An Introduction to Two-Dimensional Magnetic Materials

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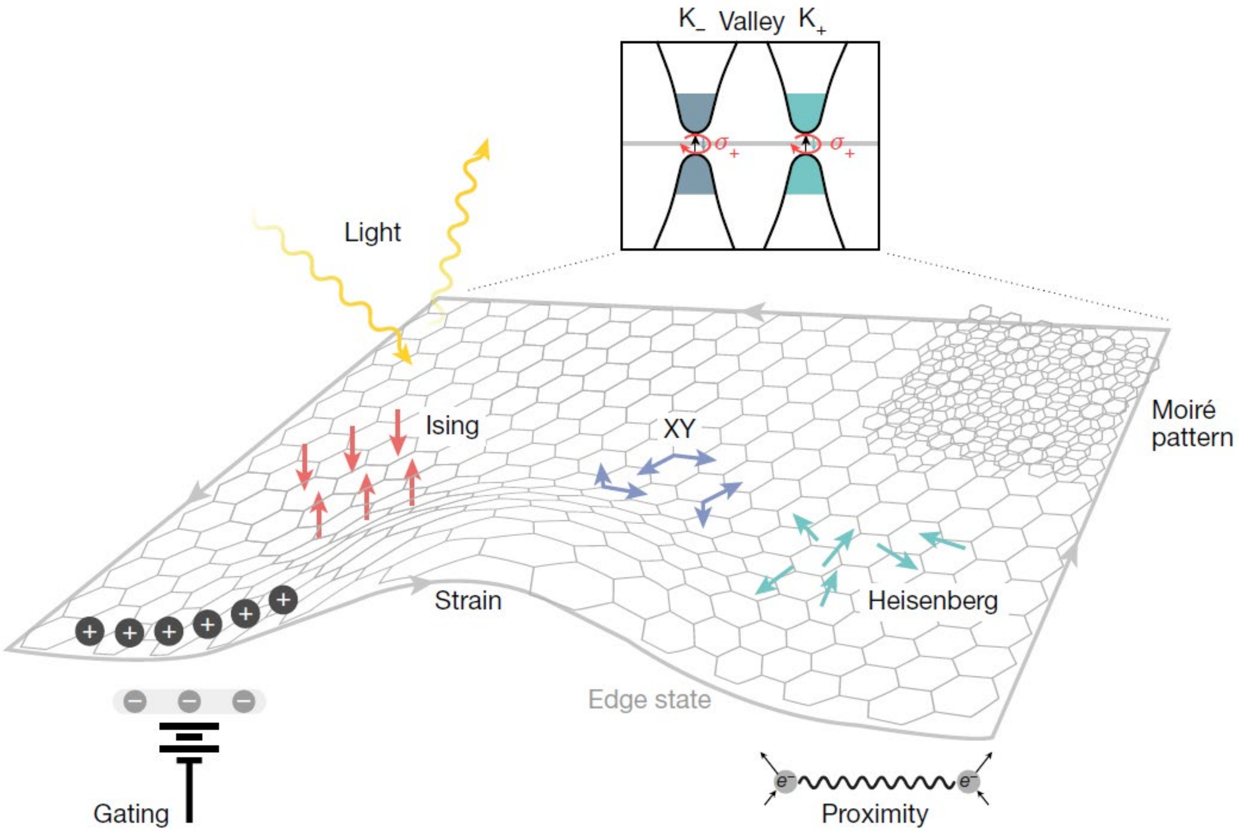
Magnetism in Two-Dimensions



- Show model Hamiltonians for magnetism
- Can be combined with other 2D materials
- Attractive to theoreticians (simple systems)

Some nice Reviews: Nature **563**, 47 (2018)
Nature Nanotech. **14**, 408 (2019)
Science **363**, 706 (2019)

Question

Did you have an introductory (or advanced) course in magnetism?

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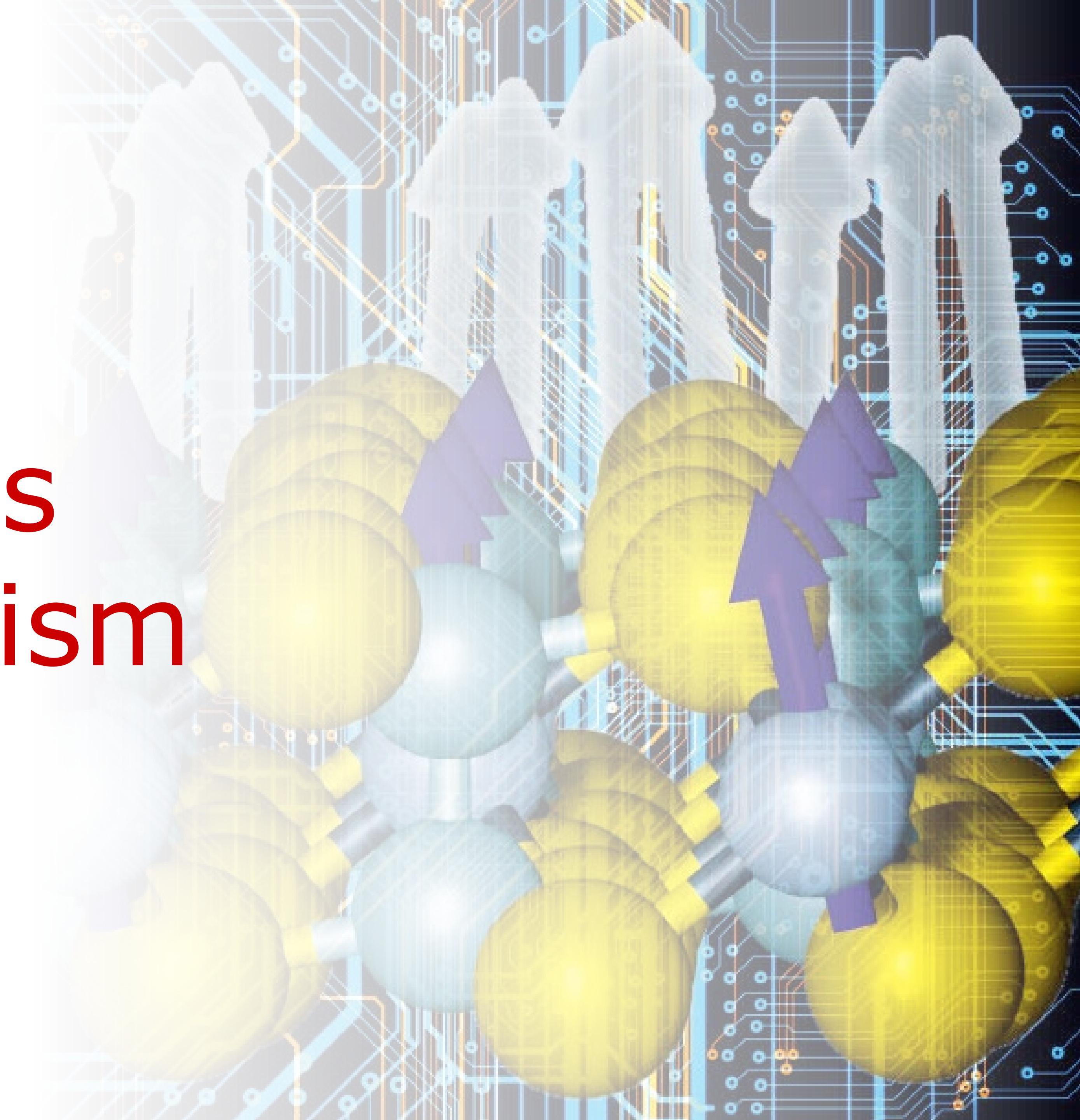
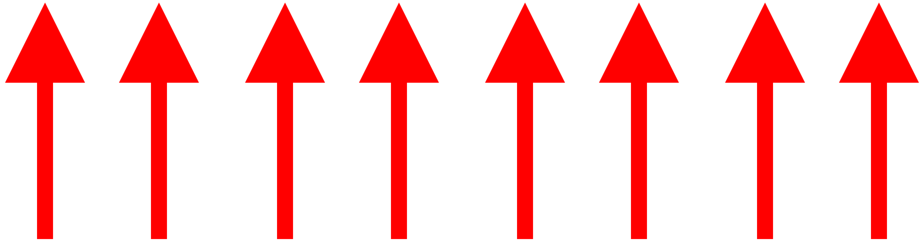
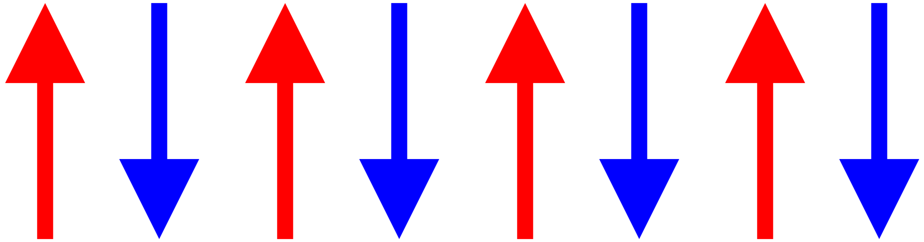
Fundamentals of (2D) Magnetism



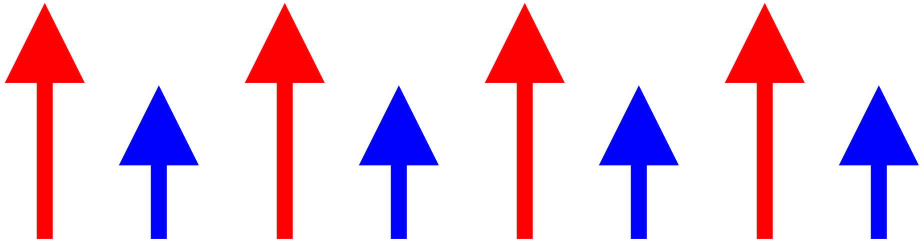
Recap: Magnetic Ordering



Ferromagnet



Antiferromagnet



Ferrimagnet

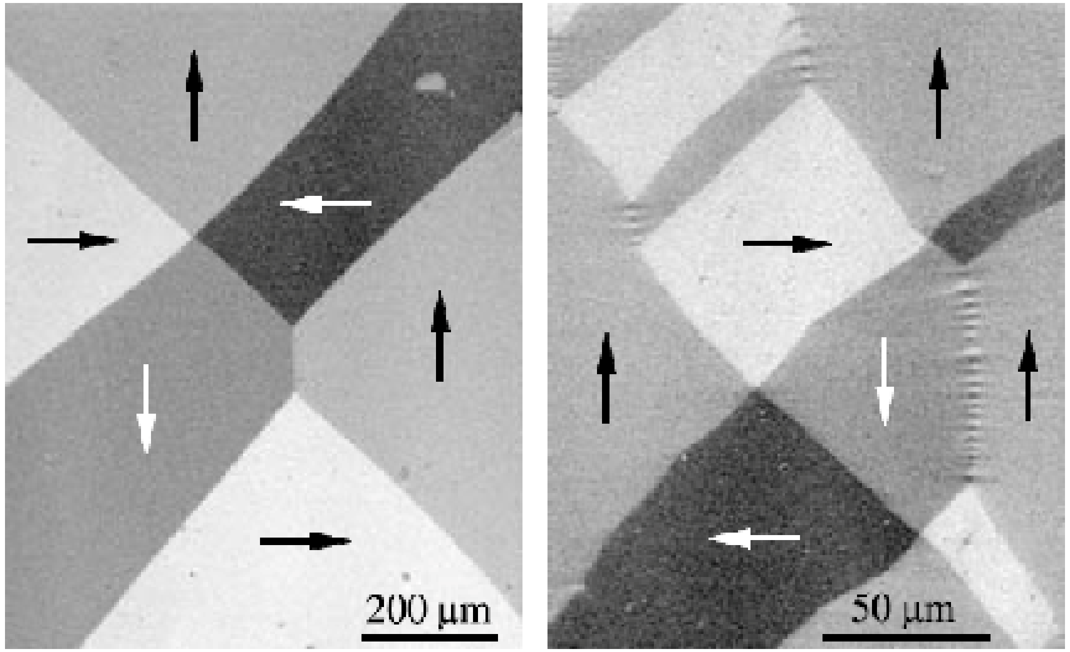
Ferromagnet

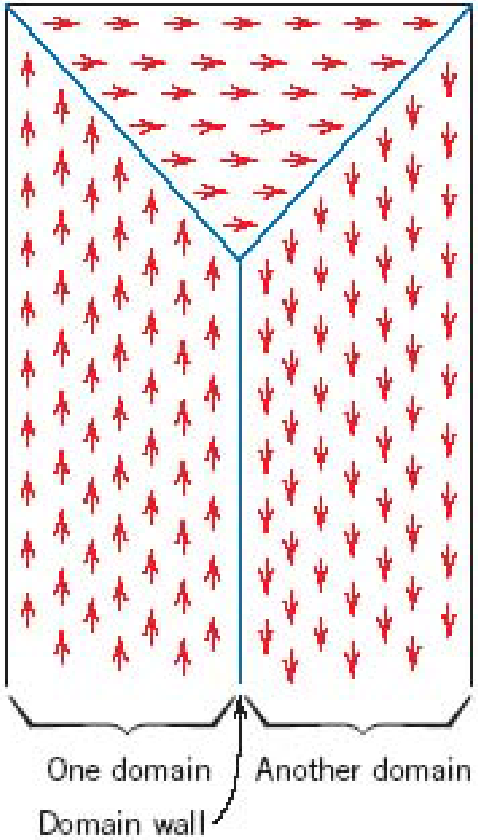
$M \neq 0$

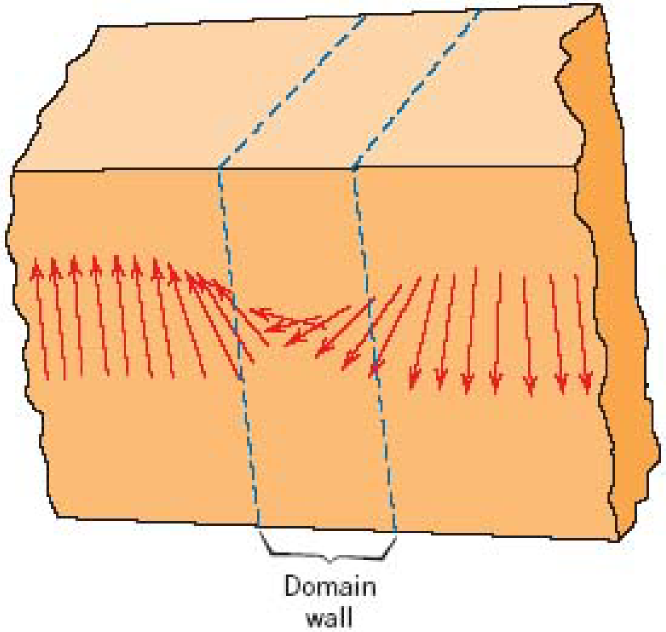
Antiferromagnet

$M = 0$

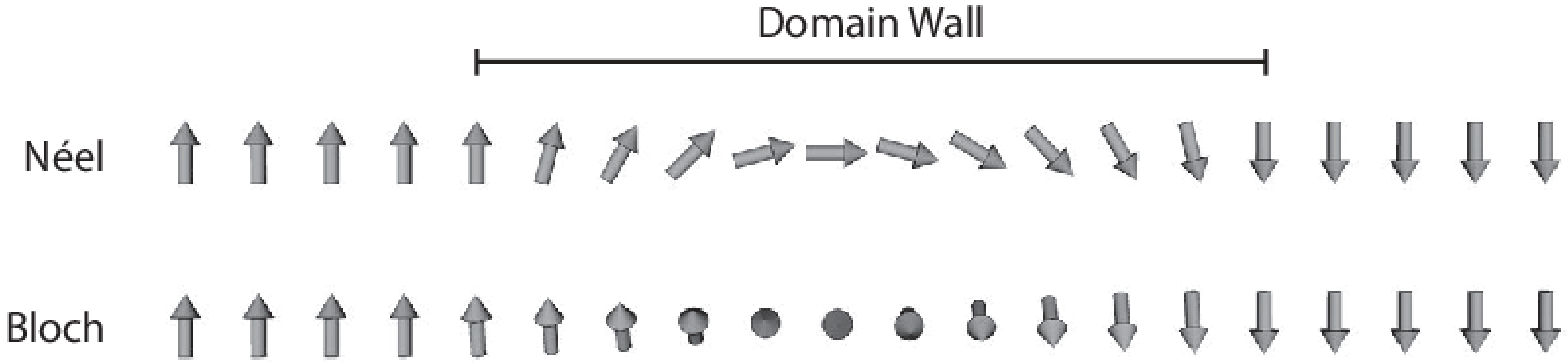
Magnetic Domains







Magnetic Domain Walls

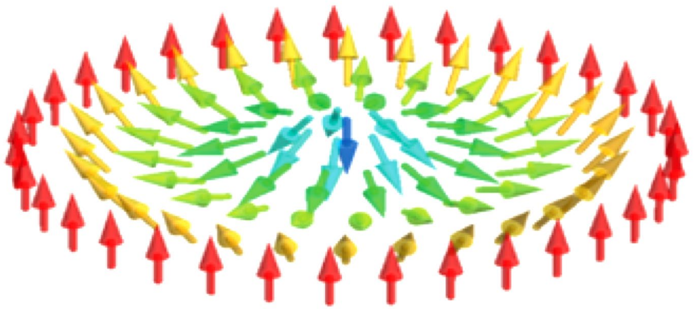


J. Franken, PhD Thesis (2014)

Important concept for topological magnetic structures,

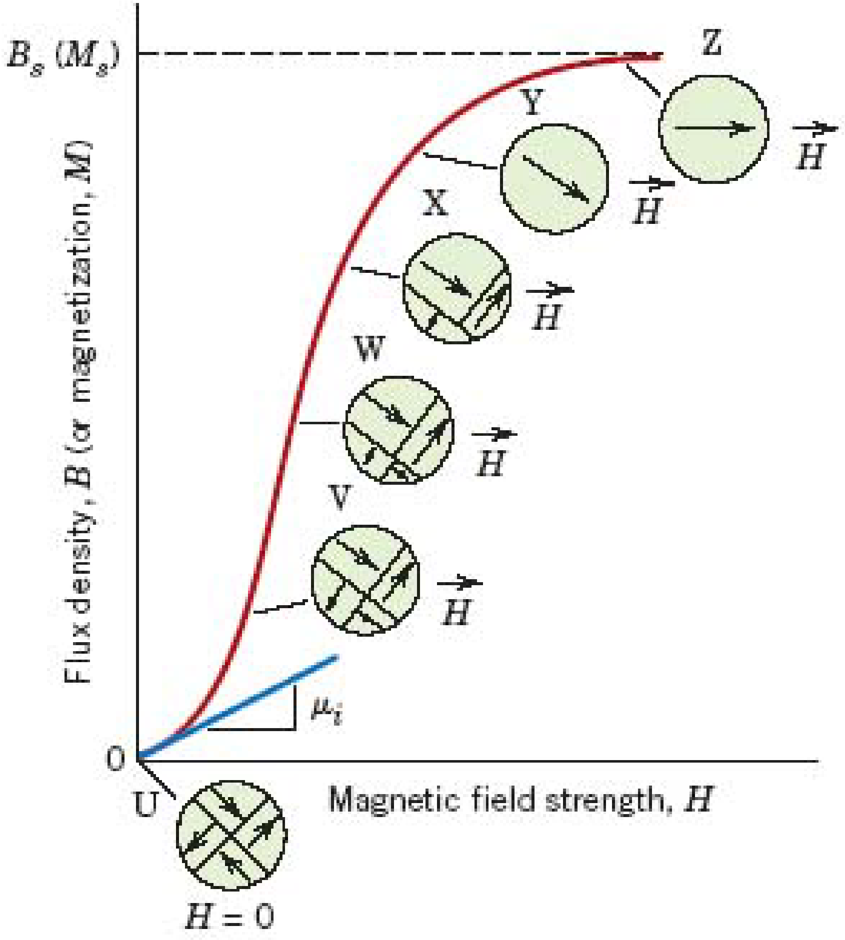
e.g. skyrmions and chiral spin spirals.

Néel-type skyrmion

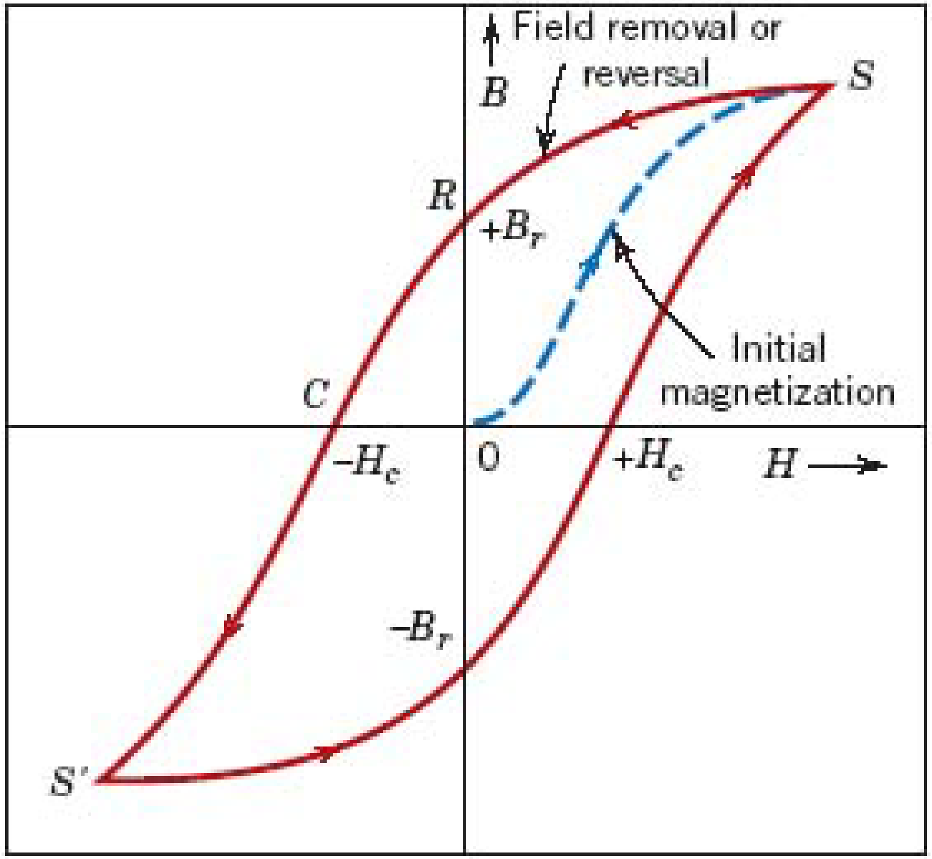


Fert et al., Nat. Rev. Mat. (2017)

M-H Loops



B_s (M_s)
Z
$\vec{H}$
Y
$\vec{H}$
X
$\vec{H}$
W
$\vec{H}$
V
$\vec{H}$
μ_i
0
U
$H = 0$
Magnetic field strength, H
Flux density, B (or magnetization, M)

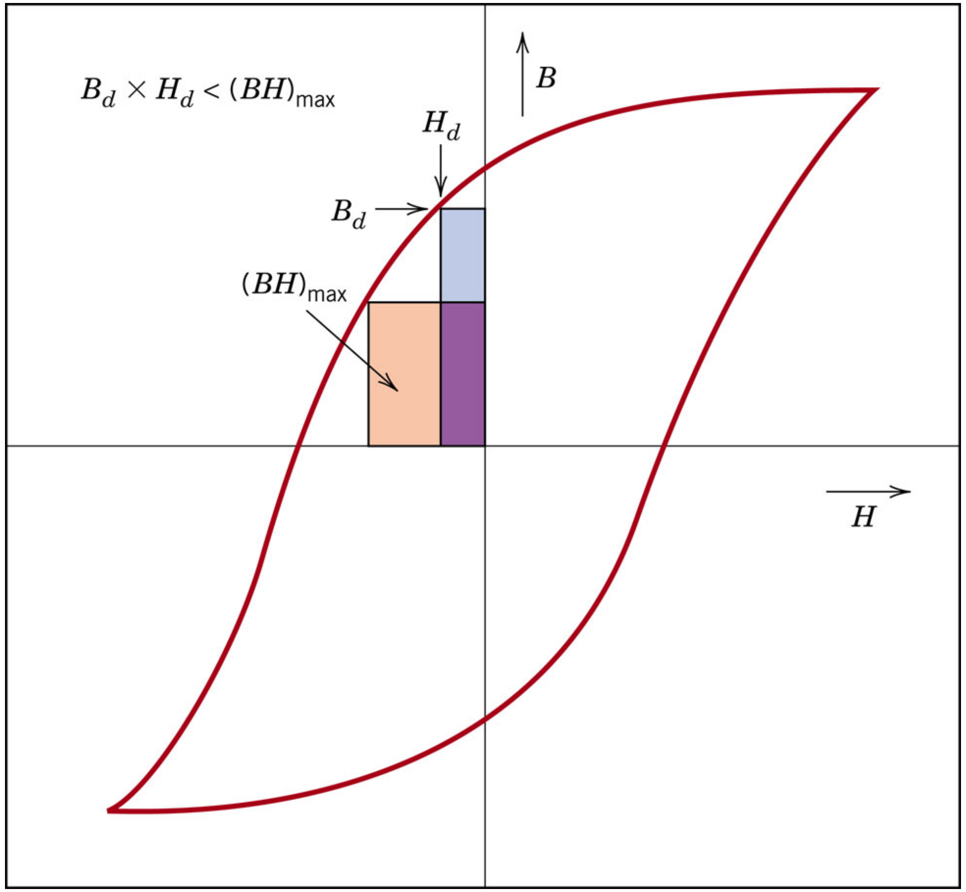


Field removal or reversal
B
S
R
$+B_r$
Initial magnetization
C
$-H_c$
0
$+H_c$
H
$-B_r$
S'

M-H Loops

The area enclosed by the hysteresis loop corresponds to energy loss associated with hysteresis;

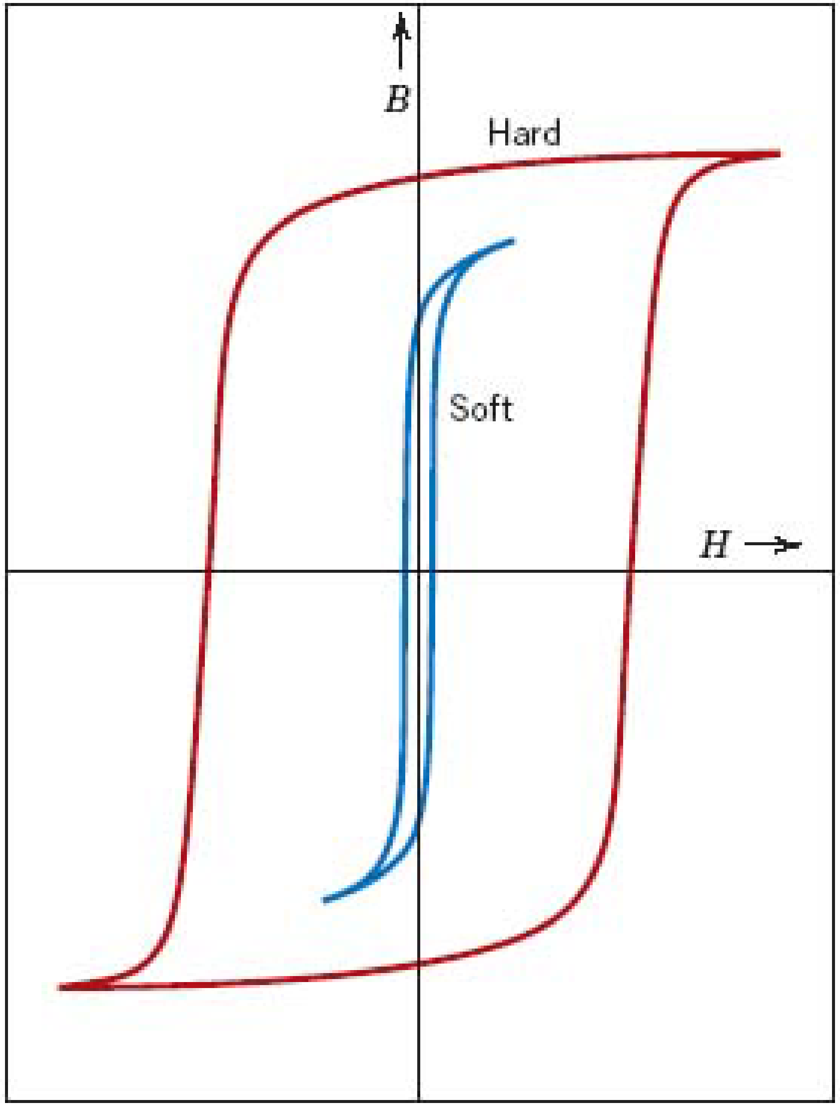
Moving, removing and introducing domain walls and reorienting magnetization cost energy!



Hard and soft magnets

A magnet having a hysteresis loop enclosing a large or small area is called hard or soft, respectively.

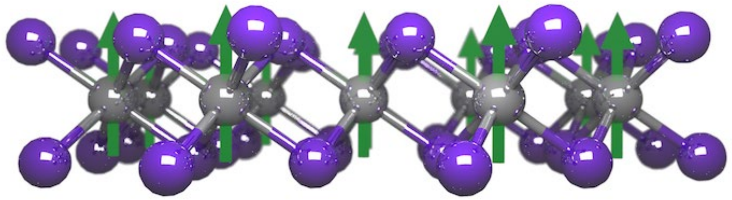
For applications the difference is particularly in the coercivity, because both saturation and remnant magnetism are (usually) desired large.



