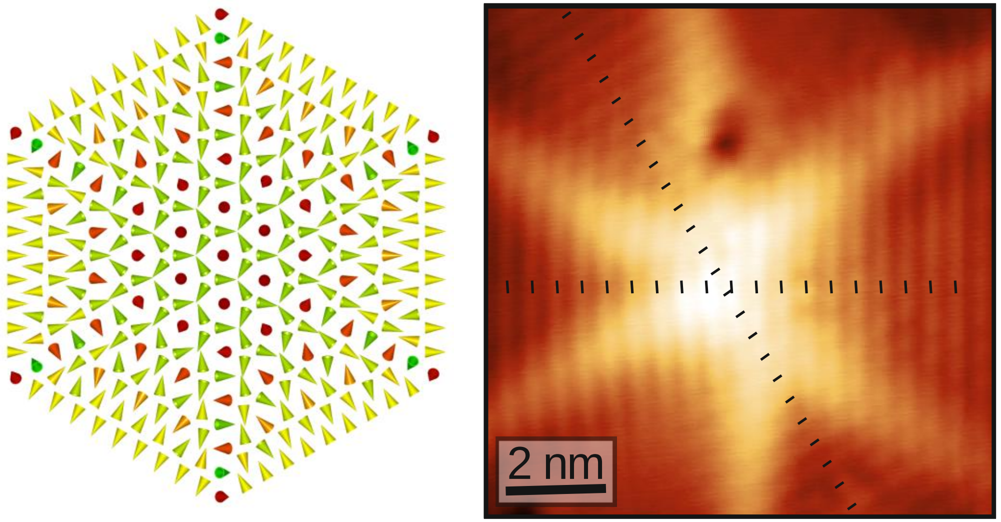


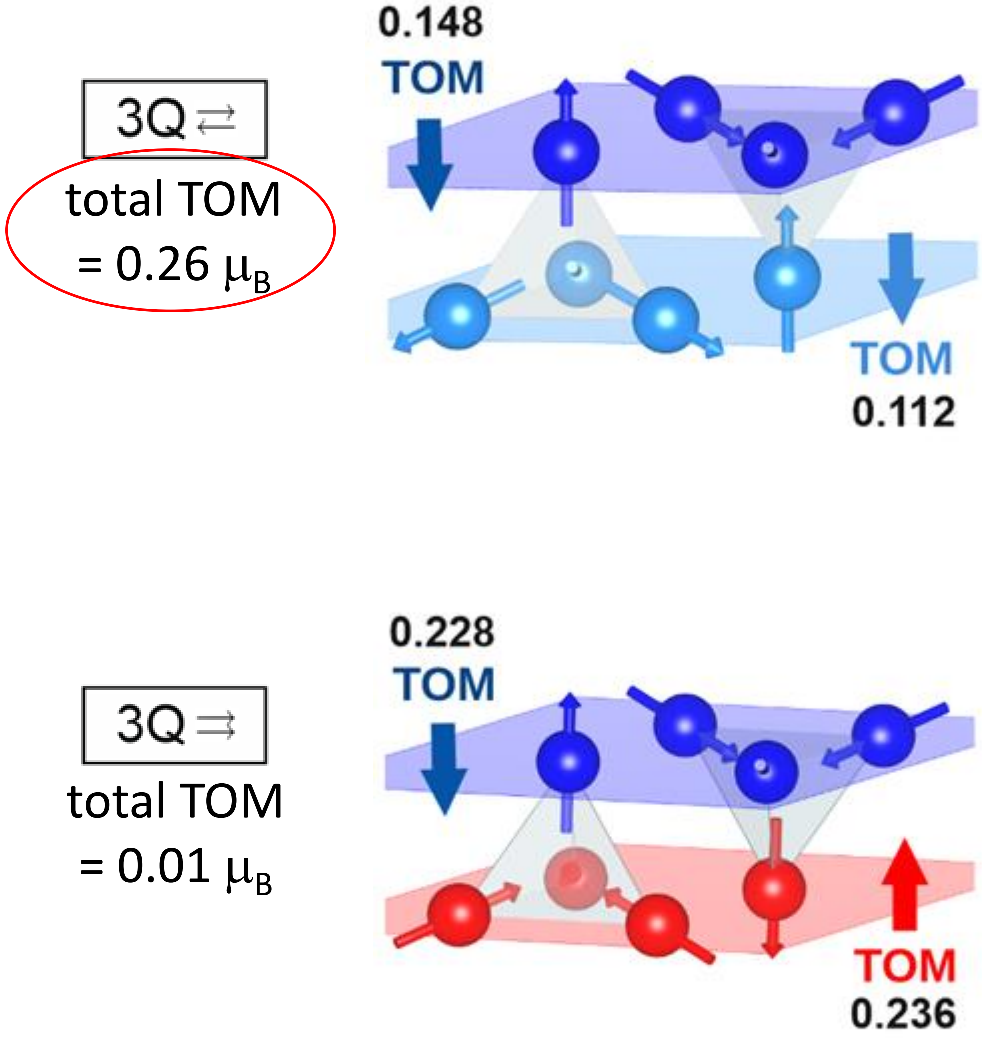


# Hexa-junctions

Mn double layer on Ir(111)



