IBM Research

Aug 5th, 2020
SPICE Workshop: 2D van der Waals Spin Systems

Observation of skyrmions and their current-driven dynamics in van der Waals heterostructures

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IBM Thomas J. Watson Research Center

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Korea Institute of Science and Technology (KIST): **Tae-Eon Park**, Kyung Mee Song [Exp.]

RIKEN Center for Emergent Matter Science: **Licong Peng**, Fehmi Yasin, **Xiuzhen Yu** [Exp.]

Chinese Academy of Sciences: Jinghua Liang, **Hongxin Yang** [Theory - DFT]

CEA/CNRS/UFG-Grenoble & CNRS Thales: Ali Hallal, **Mairbek Chshiev**, **Albert Fert** [Theory - DFT]

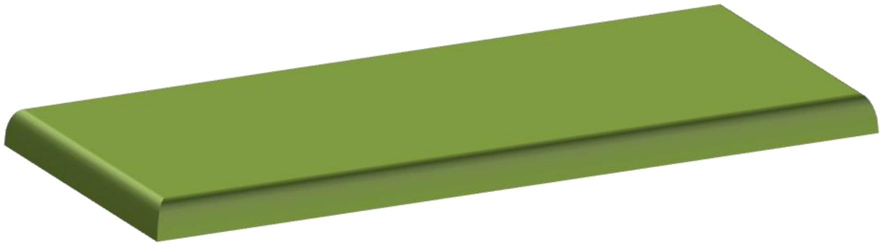
Chinese University of Hong Kong: Xichao Zhang, Yan Zhou [Simulations]

Contents

- Magnetic Skyrmions: concept, history and recent progress
- Experimental observation of skyrmions in 2D vdW system

Magnetic textures

Magnetic Film



A_{ex} - Exchange

K_u - Anisotropy

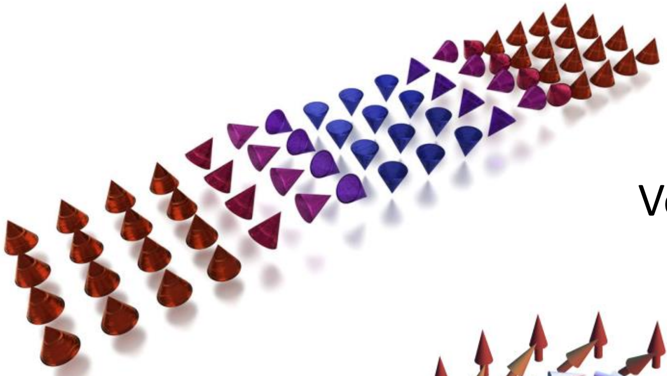
M_s - Saturation Magnetization

D - Dzyaloshinskii-Moriya exchange

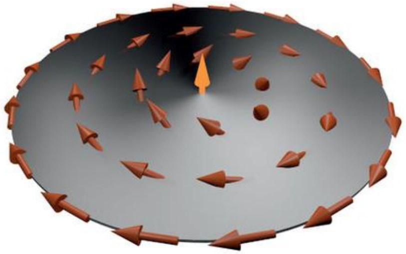
+ external field, dipole field etc.

In magnetic materials, we observe a variety of magnetic textures, and all these textures are the result from internal energetic competition

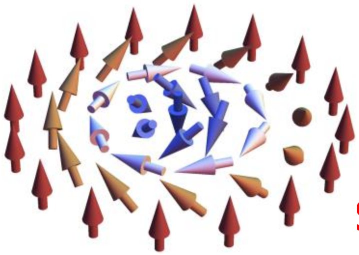
Uniform Magnetization



Domain wall



Vortex



Skyrmion

S. Seki *et al.*, ***Science*** 13, 336 (2012)
S. Wintz *et al.*, ***Nat. Nanotech***. 11, 948-953 (2016)

Dzyaloshinskii-Moriya Interaction (DMI)

1. Heisenberg Exchange

$$H_{exchange} = \sum_{i,j} J_{ex}[\mathbf{S}_i \cdot \mathbf{S}_j]$$

: Ferromagnet, Anti-ferromagnet,
Non-Uniform texture by dipolar interaction

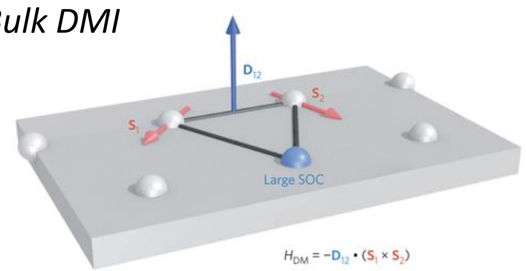
Materials with broken inversion-symmetry & strong spin-orbit coupling

2. Dzyaloshinskii-Moriya Exchange

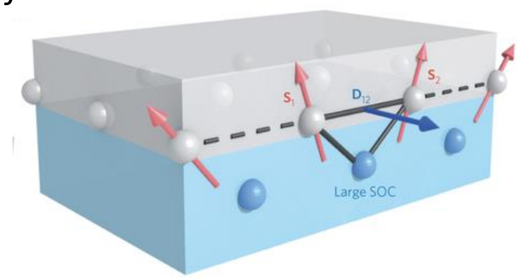
$$H_{DM-exchange} = -\sum_{i,j} \mathbf{D}_{ij} \cdot [\mathbf{S}_i \times \mathbf{S}_j]$$

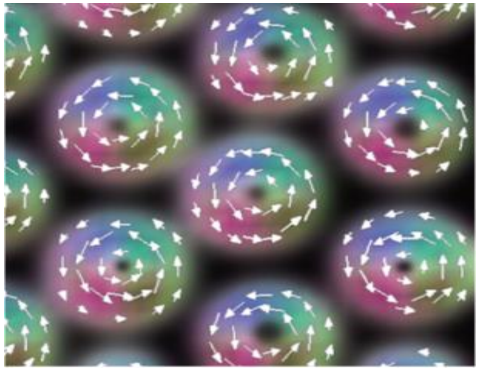
: Non-uniform texture with fixed chirality
by DM-interaction & dipolar field

Bulk DMI



Interface DMI



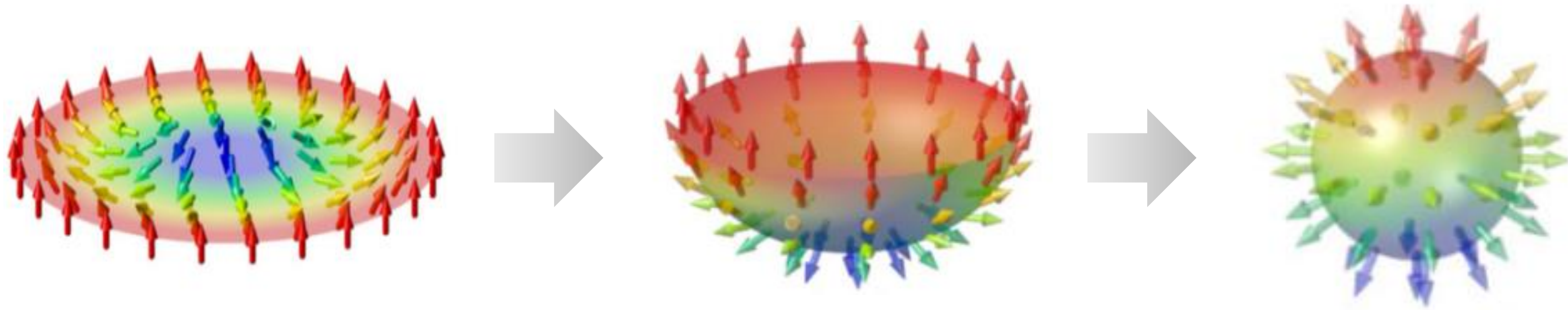


Magnetic Skyrmion

X.Z. Yu *et al*. ***Nature*** (2010), A. Fert *et al*., ***Nat. Nanotech***. (2013)

Topology of magnetic skyrmions

"Skyrmion" – Particle-like object protected by continuous vector field



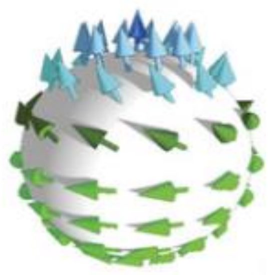
2D (in real magnet) **3D vector space**

Topological number : $n = \frac{1}{4\pi}\int \mathbf{M}\cdot\left(\frac{\partial \mathbf{M}}{\partial x}\times\frac{\partial \mathbf{M}}{\partial y}\right)dxdy$

Ferro-magnet

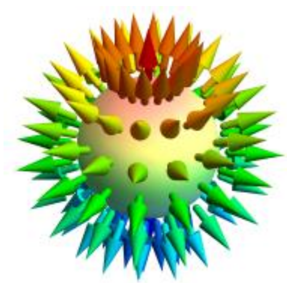
$Q = 0$

Magnetic Vortex



$Q = 1/2$

Magnetic Skyrmion:

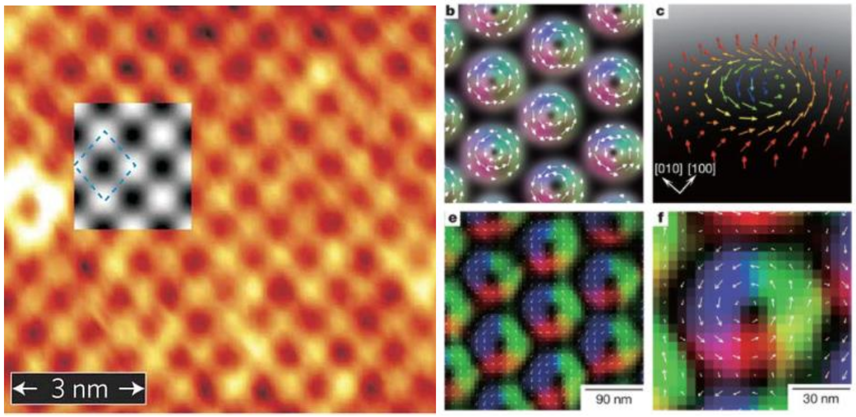


$Q = \pm 1$

Dynamics of magnetic skyrmions determined by unique topological number

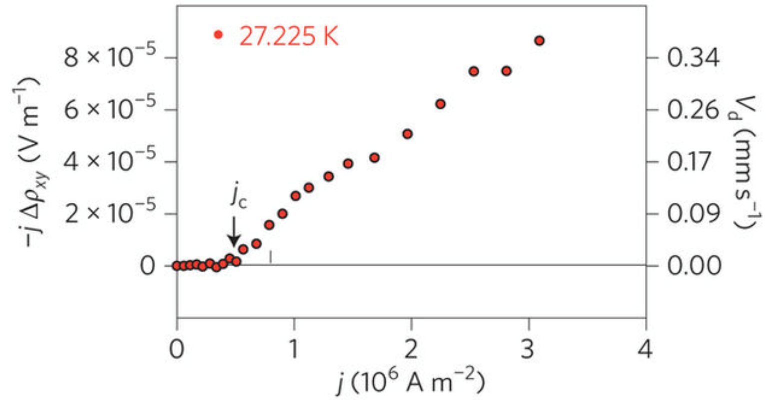
Why Skyrmions?

Topological Protection + Atomic Scale (few nm)



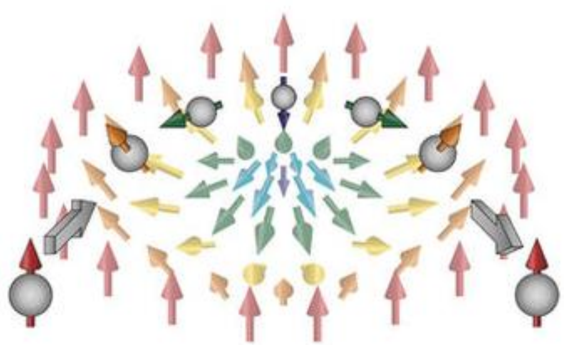
S. Heinze *et al.*, ***Nat. Phys.*** 7, 713-718 (2011)
X.Z. Yu *et al*. ***Nature*** 465, 901-904 (2010)

Extremely Low j_{motion} threshold



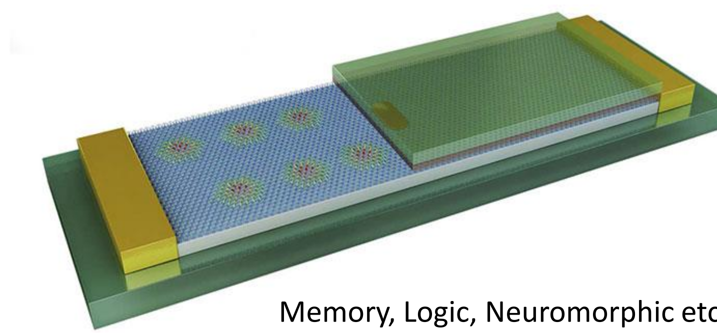
T. Schulz *et al.*, ***Nat. Phys.*** 8, 301-304 (2012)

Emergent electrodynamics – Topological Hall effect



N. Nagaosa *et al.*, ***Nat. Nanotech.*** 8, 899-911 (2013)
A. Soumyanarayanan *et al.*, ***Nat. Mater***. 16, 898-904 (2017)