Magnetic Layered Materials

CrX_3

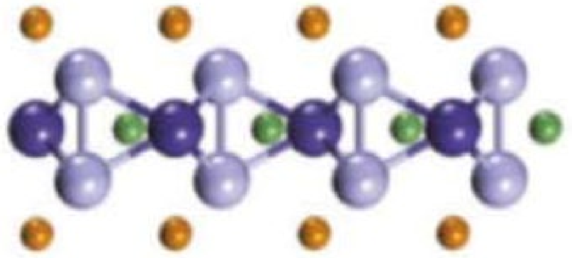
- Semiconducting (E_{BG} ~ 1.0 eV)
- Really Air Unstable
- T_c ~ 10's K
- (Anti)Ferromagnetic



- CrI_3
- $CrBr_3$
- $CrCl_3$

$M_xGe_yTe_z$

- Metals (Fe/Co/Ni-based) and Semiconducting (Cr-based)
- A little Air Unstable
- T_c ~ 100's K
- Ferromagnetic (usually)

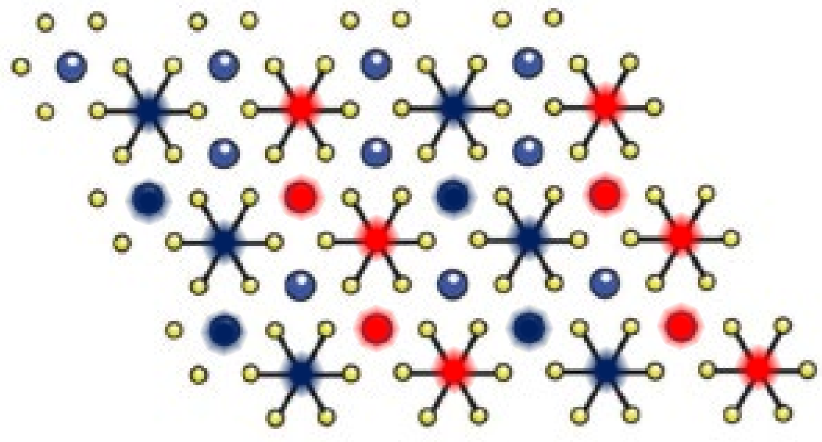


- Fe_xGeTe_2
- Ni_3GeTe_2
- $Cr_2Ge_2Te_6$
- $Co_xFe_{3-x}GeTe_2$
- …

Magnetic Layered Materials

$MPS_{3/4}$

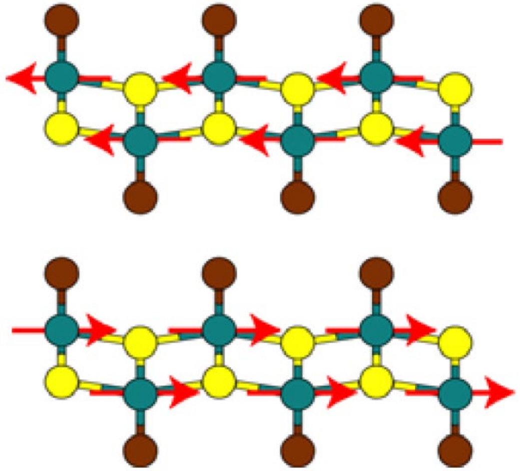
- Semiconductor (E_{BG} ~ 1-2 eV)
- Air Stable
- T_c ~ 100's K
- Antiferromagnetic



- $NiPS_3$
- $FePS_3$
- $CrPS_4$
- $MnPSe_3$
- …

CrSBr

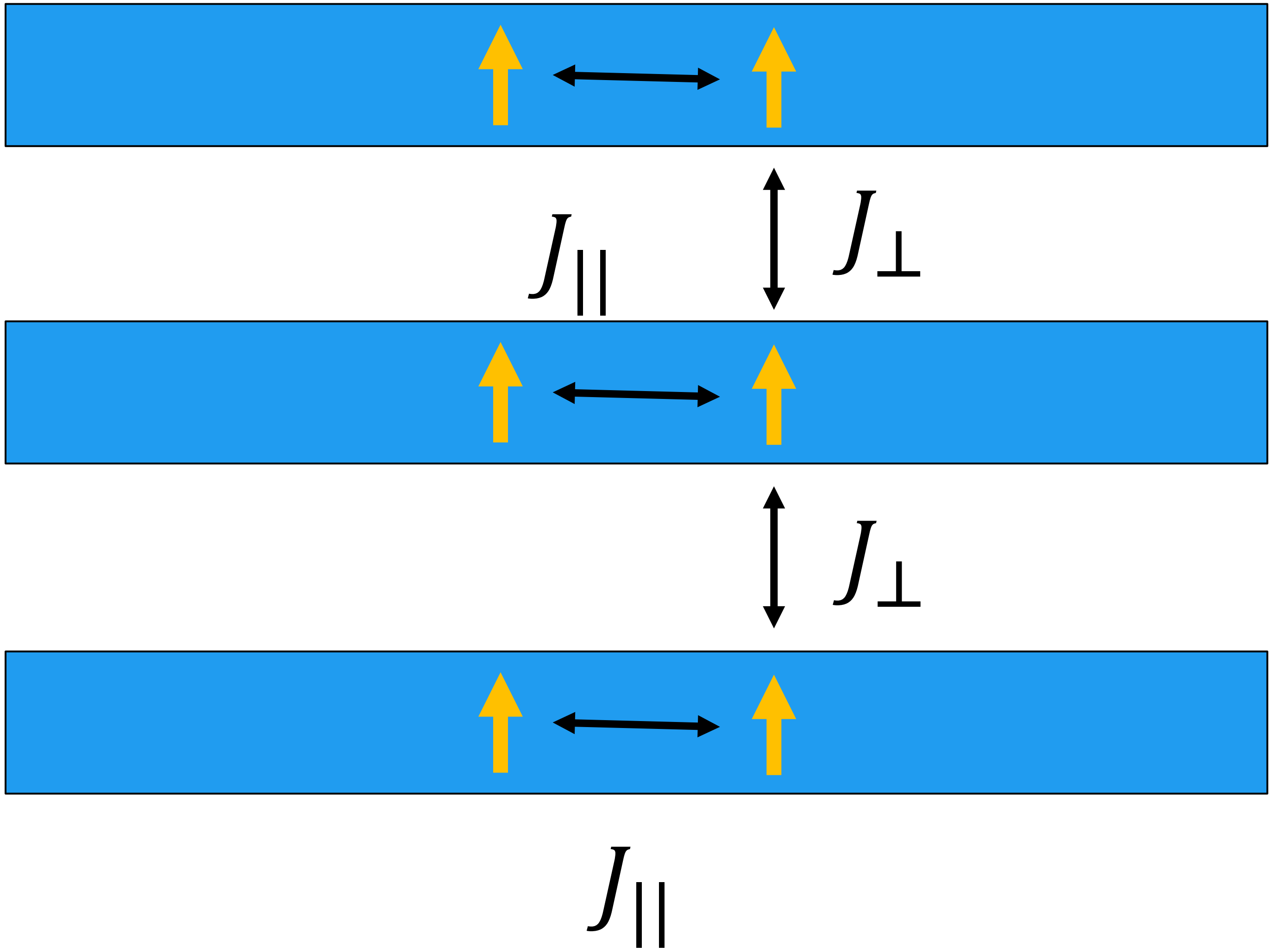
- Semiconductor (E_{BG} ~ 1.5 eV)
- Air Stable
- T_c ~ 150 K
- Antierromagnetic



Magnetic Exchange

$$H_S = -J_{ij}\left(\boldsymbol{S}_i \cdot \boldsymbol{S}_j\right)$$

Anisotropic Exchange



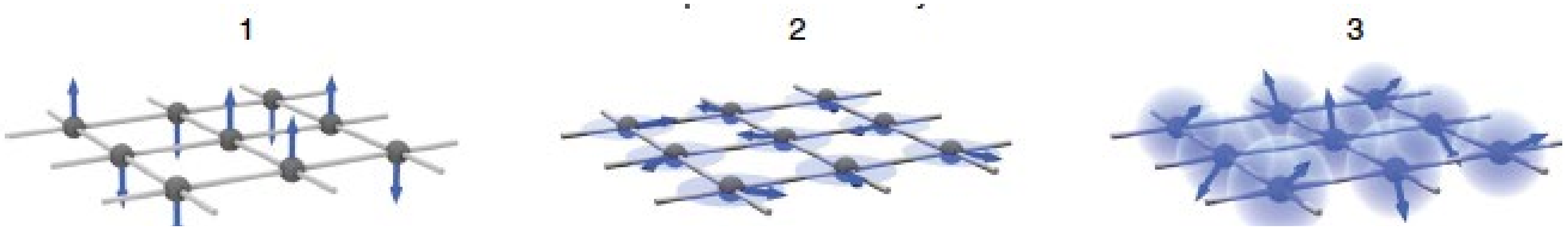
Typical
Van der Waals
Magnets

$$\boxed{J_\perp \approx 1 - 0.1\, J_{||}}$$

Spin Dimensionality

Dictated by the anisotropy term:

Strong out-of-plane anisotropy → 1D
Easy-plane anisotropy → 2D
Weak anisotropy → 3D



Fe_3GeTe_2
CrI_3
$FePS_3$

$NiPS_3$

$Cr_2Ge_2Te_6$

$$H_{Heis} = -J(\boldsymbol{S}_1 \cdot \boldsymbol{S}_2)$$

$$H_{Ising} = -J(S_1.S_2)$$

$$H_{XY} = -J_{xy}\left(S_1^x.S_2^x + S_1^y.S_2^y\right)$$

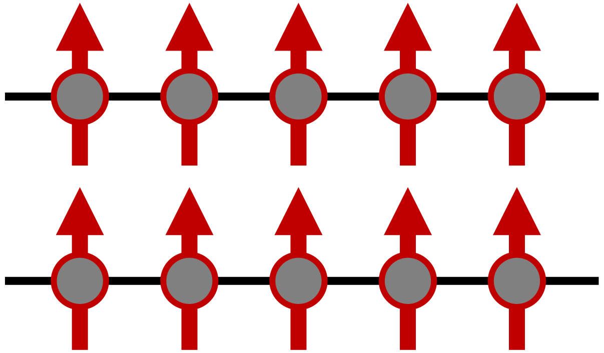
Gibertini et al., Nat. Nanotech. **14**, 408 (2019)

Magnetic Exchange

$J_\perp > 0$

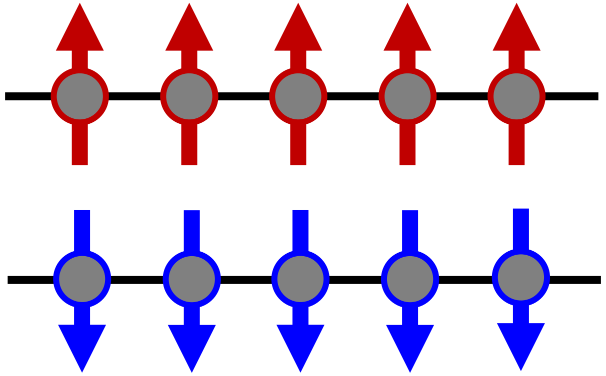
$J_\perp < 0$

$J_{||} > 0$



Fe_xGeTe_2
$Cr_2Ge_2Te_6$
$CrBr_3$

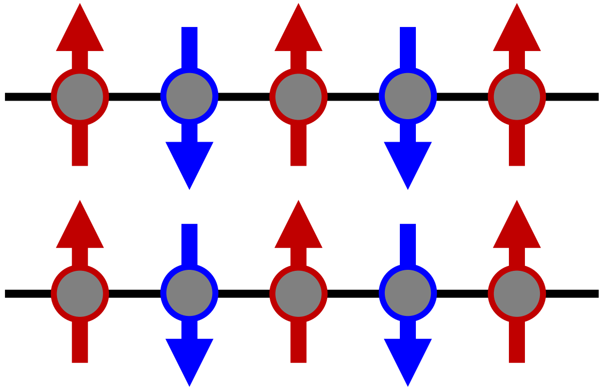
Ferromagnet



CrI_3
CrSBr
$CrPS_4$

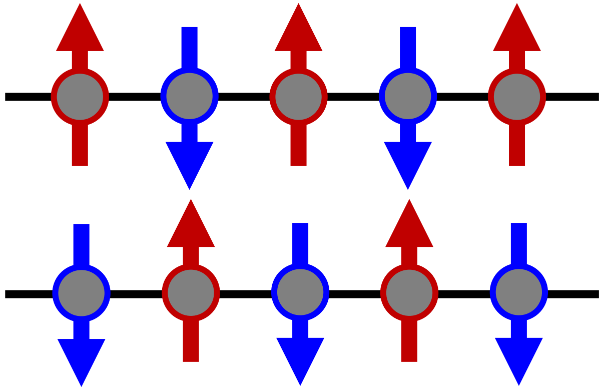
A-Type or Layered Antiferromagnet

$J_{||} < 0$



$NiPS_3$
$MnPS_3$

C-Type Antiferromagnet



$FePS_3$
$MnPSe_3$

G-Type Antiferromagnet

Mermin-Wagner Theorem

Mermin and Wagner, *PRL* **17**, 1133–1136 (1966)

States that two-dimensional with a continuous symmetry cannot be ordered, or
long-range order cannot exist in an *infinite* two-dimensional system in the absence of *anisotropy*.

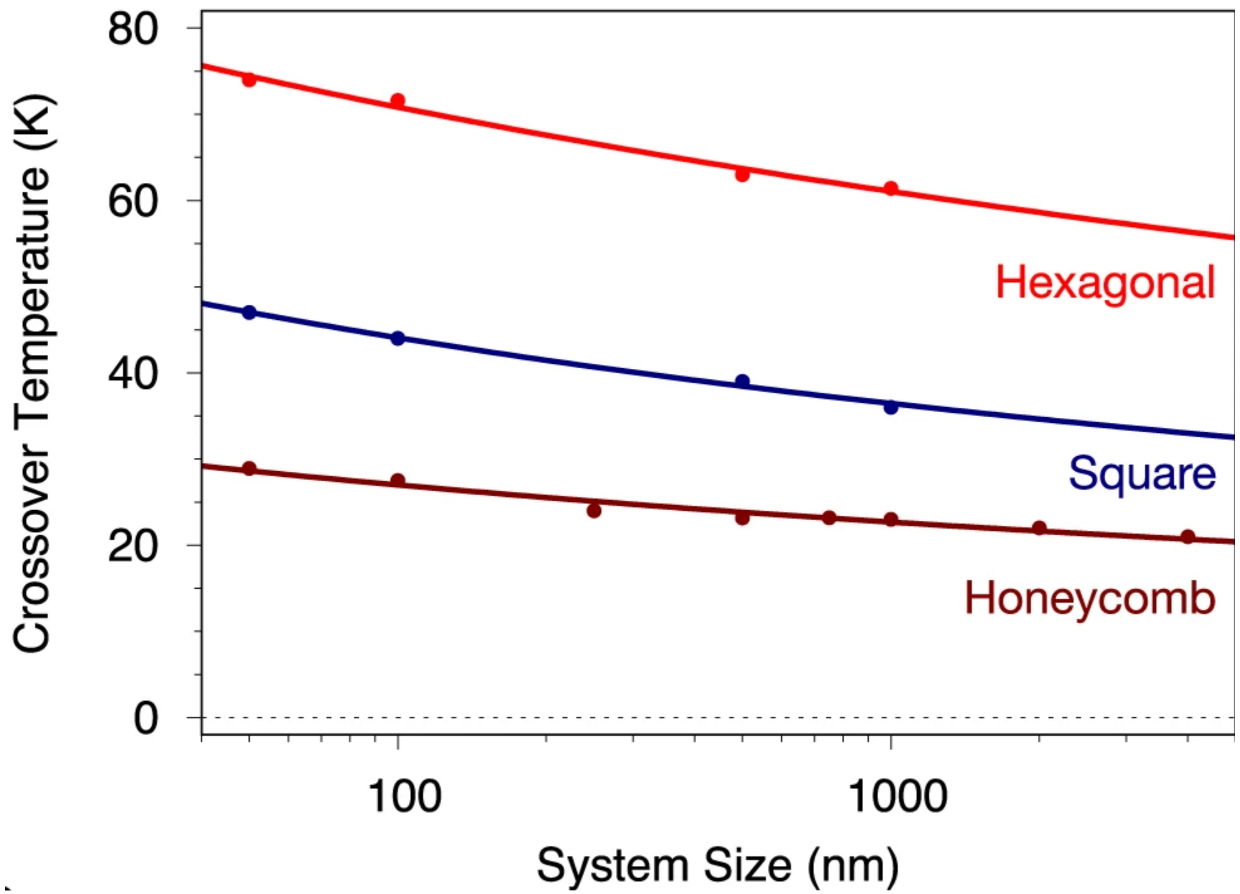
Can be calculated using the correlation between the directions of two spins:

$$c\left(\boldsymbol{r_i}-\boldsymbol{r_j}\right)=\left\langle e^{i\left(\phi(\boldsymbol{r_i})-\phi(\boldsymbol{r_j})\right)}\right\rangle$$

Or, alternatively, the average magnetization of the system:

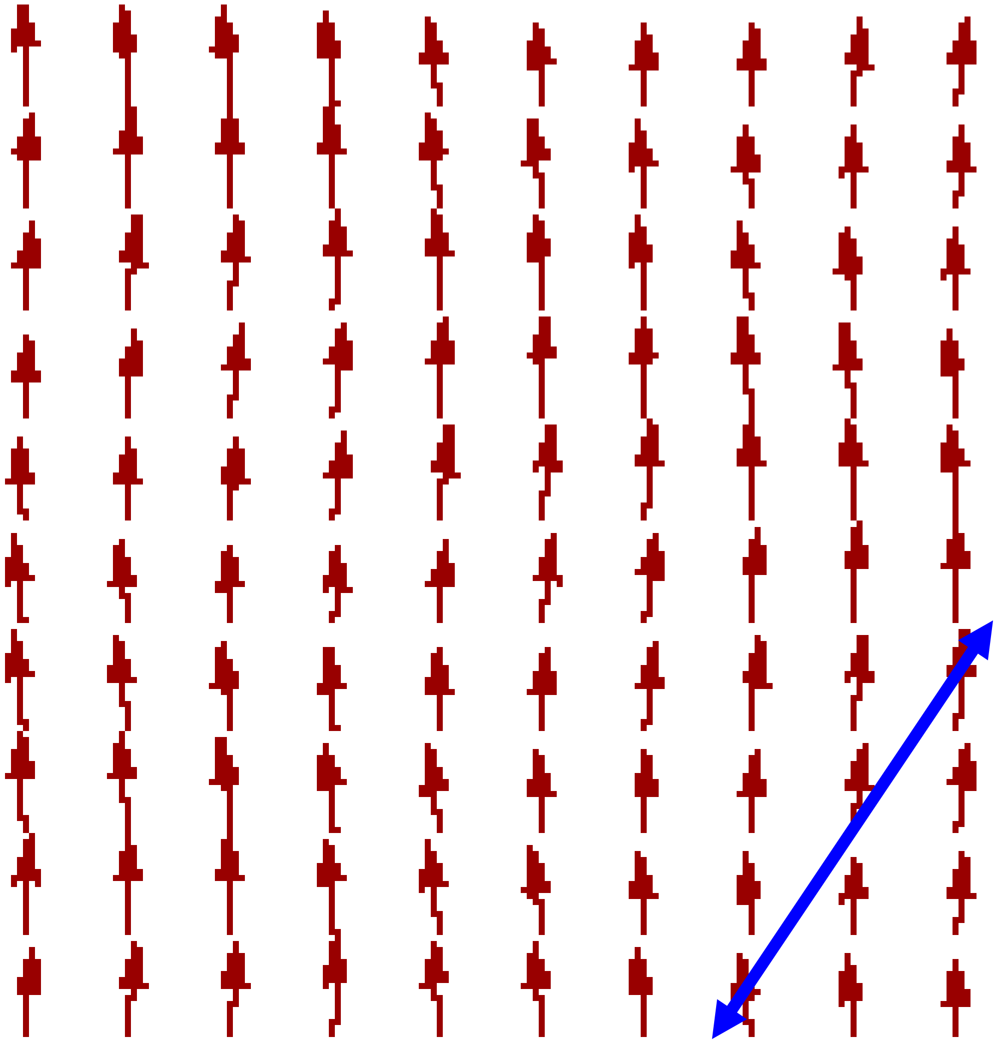
$$\langle|\boldsymbol{m}|\rangle=\left\langle\sqrt{\left(\frac{1}{N}\sum_{i}\boldsymbol{S_i}\right)^2}\right\rangle$$

Finite Size Effects



Jenkins et al., *Nat. Comm.* **13**, 6917 (2022)

A magnetic dance: spin waves (magnons)



$$\lambda = \frac{2\pi}{k}$$

Density of States

How does the density of states depend on energy for a 2D system?

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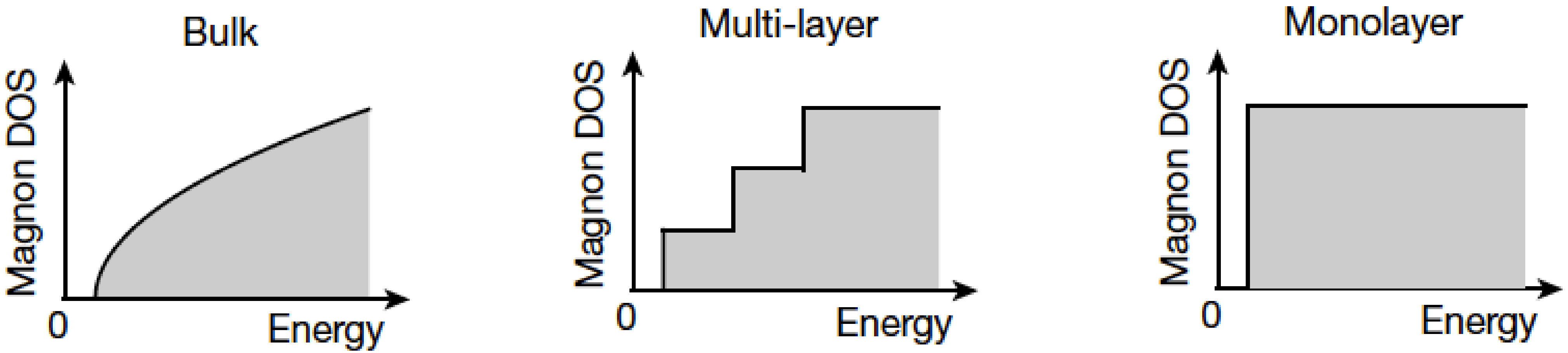
A – $g \propto E^{1/2}$

B – $g \propto E^{3/2}$

C – $g \propto constant$

D – $g \propto E^{-1/2}$

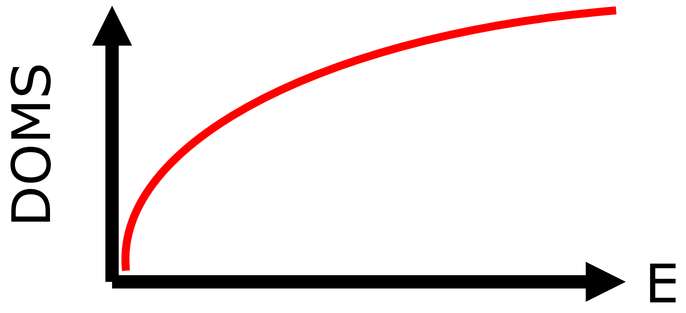
Effect of Dimensionality

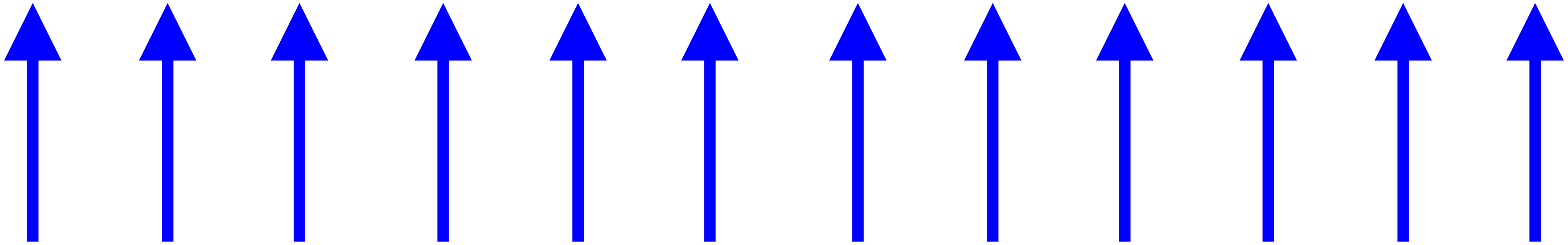


Gong et al., Science **363**, 6428 (2019)

Effect of Dimensionality

3D

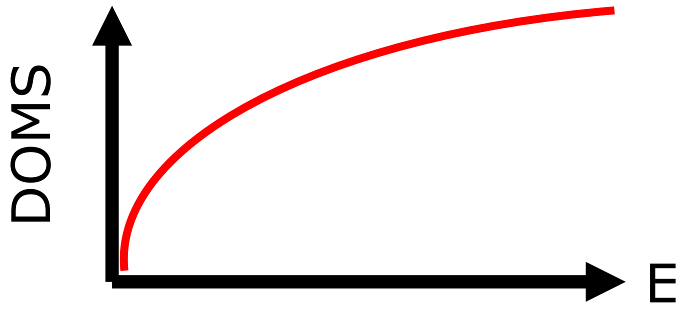


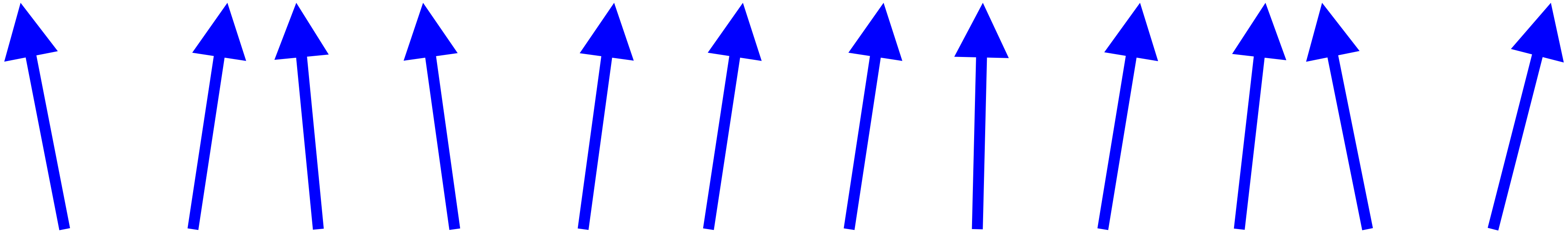


T = 0 K

Effect of Dimensionality

3D

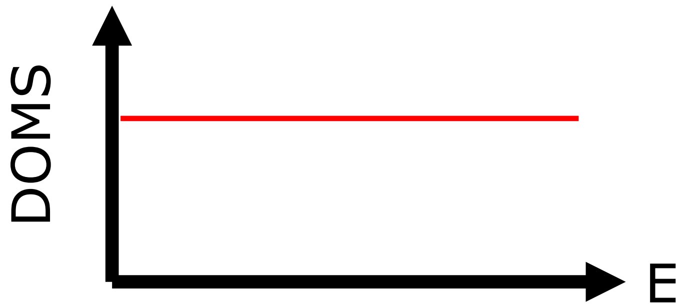


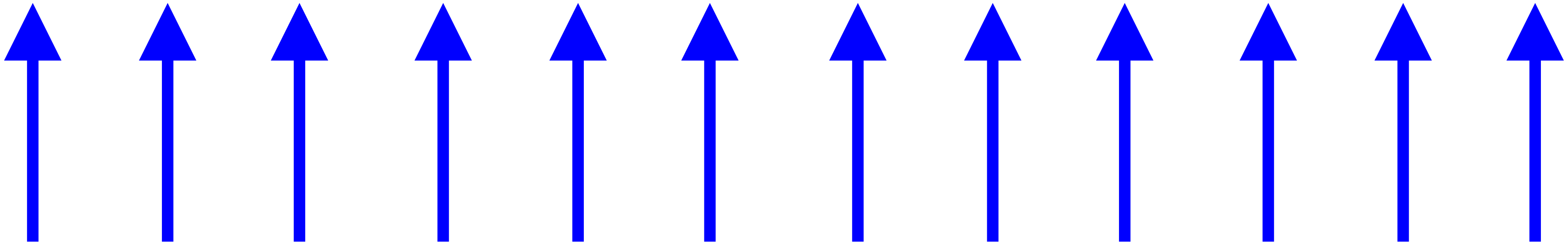


T = 10 K

Effect of Dimensionality

2D

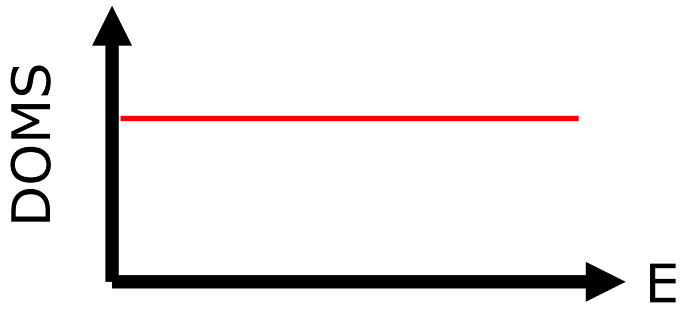


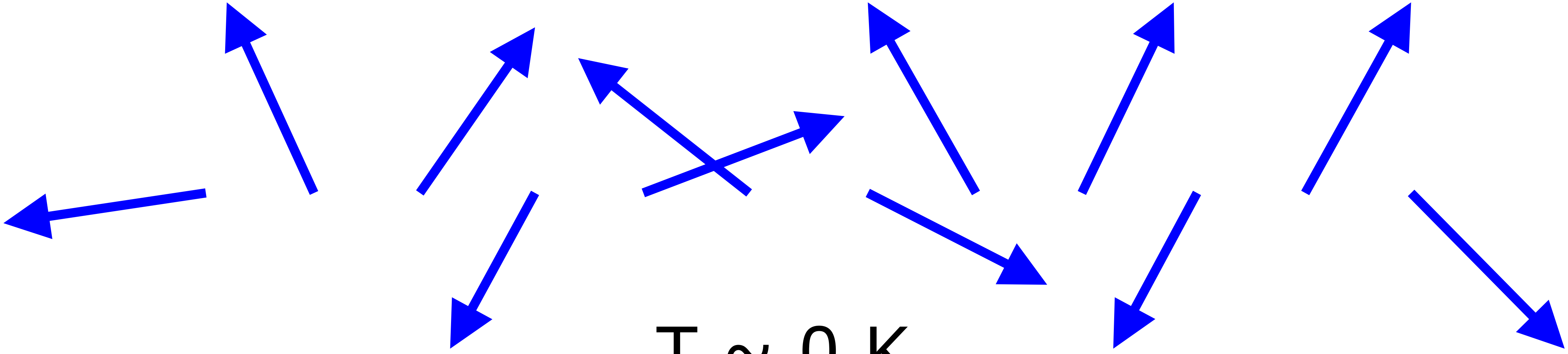


T = 0 K

Effect of Dimensionality

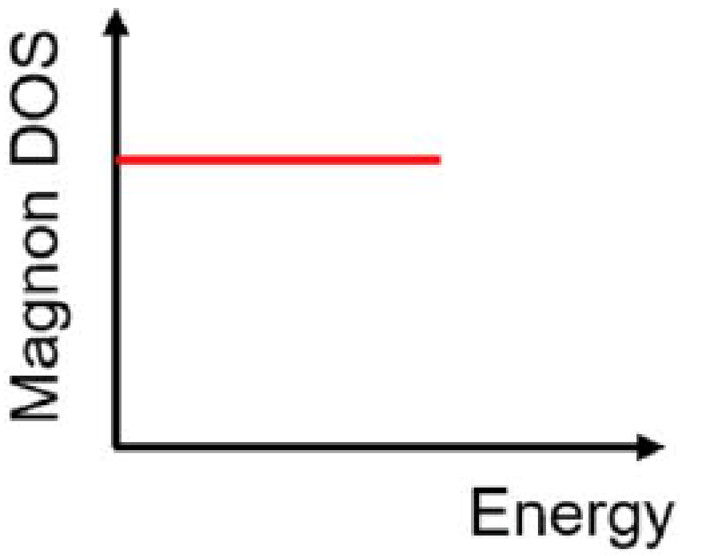
2D



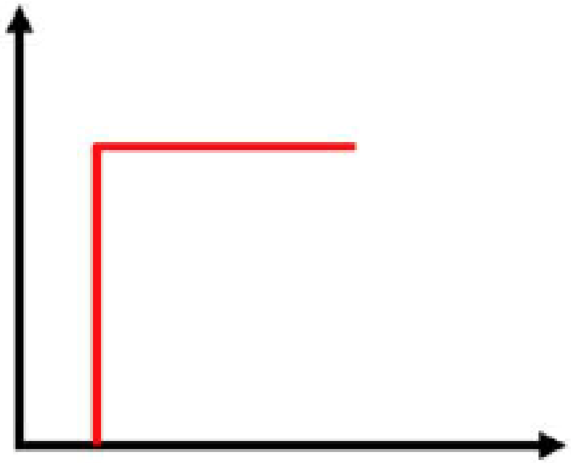


T ~ 0 K

Importance of Anisotropy



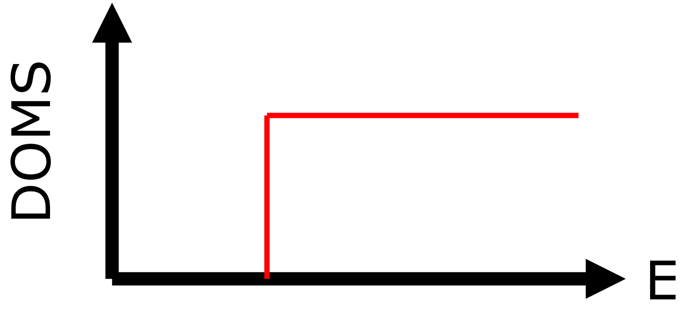
Monolayer + Isotropy

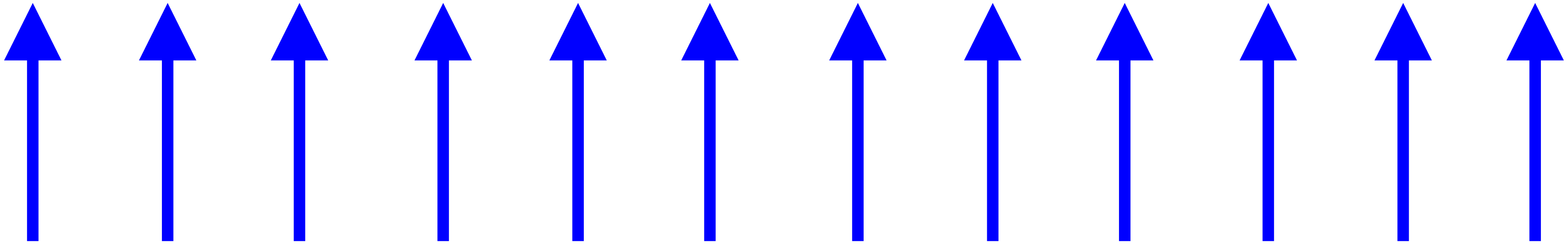


Monolayer + UMA

Dimensionality + Anisotropy

2D





T = 10 K

“Seeing” magnetism: Magneto-Optics

$$\varepsilon = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & \varepsilon_{xz} \\ \varepsilon_{yx} & \varepsilon_{yy} & \varepsilon_{yz} \\ \varepsilon_{zx} & \varepsilon_{zy} & \varepsilon_{zz} \end{pmatrix}$$