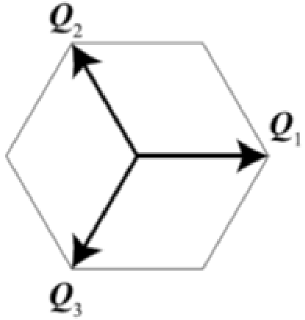



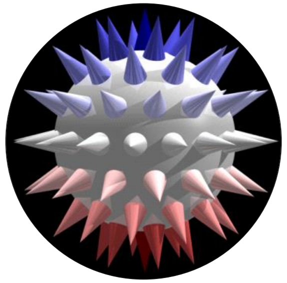

We have a domain wall network with local TOM at the junctions !

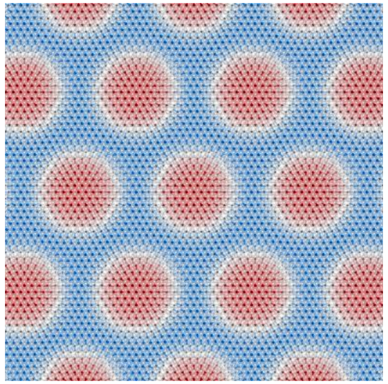
# From the skyrmion lattice to the triple-q state

**Multi-q states**




multi-q state
magnetic field induced
**Skyrmion lattice**

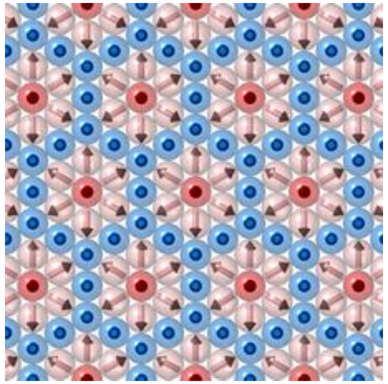




governed by
$J$, DMI, Zeeman

S. Mühlbauer et al., Science **323**, 915 (2009).
X.Z. Yu et al., Nature **465**, 901 (2010).
N. Romming, …KvB et al., Science **341**, 636 (2013).
A.O. Leonov, M. Mostovoy, Nat. Commun. 6, 8275 (2015).
C.D. Batista, et al., Rep. Progr. Phys. **79**, 084504 (2016).
N. D. Khanh et al., Nat. Nanotechnol. 15, 444 (2020).
Z. Wang et. al, Phys. Rev. Lett. **124**, 207201 (2020).
S. Hayami and Y. Motome, J. Phys.: Cond. Mat. **33**, 443001 (2021).

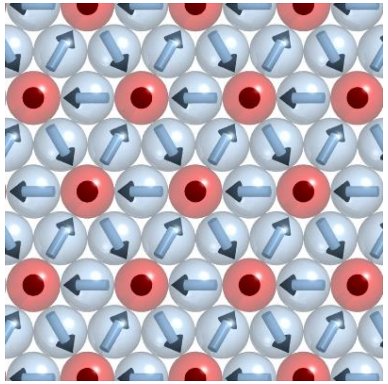
zero field HOI induced
(higher-order interactions)
**Nanoskyrmion lattice**

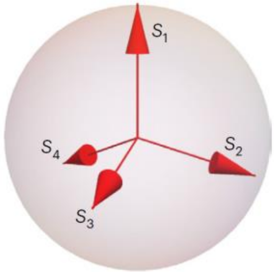


governed by
$J_{ij}$, HOI
(higher order
interactions)