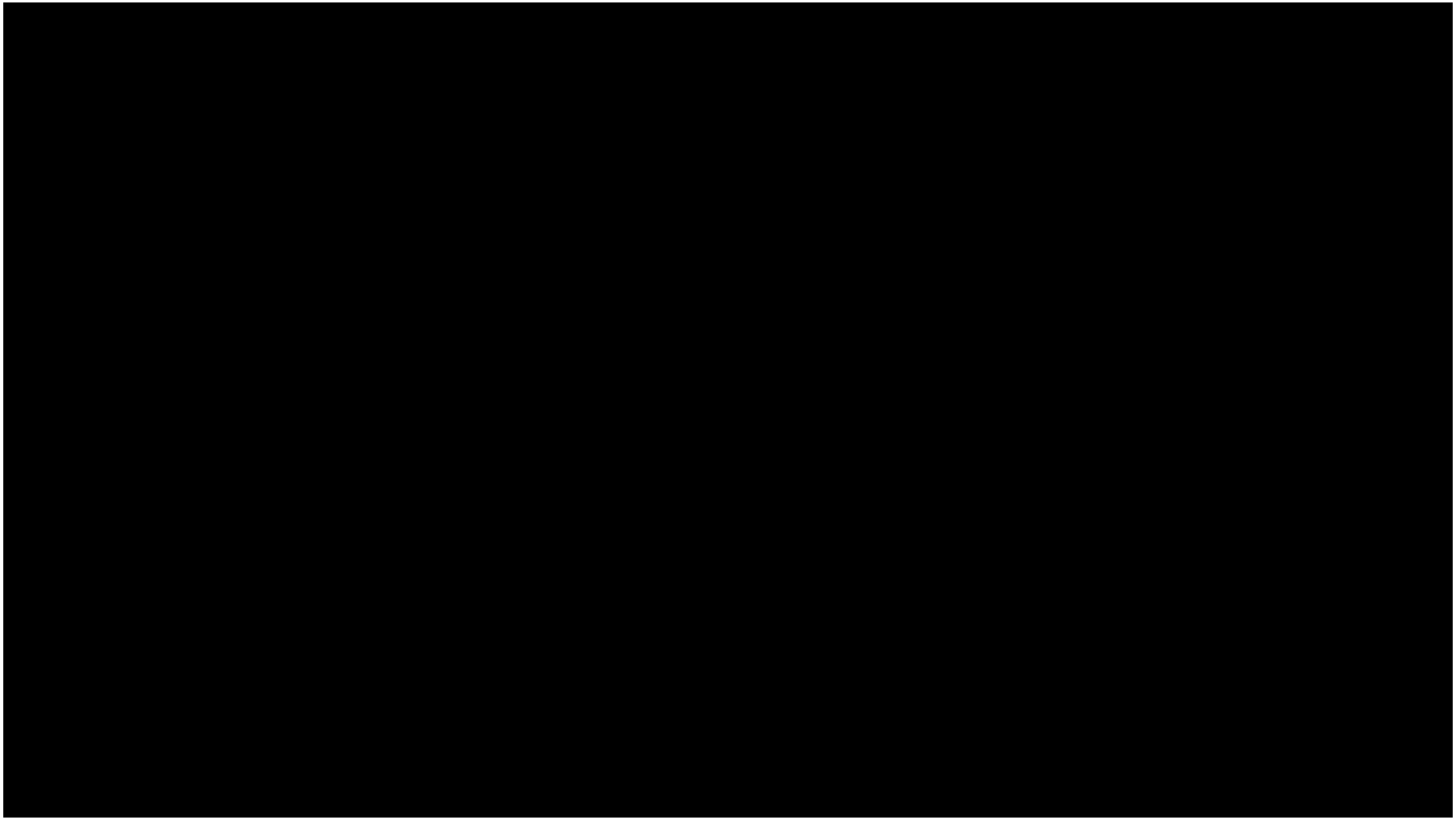
Device Opportunities



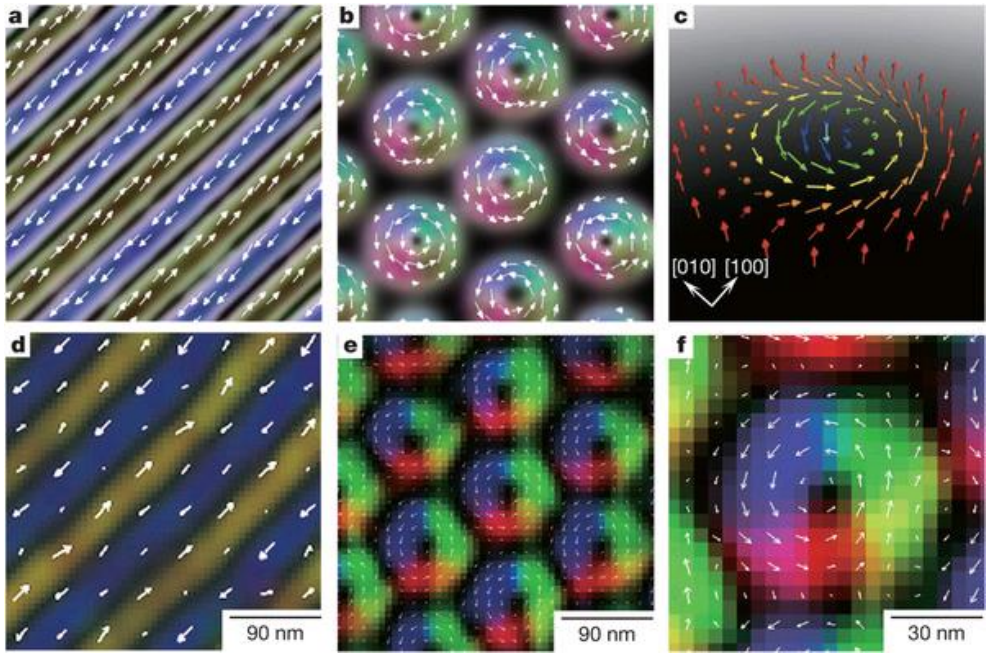
Memory, Logic, Neuromorphic etc.

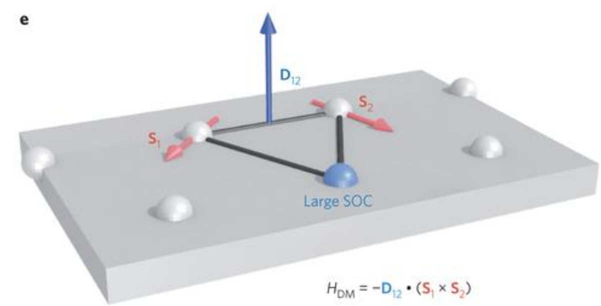
X. Zhang *et al.*, ***Sci. Rep***. 5, 9400 (2015)
Y.Q. Huang *et al.*, ***Nanotechnology***, 28 (2017)

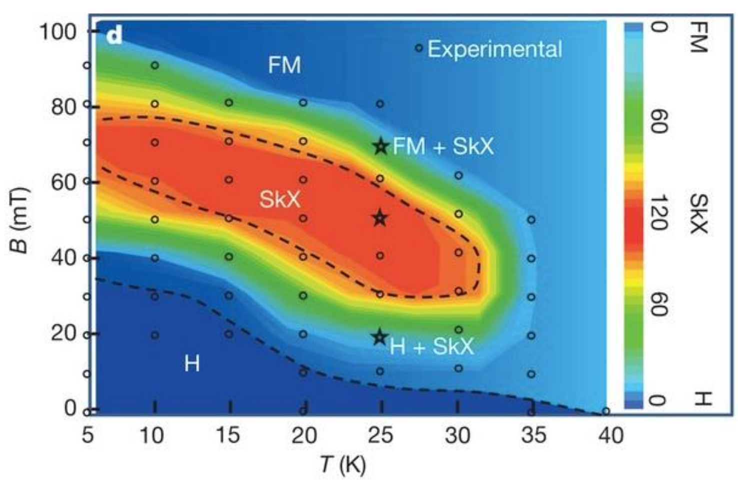
Skyrmion Memory



Skyrmions at low temperatures





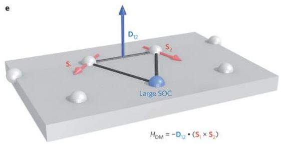


Non-centrosymmetric materials w/ bulk-DMI
(e.g. B20-type crystals like SiGe, MnSi)
to recent ferroelectric & frustrated magnetic systems

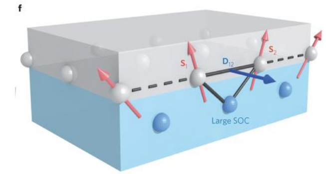
S Mühlbauer *et al.*, ***Science*** 13, 915-919 (2009)
X. Z. Yu *et al.* ***Nature*** 465, 901–904 (2010)
T. Schulz *et al.*, ***Nat. Phys***. 8, 301-304 (2012)
X. Z. Yu *et al.*, ***Nat. Commun***. 3, 988 (2012)
+ many more

Skyrmions at room temperature

Bulk-DMI
(e.g. Co-Mn-Zn)

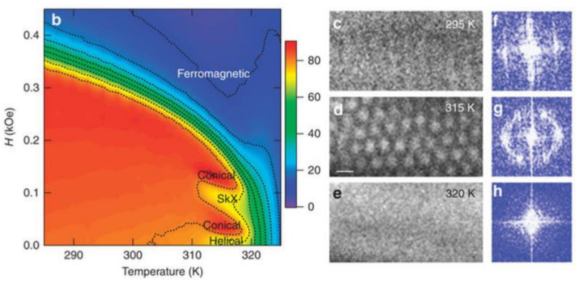


Interfacial-DMI heterostructures

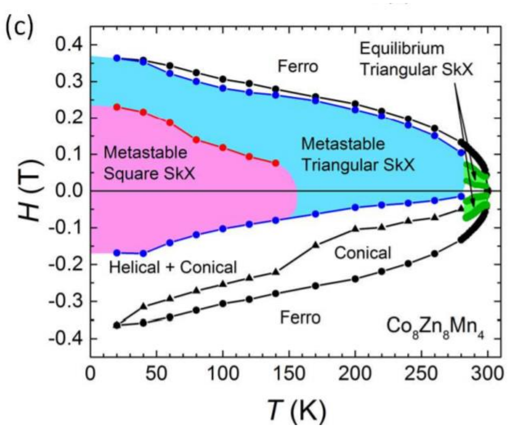


Single layer (e.g. Ta/CoFeB/TaOx)

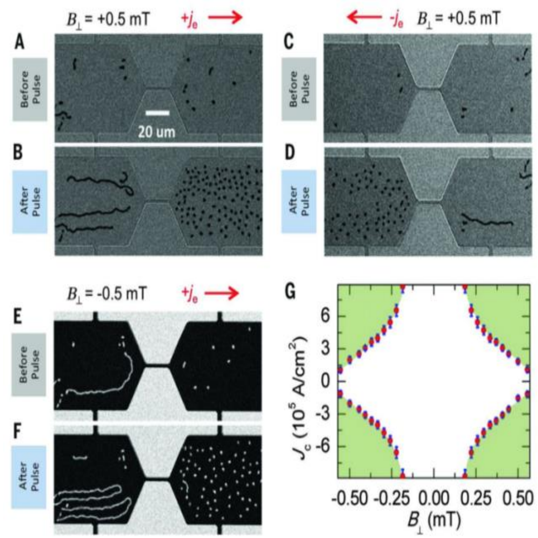
Multilayer (e.g. $[Pt/Co/Ta]_{x15\text{-}20}$)



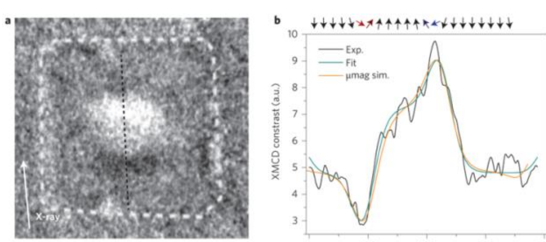
Y. Tokunaga *et al.*, ***Nat. Commun.*** (2015)



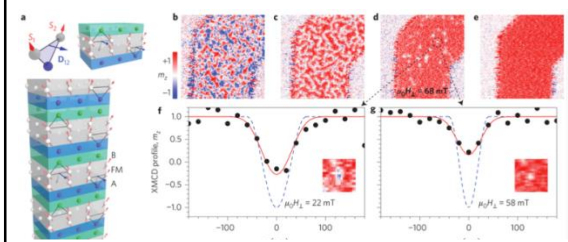
K. Karube *et al.*, ***Nat. Mater.*** (2016)



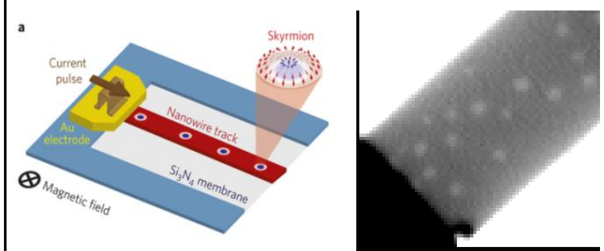
W. Jiang *et al.*, ***Science*** (2015)