Light-Matter Interaction in Magnetic Materials

$$\varepsilon = \begin{pmatrix} \varepsilon_{xx} & \varepsilon_{xy} & 0 \\ -\varepsilon_{xy} & \varepsilon_{xx} & 0 \\ 0 & 0 & \varepsilon_{xx} \end{pmatrix}$$

Eingenvectors:

$$\begin{pmatrix} E_x \\ E_y \end{pmatrix}_{\pm} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ \pm i \end{pmatrix}$$

σ_+ and σ_-

Optical conductivity:

$$\boldsymbol{\varepsilon} = \mathbf{1} + i\,\frac{4\pi}{\omega}\,\boldsymbol{\sigma}$$

Eingenvalues:

$$\varepsilon_{\pm} = \varepsilon_{xx} \pm i\varepsilon_{xy}$$

Refractive index: $n_{\pm} = n_{\pm} \pm i\eta_{\pm}$

Light-Matter Interaction in Magnetic Materials

Refractive index:

$$n_{\pm} = n_{\pm} \pm i\eta_{\pm} \qquad |V\rangle = \frac{1}{2}(|\sigma_{+}\rangle + |\sigma_{-}\rangle) \qquad |H\rangle = \frac{1}{2}(|\sigma_{+}\rangle - |\sigma_{-}\rangle)$$

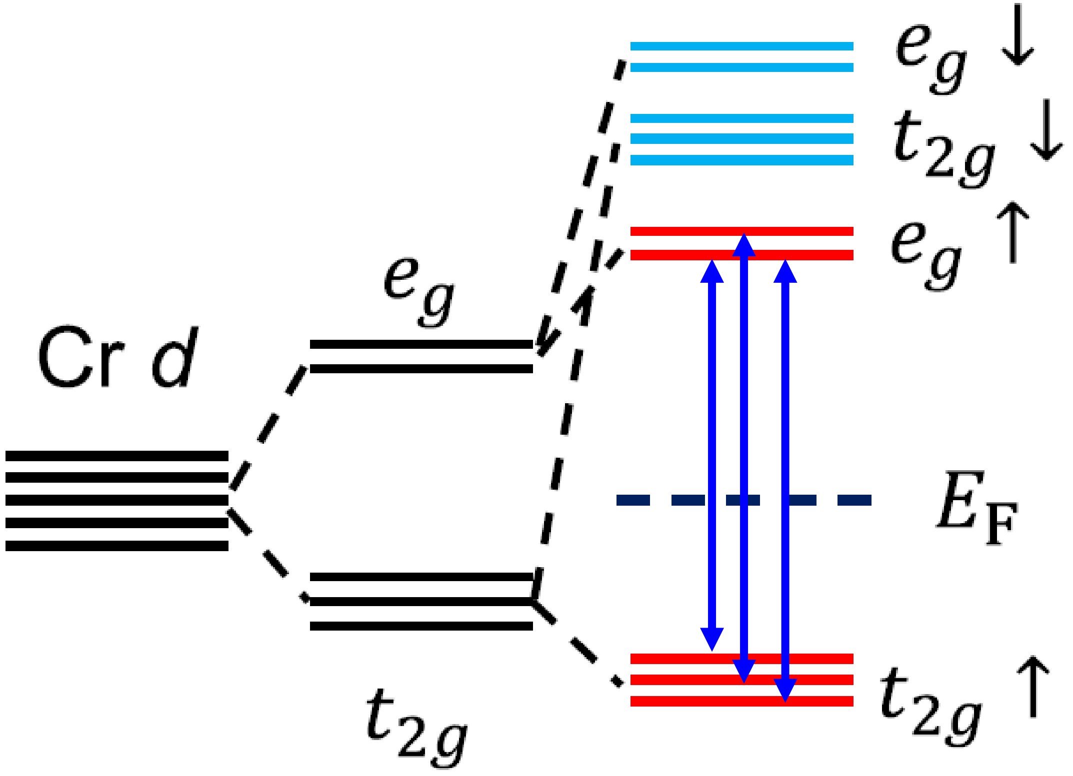
Kerr / Faraday Rotation

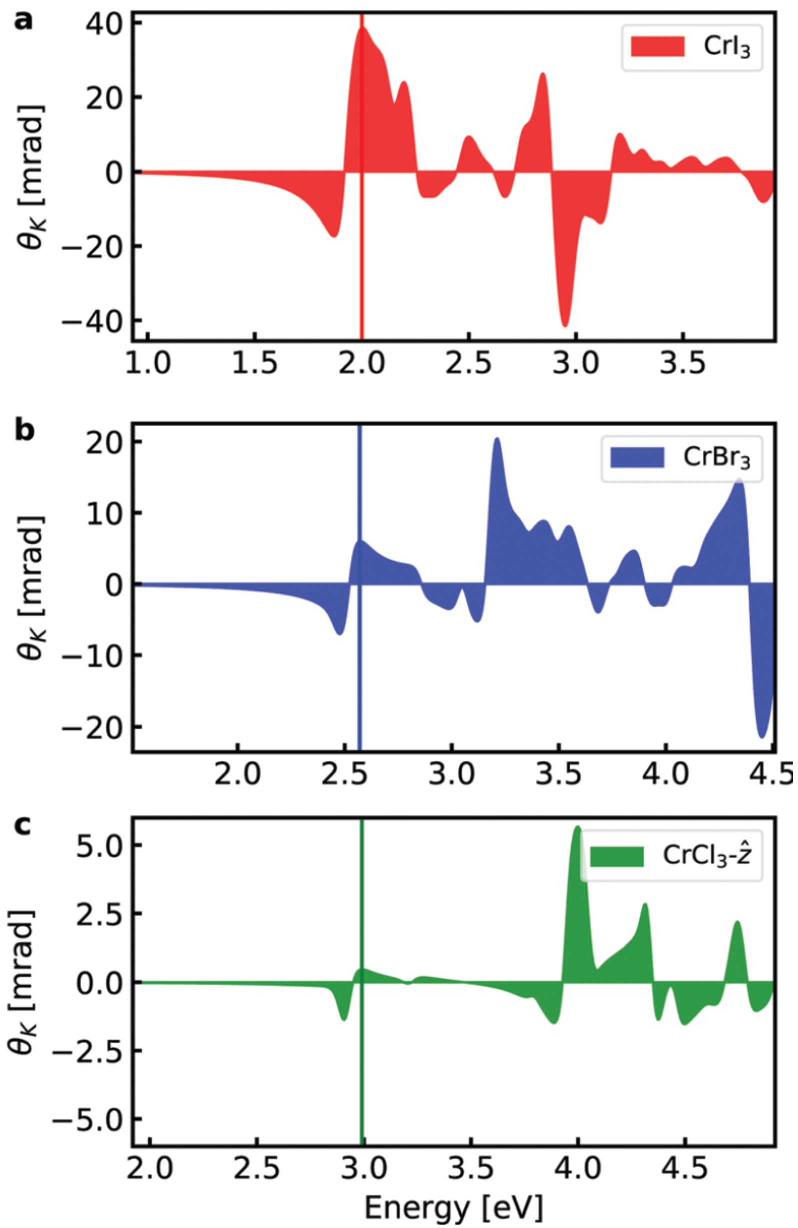
Different propagation speeds
Different phase factor
Rotation of the polarization

Kerr / Faraday Ellipticity
Magnetic Circular Dichroism

Different absorption coefficients
Different amplitudes
Ellipticity of the polarization

Light-Matter Interaction in Magnetic Materials

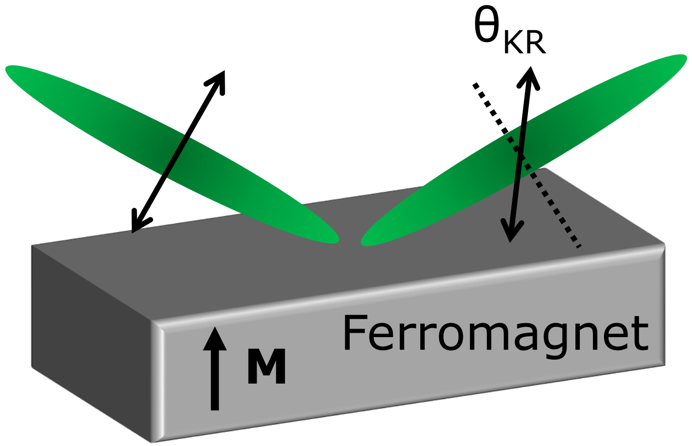


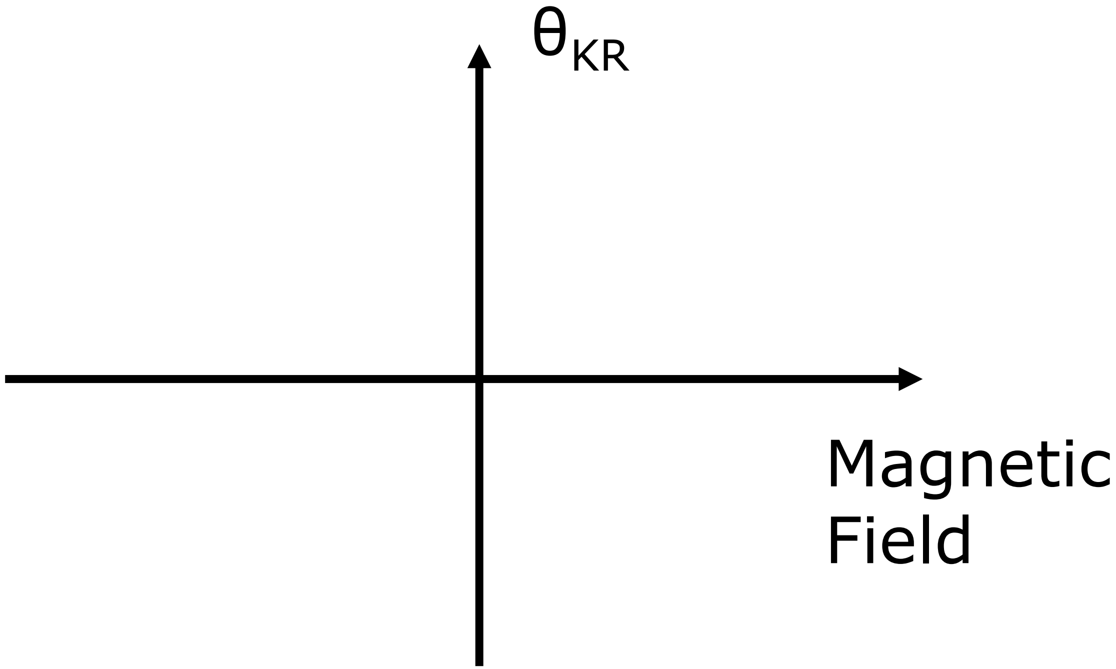


Wu et al., PR Materials **6**, 014008 (2022)

Molina-Sanchez et al., J. Mat. Chem. **8**, 8856 (2020)

"Seeing" magnetism with light: Magneto-Optic Kerr Effect



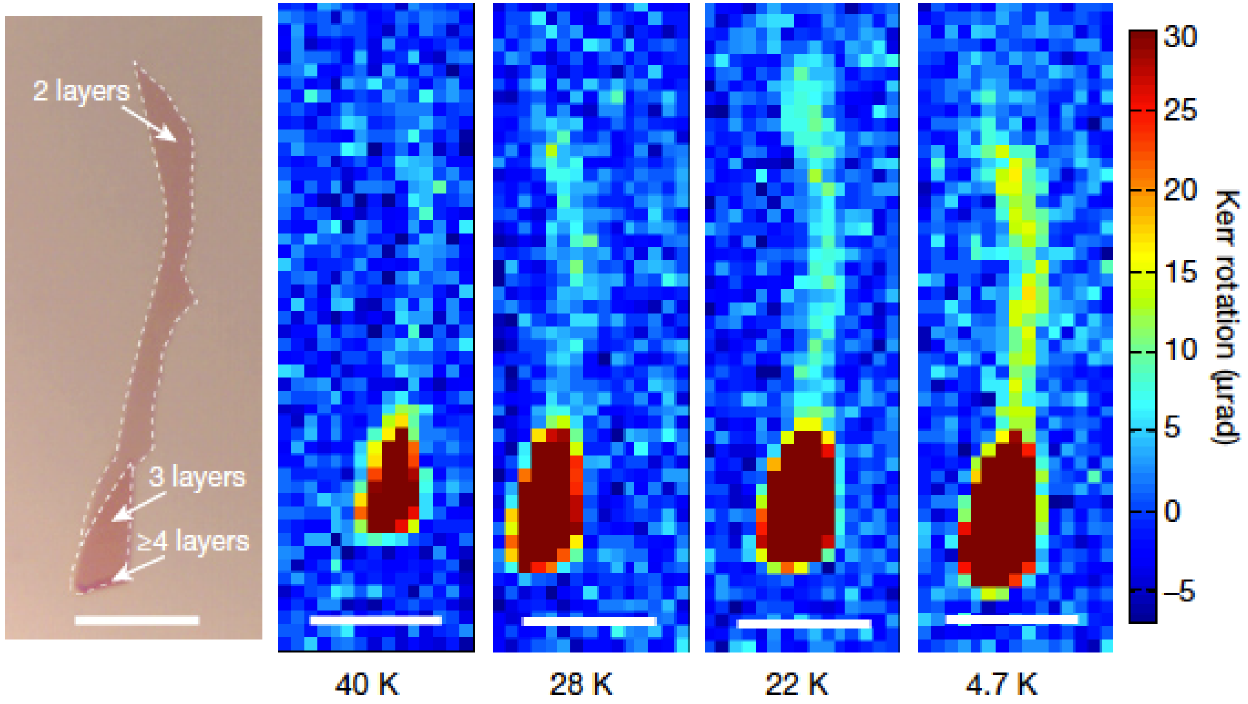


2D Magnets: Very large magneto-optical efficiencies!

From discovery to applications

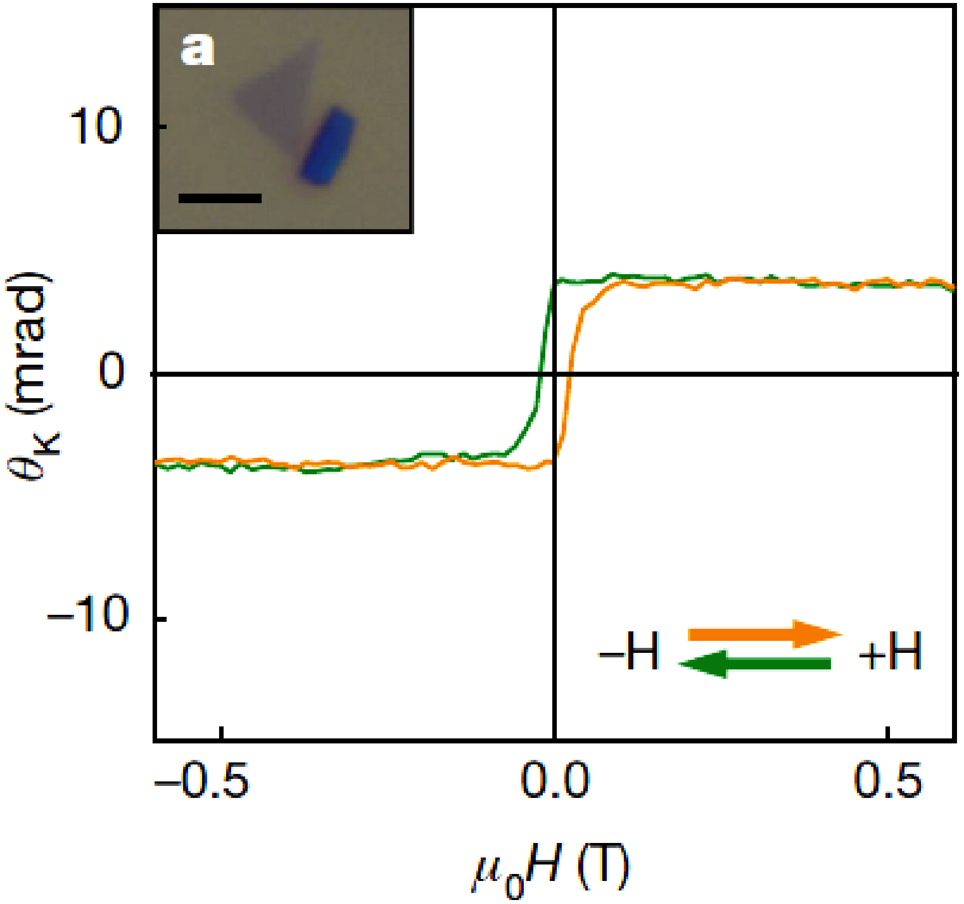
First Observations

$Cr_2Ge_2Te_6$



Burch et al., Nature **546**, 265 (2017)

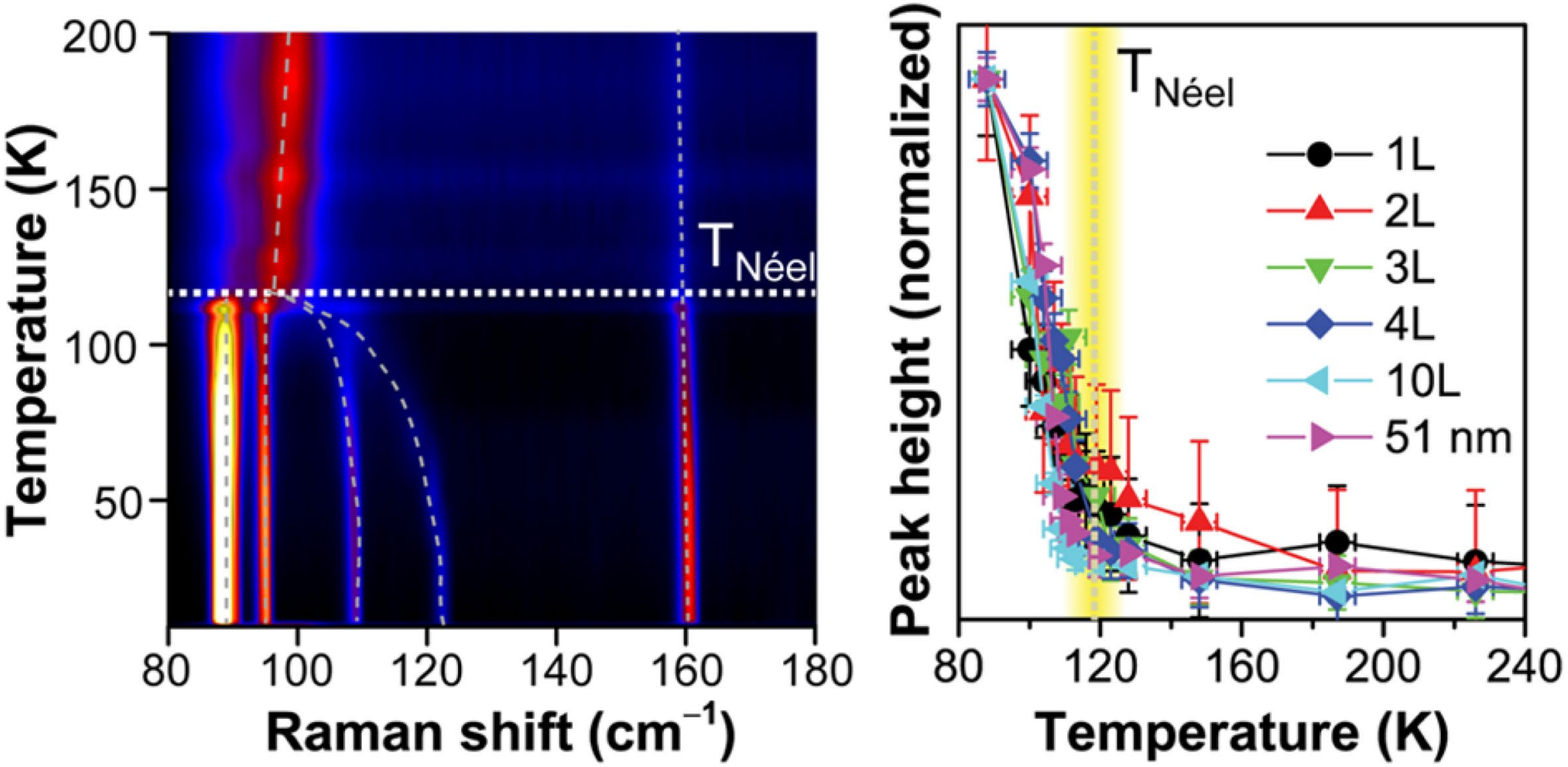
CrI_3



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

First Observations

$FePS_3$



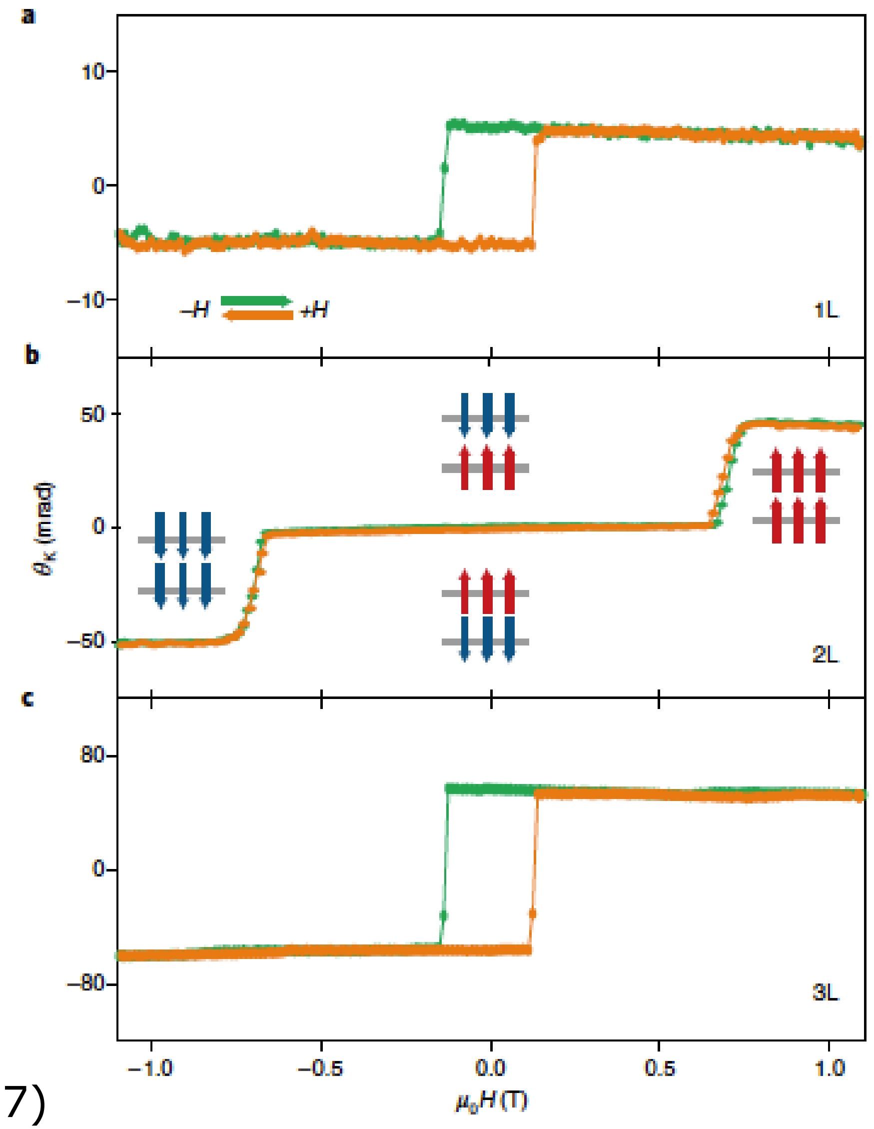
Lee et al., Nano Lett. **16**, 7433 (2016)

CrI_3 – Layered AFM

Bulk is FERROMAGNETIC

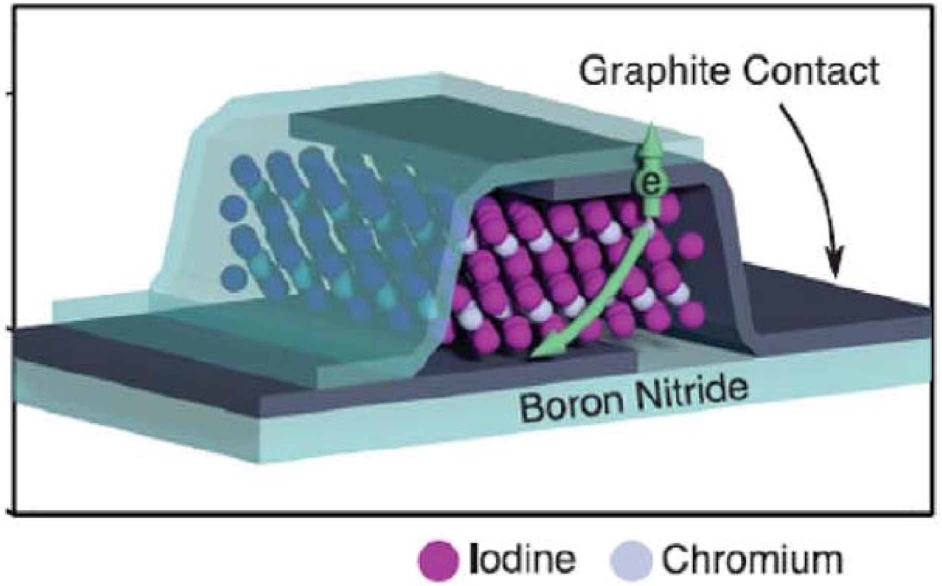
Thin layers seem to be ANTIFERROMAGNETIC!

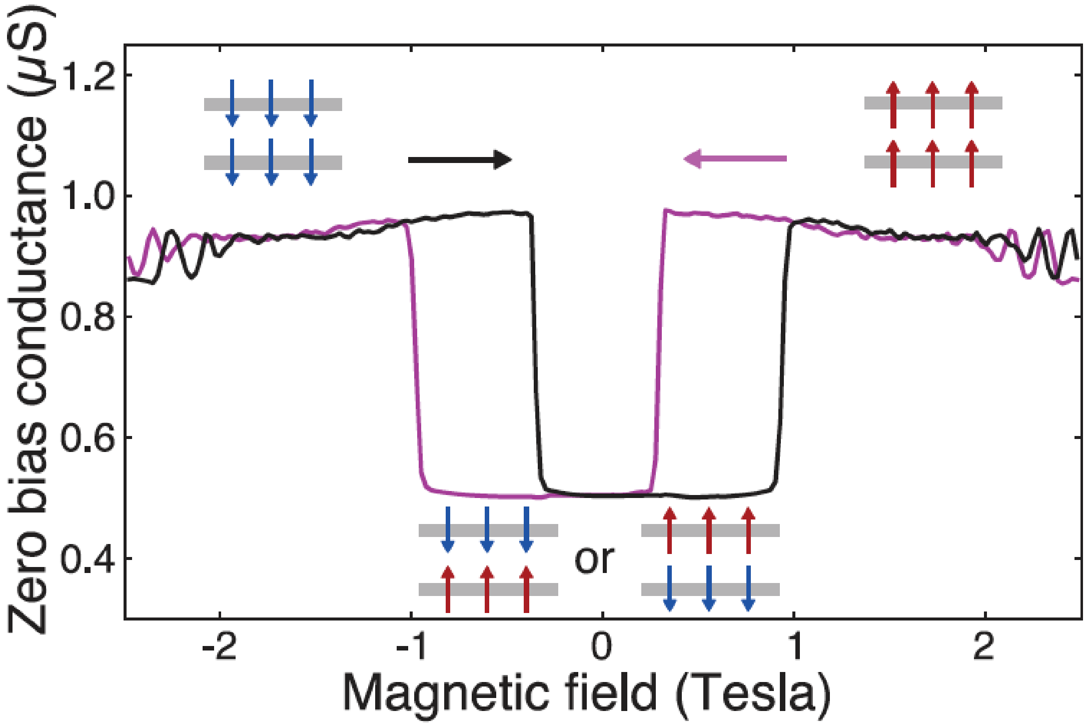
Phase transition for the crystal structure



Nature **546**, 270 (2017)