S. Heinze, KvB et al. Nature Phys. **7**, 713 (2011).
KvB et al, Nano Lett. **15**, 3280 (2015).

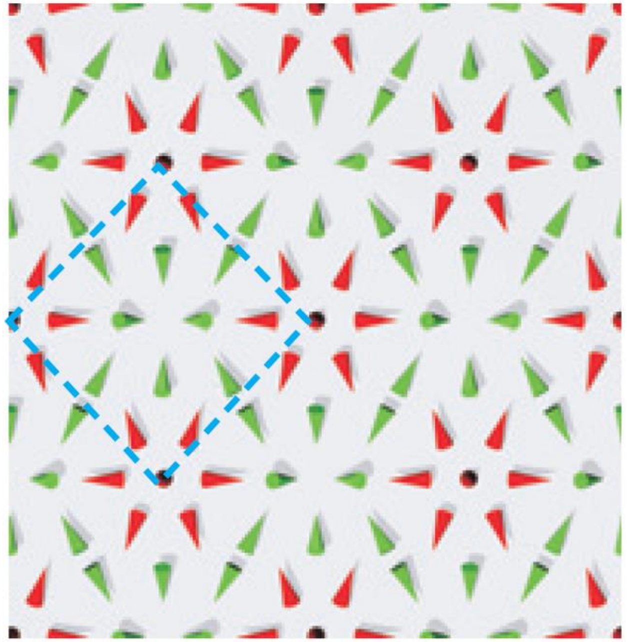
**triple-q state = 3Q**
4 atoms in the unit cell

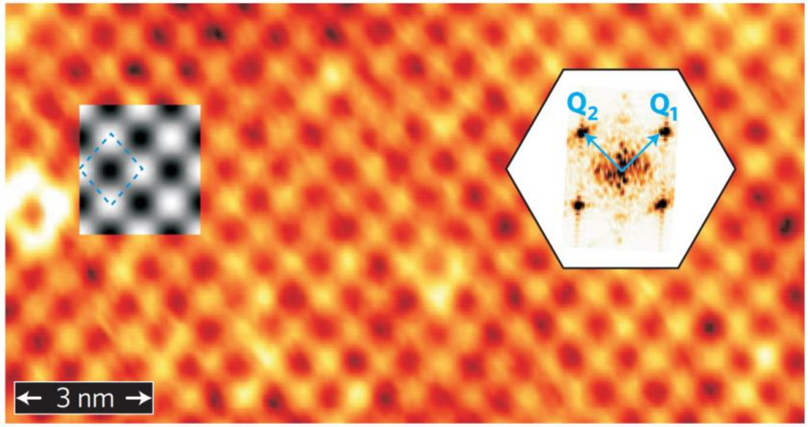





P. Kurz et al., PRL **86**, 1106 (2001).
J. Spethmann, ... KvB et al., PRL **124**, 227203 (2020).
H. Takagi et al., Nature Phys. **19**, 961 (2023).
P. Park et al., Nat. Commun. **14**, 8346 (2023).
F. Nickel, ... KvB, Phys. Rev. B **108**, L180411 (2023).