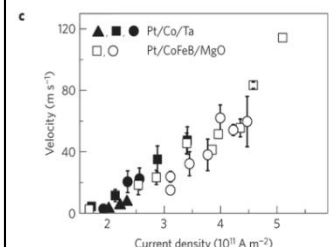


O. Boulle *et al.*, ***Nat. Nanotech.*** (2016)



C. Moreau-Luchaire *et al.*, ***Nat. Nanotech.*** (2016)

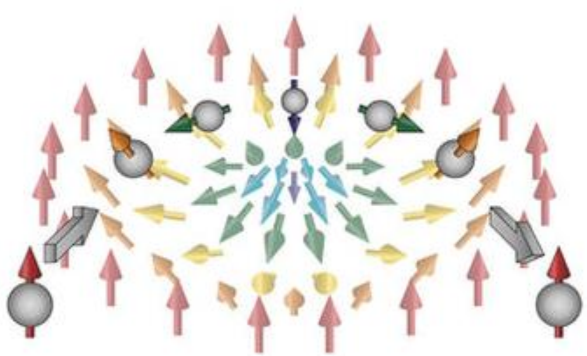




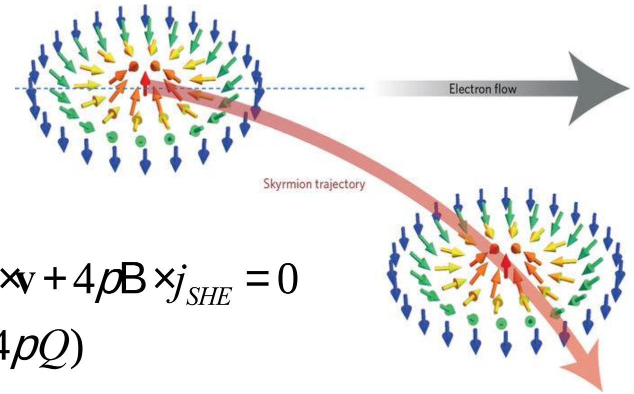
S. Woo *et al.*, ***Nat. Mater*** (2016)
S. Woo* *et al.*, ***Nat. Commun.*** 8, 15573 (2017)

Topological Hall effects of FM skyrmions

Topological Hall effect – Skyrmion Hall effect



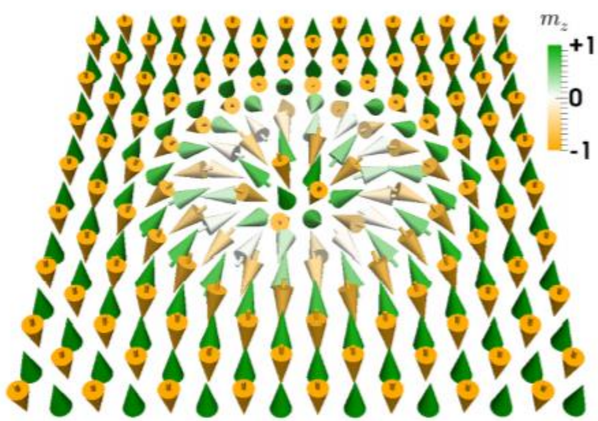
N. Nagaosa *et al.*, ***Nat. Nanotech*.** 8, 899 (2013)
A. Soumyanarayanan *et al.*, ***Nat. Mater***. 16, 898 (2017)



$$\mathbf{G}\times\mathbf{v}-\alpha\mathcal{D}\cdot\mathbf{v}+4\pi\mathcal{B}\cdot j_{SHE}=0$$

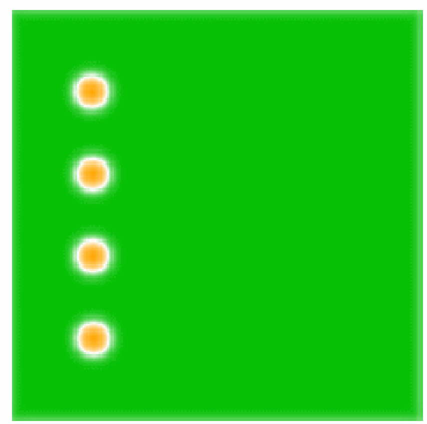
$$\mathbf{G}=(0,0,-4\pi Q)$$

Jiang, W. *et al.*, ***Nat. Physics,* 13**, 162 (2017)
Litzius, K. *et al.*, ***Nat. Physics,* 13**, 170 (2017)

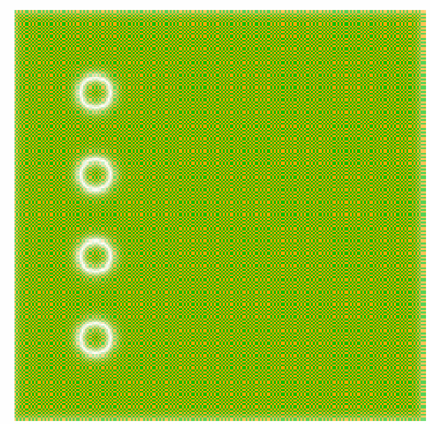


X. Zhang *et al.*, ***Sci. Rep***. 6, 24795 (2016)
Barker J. *et al.*, ***Phys. Rev. Lett.*** 116, 147203 (2016)

FM Skyrmion



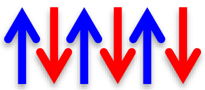
AFM Skyrmion



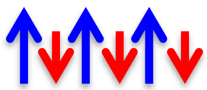
Skyrmions in Ferri-magnets

FM: Q=1

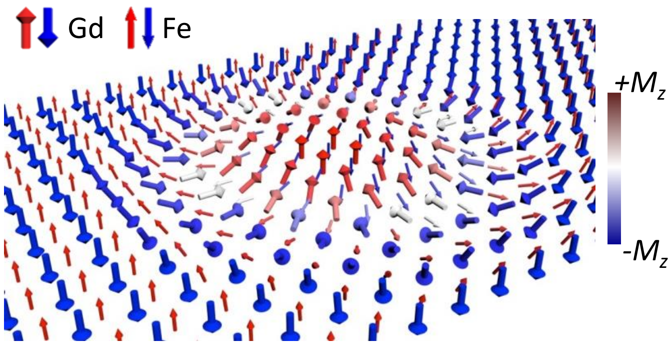
AFM: Q=0



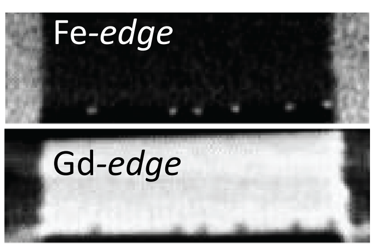
Ferrimagnet!
(RE-TM)
Q~0 and material!

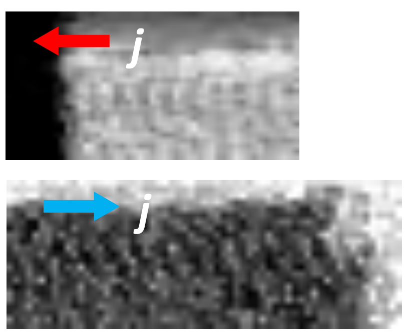


Material: [Pt/GdFeCo/MgO]$_{20}$



STXM images



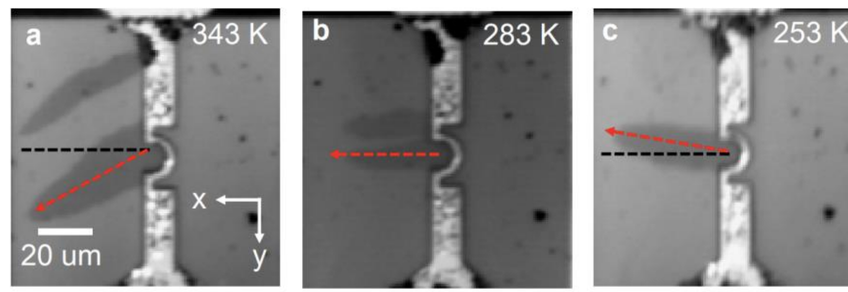


S. Woo* *et al.*, ***Nat. Commun.*** 9, 959 (2018)

Electrical writing and deleting of ferri-skyrmions

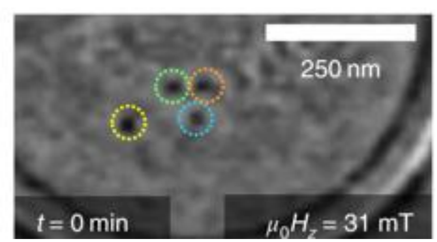
S. Woo* *et al.*, ***Nat. Electron.*** 1, 288-296 (2018)

Zero skyrmion Hall effect at T_A



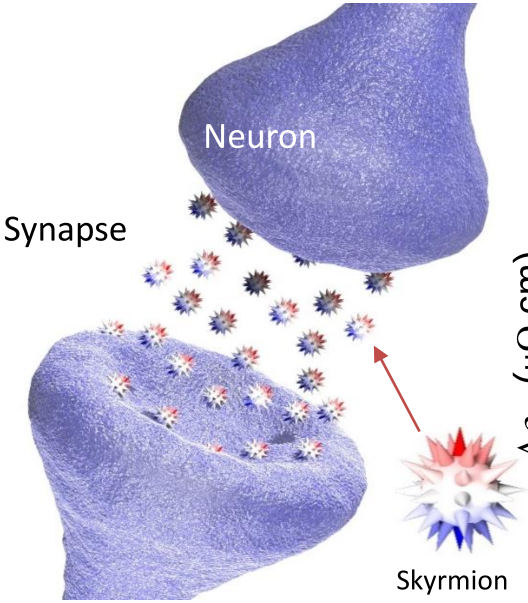
Y. Hirata *et al.*, ***Nat. Nano.*** *14, 232* (2019)

Small (<10 nm) & Fast (> 1km/s) skyrmions at T_M/T_A



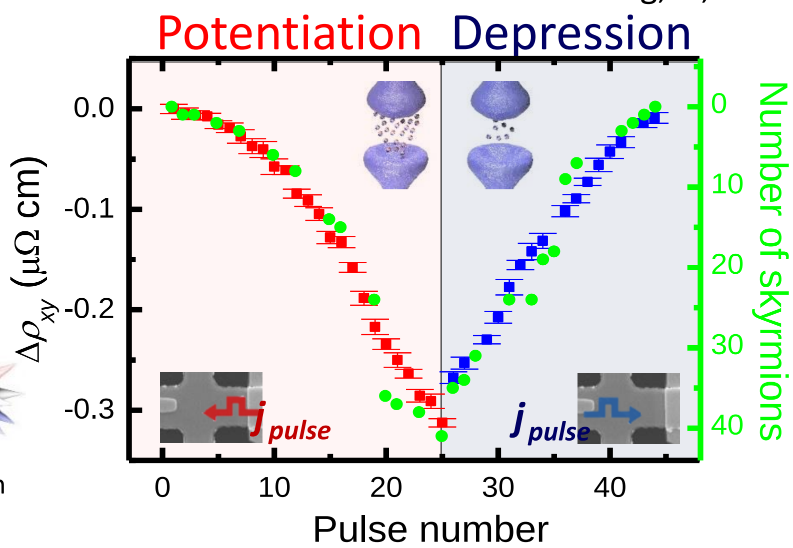
L. Caretta *et al.*, ***Nat. Nano.*** *13, 1154* (2018)

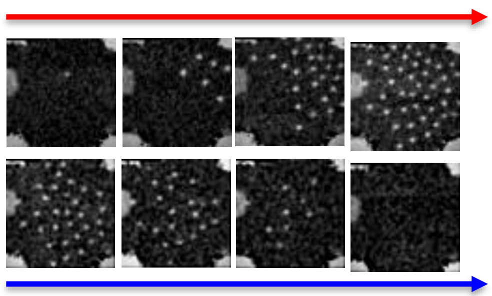
Skyrmion-based neuromorphic computing



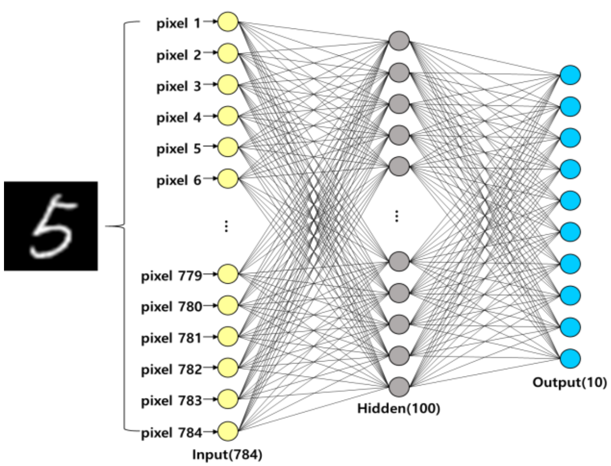
Skyrmion-based artificial synaptic device

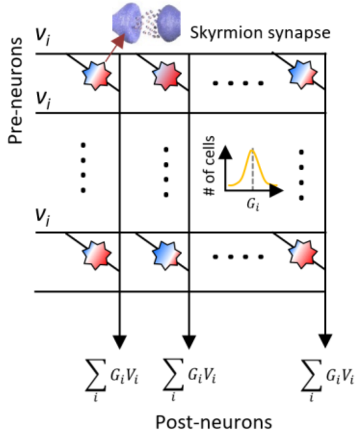
K.M. Song, …, S. Woo*, ***Nat. Electron.*** 3, 148-155 (2020)