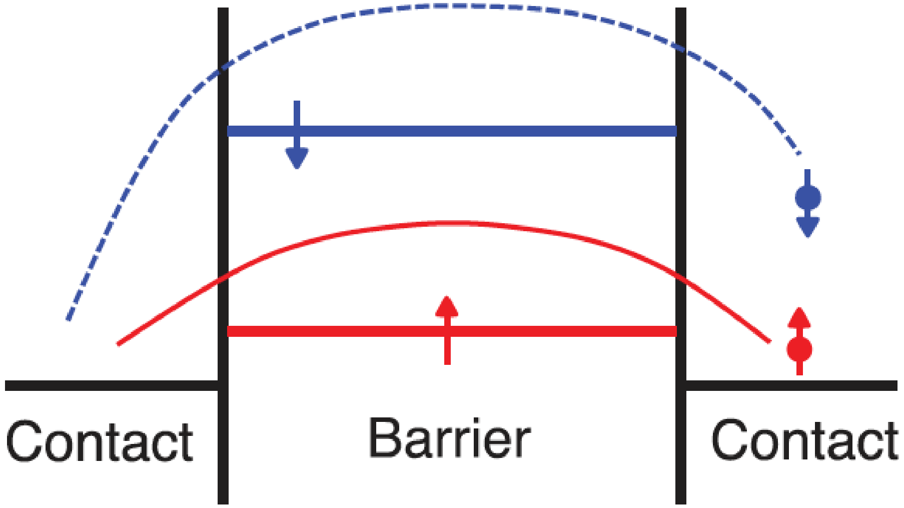
Tunneling Through CrI_3 Layers

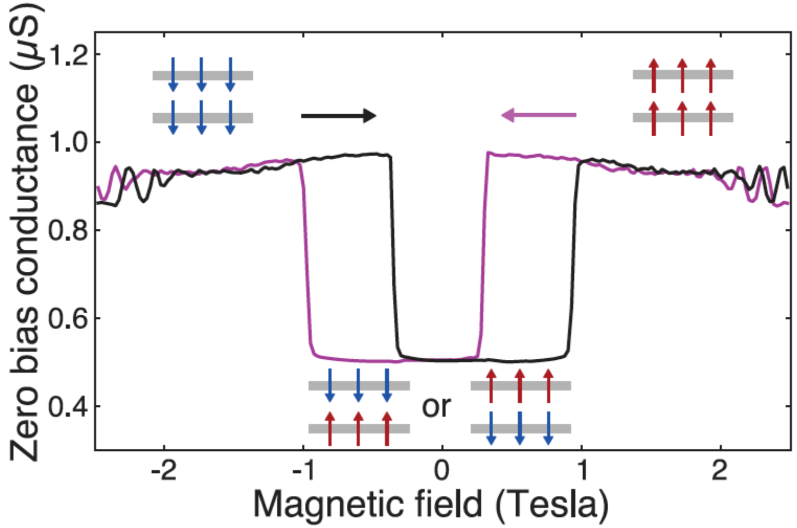


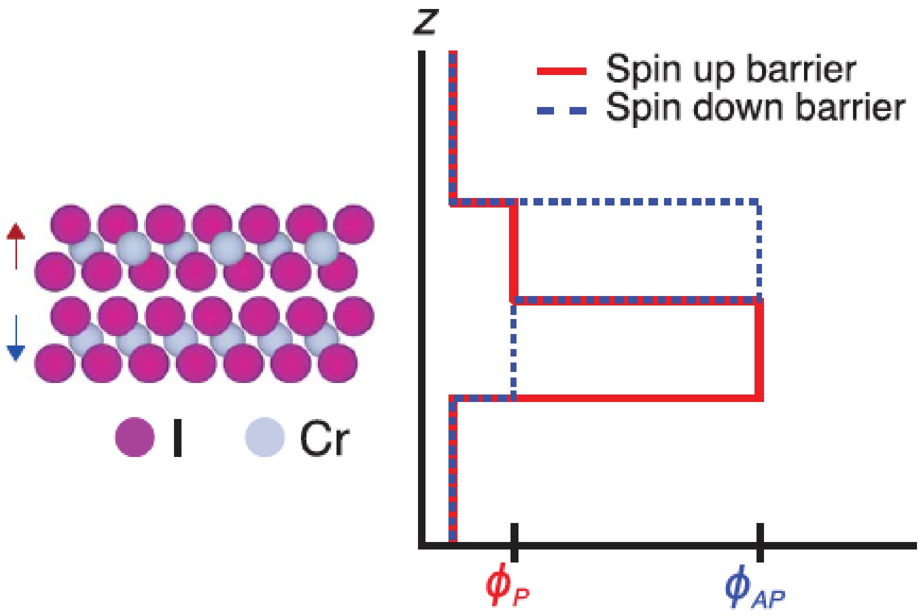


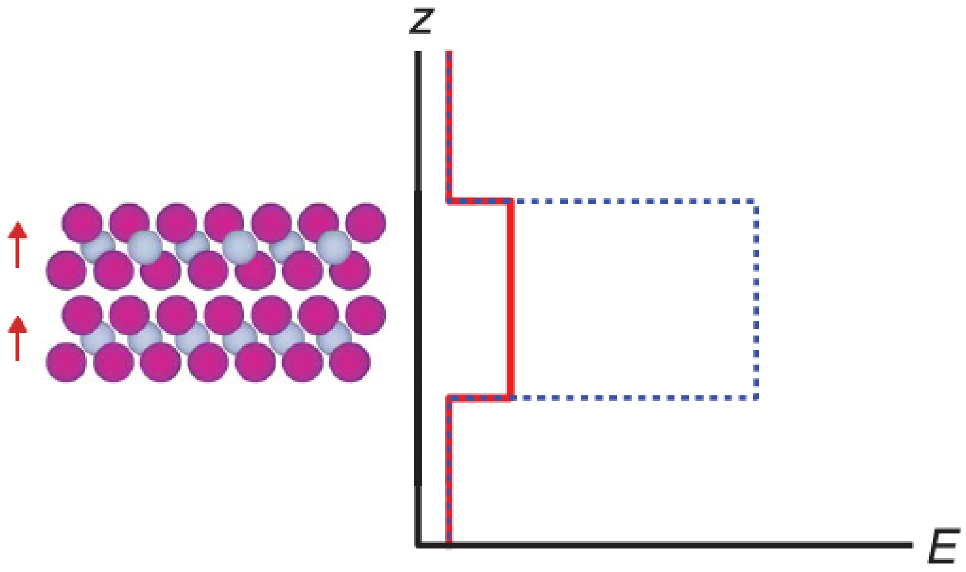
Klein et al, Science **360**, 1218 (2018)

Tunneling Through CrI$_3$ Layers



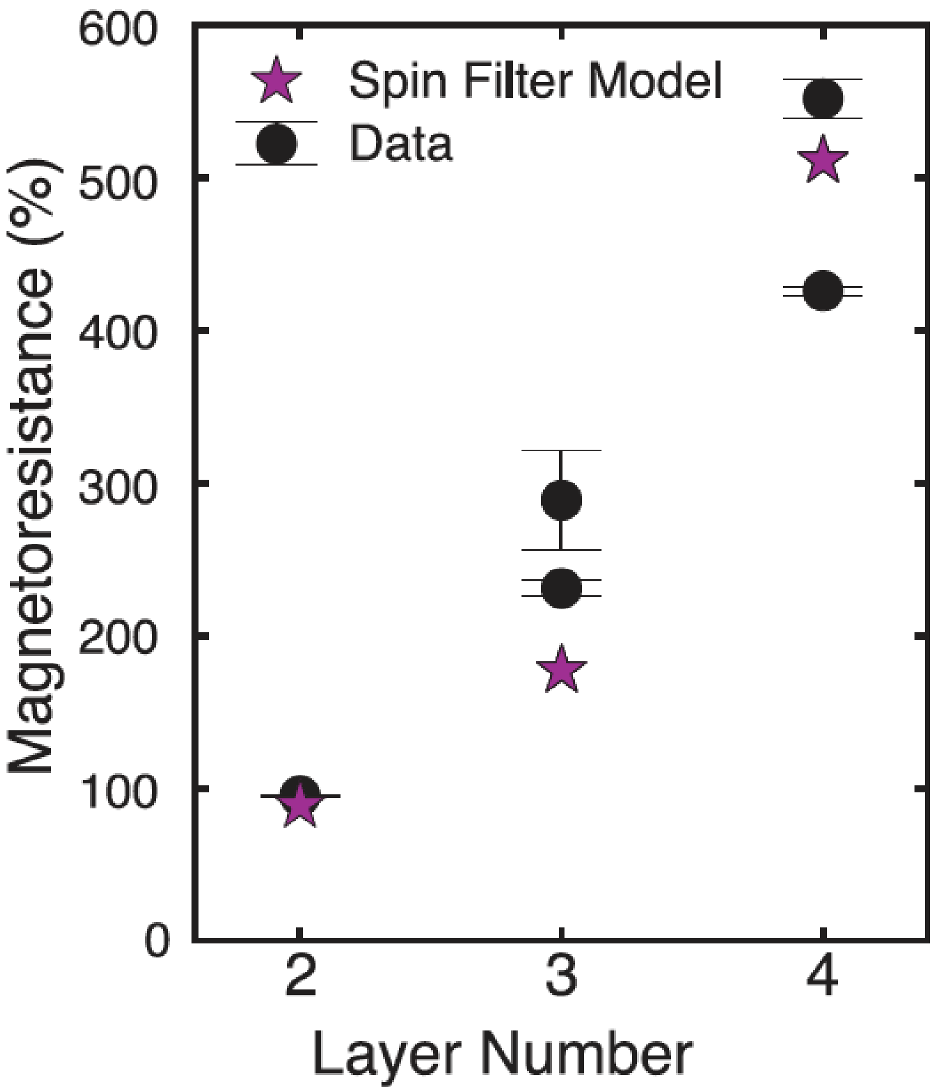






Science **360**, 1218 (2018)

Tunneling Through CrI_3 Layers



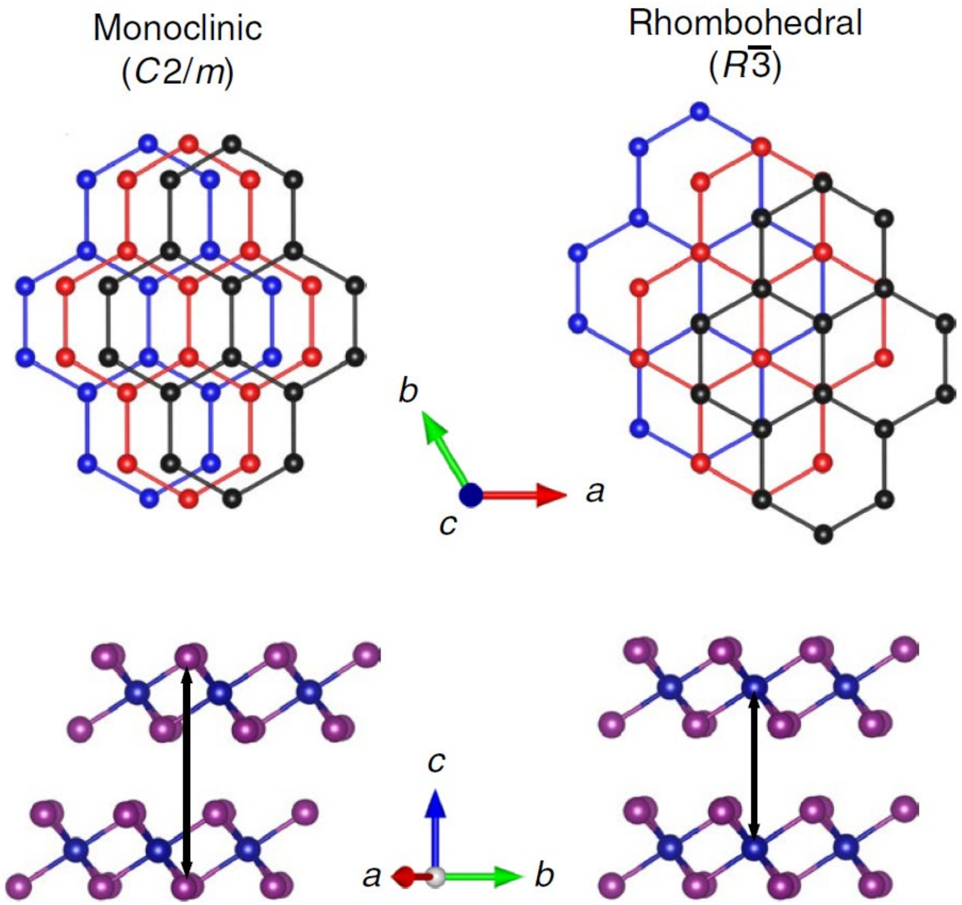
Science **360**, 1218 (2018)

Coffee, please!

Tuning the Interlayer Exchange in CrI_3

$J_\perp < 0$ $J_\perp > 0$

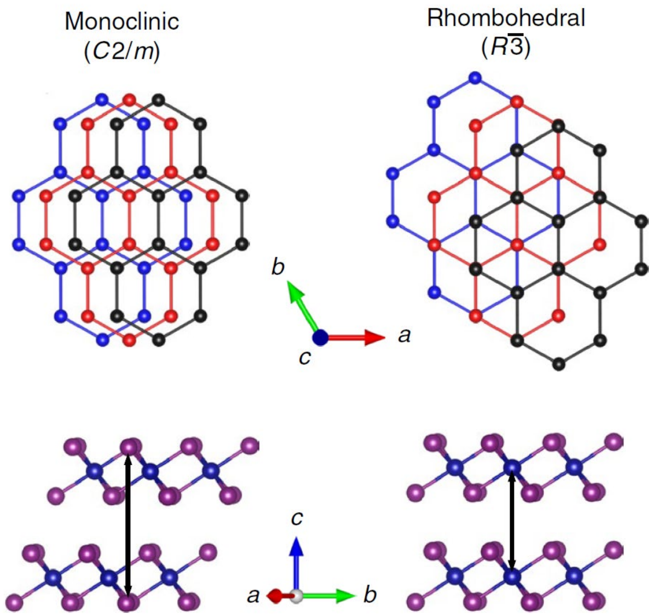
Low temperature phase

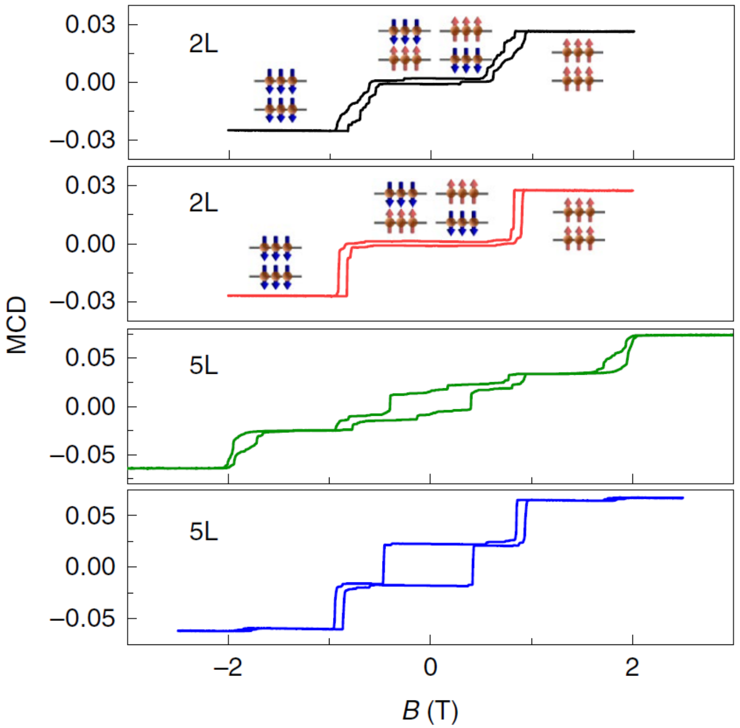


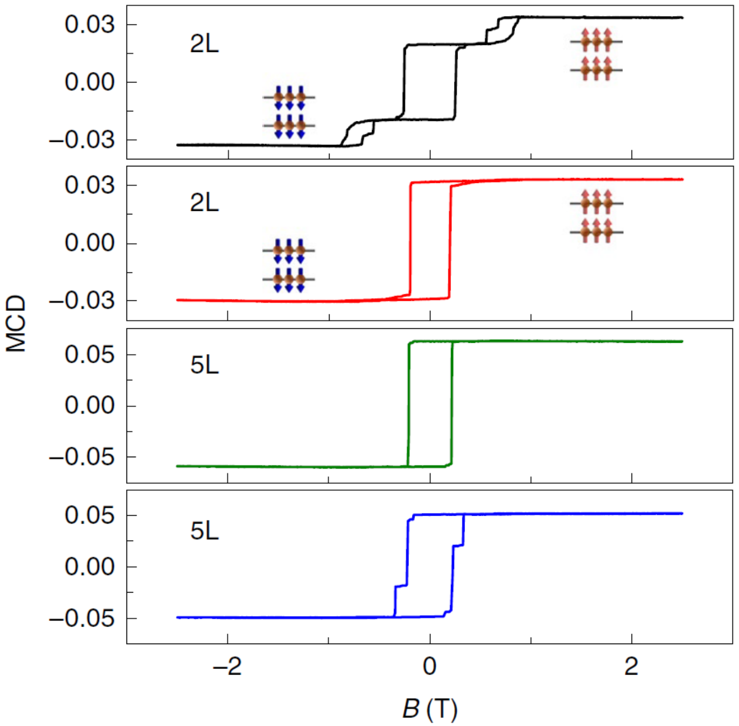
High temperature phase

Nat. Mat. **18**, 1303 (2019)

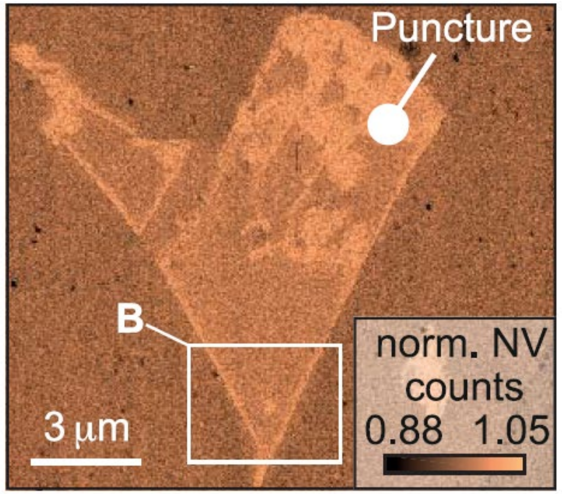
Tuning Magnetism in CrI_3 Layers with Pressure

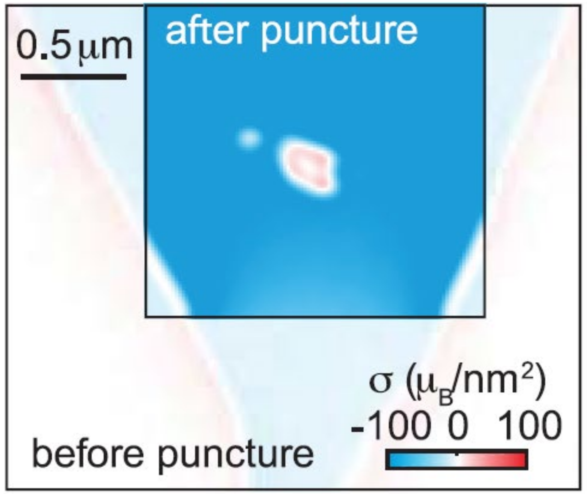






Nat. Mat. **18**, 1303 (2019)





Science **364**, 973 (2019)

How thin is ‘thin’?

What determines the dimensionality of our magnetic system?

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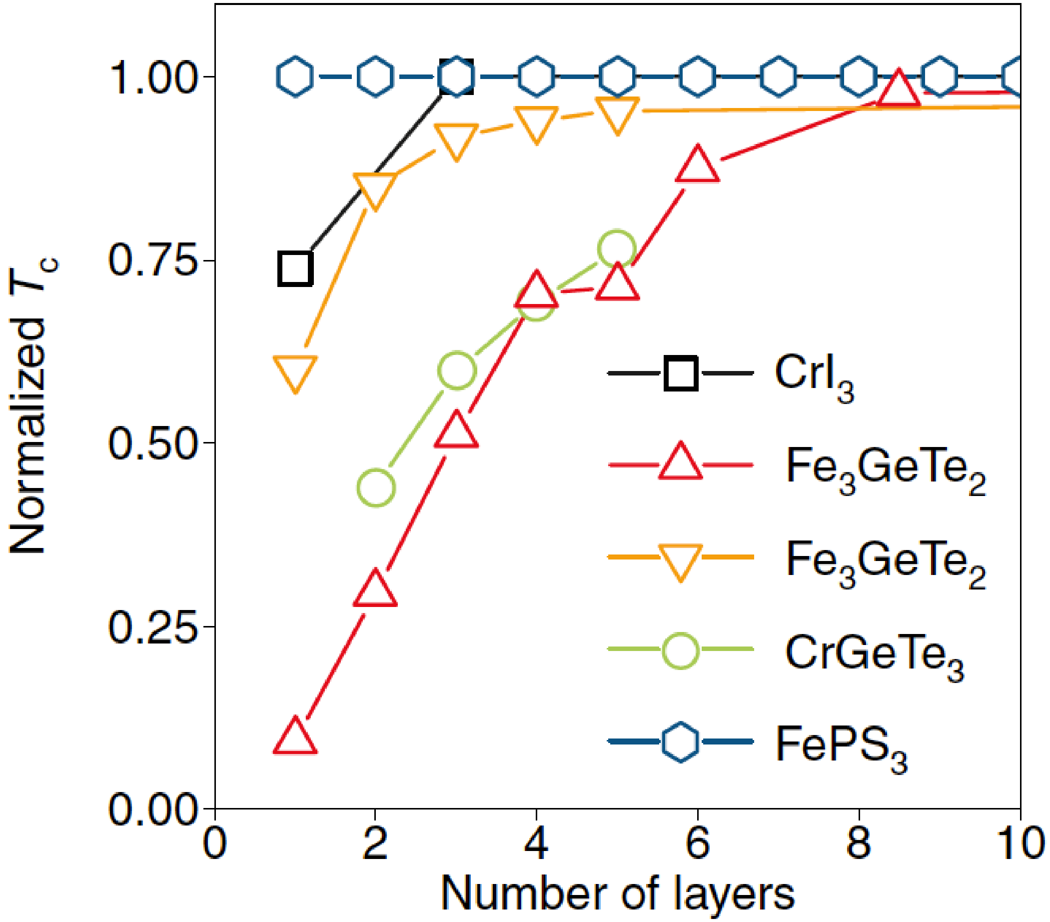
A - $J_{||}$
B - $J_{\perp}$
C – K (magnetic anisotropy)
D – All of the above

How thin is ‘thin’?

It depends on $J_\perp$

Larger $J_\perp$ will result on lower T_C when thinning

Normalized T_C: T_C / T_C^{Bulk}

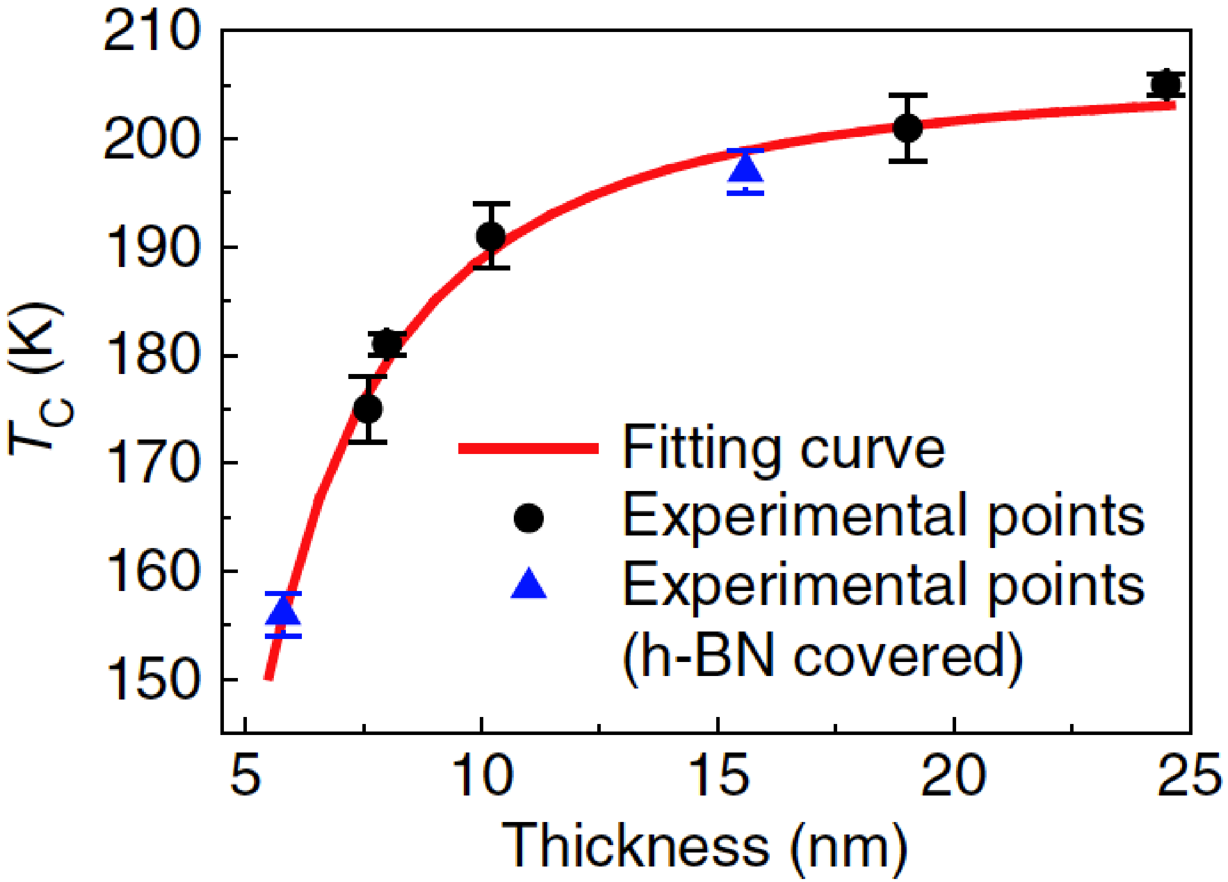


Nature Nanotech. **14**, 408 (2019)

How thin is ‘thin’?

$$\frac{T_C(n)}{T_C^{Bulk}} = 1 - \left[\frac{(N_0+1)}{2n}\right]^{\lambda}$$

For FGT:
N_0 ~ 5 monolayers
λ ~ 1.66 (3D Heisenberg)



Nature Comm. **9**, 1554 (2018)

CrI_3 – Electric Field Effects

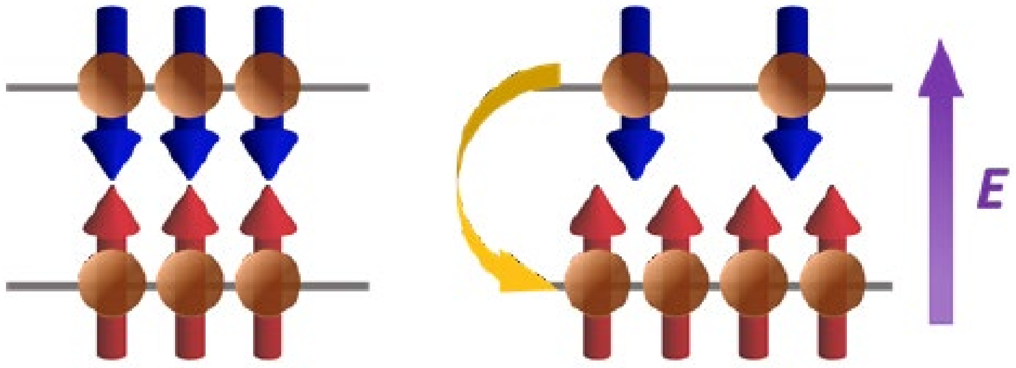

graphene

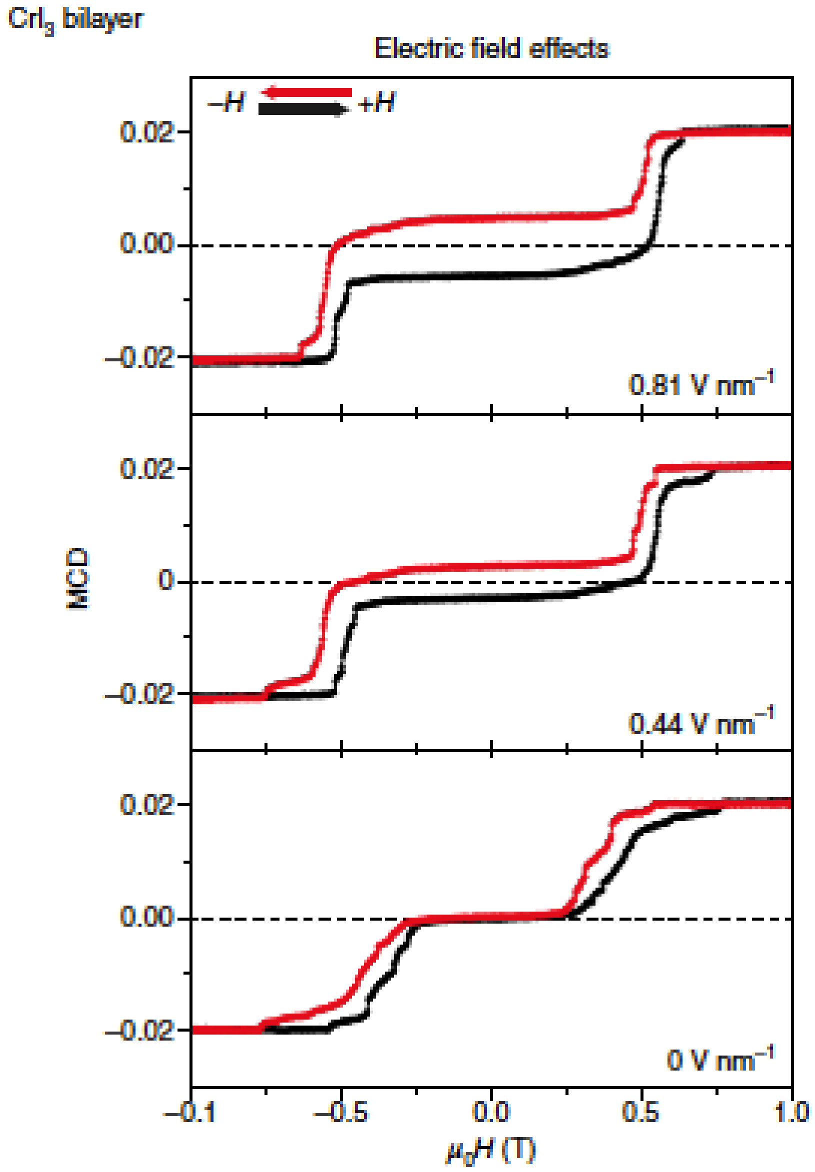
hBN

CrI_3

hBN

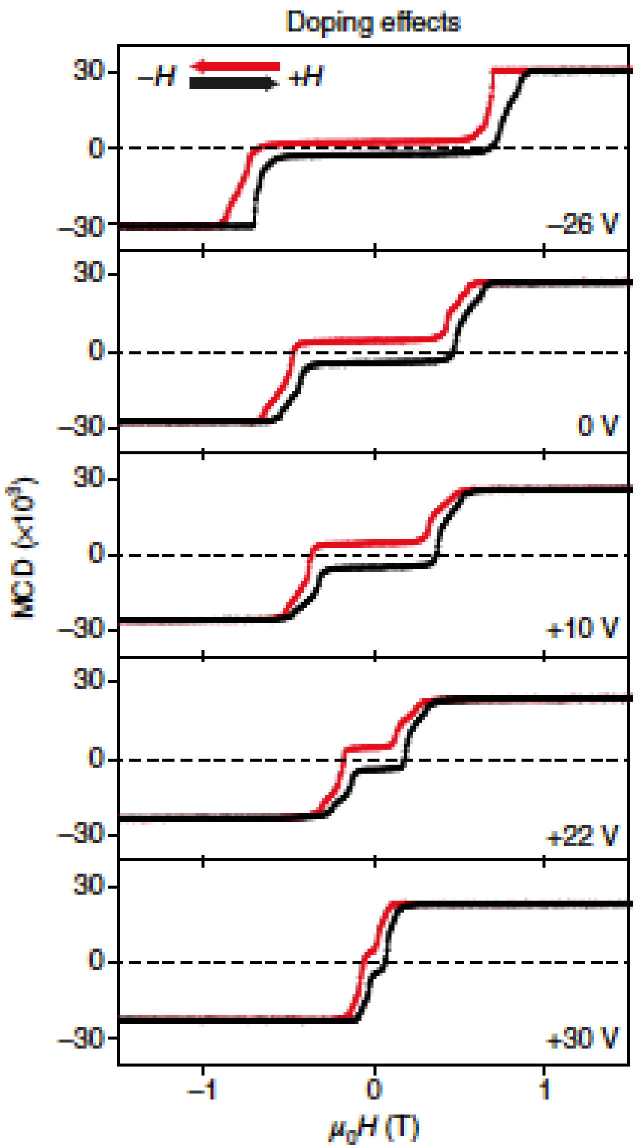
graphene

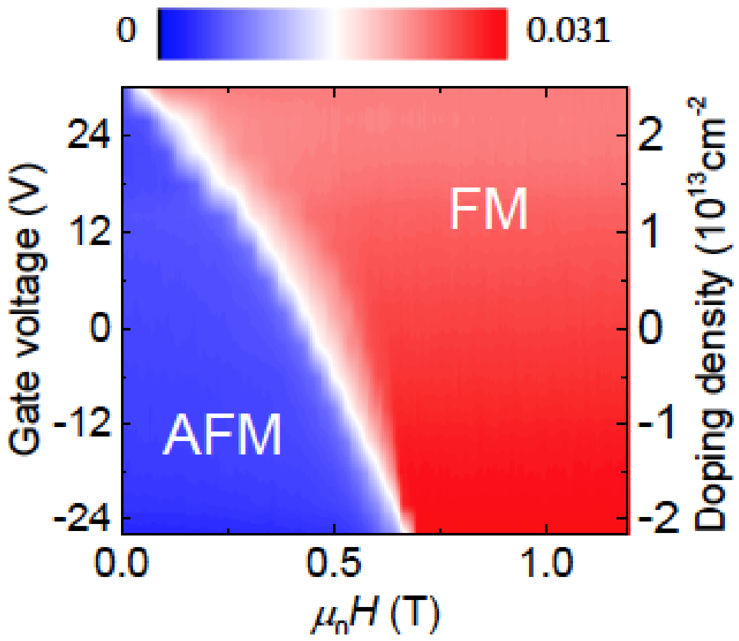


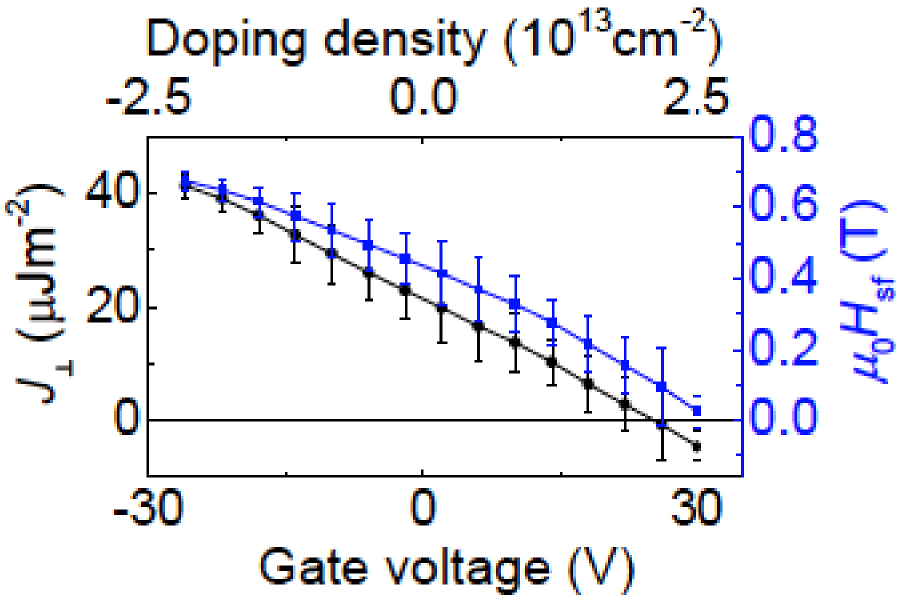


Nature Mat. **17**, 406 (2018)