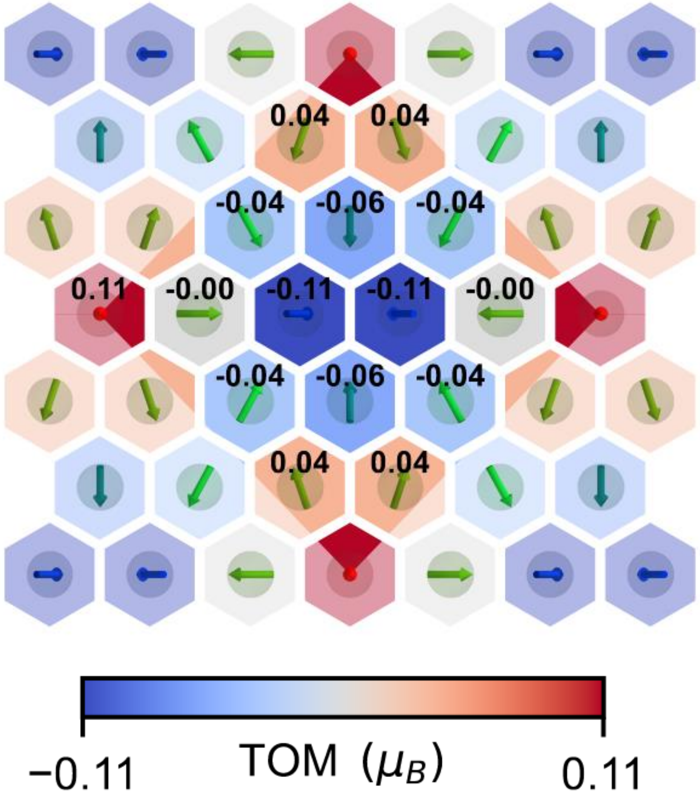
# TOMs of nanoskyrmion lattices





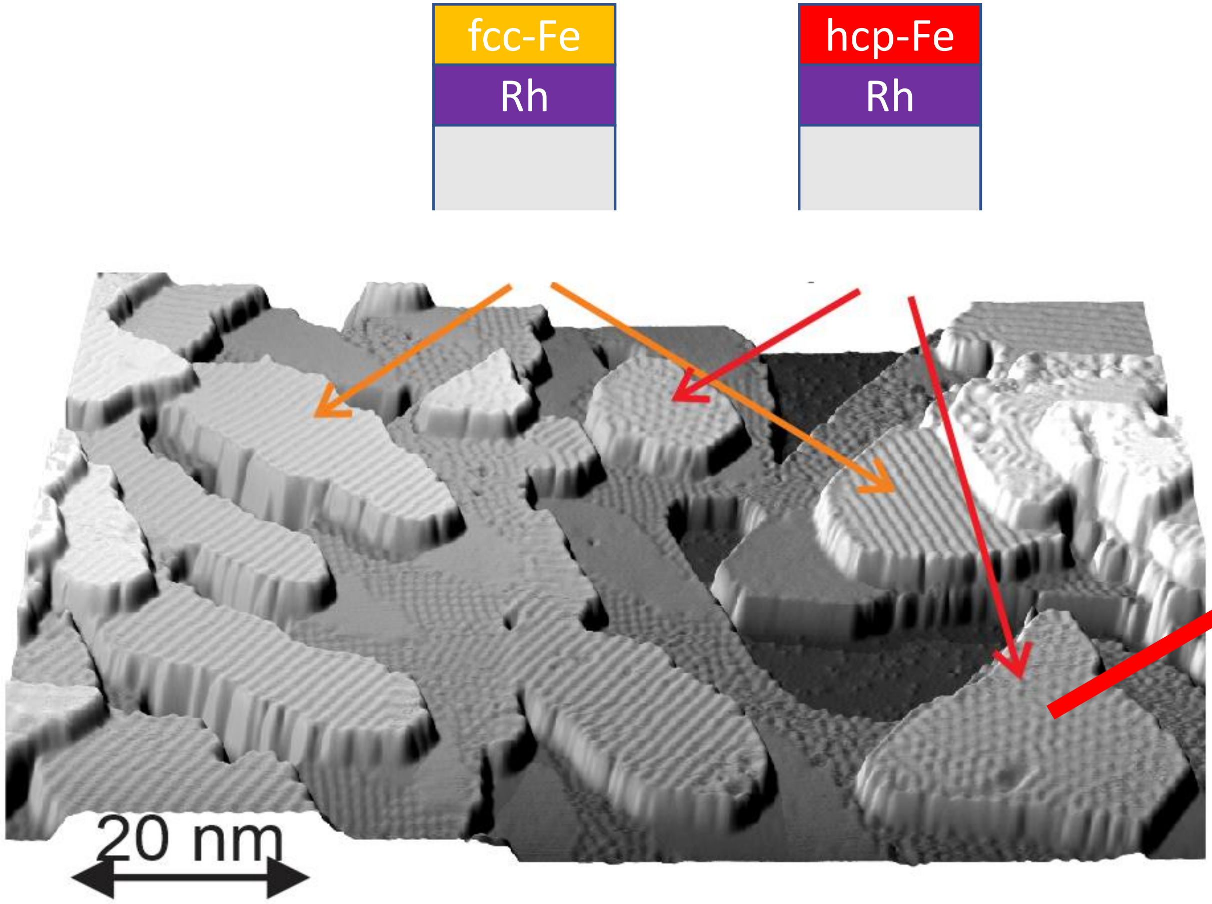

The SkXs have local TOMs that are arranged in an antiferromagnetic fashion. Topological orbital AFM.

The nano-SkX has a net TOM of 0.24 $\mu_B$ per magnetic unit cell.