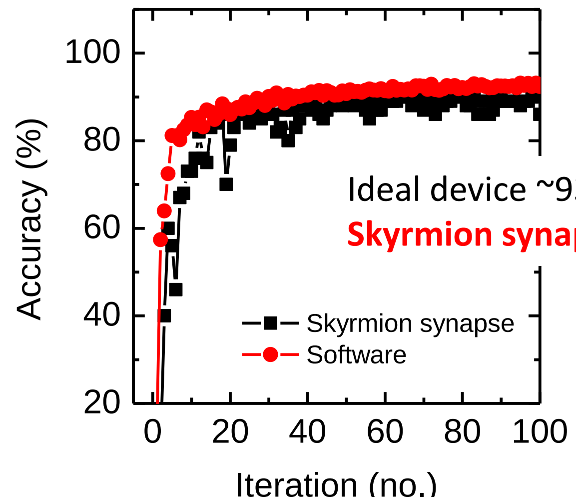




Pattern-recognition task







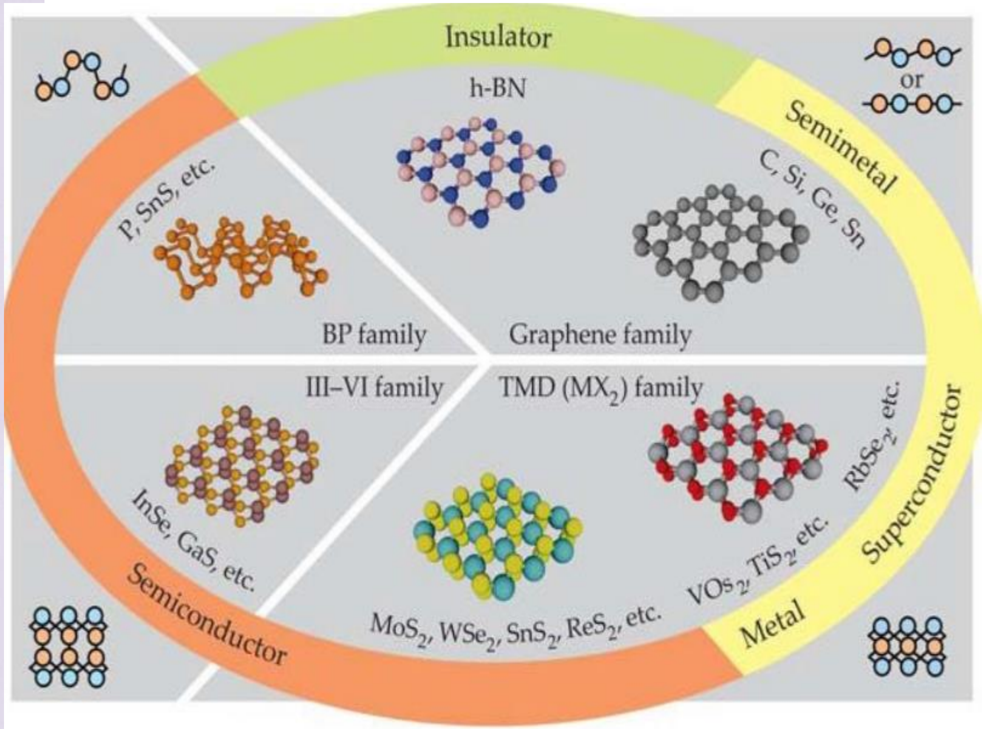
Contents

- Magnetic Skyrmions: concept, history and recent progress

- Discovery of skyrmions in 2D vdW system

  Ref. T.-E. Park, …, H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

2D materials and their heterostructures



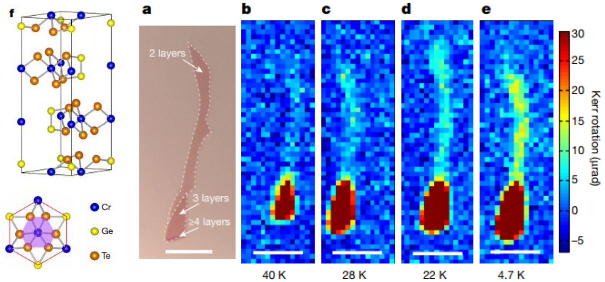
Ajayan, Kim and Banerjee, ***Physics Today*** 69, 9 (2016)

- Metallic materials: Graphene, ..
- Insulating materials: hBN, ..
- Semiconducting materials: WSe_2, MoS_2, ..
- Complex-metallic compounds : $TaSe_2$, TaS_2, ..
- Superconducting: $NbSe_2$, ..
- **Magnetic materials**: $Cr_2Ge_2Te_6$, Fe_3GeTe_2, CrI_3

2D magnets for spintronics

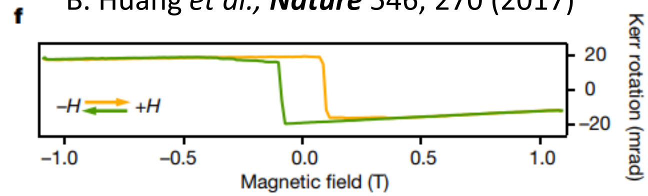
Discovery of intrinsic 2D magnetism at BL/ML (Large SoC)

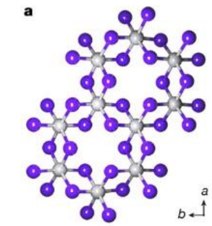
$Cr_2Ge_2Te_6$



C. Gong *et al.*, ***Nature*** 546, 265 (2017)

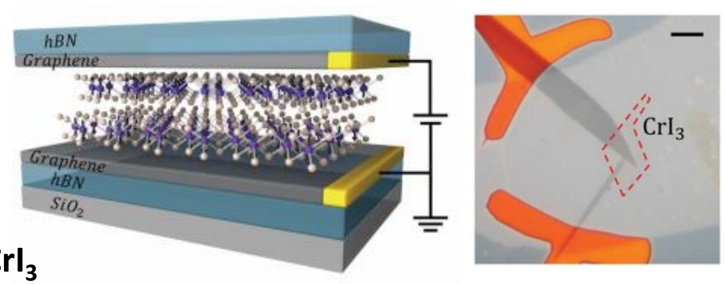
B. Huang *et al.*, ***Nature*** 546, 270 (2017)



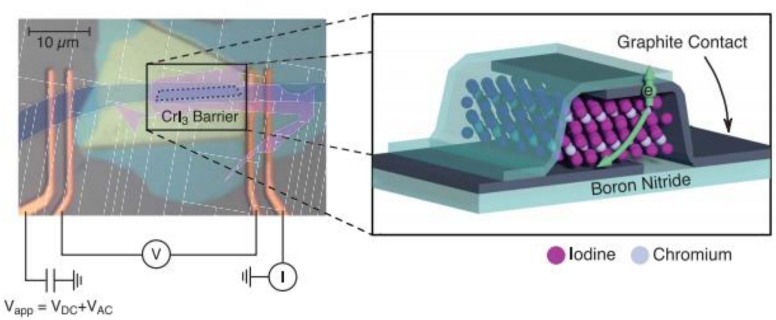


CrI_3

Tunneling magnetoresistance



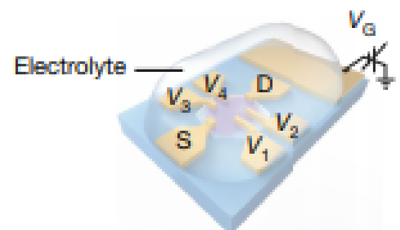
CrI_3

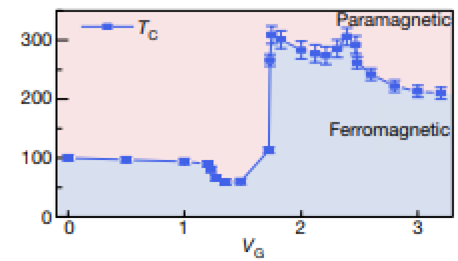


T. Song *et al.*, ***Science*** 360, 1214 (2018)

D.R. Klein *et al.*, ***Science*** 360, 1218 (2018)

Gate tunable room temperature 2D magnet

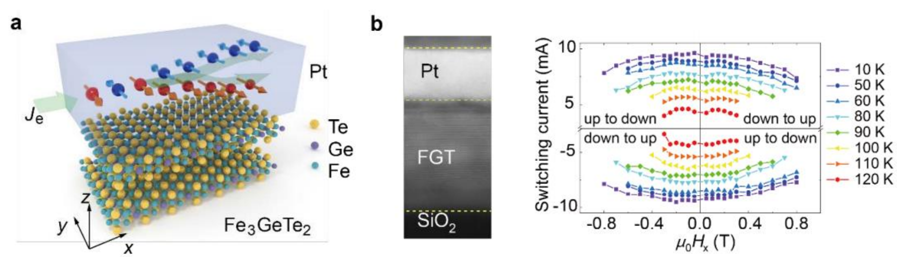




Fe_3GeTe_2

Y. Deng *et al.*, ***Nature*** 563, 94 (2018)

Current-driven magnetization switching

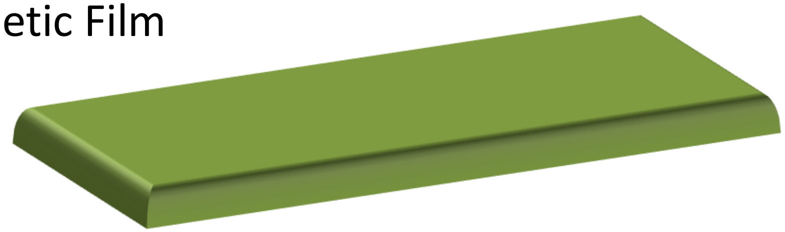


Fe_3GeTe_2

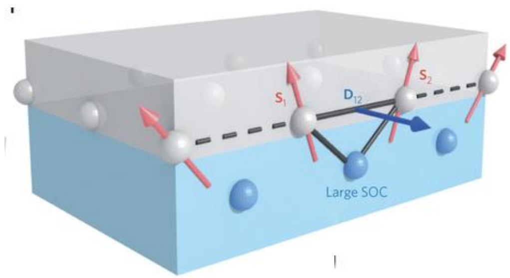
X. Wang *et al.*, ***Sci. Adv.*** 5, eaaw8904 (2019)

2D vdW materials for skyrmion

Magnetic Film



Interface-driven strong DMI (SoC + **symmetry breaking**)



A_{ex} - Exchange

K_u - Anisotropy

M_s - Saturation Magnetization

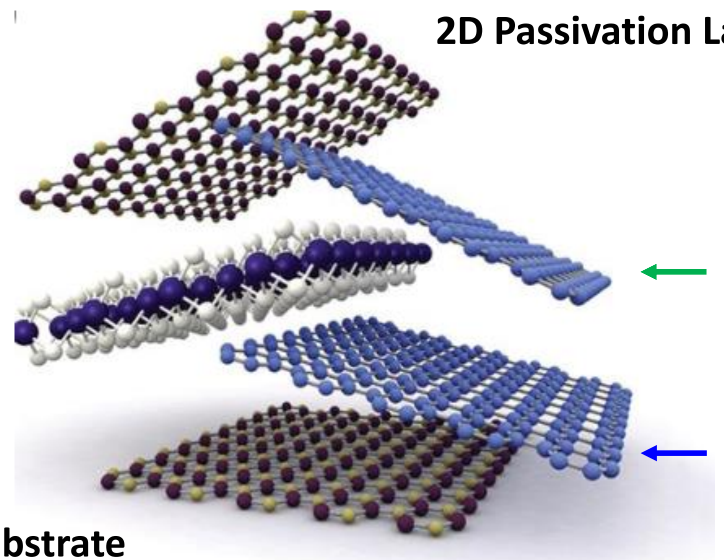
D **- Dzyaloshinskii-Moriya exchange**

+ external field, dipole field etc.

Exemplary skyrmion material design
: 2D in monolayer limit -> Strong DMI
: Crystalline 2D vdW magnet
: Tunable interfaces

2D Passivation Layer

2D Magnet →



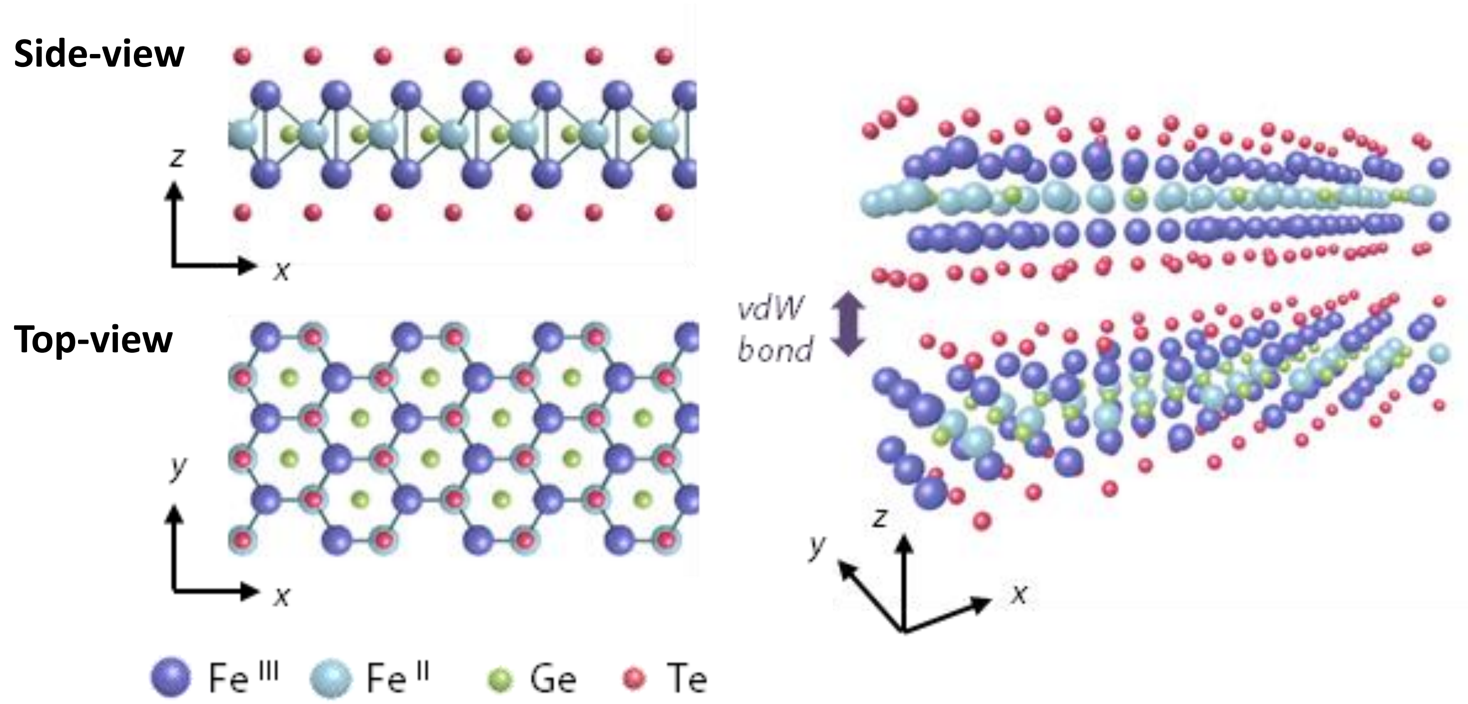
← **High SoC Material B or Sym. Breaking layer**