CrI_3 – Doping Effects

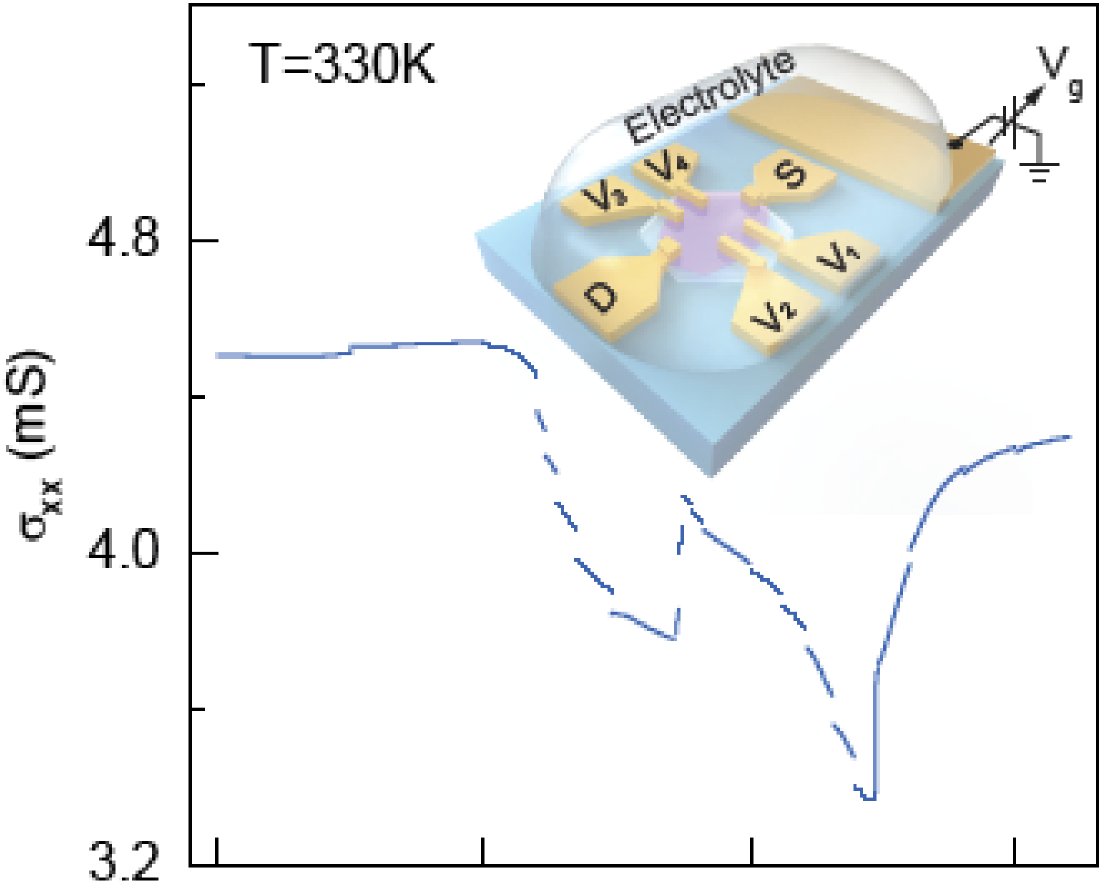


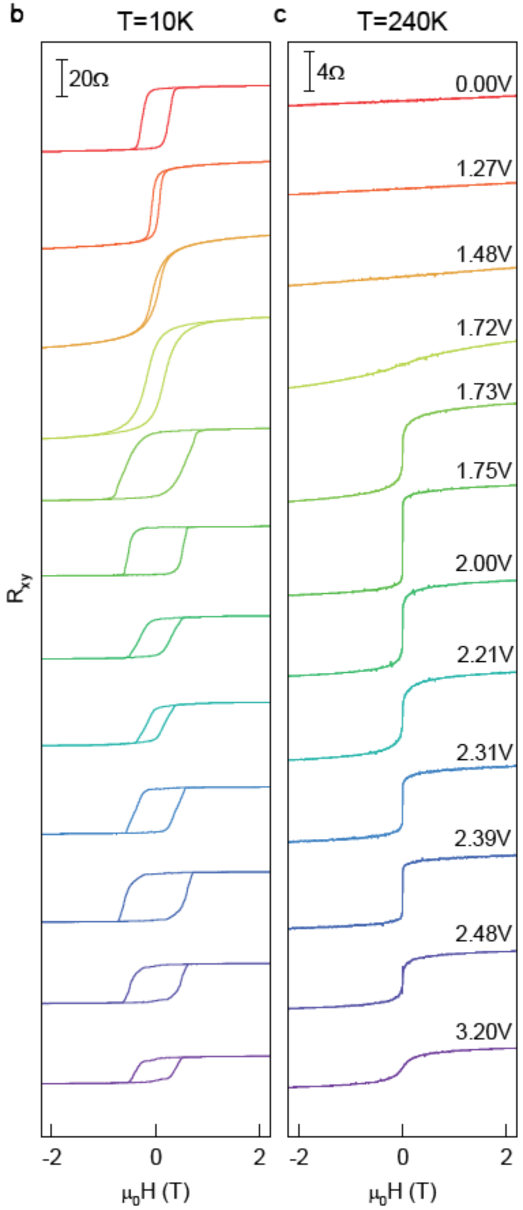




Nature Mat. **17**, 406 (2018)

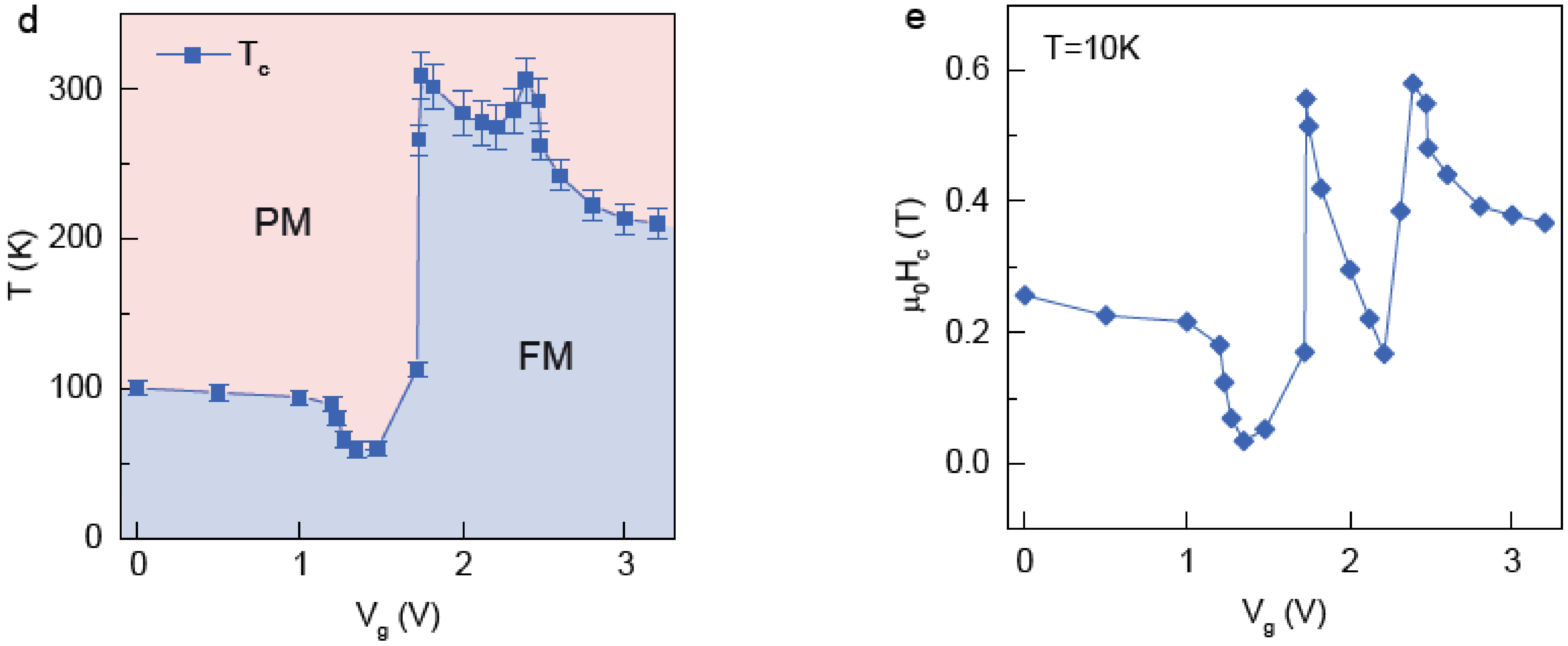
Gate Tunable Ferromagnetism in Fe_3GeTe_2





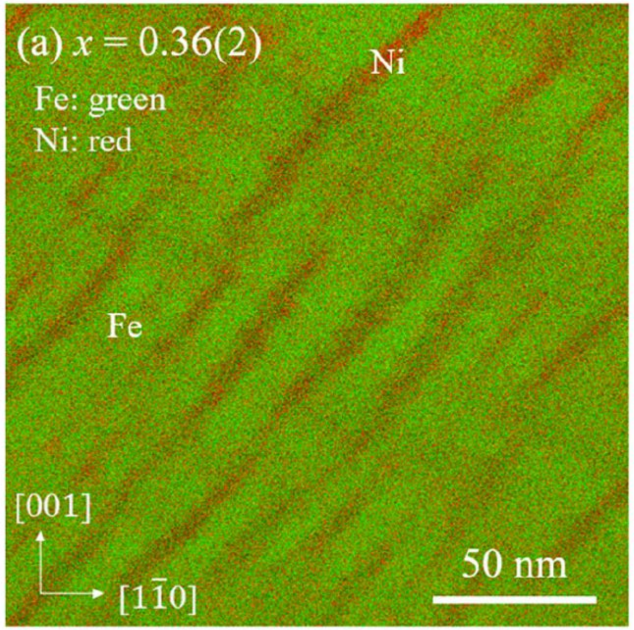
Nature **563**, 94 (2018)

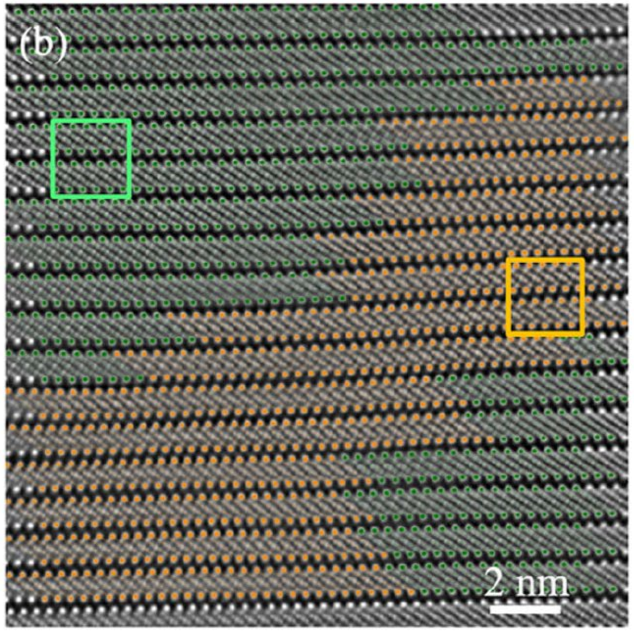
Gate Tunable Ferromagnetism in Fe_3GeTe_2

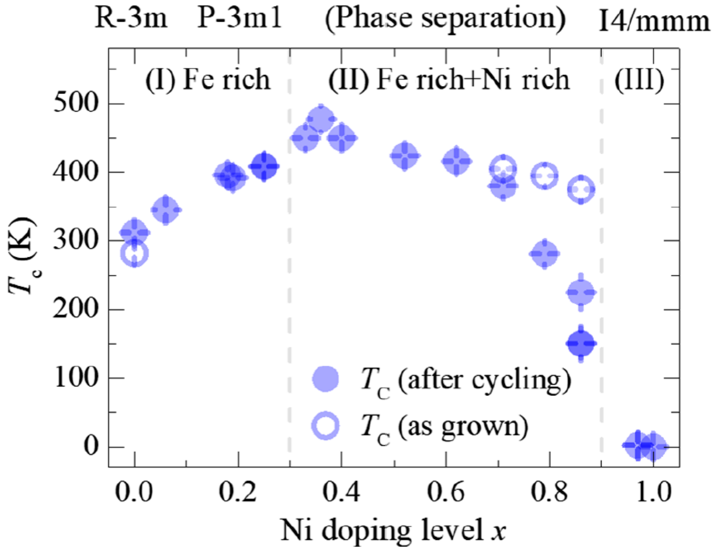


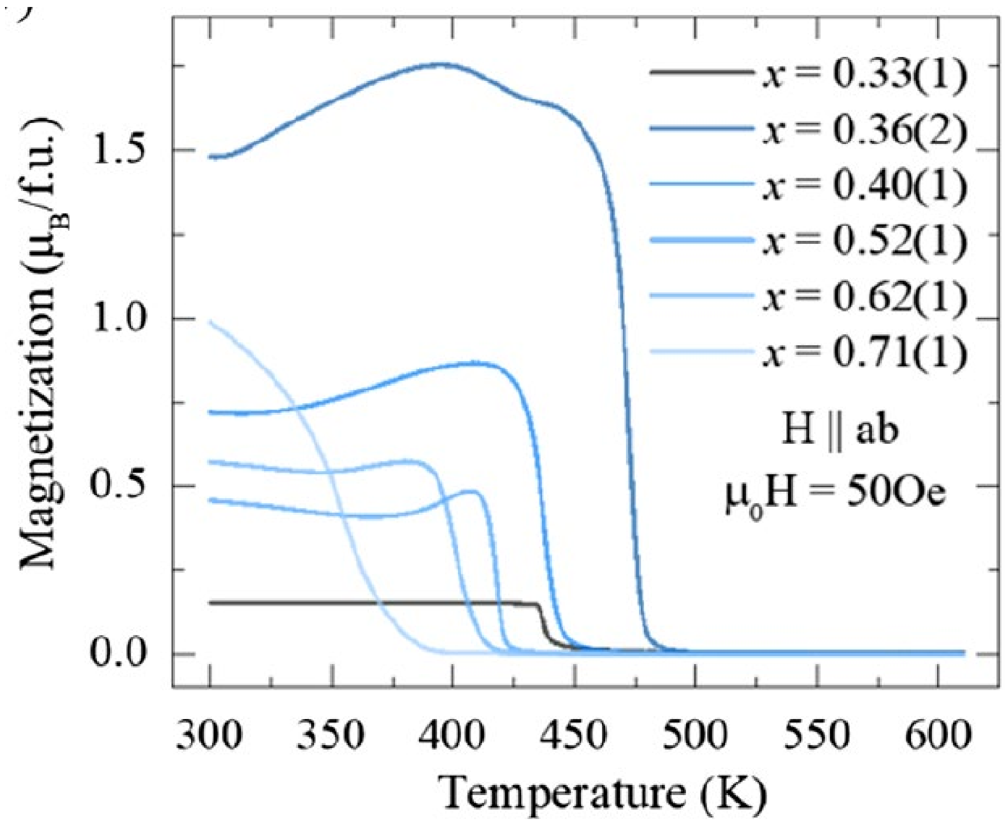
Nature **563**, 94 (2018)

Magnetism Well-Beyond Room Temperature



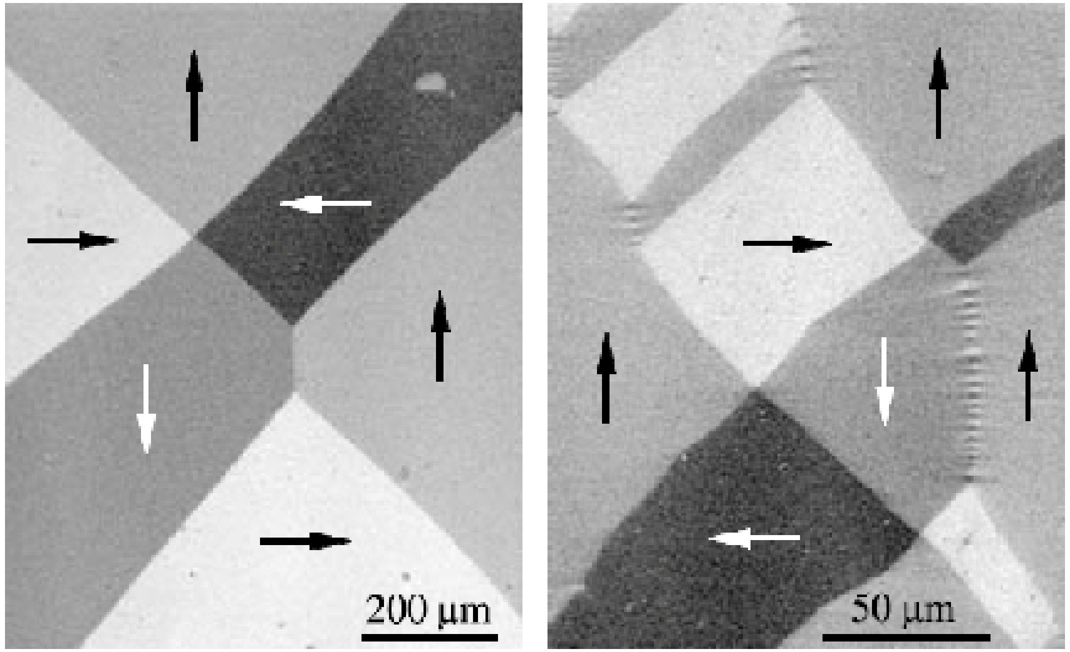


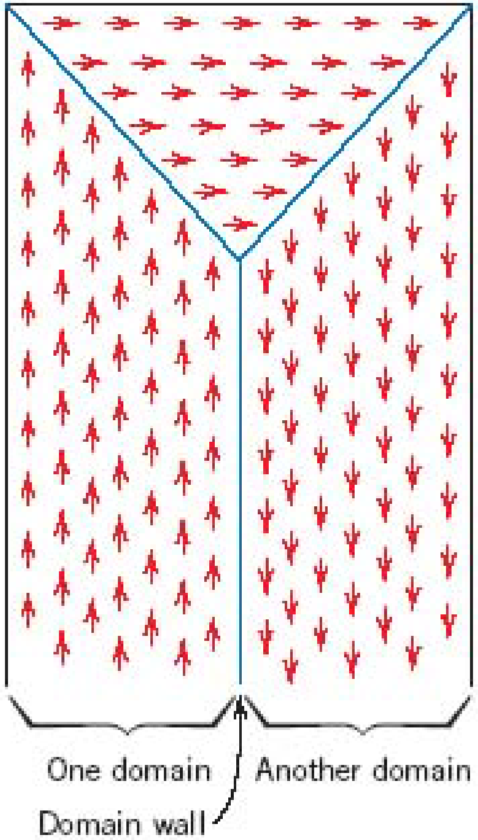


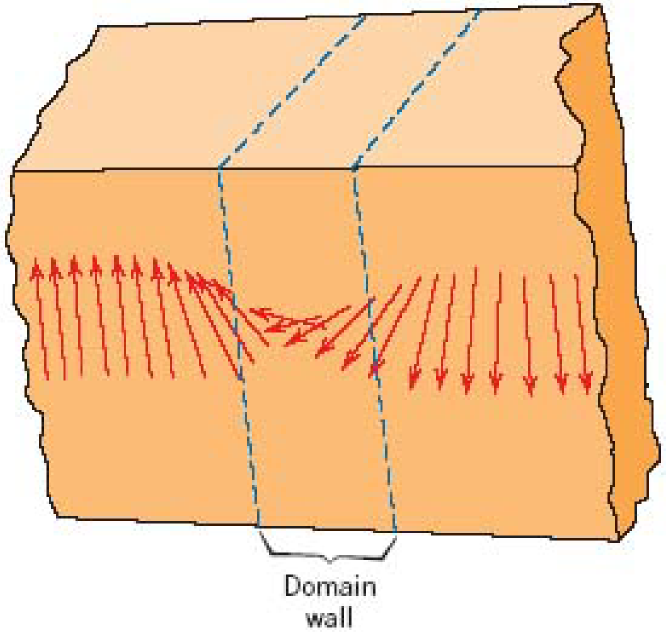


Chen et al., PRL **128**, 217203 (2022)

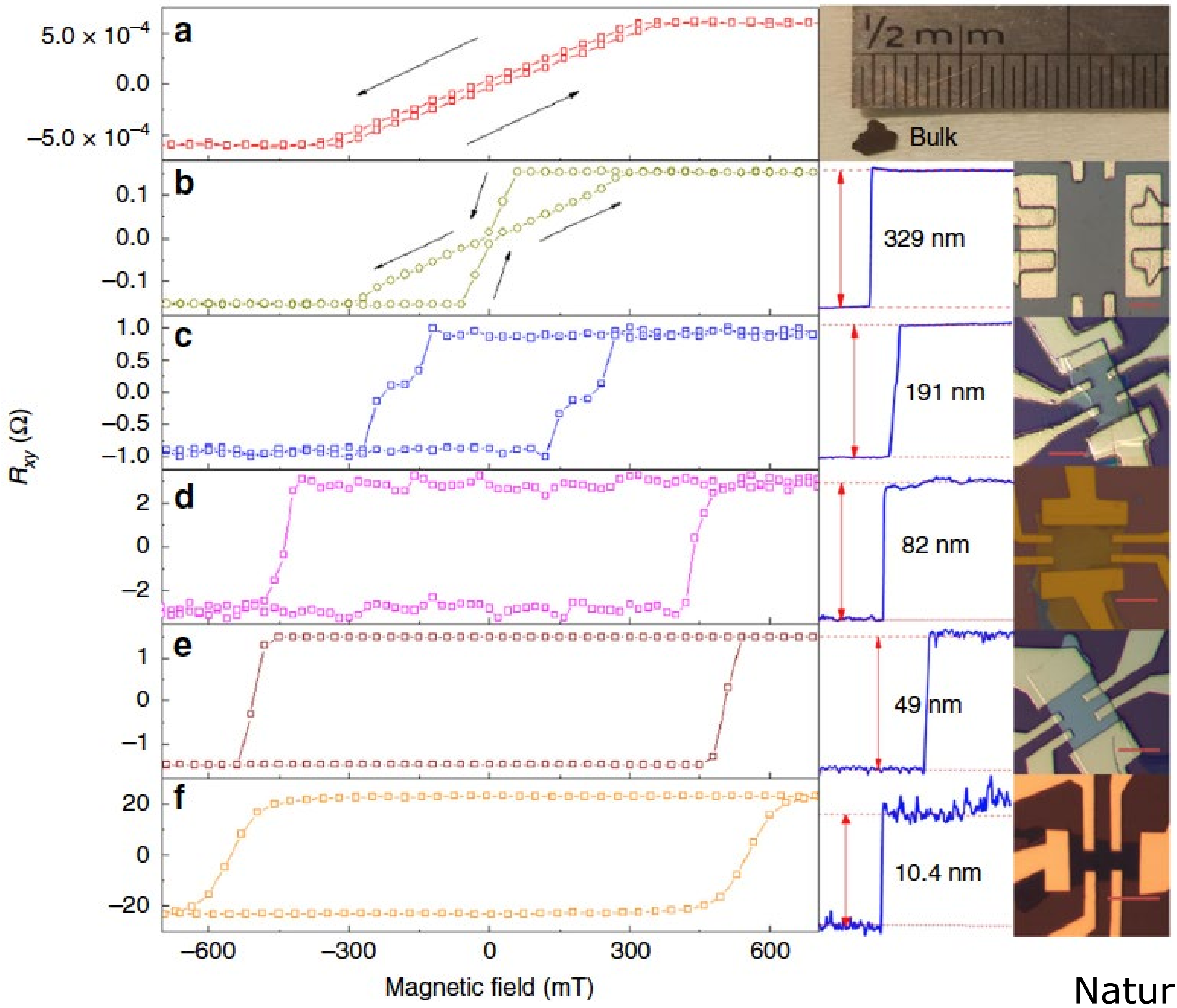
Magnetic Domains





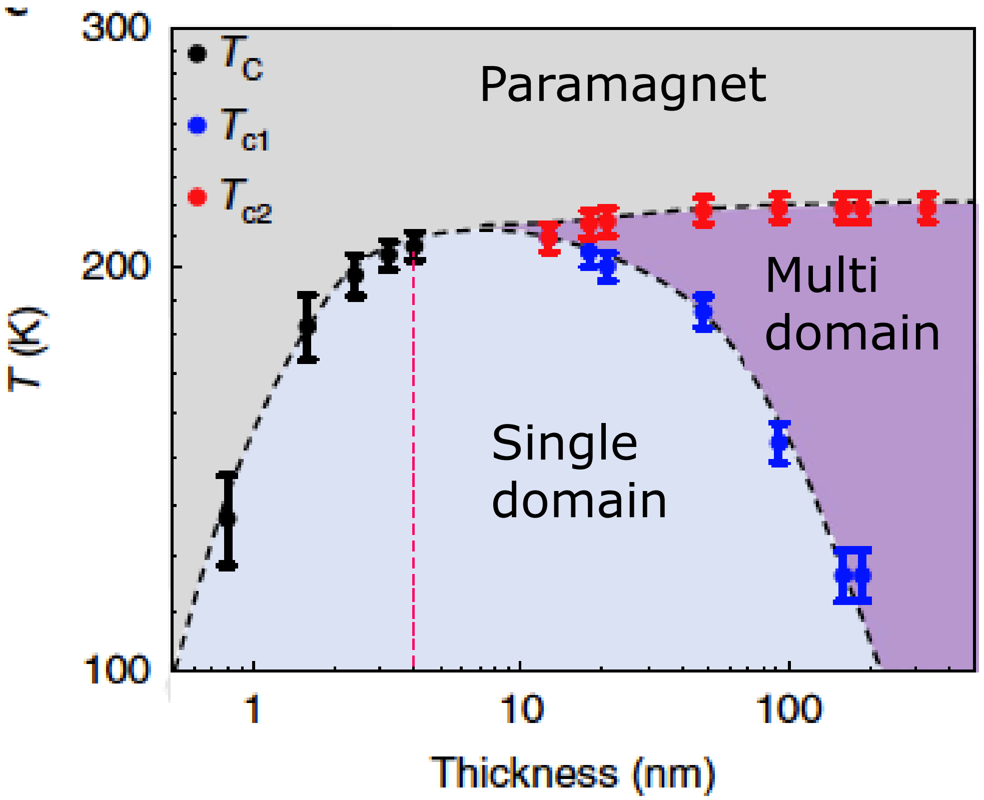


Magnetic Structure of Fe_3GeTe_2

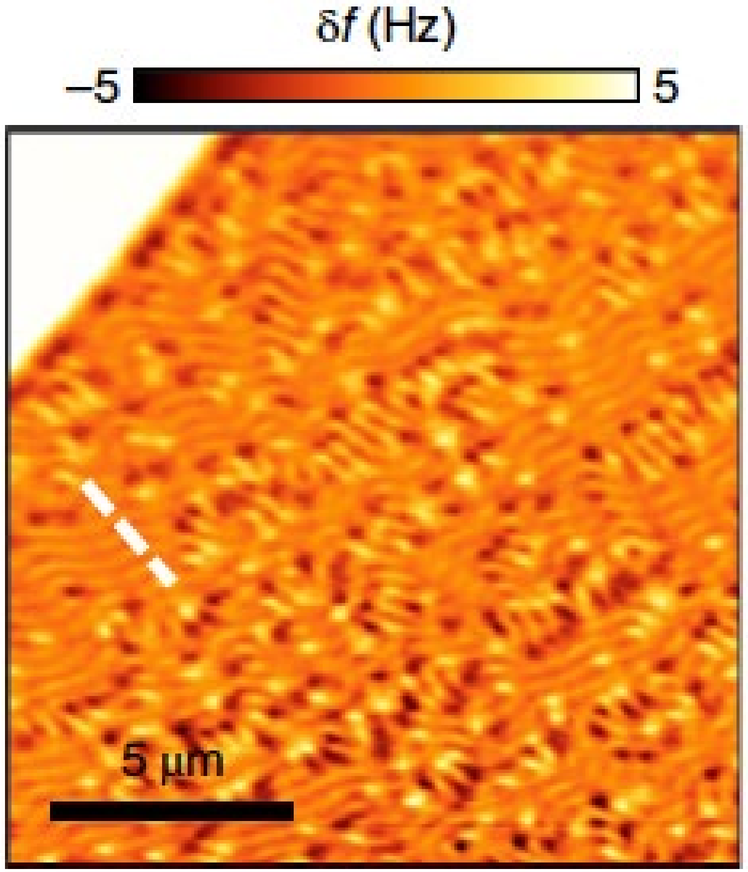


Nature Comm. **9**, 1554 (2018)

Magnetic Structure of Fe_3GeTe_2



MFM Image of Thick Flake

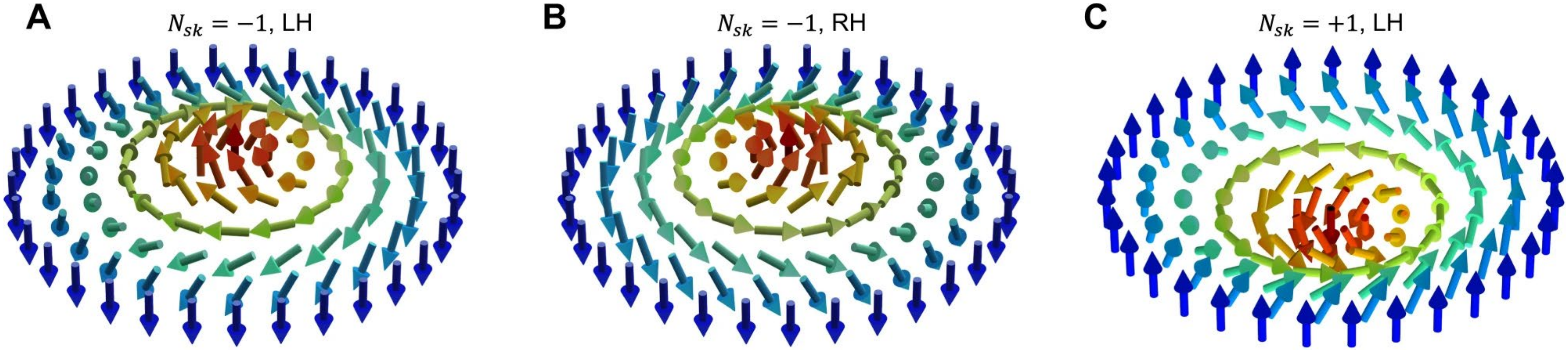


Nature Mat. **17**, 778 (2018)

Why Chiral Spin Textures?