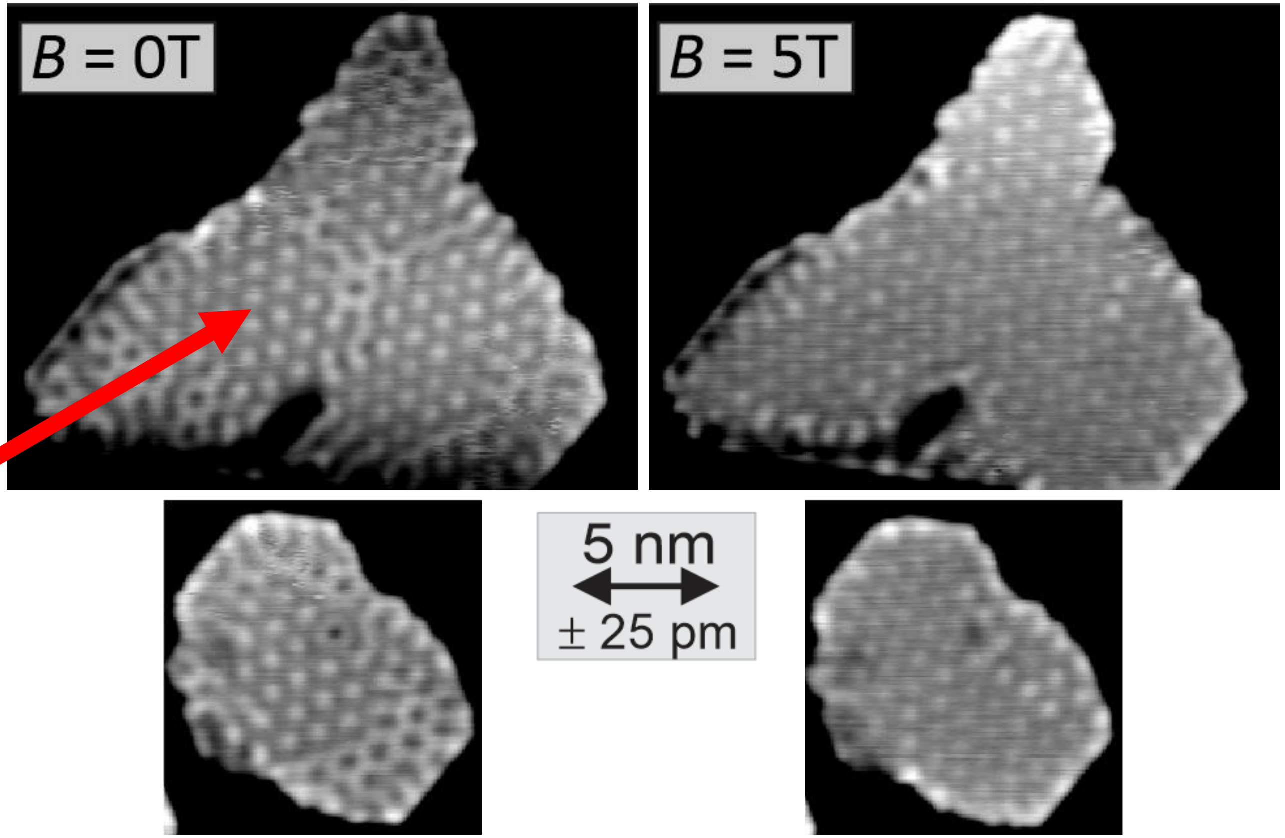



S. Heinze, KvB et al. Nature Phys. **7**, 713 (2011).
KvB et al, Nano Lett. **15**, 3280 (2015).
A. Kubetzka, ... KvB, Phys. Rev. Mater. **4**, 081401 (2020).
F. Nickel, … KvB, et al., arXiv:2405.18088.

# Fe/Rh/Ir(111)




hcp-Fe/Rh1 changes in *B*,
→ net magn moment



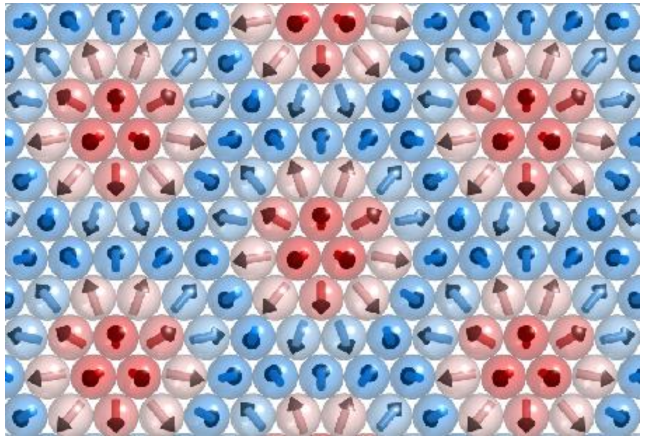

hexagonal magnetic unit cell,
with about 27 atoms



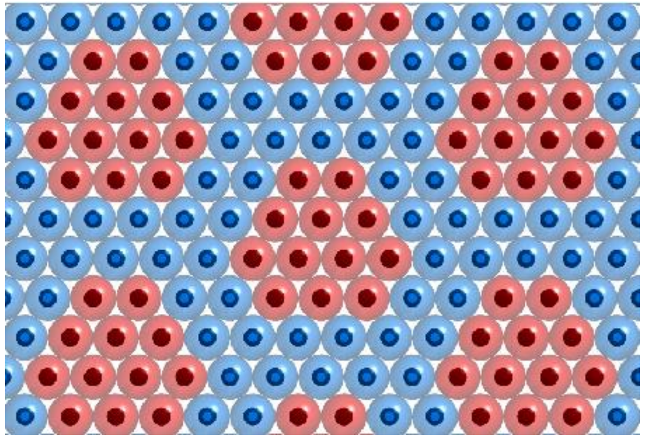
M. Gutzeit, ..., S. Heinze, and KvB, Nature Commun. **13**, 5764 (2022).