← **High SoC Material A**

2D Substrate

[Image from ***Science*** 353, 6298 (2016)]

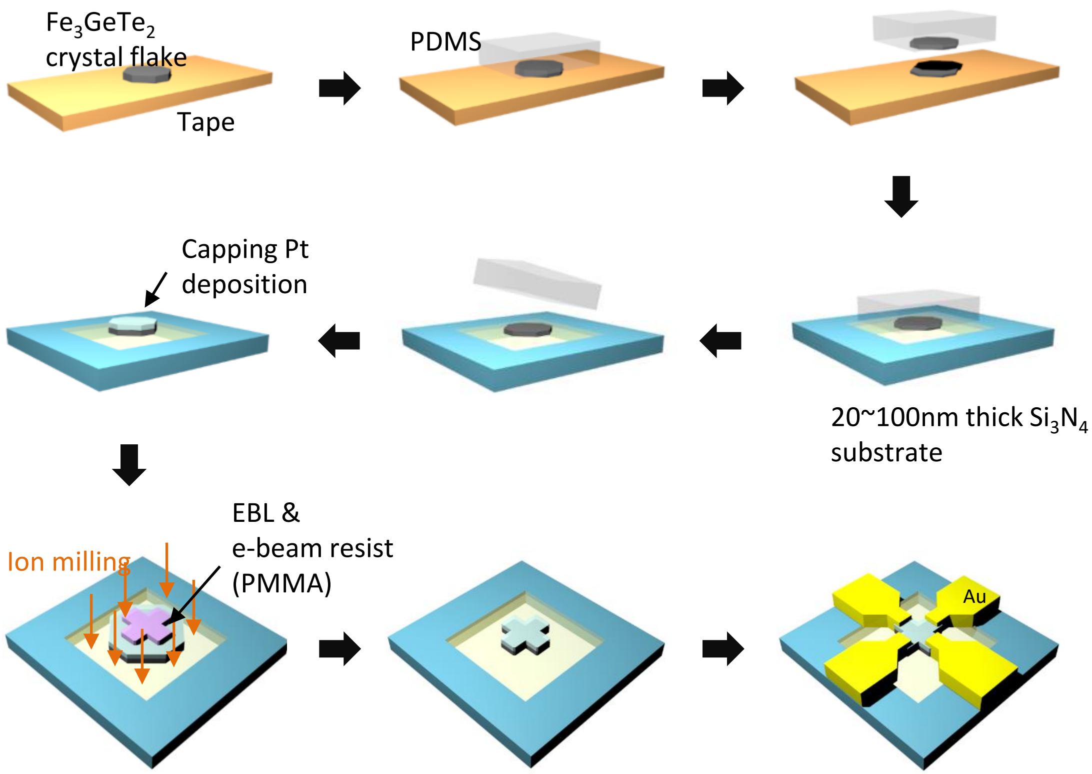
Magnetic skyrmions in Fe_3GeTe_2

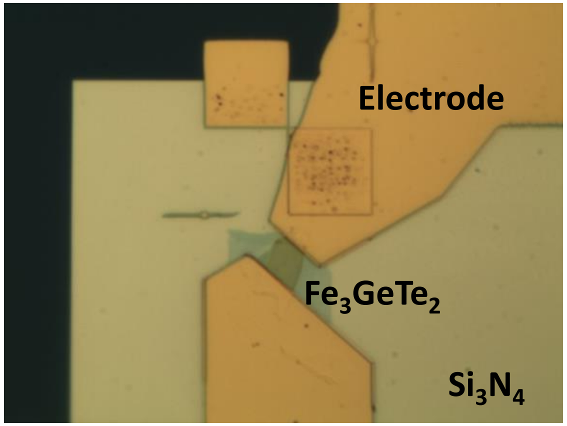


- Relatively high ferromagnetic transition temperature (T_c) > 150K
- Perpendicular magnetic anisotropy (PMA) & metallic nature
- Hexagonal system (**space group 194**, **point group 6/m2/m2/m**): ***D_{6h}***
- **No bulk Dzyaloshinskii-Moriya interaction (DMI) : No inversion symmetry**

Fabrication of Fe_3GeTe_2 devices

Mechanical exfoliation + Dry PDMS transfer + EBL lithography on SiN Membranes

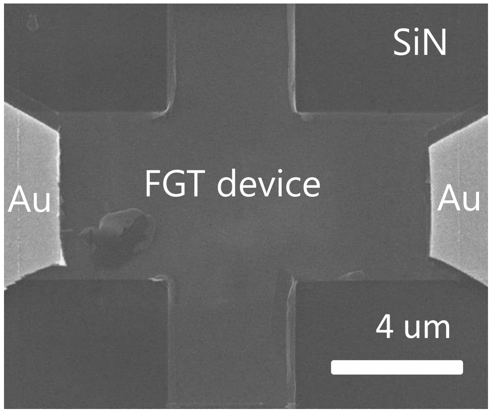




T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

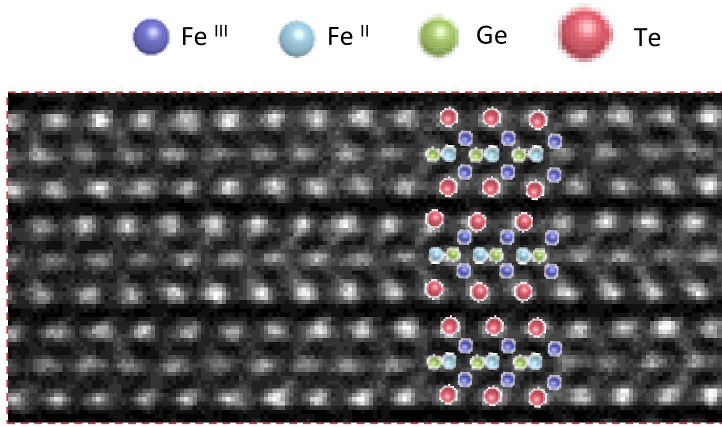
Crystal structure of Fe_3GeTe_2 device

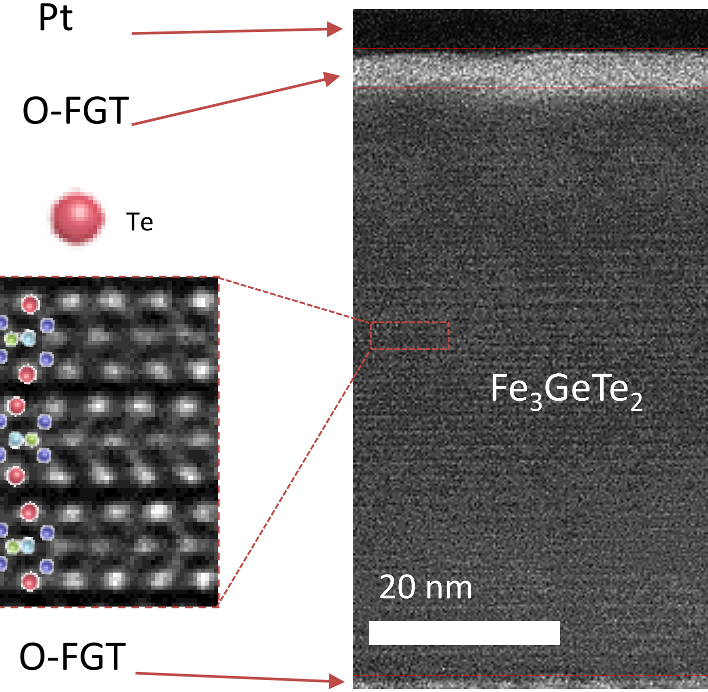
SEM Image of device



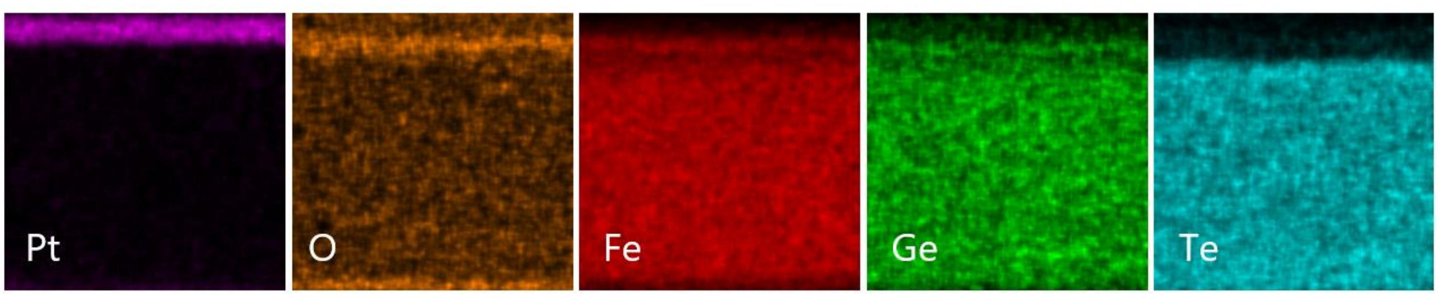
Hall bar device
on a Si_3N_4 membrane

Cross-sectional TEM





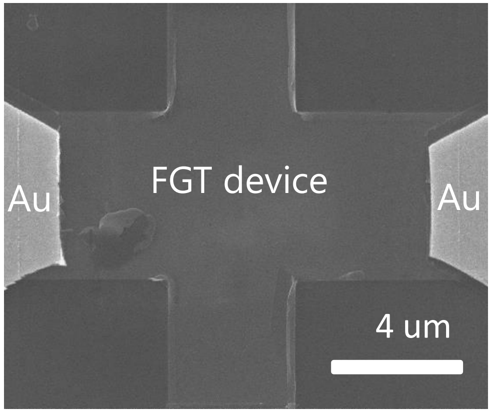
Element-mapping (STEM-EDS) of [O-FGT**|FGT|**O-FGT**|**Pt**]**



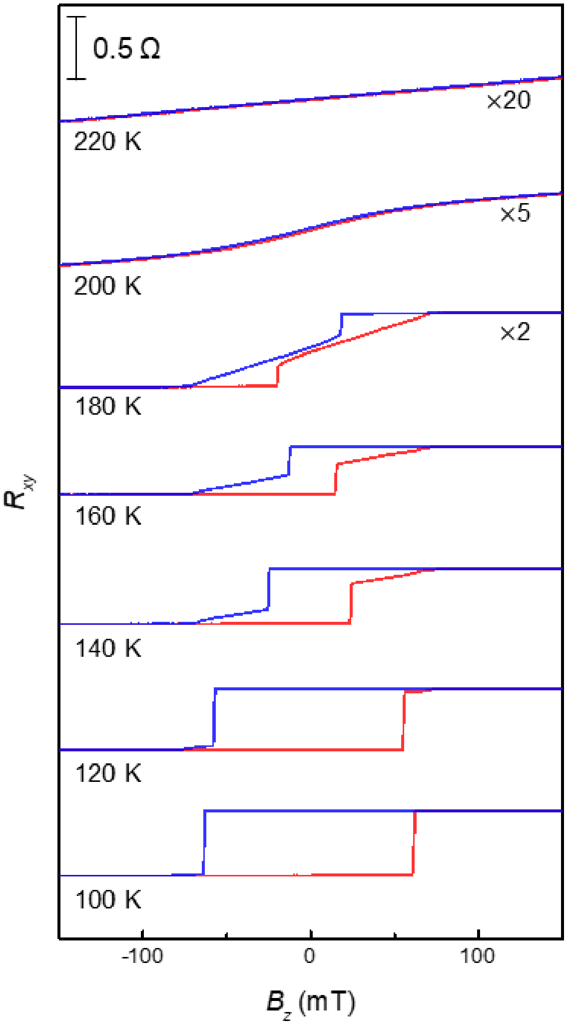
T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

Hall-measurement of Fe_3GeTe_2 device

SEM Image of device



Hall bar device
on a Si_3N_4 membrane

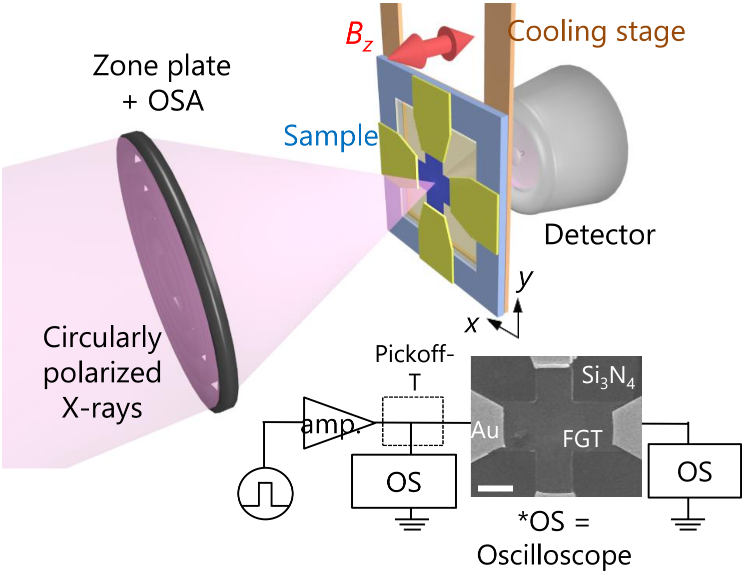


- Negligible ordinary Hall coefficient measured
- Stronger PMA at lower temperature (lower thermal fluctuation)