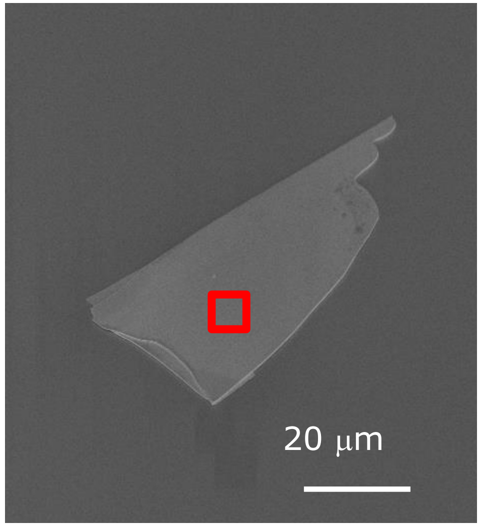
Topological charge,
or Skyrmion number:

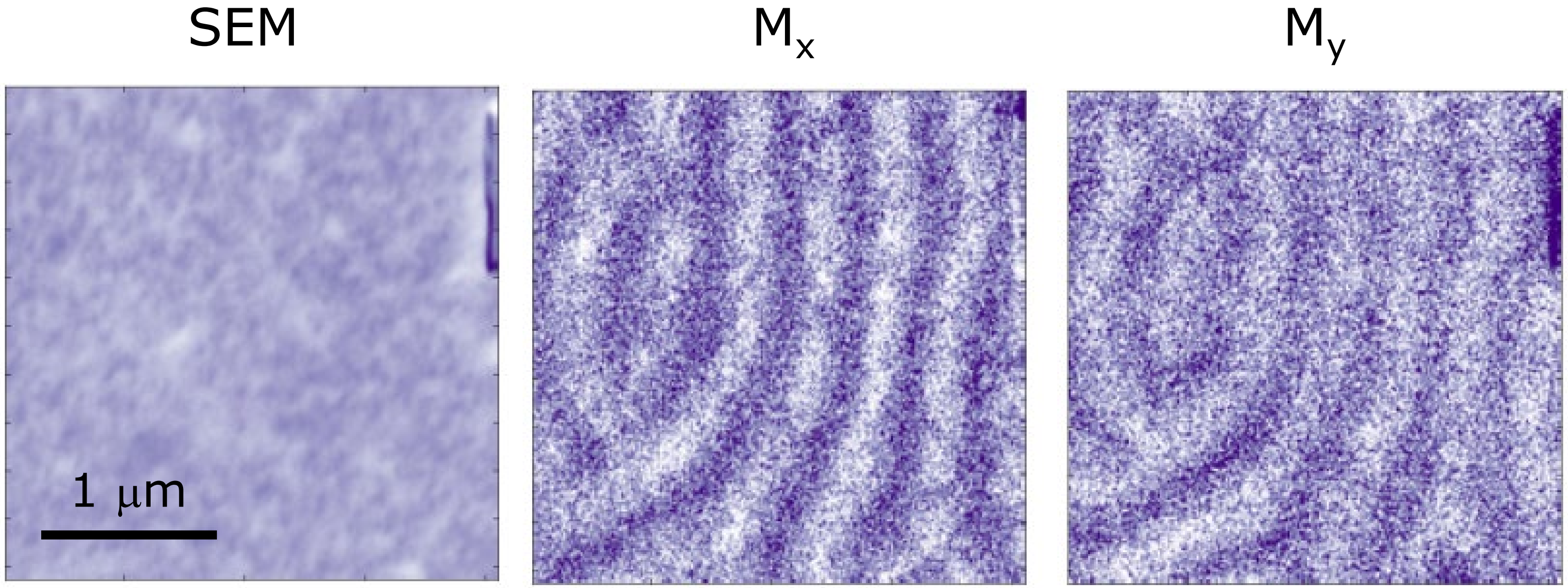
$$N_{sk} = \frac{1}{4\pi} \iint \vec{n} \cdot \left(\frac{\partial \vec{n}}{\partial x} \times \frac{\partial \vec{n}}{\partial y} \right) d^2 r$$



Shao et al., Nat. Comm. **14**, 1355 (2023)

Magnetic Structure of Fe_3GeTe_2




SEM

M_x

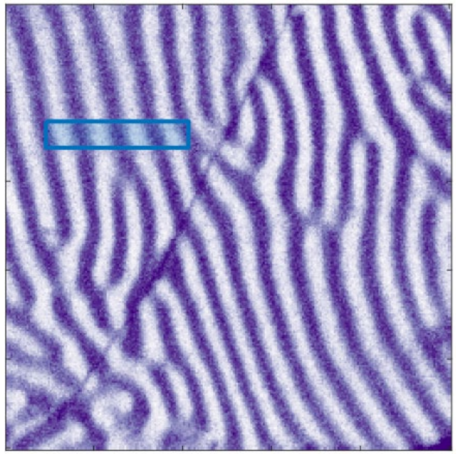
M_y

SEM with
Spin Polarization Sensitivity
(SEMPA)

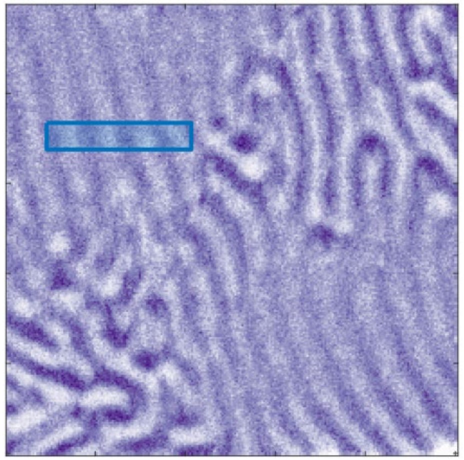
Meijer et al., Nano Lett. **20**, 8563 (2020)

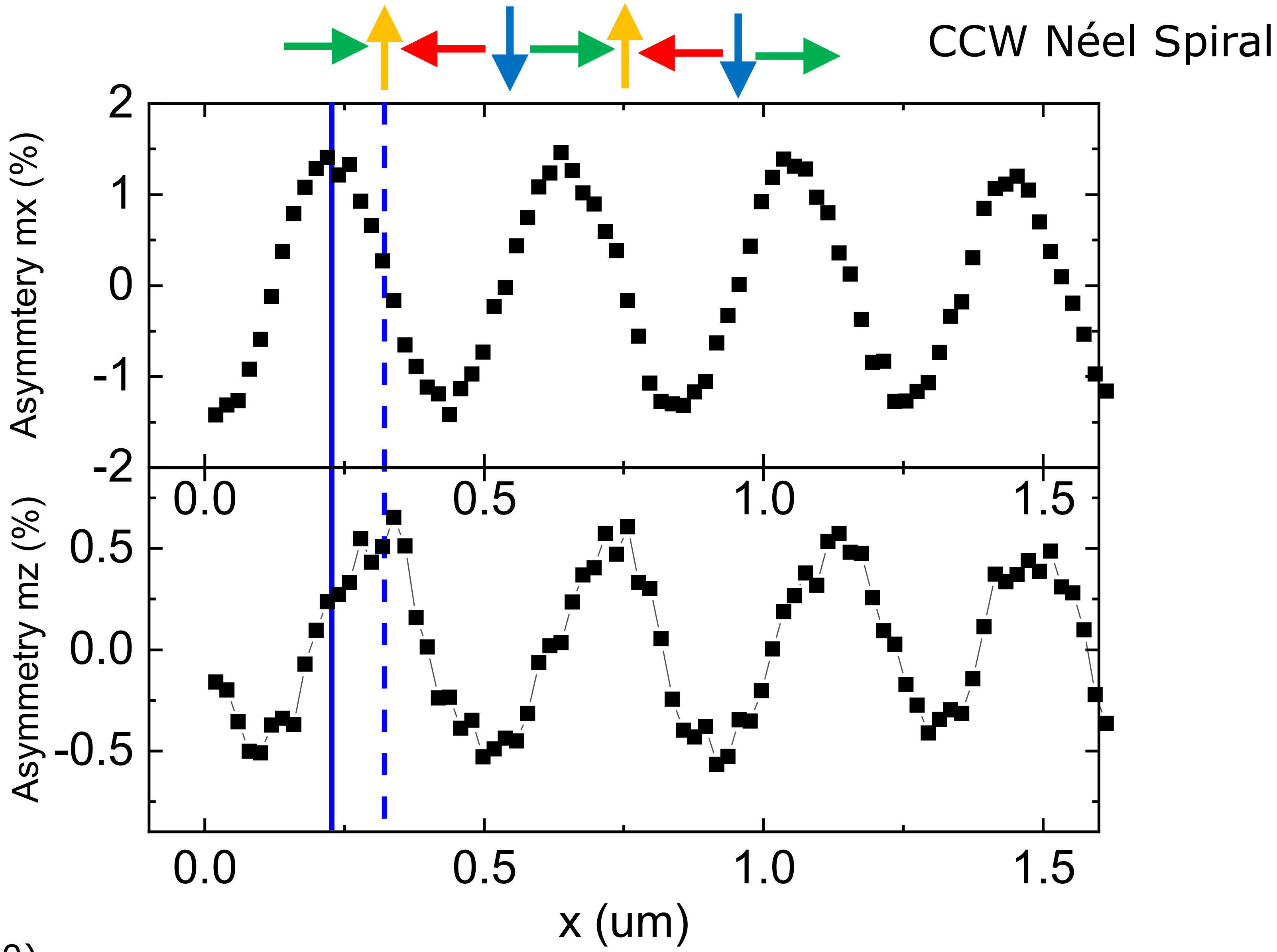
Magnetic Structure of Fe_3GeTe_2

M_x



M_z

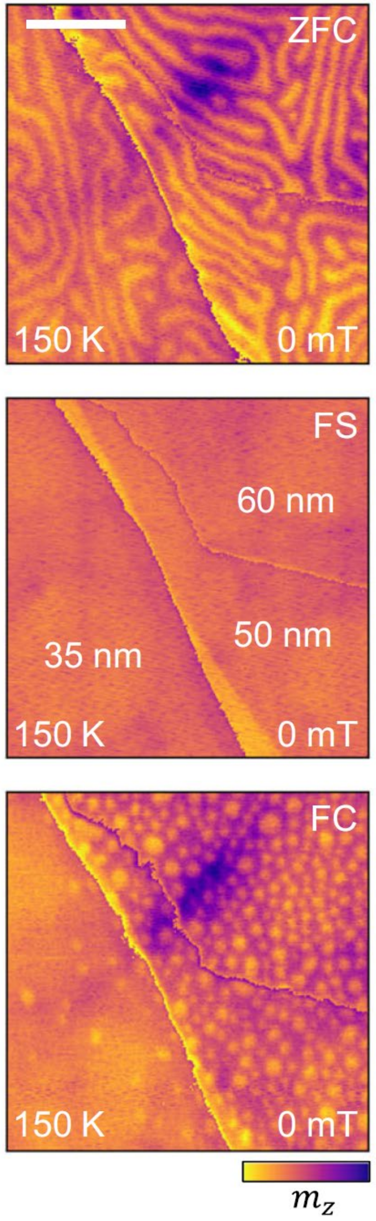




Meijer et al., Nano Lett. **20**, 8563 (2020)

Chiral Spin Textures in Fe_3GeTe_2

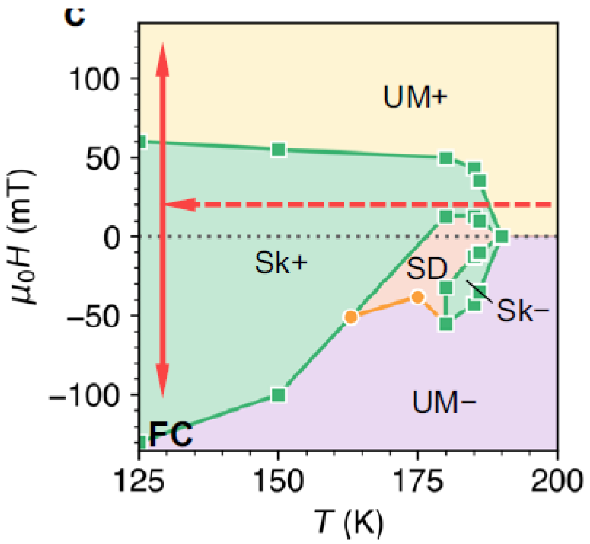
Scanning transmission x-ray microscopy (STXM)



Zero-field cooling: spin spiral state

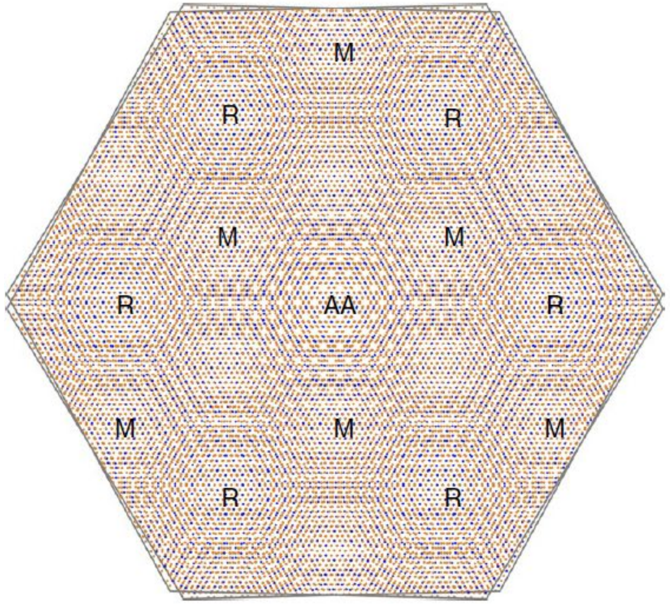
Field sweep: homogeneous magnetization state

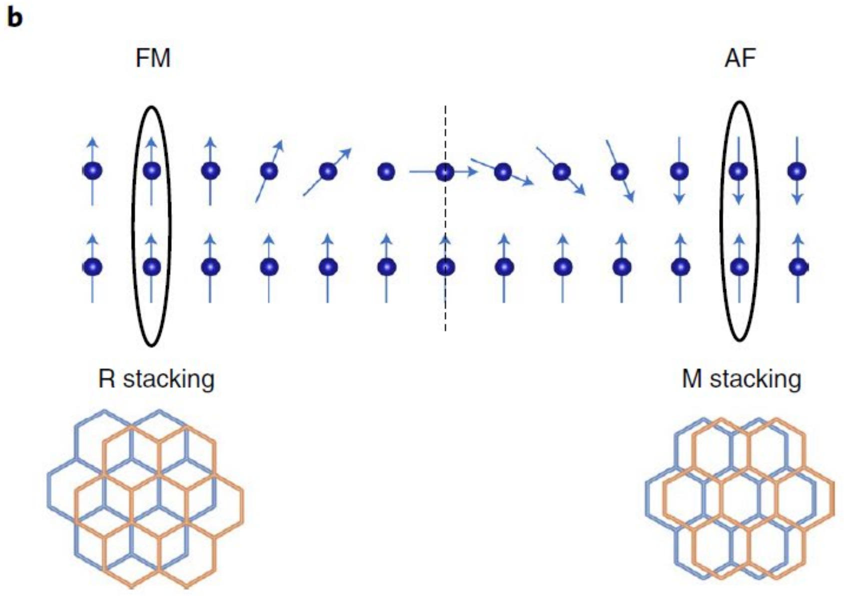
Field cooling: skyrmions

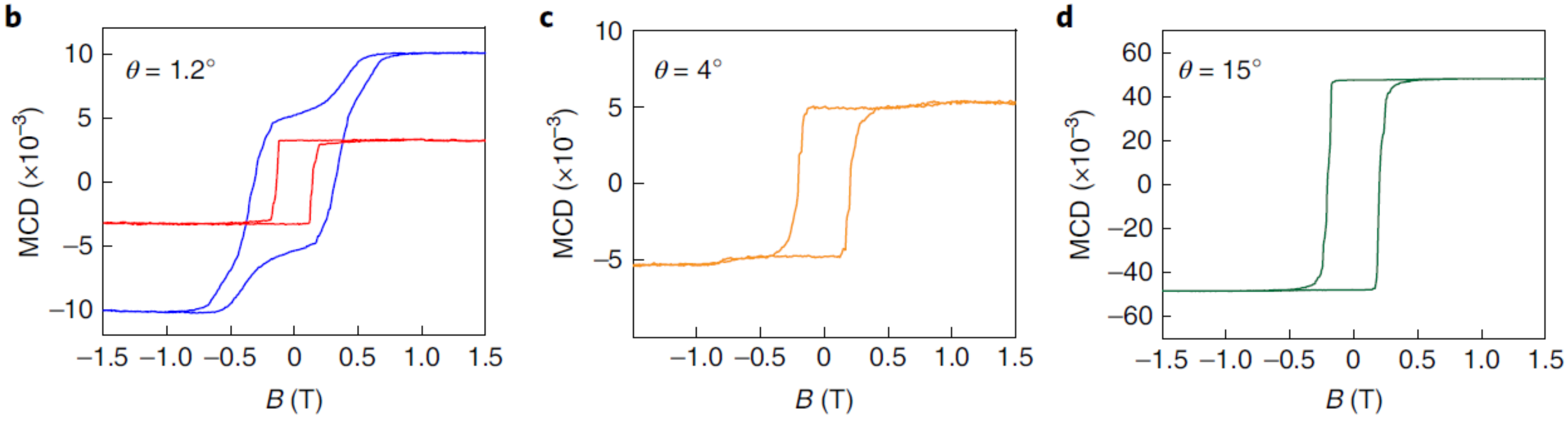


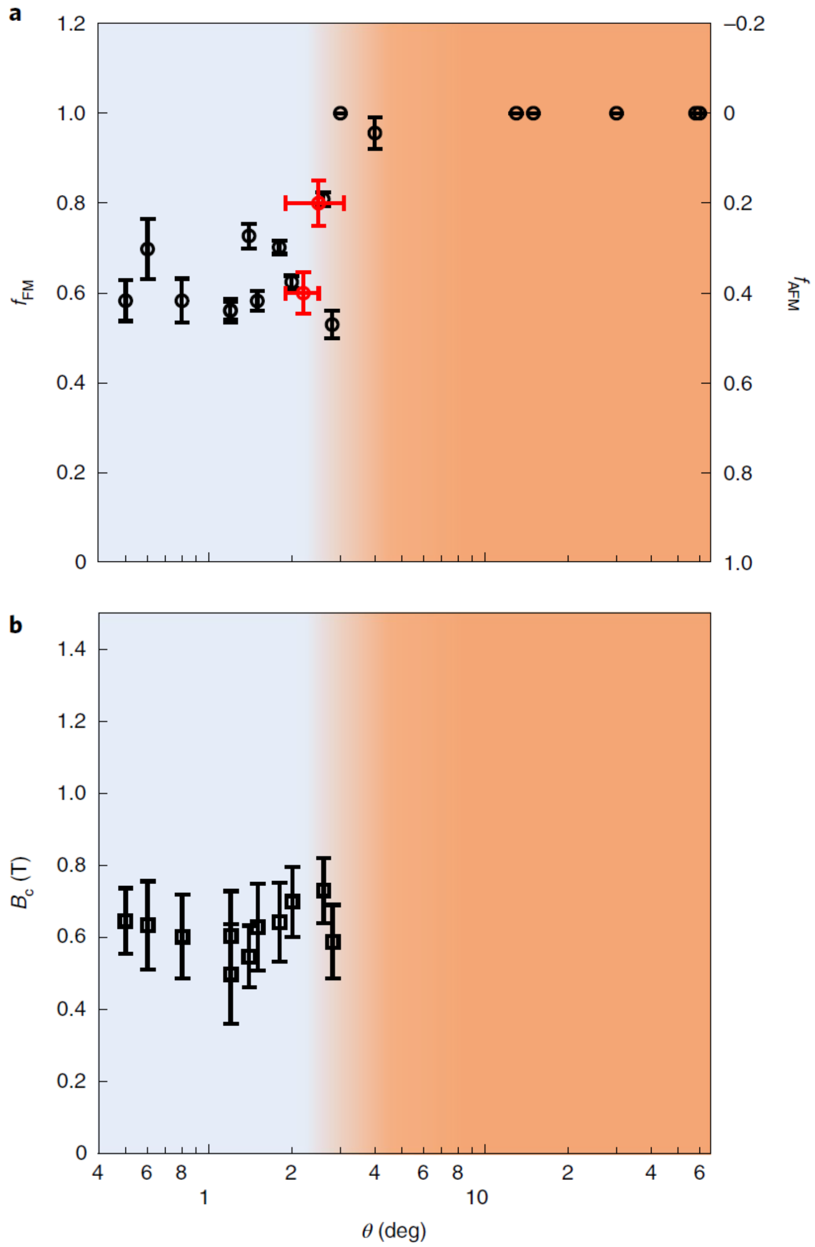
Birch et al., Nat. Comm. **13**, 3035 (2022)

Designed Spin Textures in CrI_3



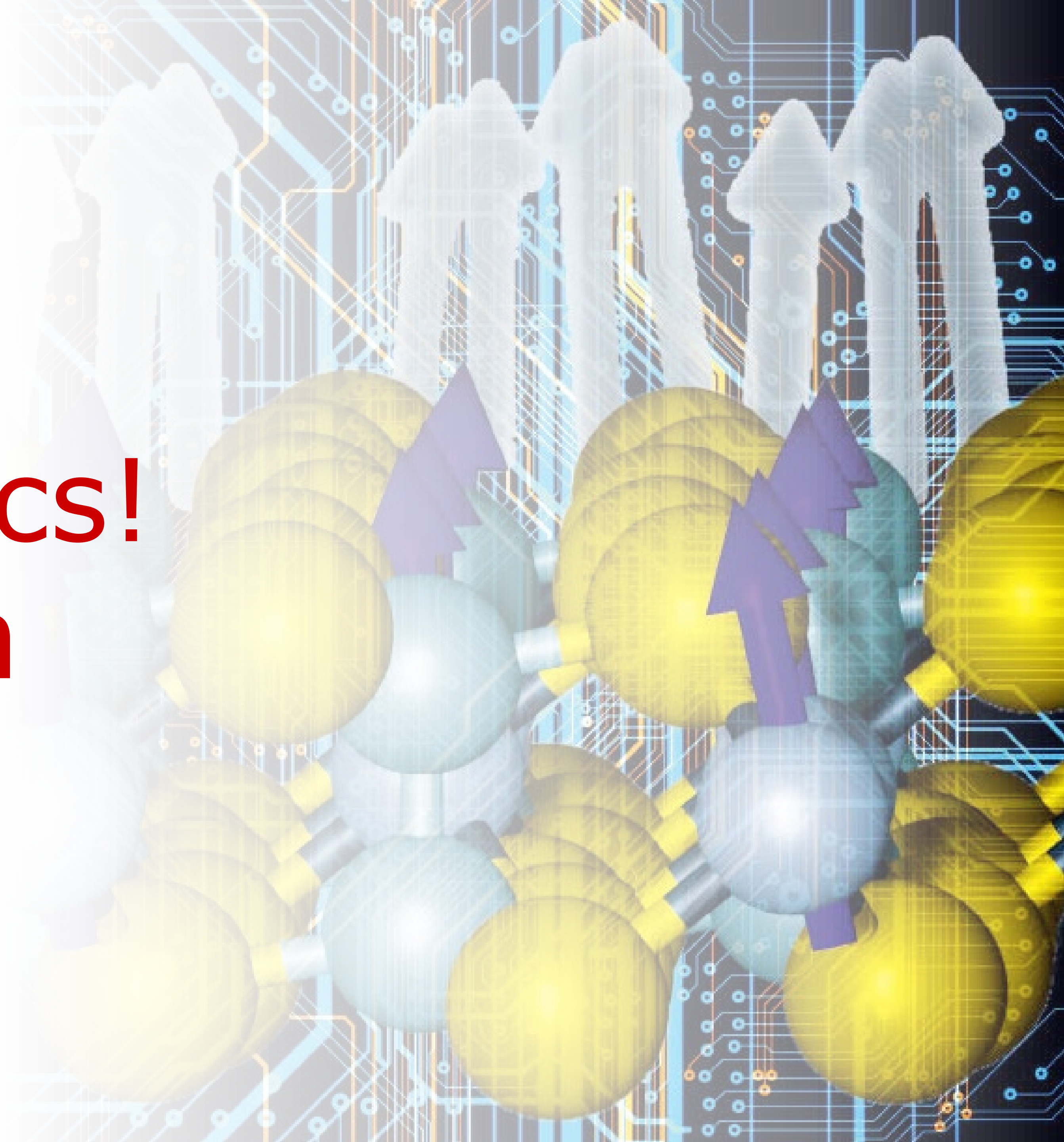






Xu et al., Nat. Nanotech. **17**, 143 (2022)

Away from statics!
Magnetization Dynamics



Ferromagnetic Resonance

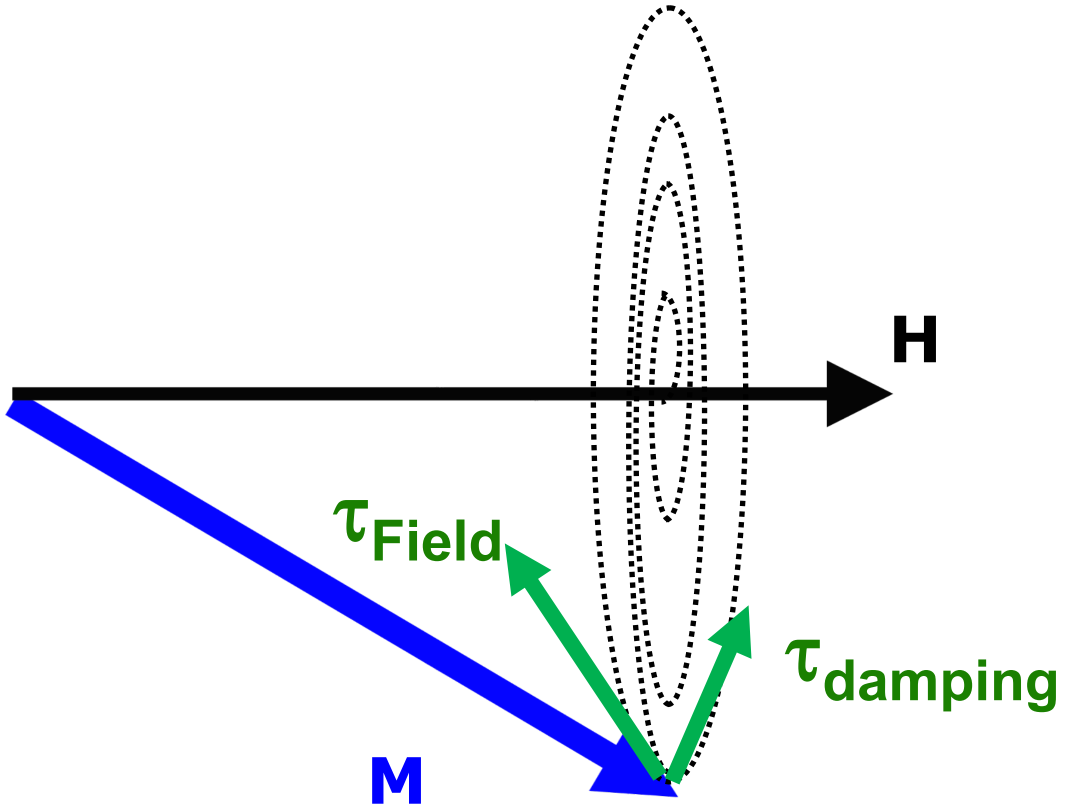
$$\frac{d\boldsymbol{m}}{dt} = -\gamma \boldsymbol{m} \times \boldsymbol{H}_{eff} + \alpha \boldsymbol{m} \times \frac{d\boldsymbol{m}}{dt}$$

Assuming H_{ext} in the z direction and M at an angle θ_M from it:

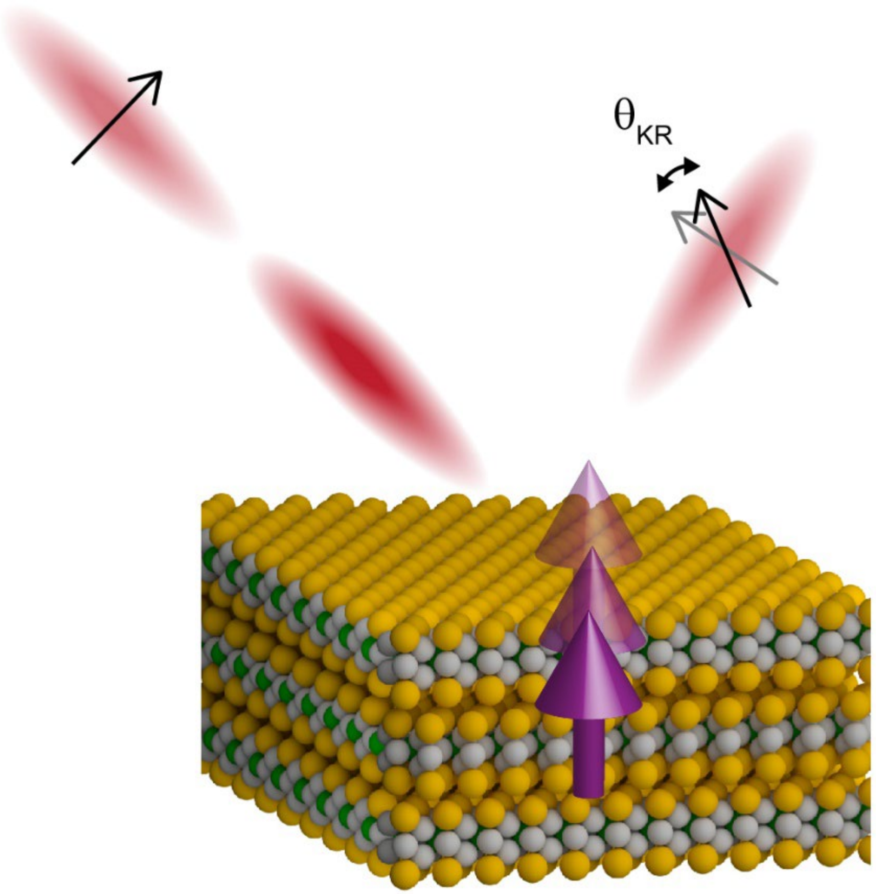
$$f_{res} = \gamma \sqrt{H_{eff}\left(H_{eff} - H_{int}\sin^2(\theta_M)\right)}$$