# Fe-ML/Rh-ML/Ir(111)

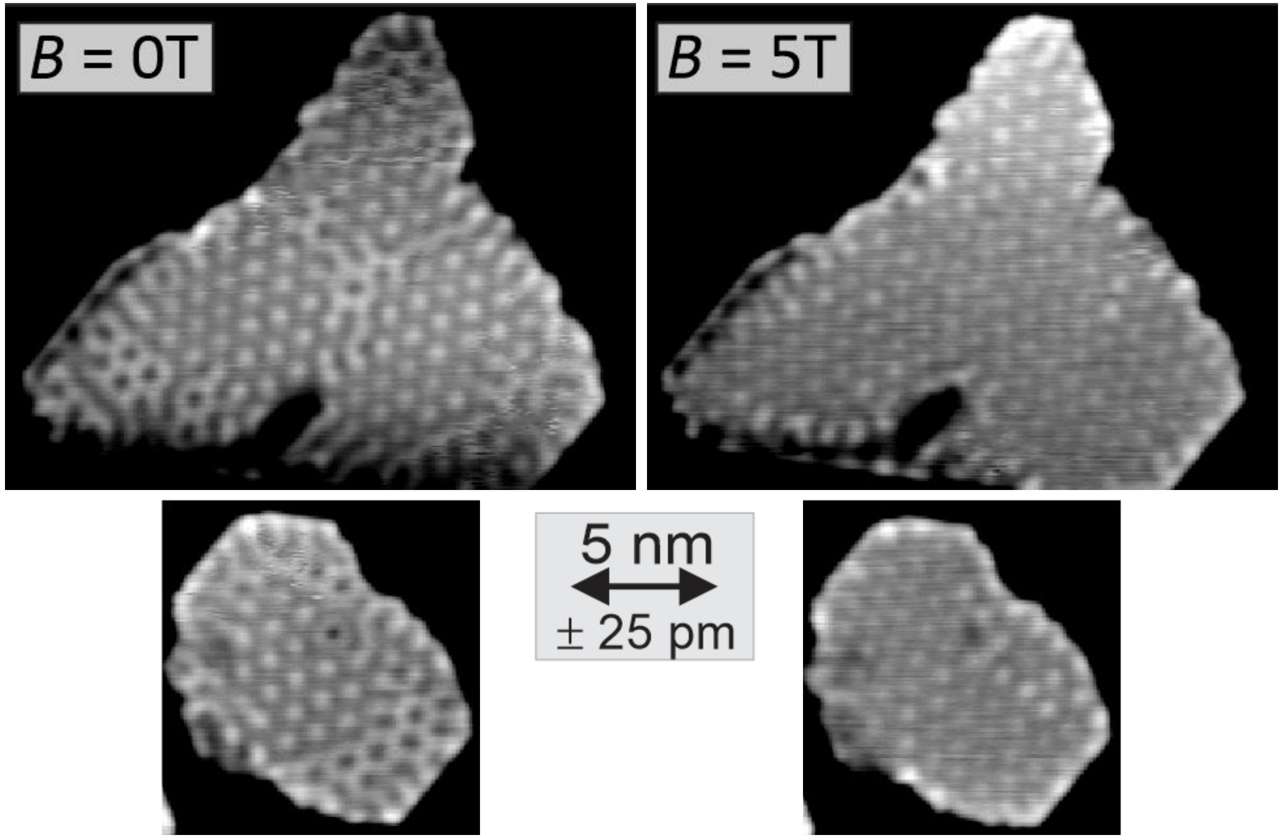
non-collinear
skyrmion lattice
27-SkX
(multi-Q state)



collinear
mosaic state
12:15-MS
(multi-Q, only $m_z$)