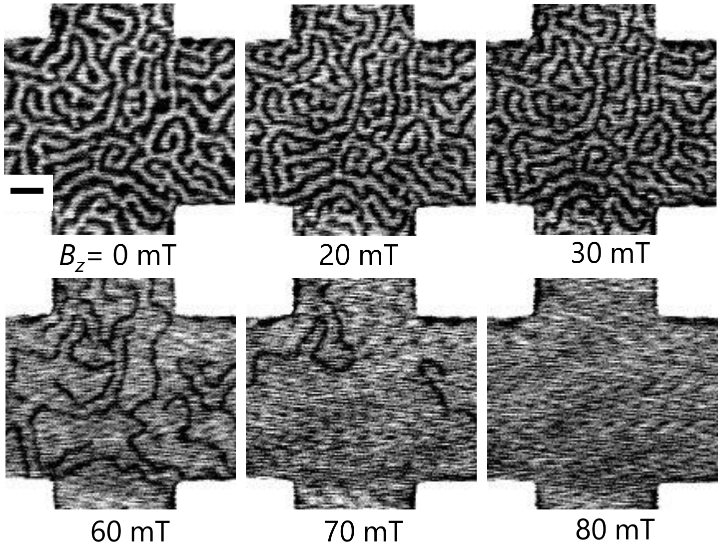
T.-E. Park, …, H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

X-ray imaging (STXM) on Fe_3GeTe_2

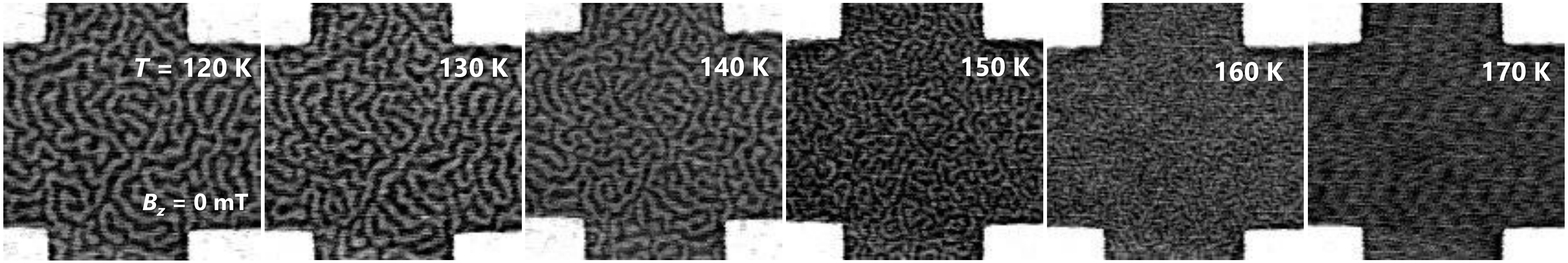
X-ray images at Fe-edge (120K)



Imaging at Fe-edge, Black: down & White: up

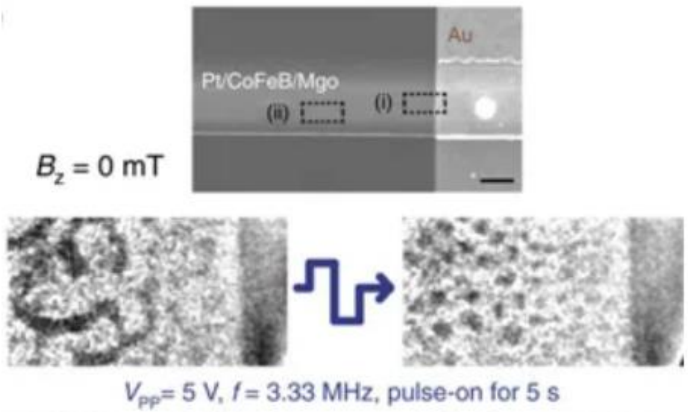


Temperature-dependent domain images using STXM



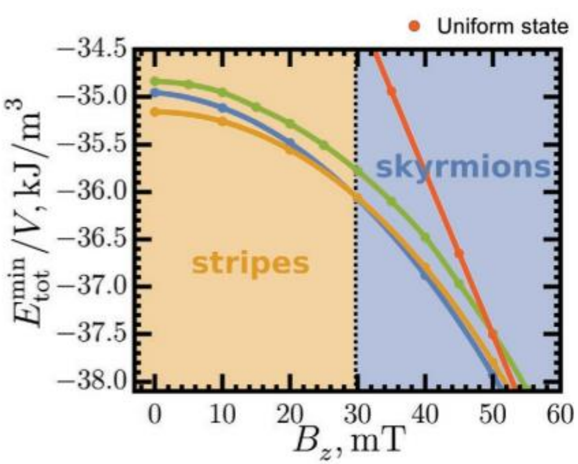
- Clear magnetic contrast upto ~ 150K
- Strong reduction in saturation magnetization yields thinner domains

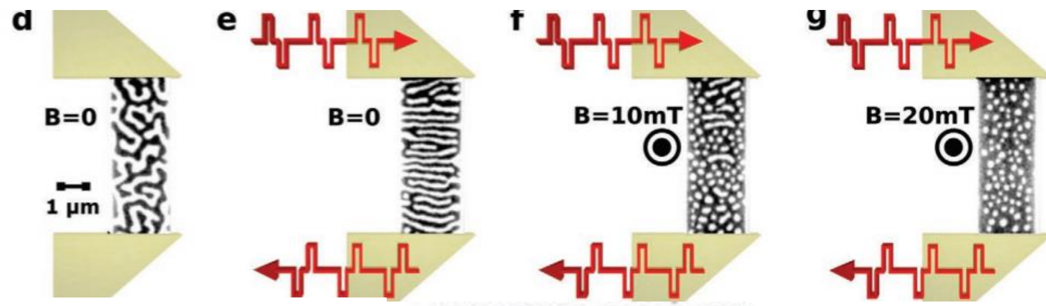
Skyrmion generation in Fe_3GeTe_2

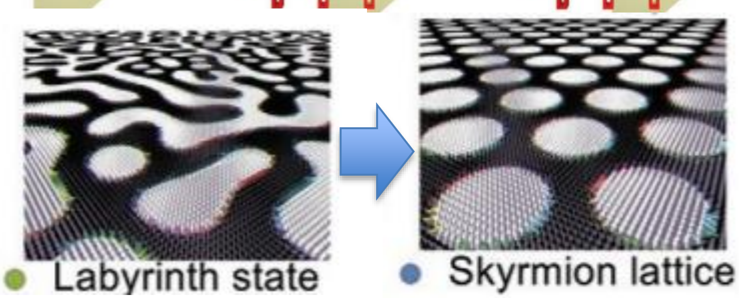


S. Woo* *et al.*, ***Nat. Commun.*** 8, 15573 (2017)

Current-induced skyrmion generation.
: Thermal transformation into skyrmions, by reducing the energy barrier towards the global skyrmionic ground state







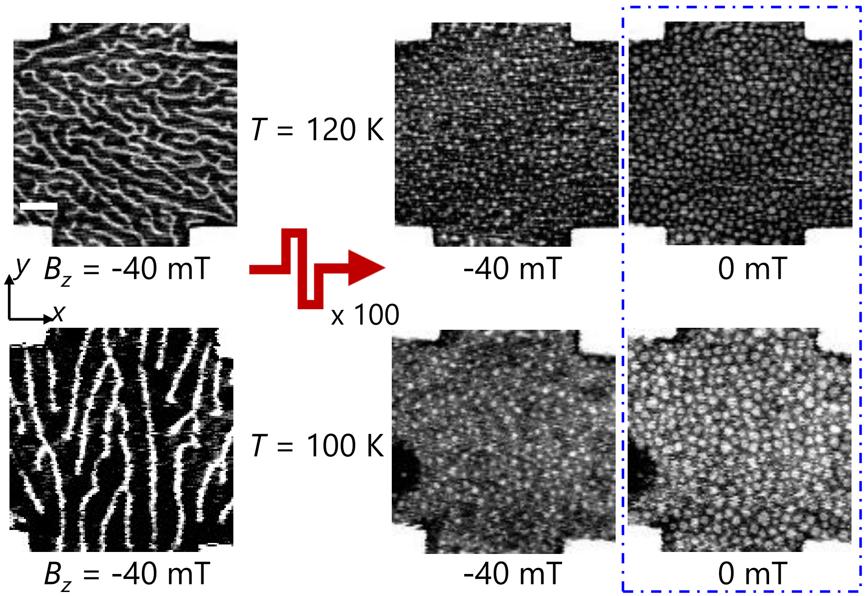
Current-induced skyrmion generation

I. Lemesh *et al.,* ***Adv. Mater.*** 30, 49 (2018)

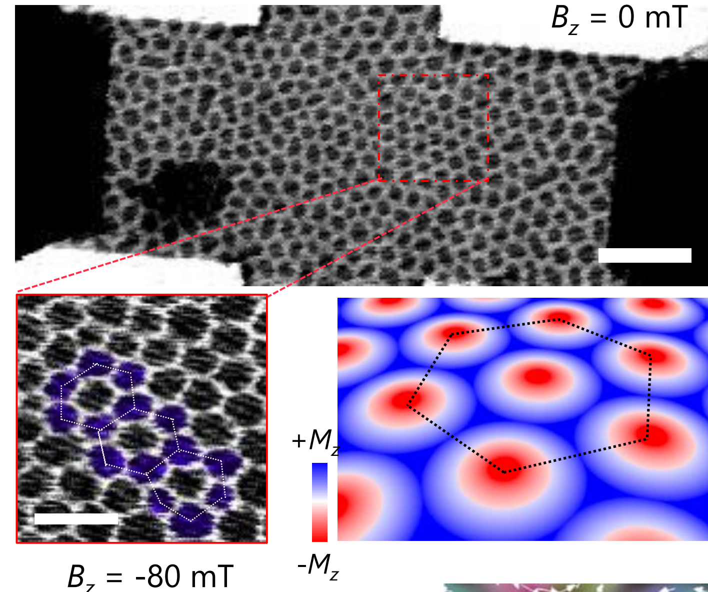
Skyrmion generation in Fe_3GeTe_2

Device 1. [O-FGT/**FGT**/O-FGT/Pt]

Current-induced skyrmion generation.

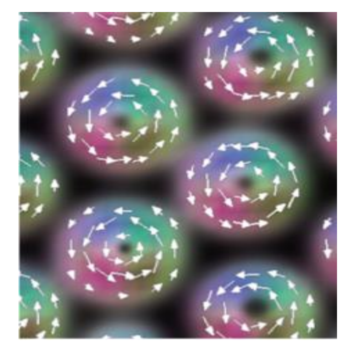


Field-induced relaxation.



Yu, X. Z. et al. ***Nature*** (2010)

SkX at low temperature & B20

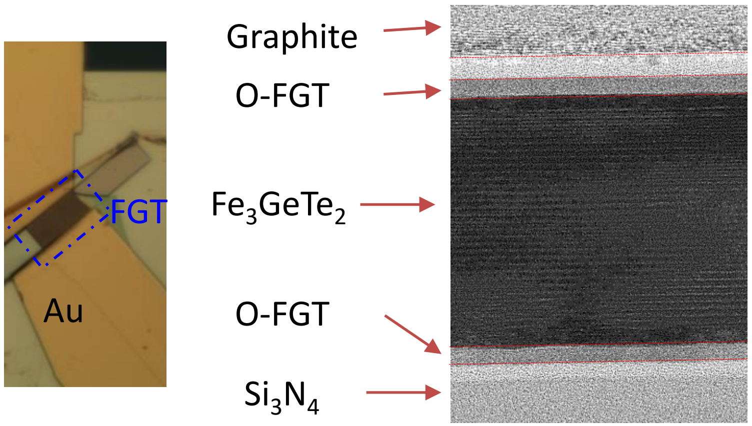


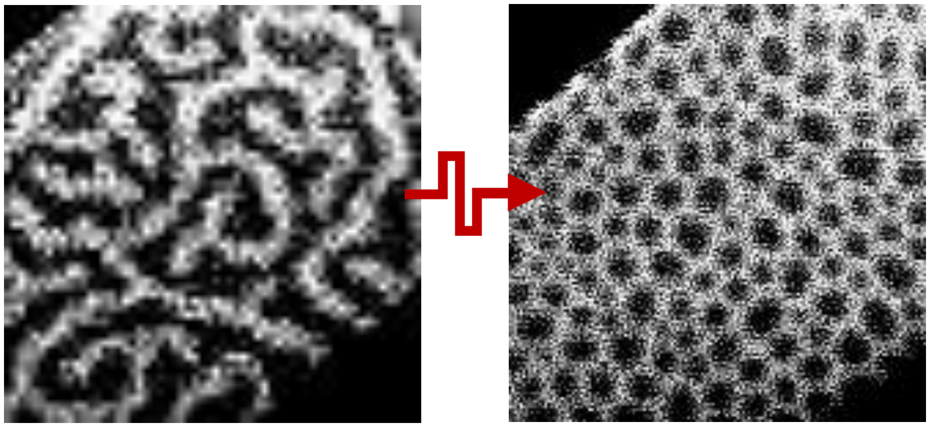
Step 1. Bipolar pulses to transformation from labyrich domains into multi-domains
Step 2. Alternative field sweep for domain relaxation

- T.-E. Park, …, H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

Skyrmion generation in Fe_3GeTe_2

Device 2. [O-FGT/**FGT**/O-FGT/**Graphite**]