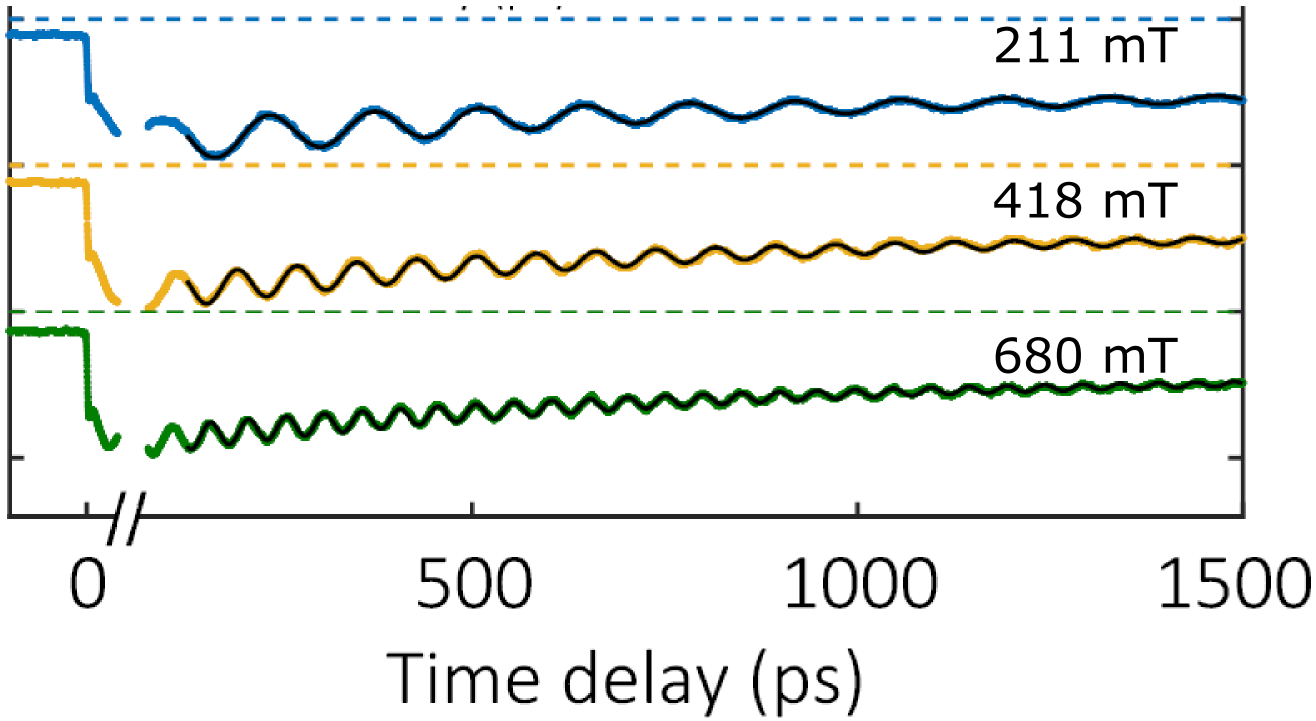
$$\boldsymbol{H}_{\boldsymbol{eff}} = \boldsymbol{H}_{\boldsymbol{ext}} + H_{int}\cos(\theta_M)\,\hat{\boldsymbol{z}}$$

$$H_{int} = \frac{2K_u}{\mu_0 M_s} - M_s$$



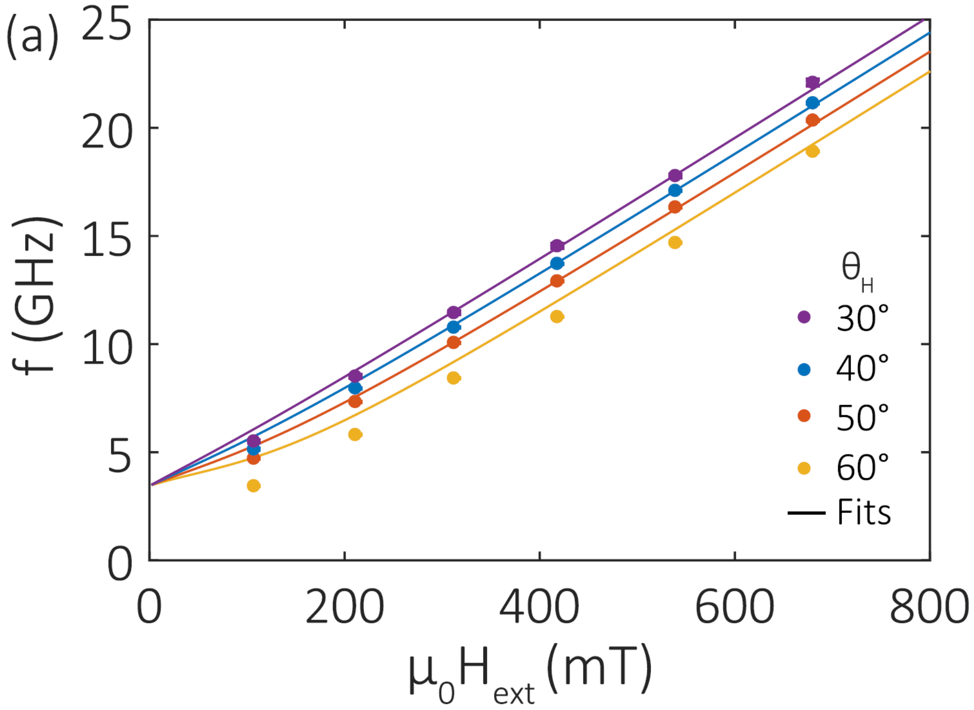
Optical-Excitation of Magnetization Dynamics in a 2D ferromagnet: $Cr_2Ge_2Te_6$

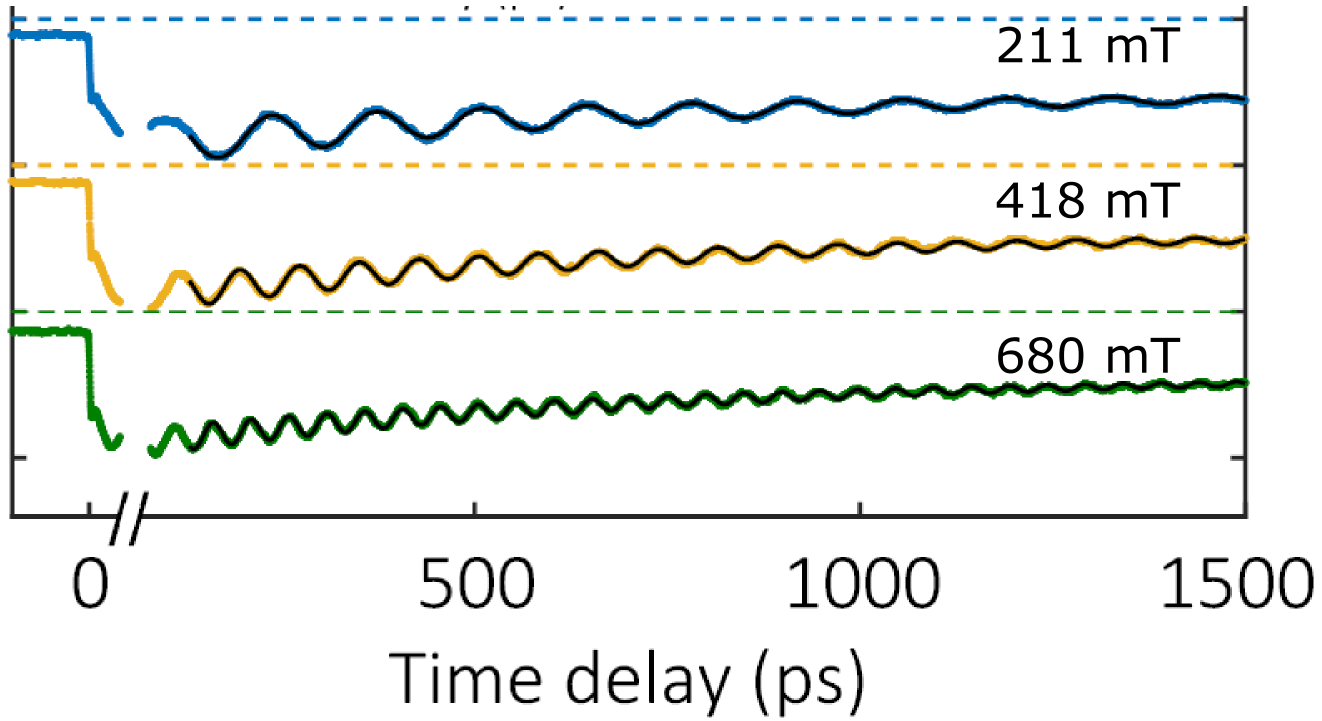




Zhang et al., APL **116**, 223103 (2020)

Optical-Excitation of Magnetization Dynamics in a 2D ferromagnet: $Cr_2Ge_2Te_6$

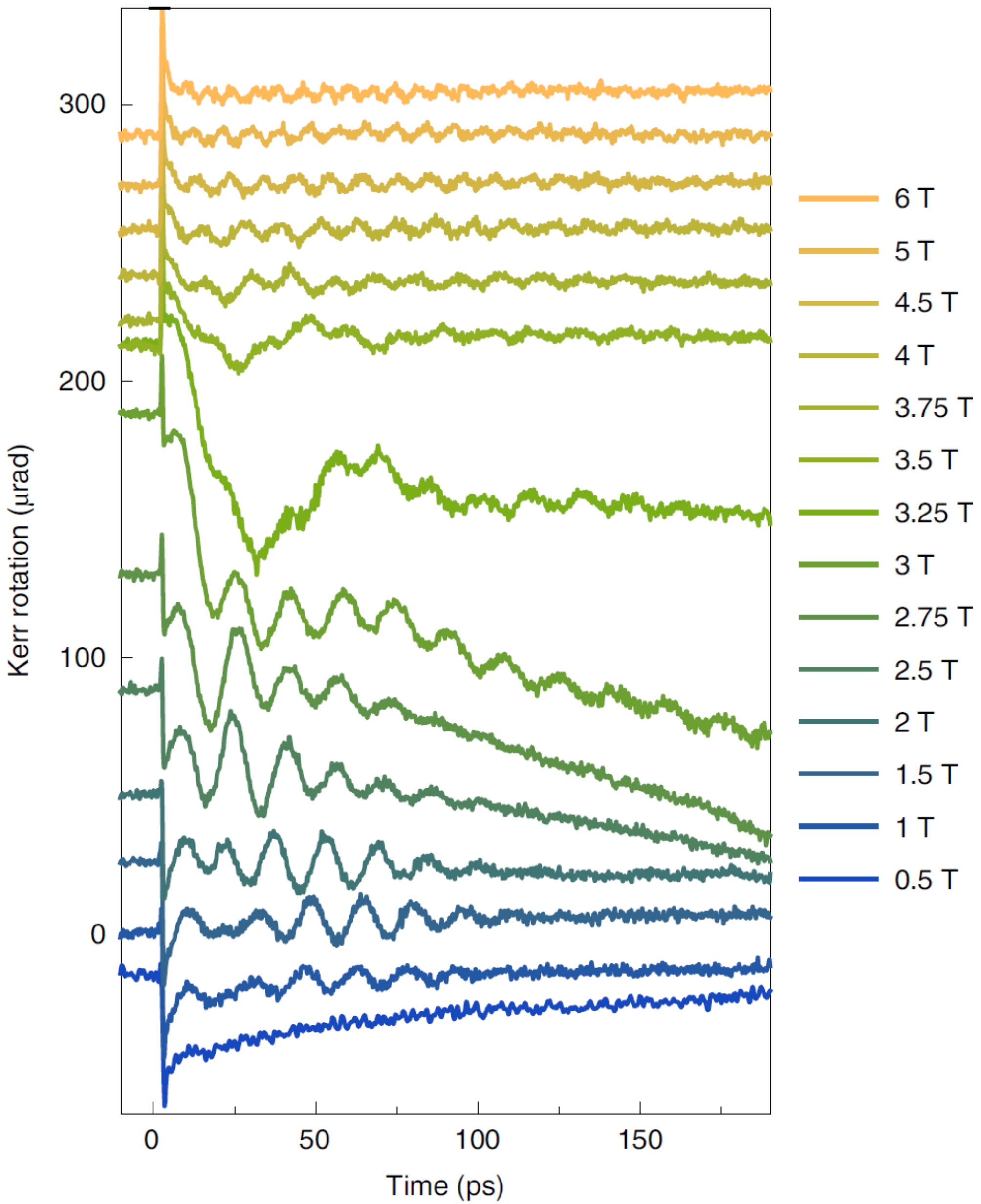


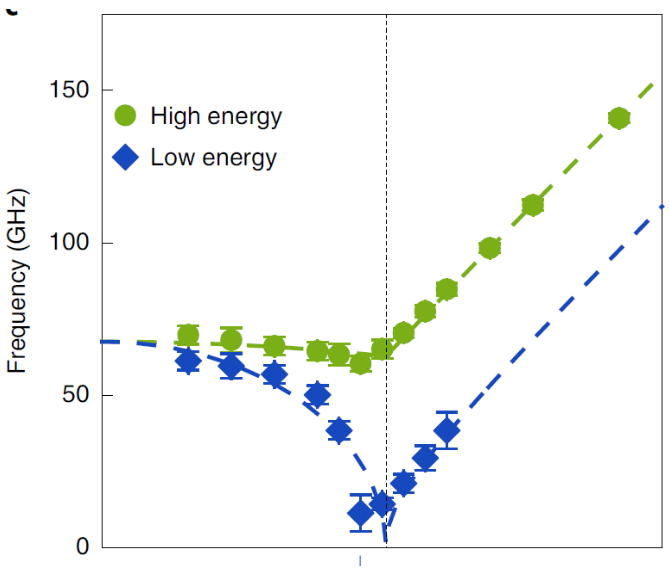


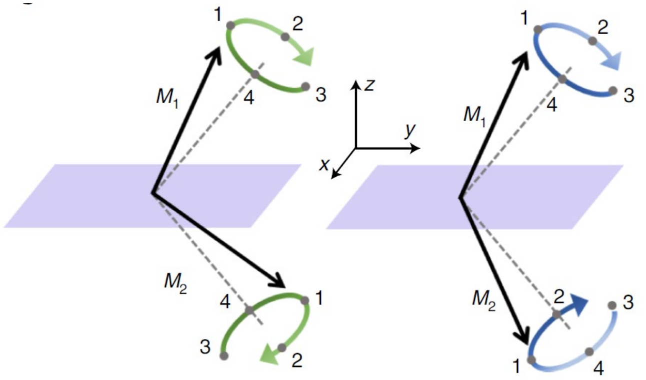
$$M_s \approx 192\ \mathrm{emu/cm^3}$$
$$K_u \approx 3.4 \times 10^5\ \mathrm{erg/cm^3}$$
$$\alpha \approx 0.005$$

Zhang et al., APL **116**, 223103 (2020)

Magnetization Dynamics in bilayer CrI_3







Zhang et al., Nature Nanotech. **19**, 838 (2020)

What happens?

What changes upon doping of the system?

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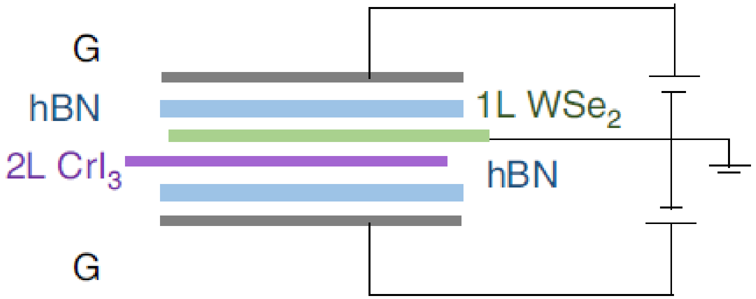
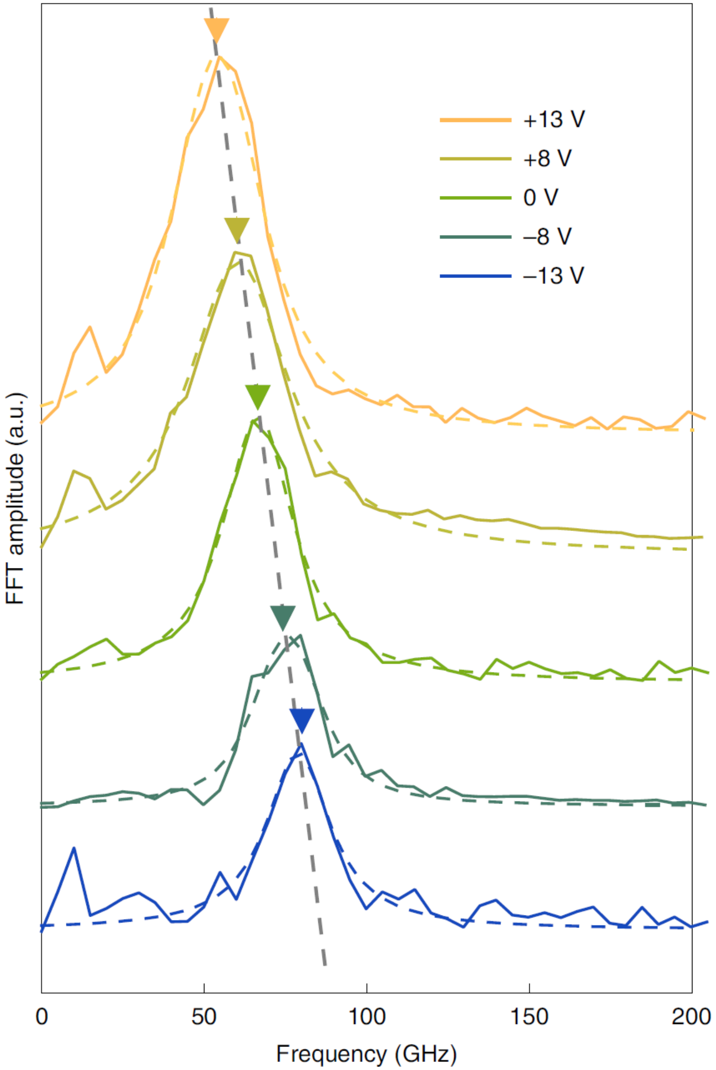
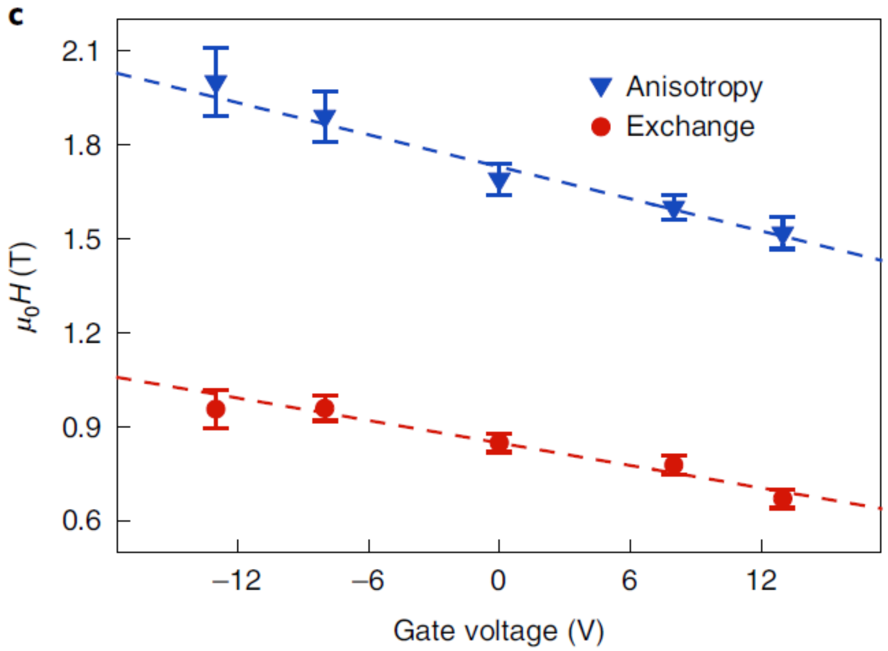
A – Anisotropy and exchange change, hence the frequency changes.
B – Only the anisotropy changes, hence the amplitude of oscillation changes.
C – Only the exchange changes, hence the frequency changes.
D – None of the above.

Electric Control Over the Magnetization Dynamics

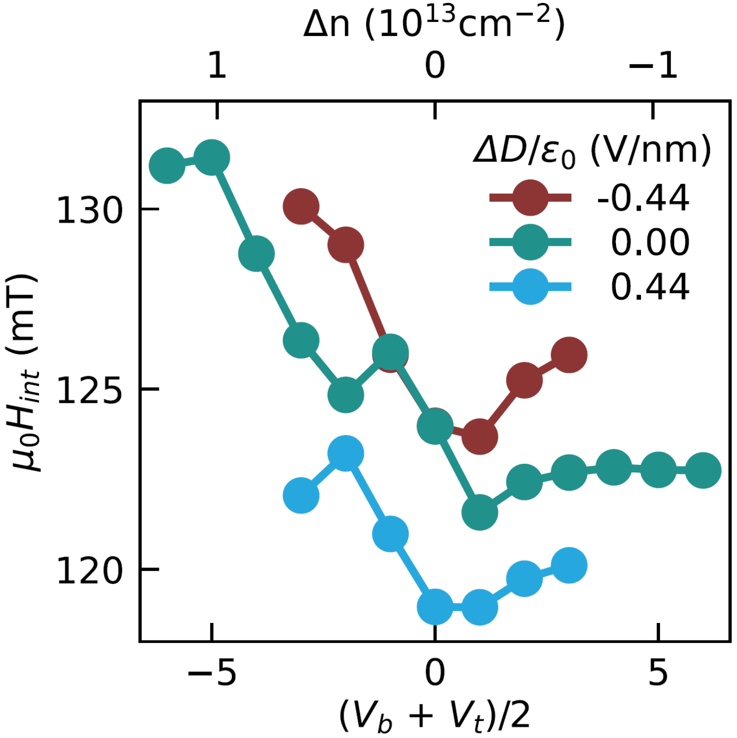


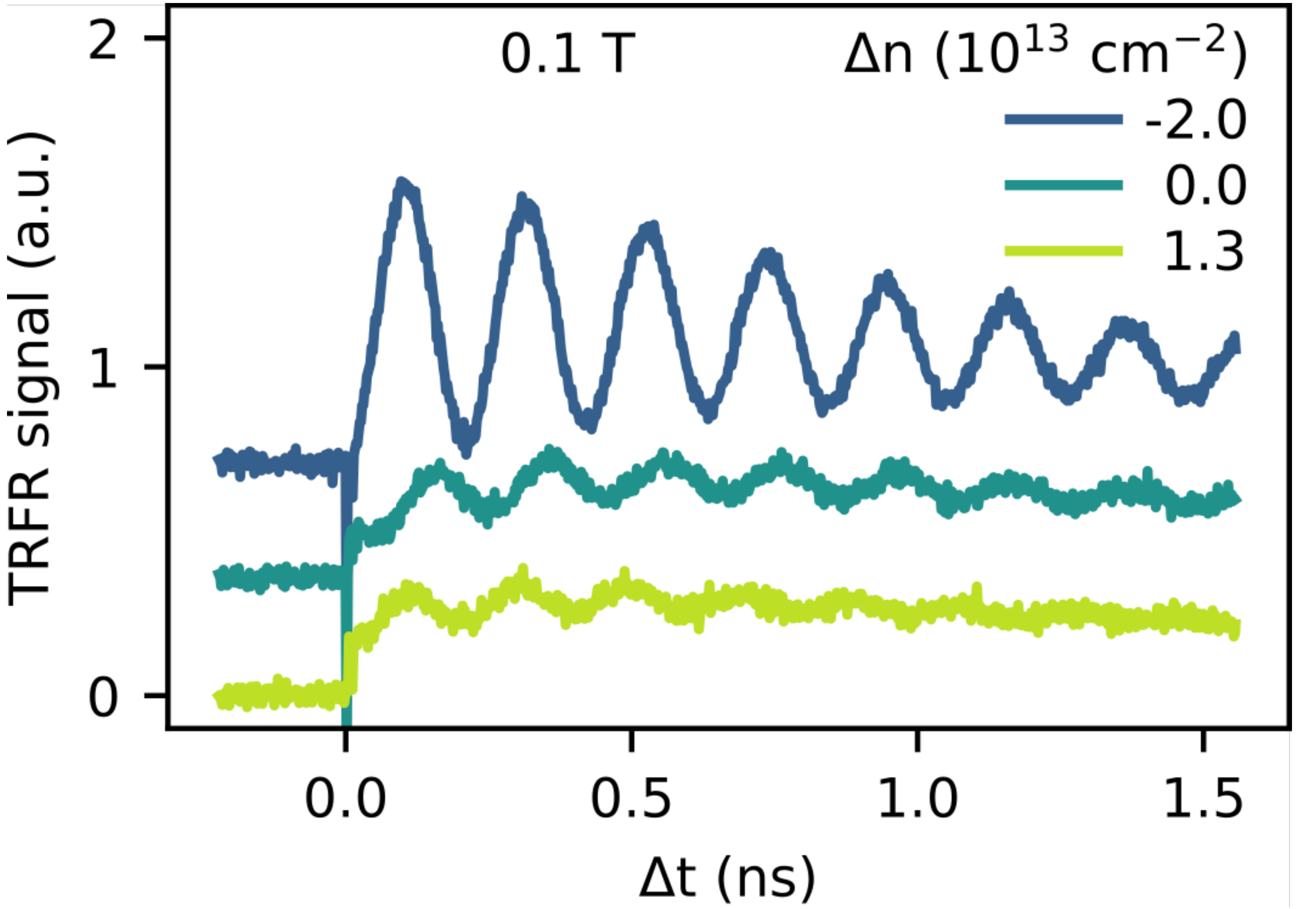




Zhang et al., Nature Nanotech. **19**, 838 (2020)

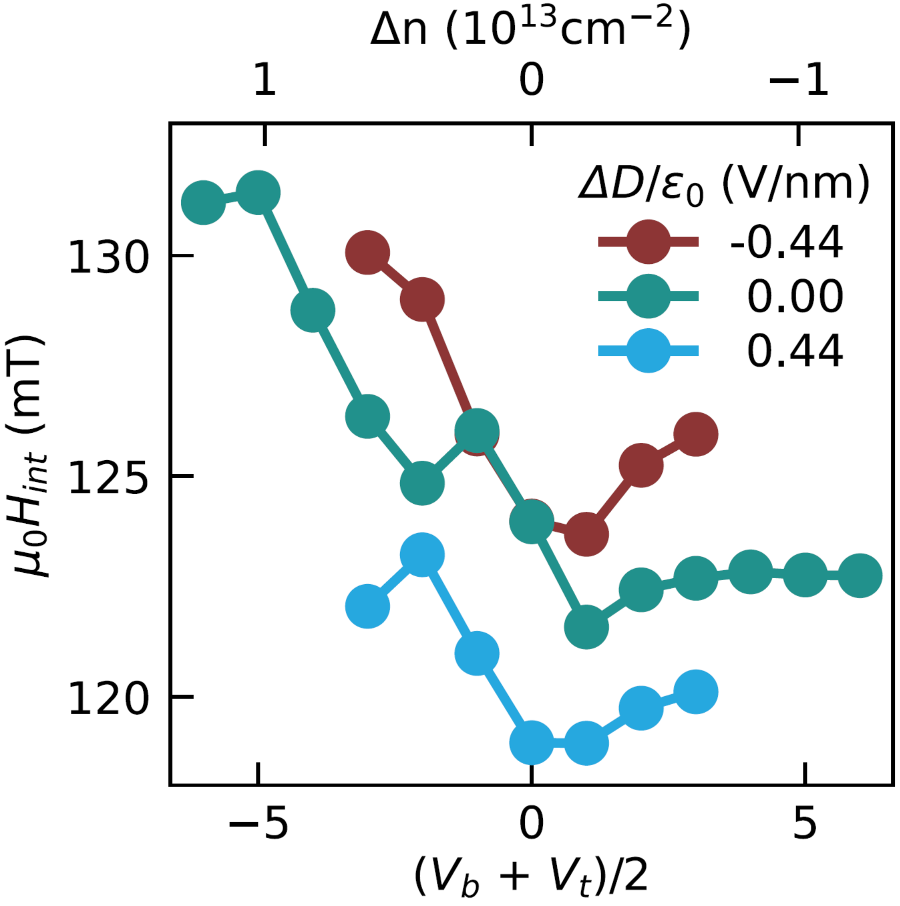
Electric Control Over the Magnetization Dynamics

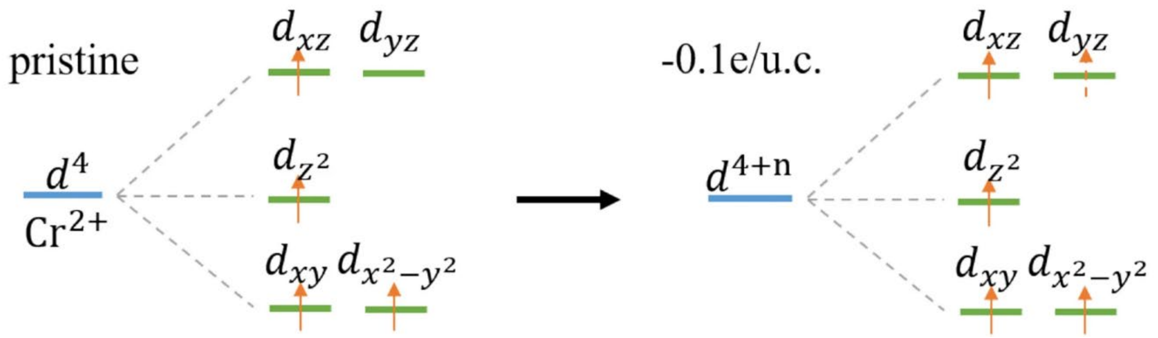


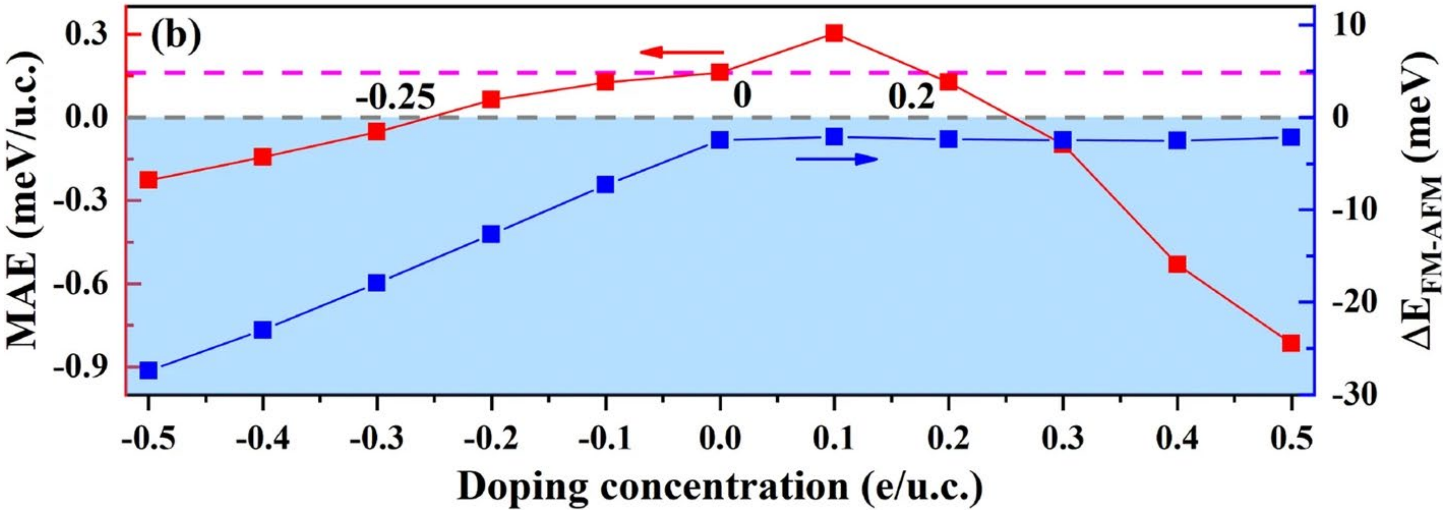


Hendriks et al., Nat. Comm. **15**, 1298 (2024)