hcp-Fe/Rh1 change in *B*,
$\rightarrow$ net magn moment

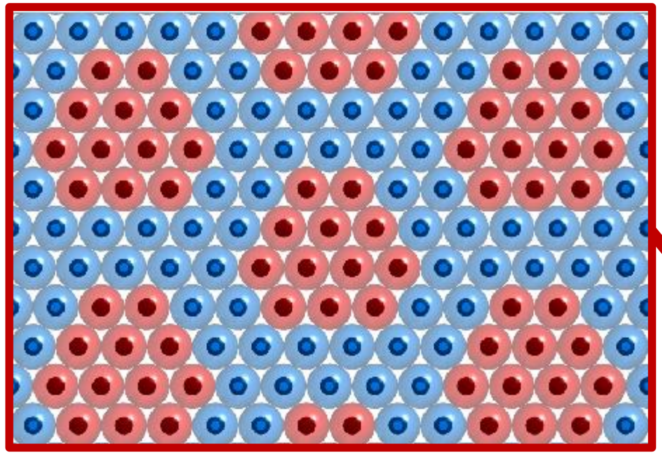


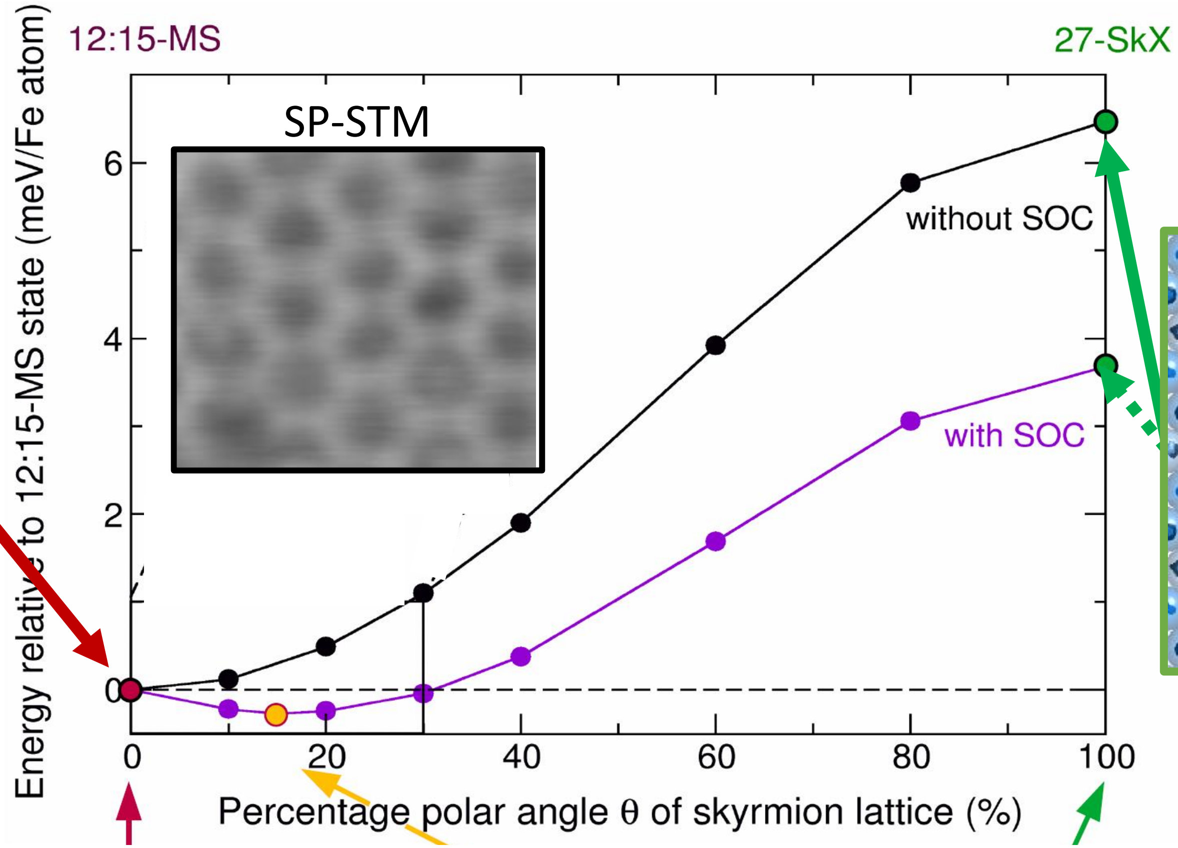

hexagonal magnetic unit cell,
with about 27 atoms

M. Gutzeit, …, S. Heinze, and KvB, Nature Commun. **13**, 5764 (2022).

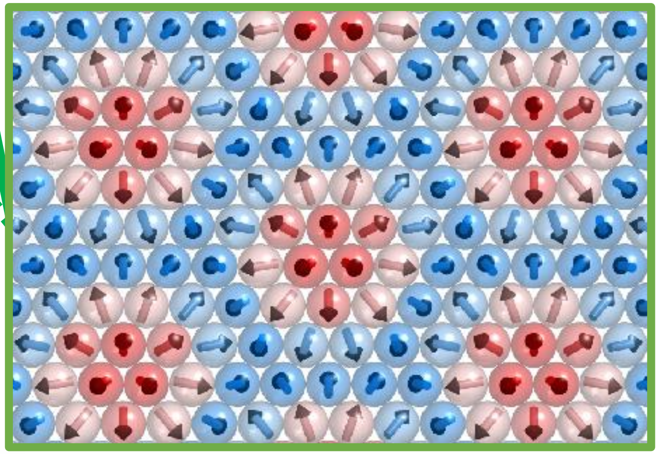
# DFT: hcp-Fe/Rh-ML/Ir(111)