B_z =0 mT, T =120 K

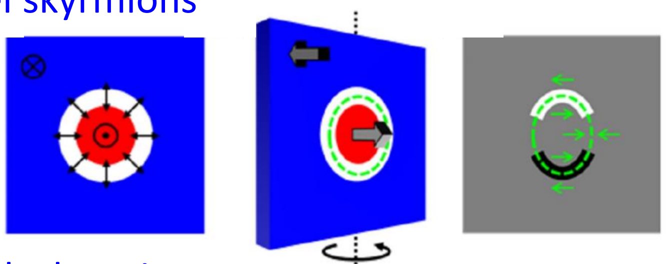
Graphite-capped device show the same qualitative domain transformation
: No DMI from Pt cap, then **maybe O-FGT?** or **observed domains are *achiral* bubbles?**

- T.-E. Park, …, H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

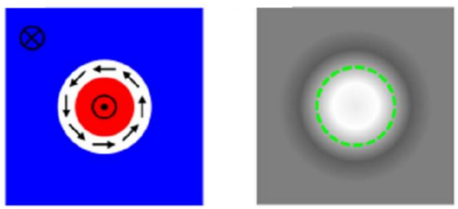
Magnetic skyrmions in Fe_3GeTe_2

Direct skyrmion observation using Lorentz-TEM

Néel skyrmions

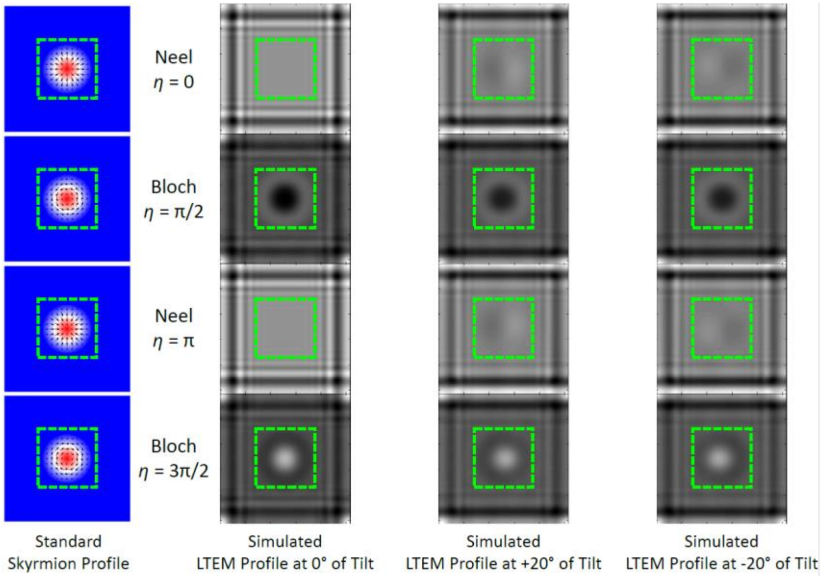


Bloch skyrmions

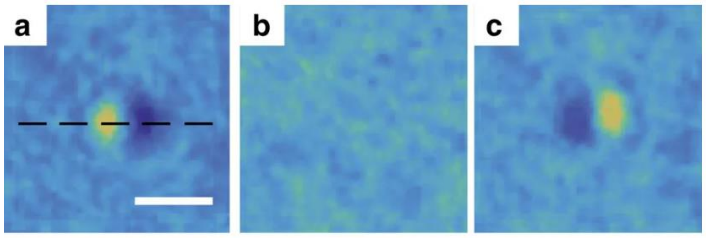


S. McVitie *et al.*, ***Sci. Rep.*** *8*, 5703 (2018)

No-tilt **+ tilt** **- tilt**



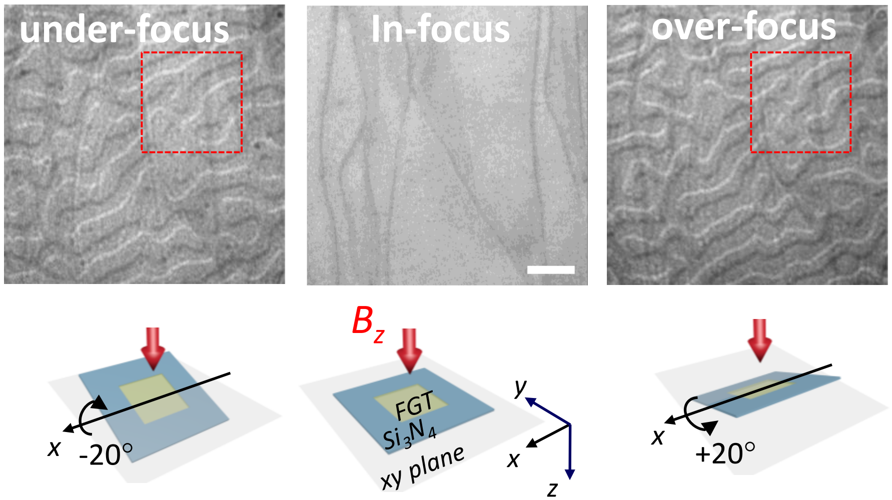
Experiment. Pt/Co/Pd multilayer

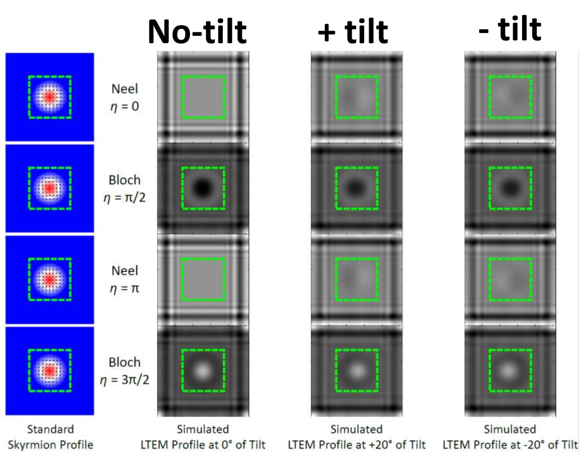


S. D. Pollard *et al.*, ***Nat. Comm.*** *8*, 14761 (2017)

Magnetic skyrmions in Fe_3GeTe_2

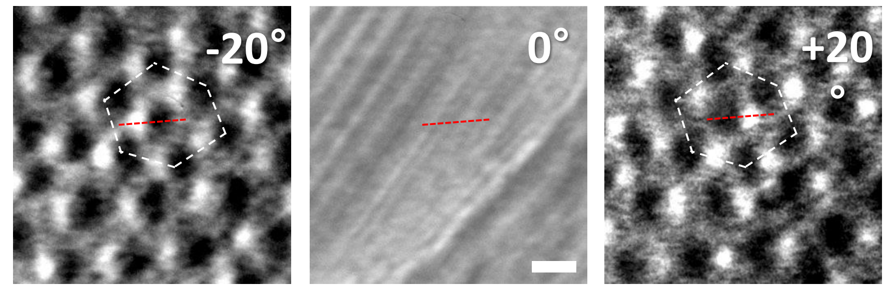
LTEM measurements on FGT device 1





T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

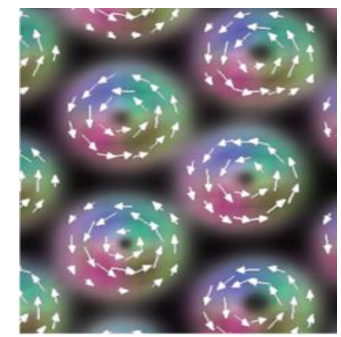
After field-cooling (FC)

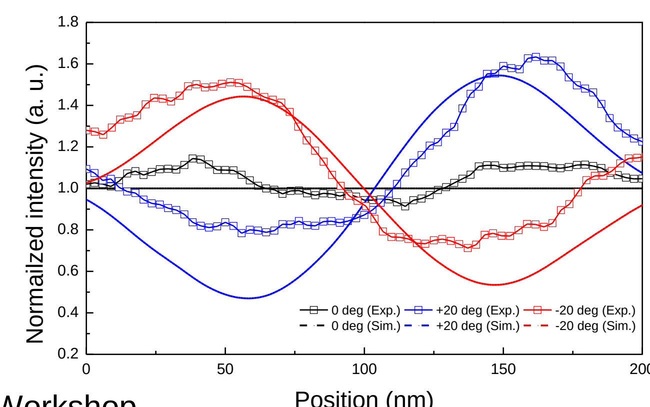


Chiral Neel-type skyrmions in our device!

Yu, X. Z. et al. ***Nature*** (2010)

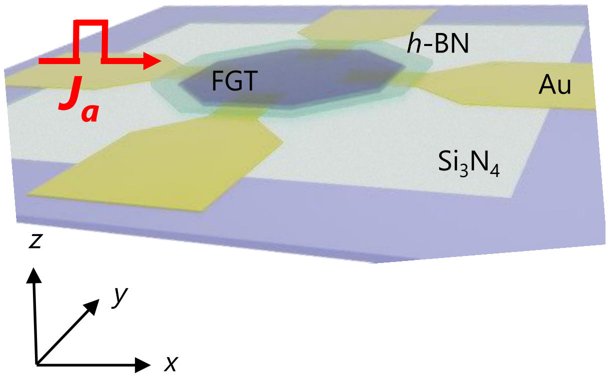
SkX at low temperature & B20

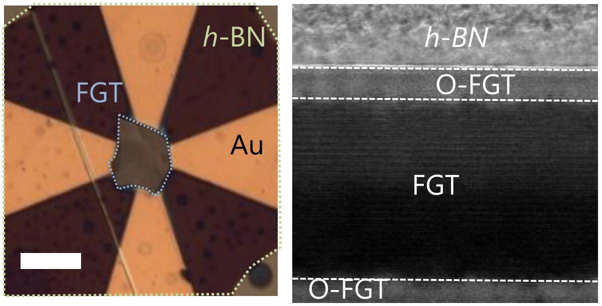




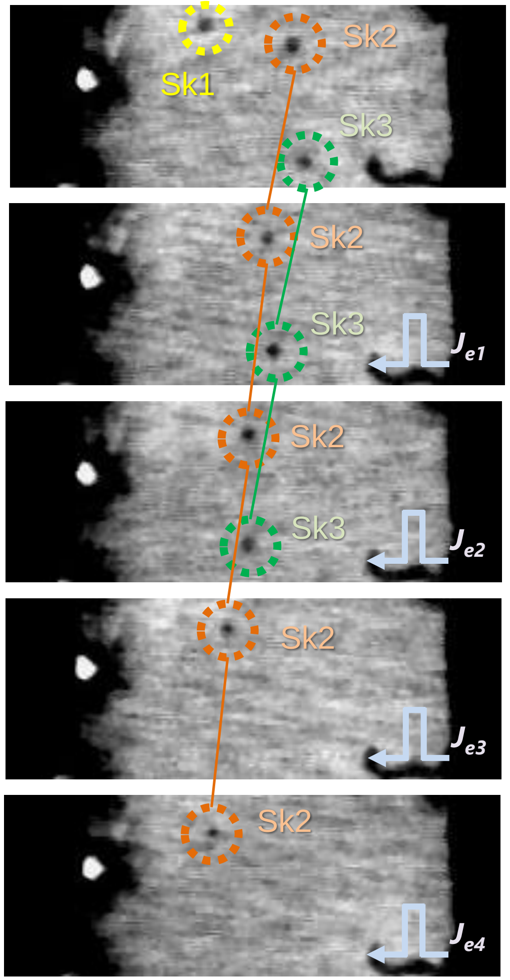
Current-driven motion of skyrmions

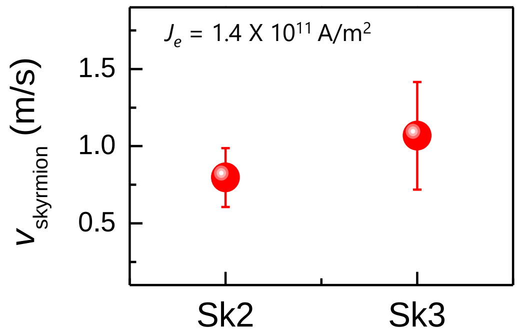
FGT/h-BN bilayer heterostructure



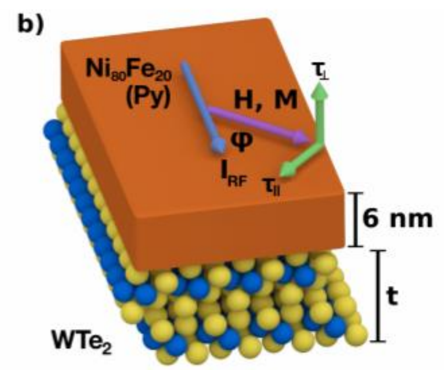


[O-FGT/**FGT**/O-FGT/h-BN]





Velocity is rather limited to ~ m/s
Skyrmion along *electron-flow*
: **STT-driven motion**



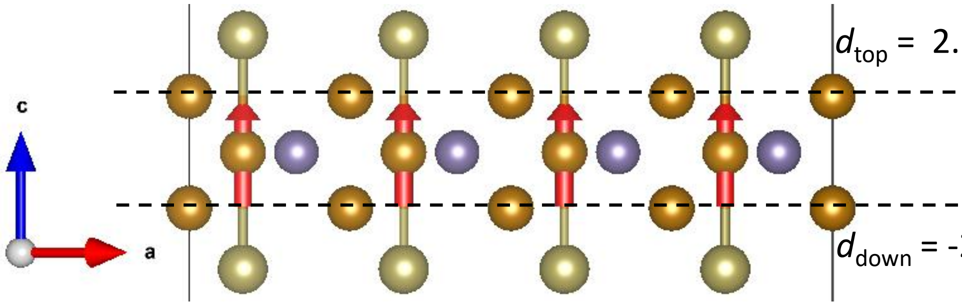
D. MacNeil *et al.*, ***Nat. Phys***. 13 (2017)

SOT-driven motion in WTe2/FGT could significantly enhance the skyrmion controllability

T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

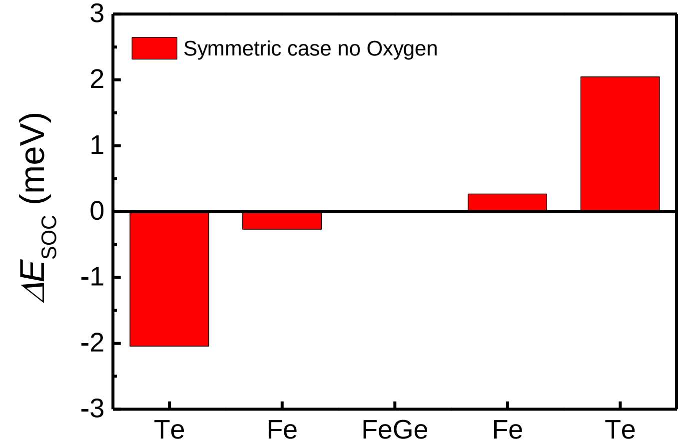
Origin of iDMI in Fe_3GeTe_2 heterostructure

No DMI from crystal symmetry!



d_{top} = 2.5033 meV

d_{down} = -2.5075 meV

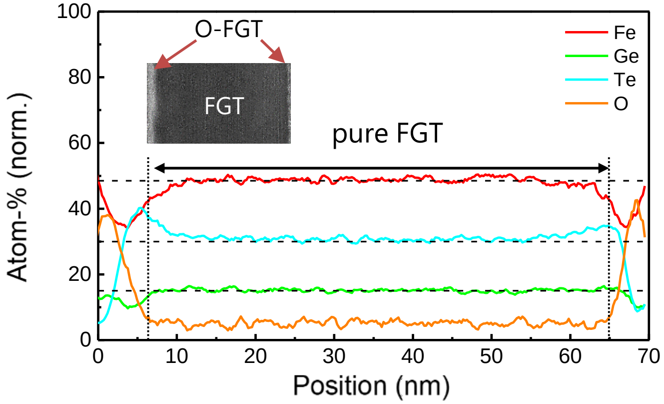


Sublayer	Path	Symmetry	Sublayer DMI	Cancellation	Cancelled w/
Fe1	Fe1-Te1-Fe1	$\mathbf{C_{3v}}$	Yes	Yes	Fe3-Te2-Fe3
Fe1	Fe1-Ge1-Fe1	$\mathbf{C_{3v}}$	Yes	Yes	Fe3-Ge1-Fe3
Fe2	Fe2-Ge1-Fe2	D_{3h}	No		
Fe3	Fe3-Te2-Fe3	$\mathbf{C_{3v}}$	Yes	Yes	Fe1-Te1-Fe1
Fe3	Fe3-Ge1-Fe3	$\mathbf{C_{3v}}$	Yes	Yes	Fe1-Ge1-Fe1
Fe1 & Fe2	Fe1-Te1-Fe2	C_{3v}	Yes	Yes	Fe2-Te2-Fe3
Fe1 & Fe2	Fe1-Ge1-Fe2	C_{3v}	Yes	Yes	Fe2-Ge1-Fe3
Fe2 & Fe3	Fe2-Te2-Fe3	C_{3v}	Yes	Yes	Fe1-Te1-Fe2
Fe2 & Fe3	Fe2-Ge1-Fe3	C_{3v}	Yes	Yes	Fe1-Ge1-Fe2
Fe1 & Fe3	Fe1-Ge1-Fe3	D_{3h}	No		

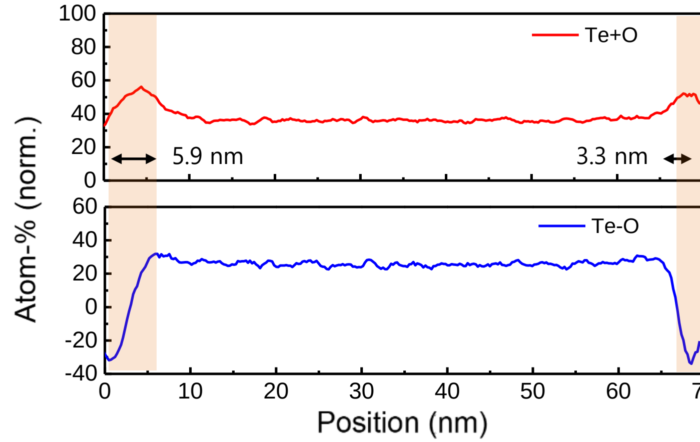
T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

iDMI from oxidized interfaces

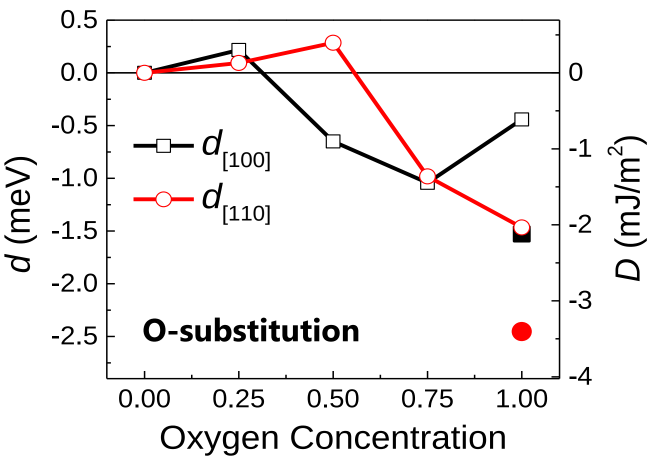
All elements.

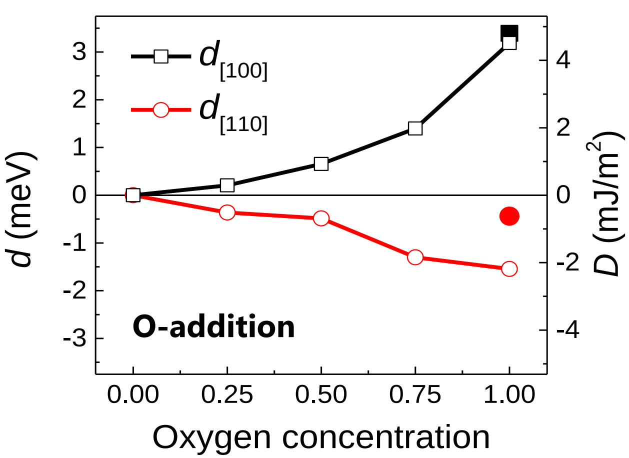


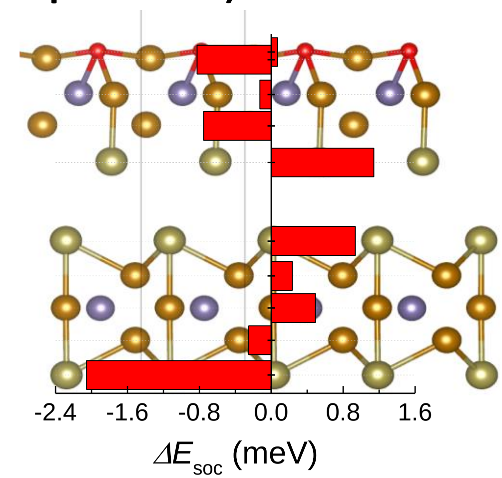
Te-O only.



Element mapping suggests probably i) O-substitution at Te sites & possible ii) O-addition





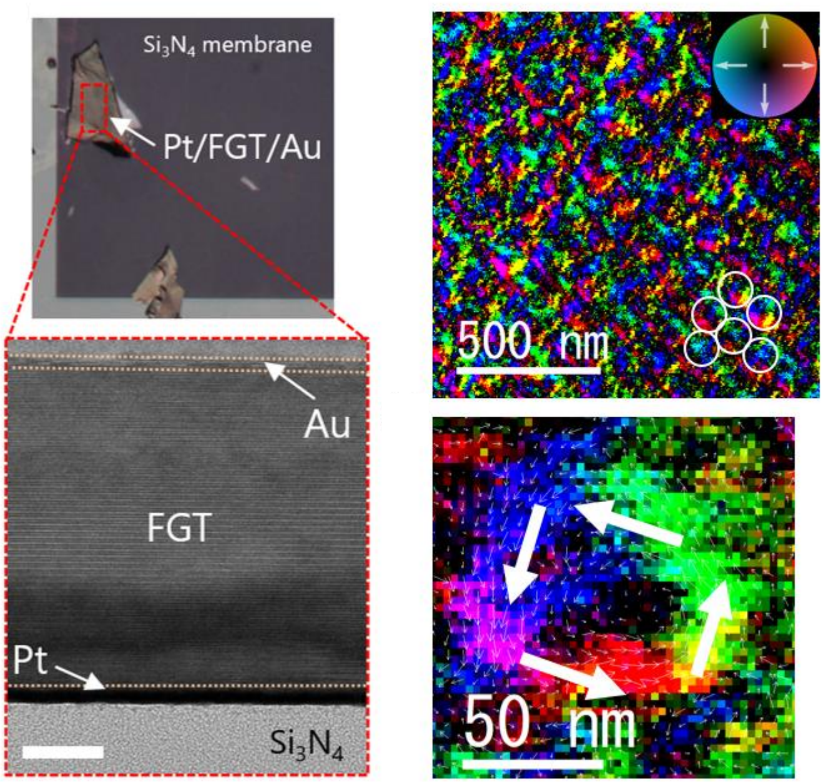


First principle calculations reveals strong emergence of DMI at Oxidized interfaces

What if we do not have O-FGT layer at interfaces?

iDMI from oxidized interfaces

LTEM observation on non-oxidized FGT (DPCM imaging)



Bloch-type magnetic textures and skyrmions are observed in FGT without oxidized interfaces

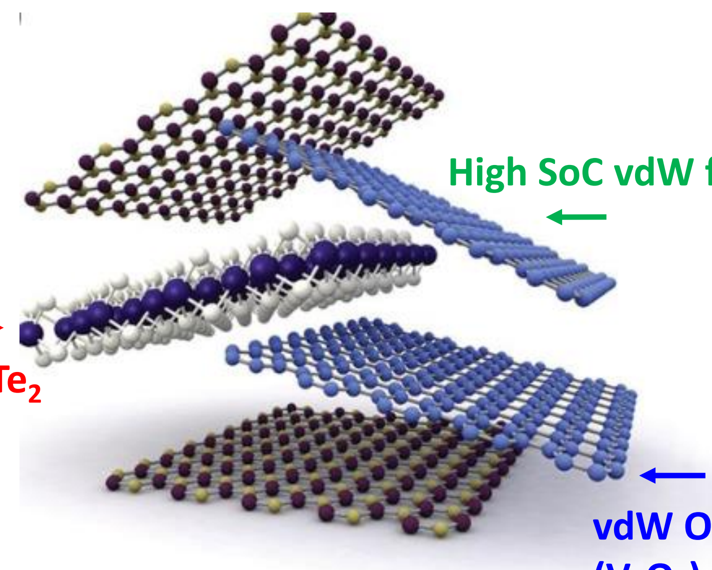
In FGT-heterostructures, we have demonstrated

1] Chiral Néel-type skyrmions & their lattice (SkX)

2] Current-driven skyrmion motion

3] Origin of i-DMI from oxidized layers

"Ideal vdW heterostructure for skyrmions"



[Image from ***Science*** 353, 6298 (2016)]

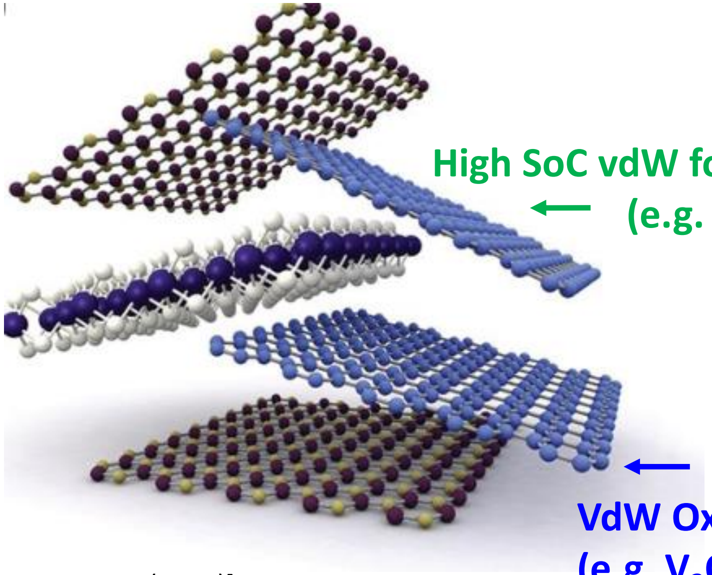
T.-E. Park, ..., H. Yang*, X. Yu*, S. Woo* ***arXiv:1907.01425*** (2019)

Conclusion

- We have observed Néel-type magnetic skyrmions, their lattice phase (SkX) and also demonstrated the current-induced motion of skyrmions in FGT-based heterostructures
- First-principle calculation with corroborating experiments reveal the presence of strong interfacial DMI at oxidized interfaces (FGT|O-FGT)
- Our study presents novel vdW heterostructures as a new skyrmion platform

More information: T.-E. Park *et al.*, ***arXiv:1907.01425* (2019)

"Ideal vdW heterostructure for skyrmions"



[Image from ***Science*** 353, 6298 (2016)]