12:15-MS






27-SkX



collinear state has lower energy due to HOIs

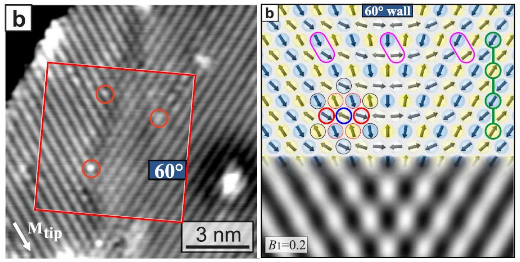
non-collinear state gains DMI energy when SOC is included

lowest energy for HOI + DMI

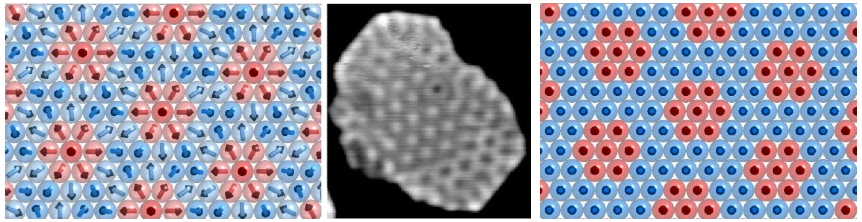


M. Gutzeit, ..., S. Heinze, and KvB, Nature Commun. **13**, 5764 (2022).

# summary

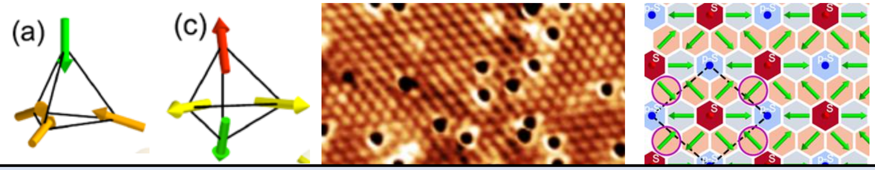



Higher order interactions (HOI)
- select between 1Q or multi-Q state
- induce superposition domain walls
- select between non-collinear SkX and collinear MS



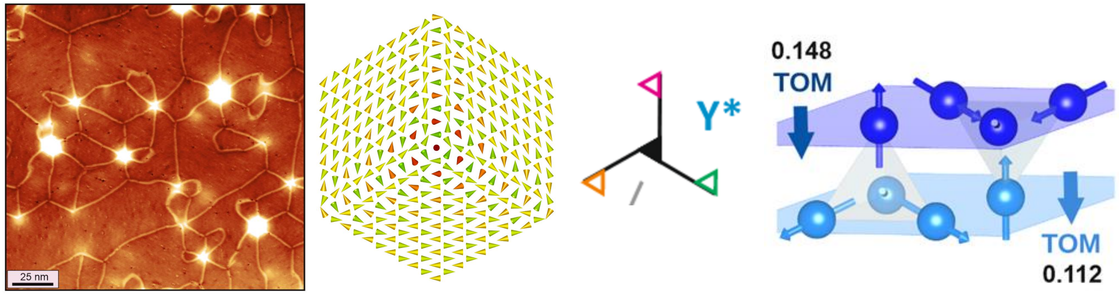
J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).
J. Spethmann, ... KvB et al., Nature Commun. **12**, 3488 (2021).

M. Gutzeit, ..., S. Heinze, and KvB, Nature Commun. **13**, 5764 (2022).




J. Spethmann, ... KvB et al., Phys. Rev. Lett. **124**, 227203 (2020).
F. Nickel, ... KvB, Phys. Rev. B **108**, L180411 (2023).
F. Nickel, ... KvB, et al., arXiv:2405.18088.

Non-coplanar magnetic order
- can lead to topological orbital moments (TOM)
- TOMs occur for the inversional domains of the triple-q state (orbital ferromagnet with zero net spin moment)
- nanoscale spin textures can show local TOMs that are aligned antiparallel (orbital antiferromagnet, can have net TOM)




Strain-induced domain wall network with 1Q, 2Q, and 3Q
- Ar bubbles induce domain walls
- Magnetism-induced shift of 1 Angström
- Chiral triple-junctions
- 3Q with additive TOM at junctions

# acknowledgements

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**DFG HOI**
**418425860**

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