Electric Control Over the Magnetization Dynamics





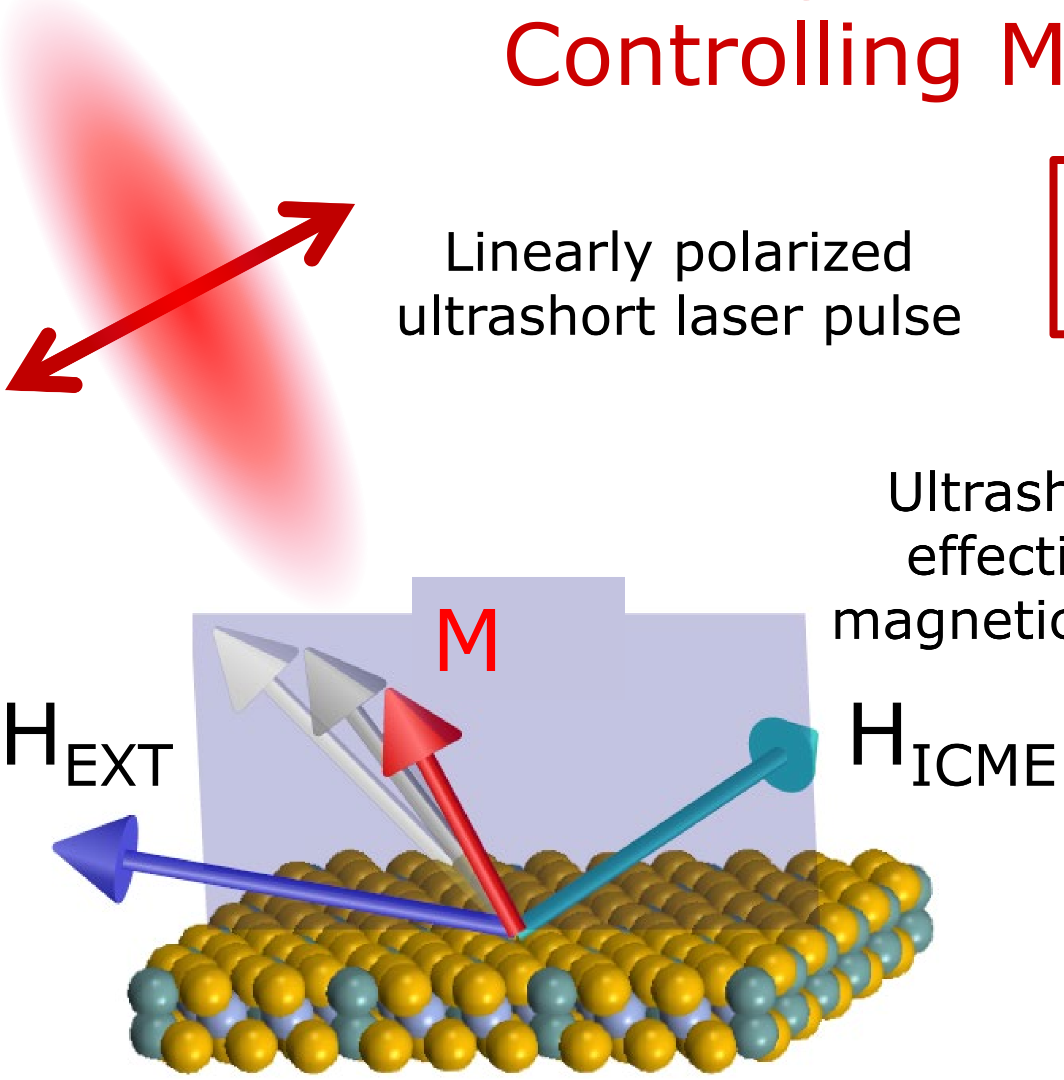


Hendriks et al., Nat. Comm. **15**, 1298 (2024)

Ren et al., Sci. Rep. **11**, 2744 (2021)

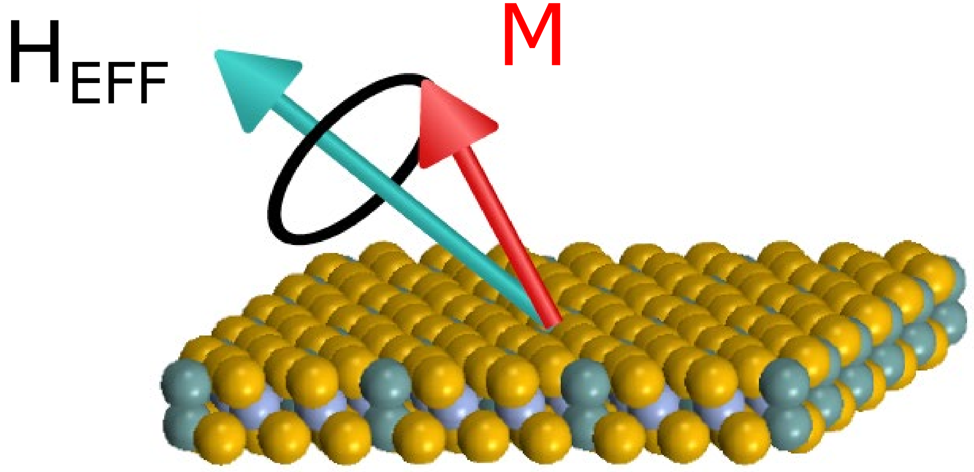
2D Magnets for Opto-Magnetics: Controlling Magnetism with Light

Coherent Opto-Magnetics:
Inverse Cotton-Mouton Effect



Linearly polarized ultrashort laser pulse

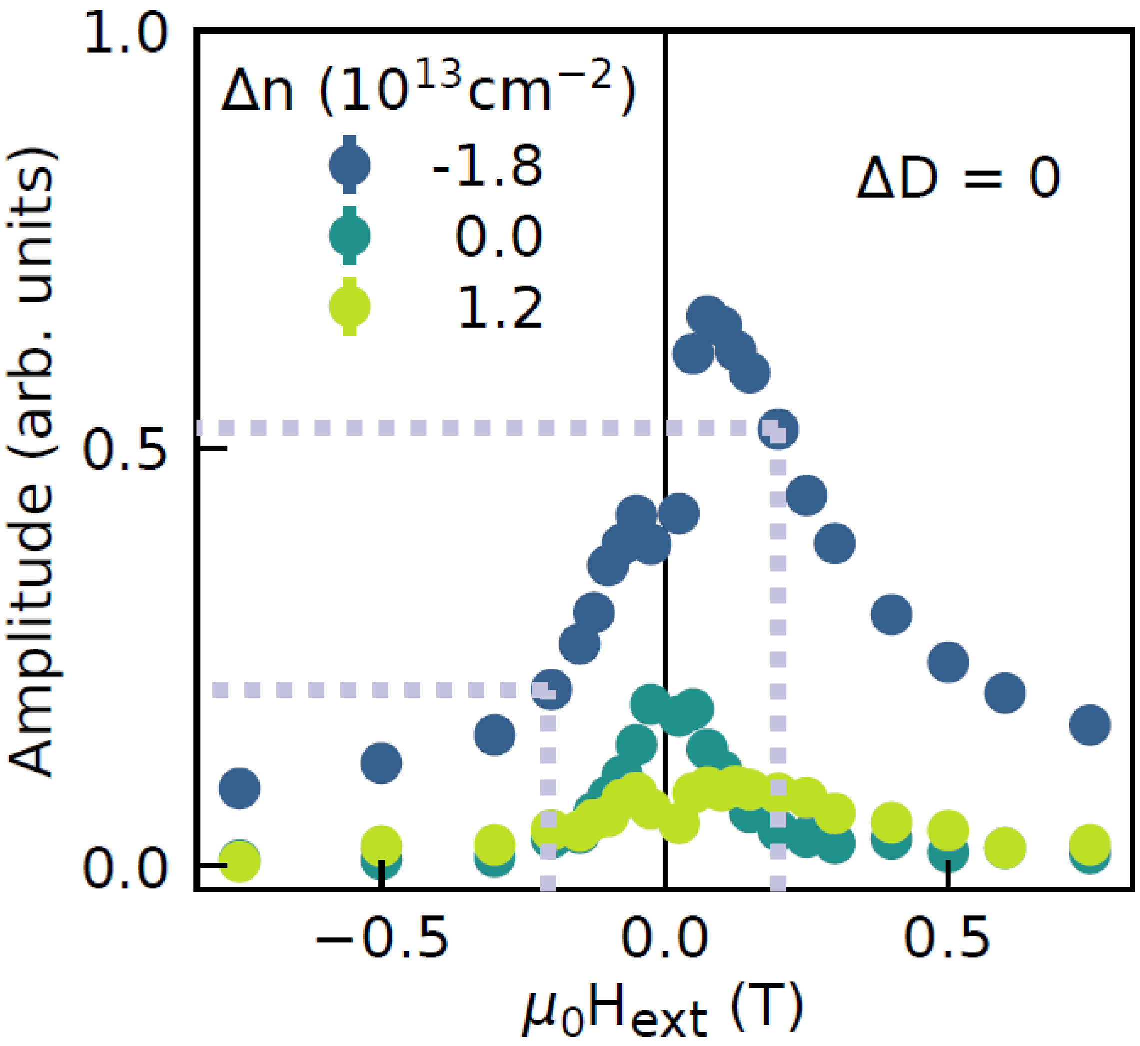
Ultrashort effective magnetic field

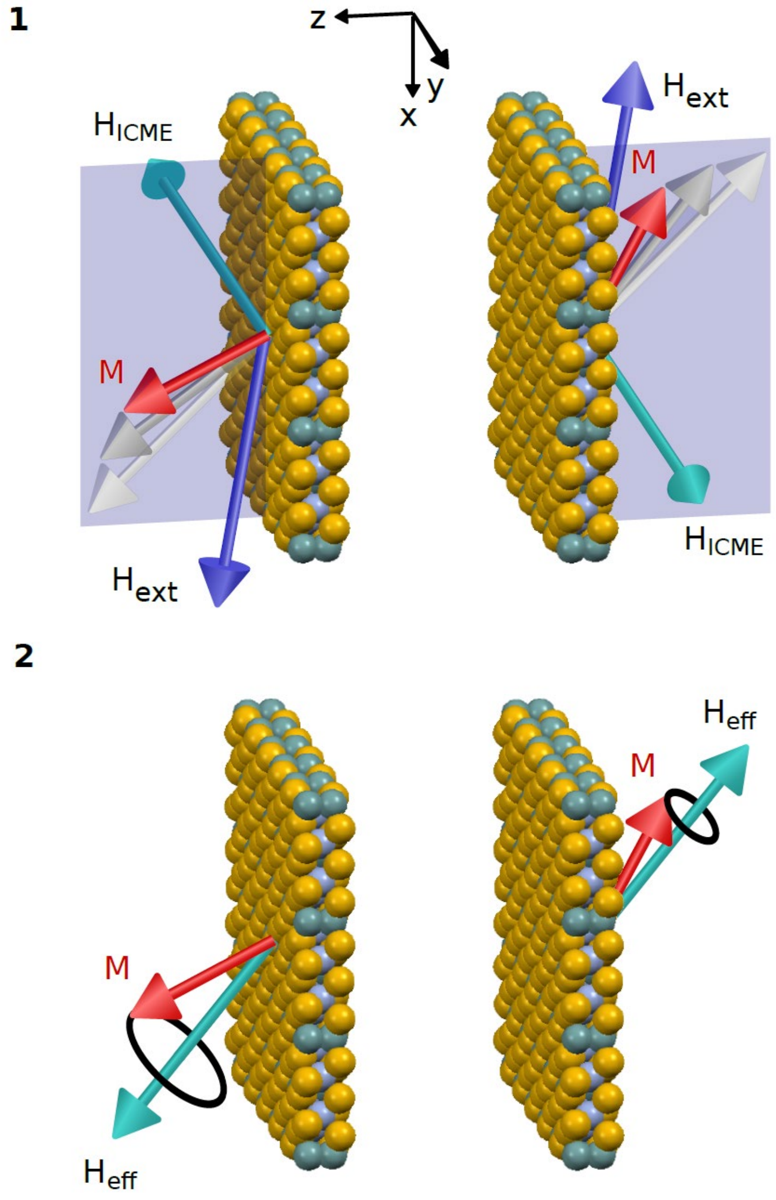


Magnetization is left to precess around the field

Hendriks et al., Nat. Comm. **15**, 1298 (2024)

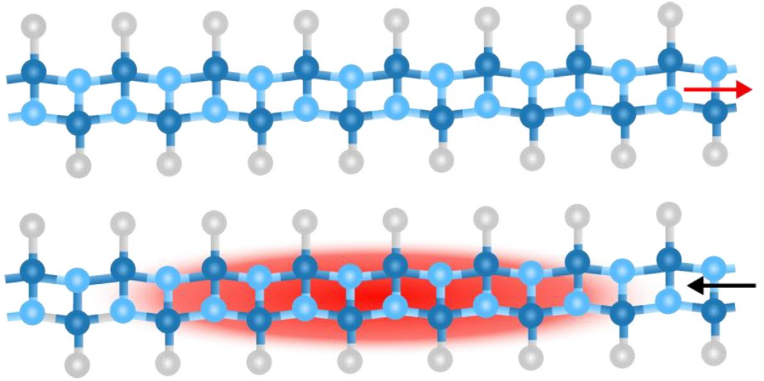
Electric control over opto-magnetic effects



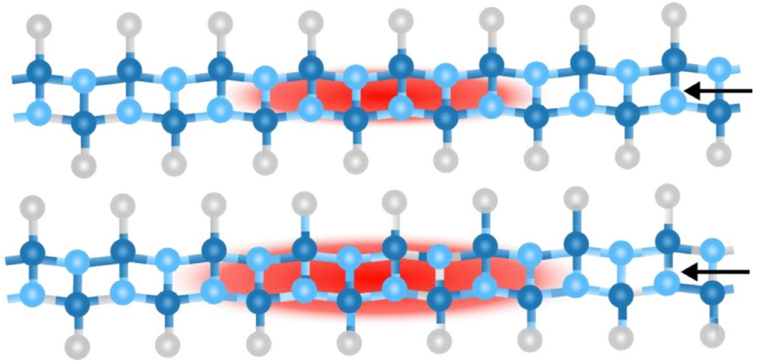


Hendriks et al., Nat. Comm. **15**, 1298 (2024)

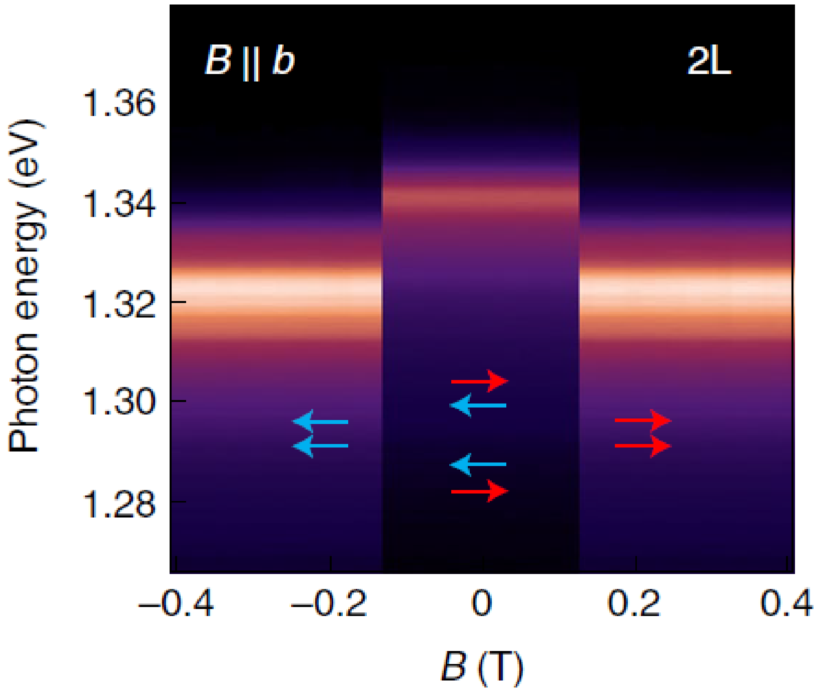
Excitons in CrSBr

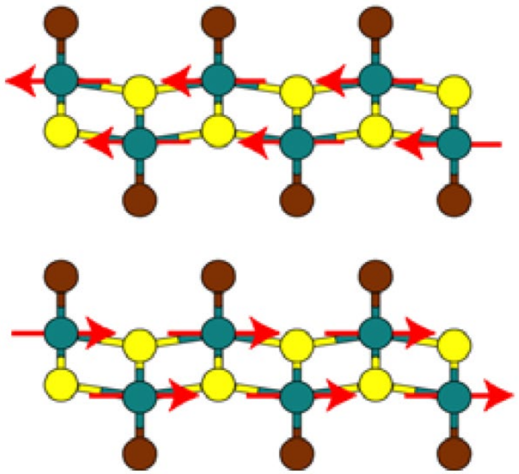


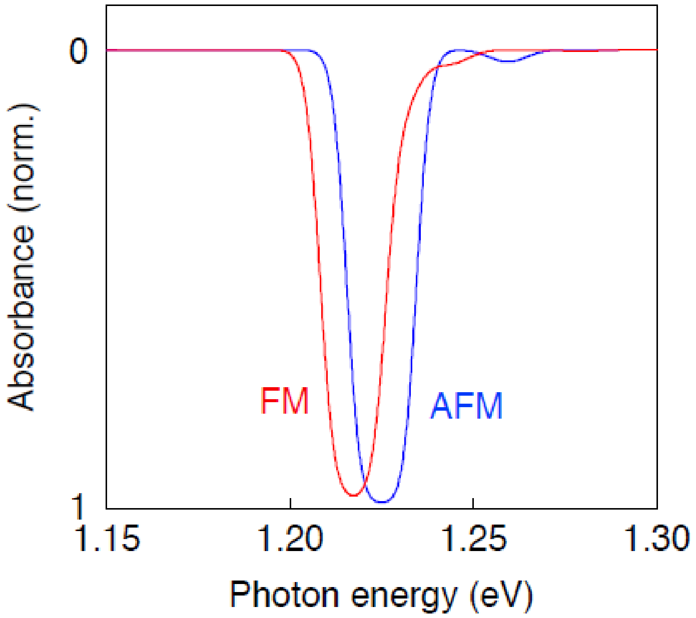
AFM ordering
Excitons localized within layer



FM ordering
Excitons delocalized between layers

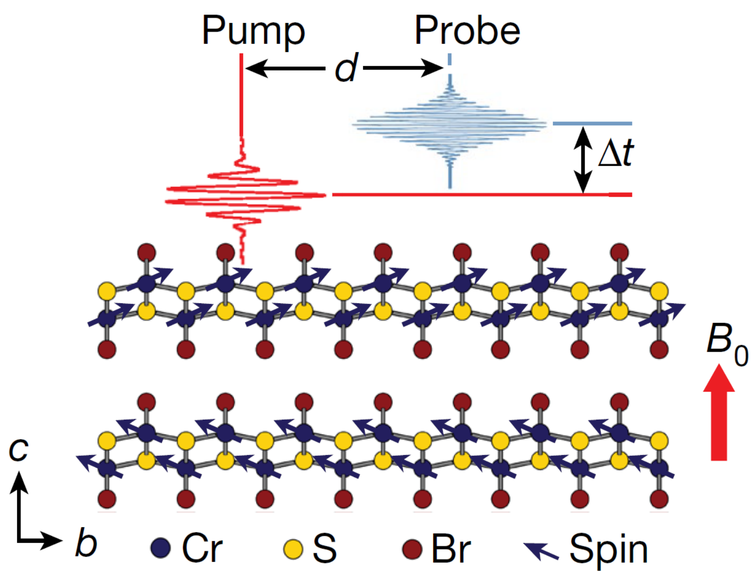


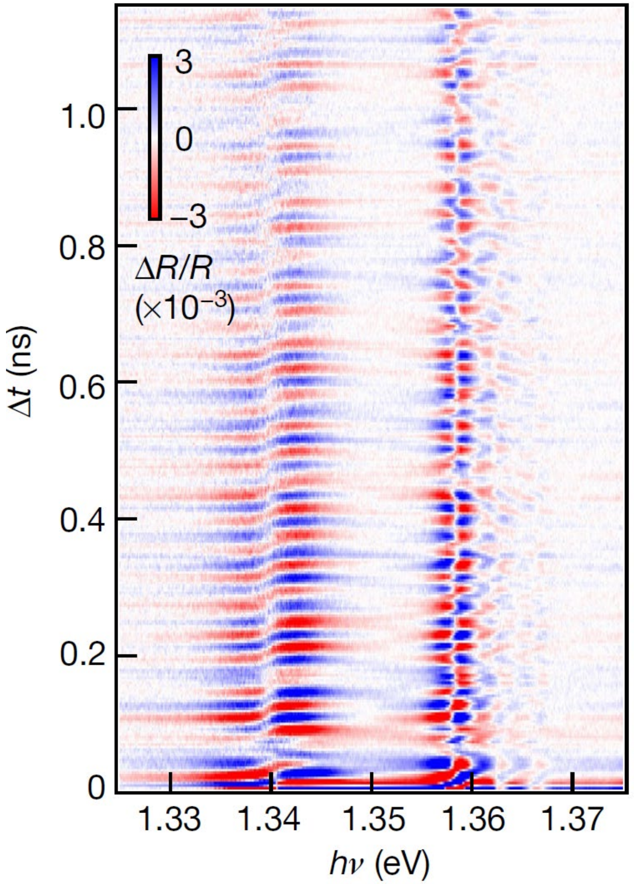


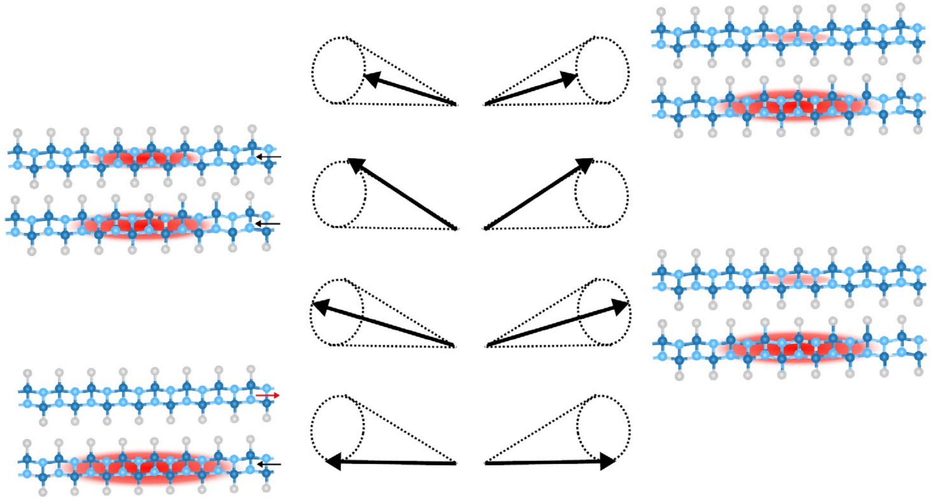


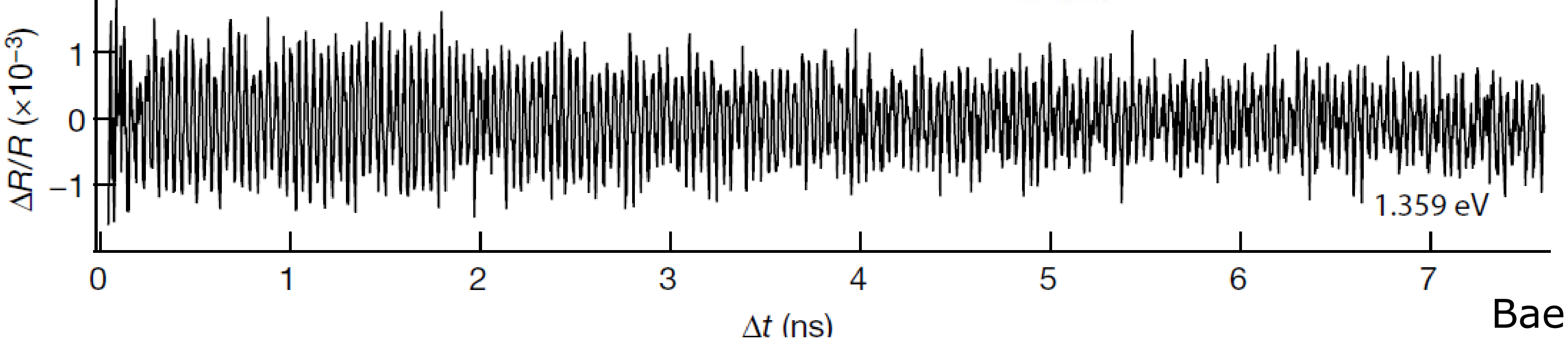
Wilson et al., Nature Mat. **20**, 1657 (2021)

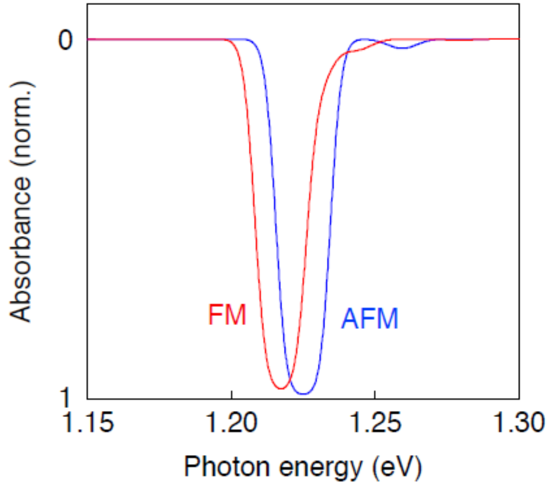
Magnon-Exciton Coupling











Bae et al., Nature **609**, 282 (2022)

2D Materials: A (very!) large family

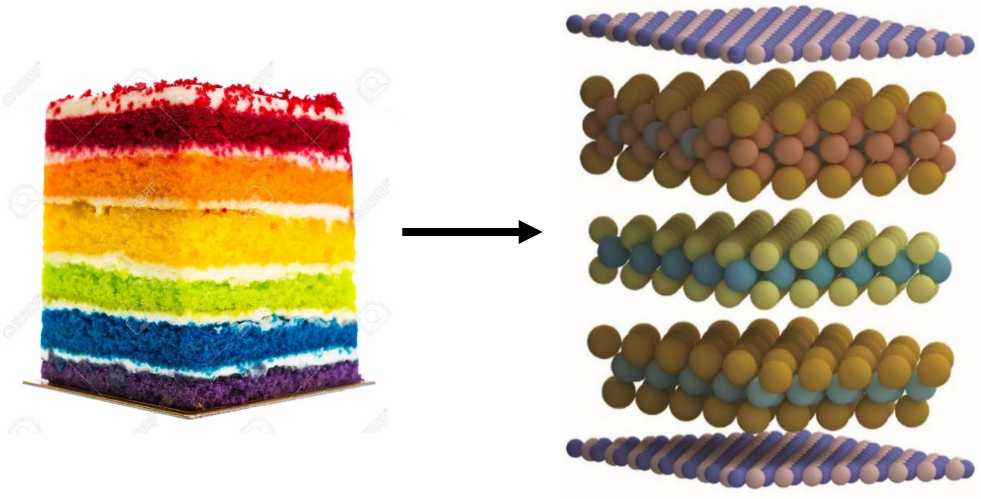
Over 2500 different materials!

Metals, Insulators, Semiconductors,

Superconductors, Semi-Metals,

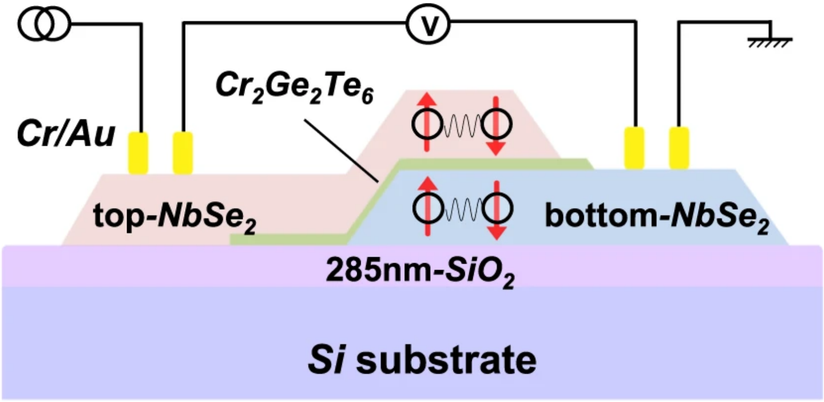
Topological Insulators,

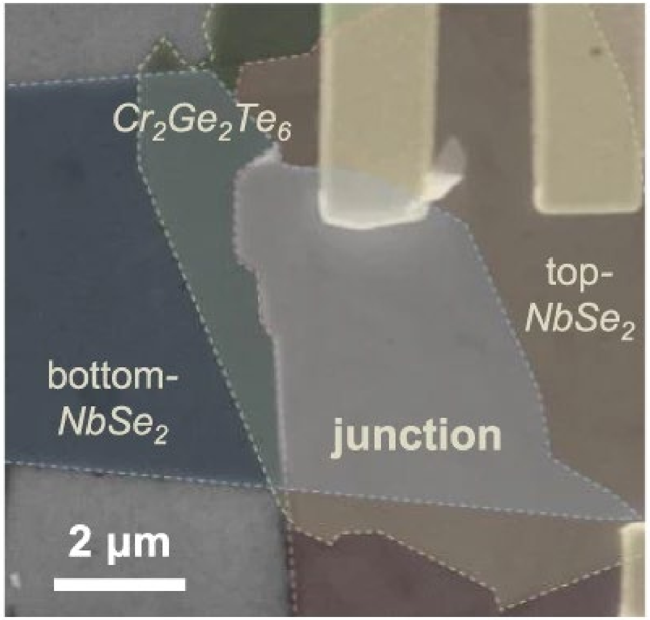
Ferroelectrics, Magnets, …



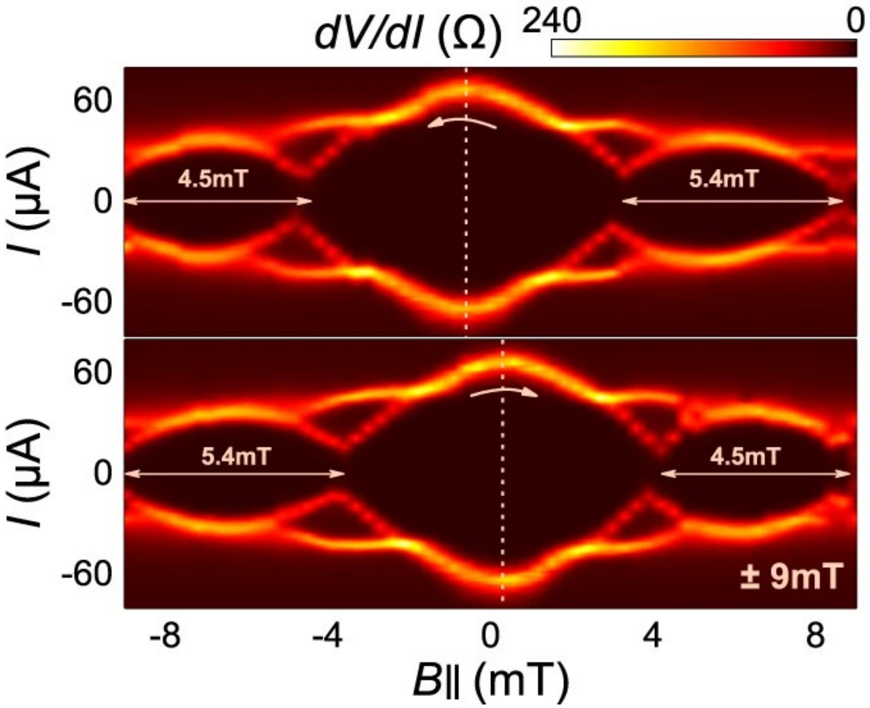
New physics and emergent phenomena → New possibilities!

Combining magnetism and superconductivity





Hysteretic Fraunhofer patters in a SC/FM/SC van der Waals heterostructure.



Ai et al., Nat. Comm. **12**, 6580 (2021)

Thanks for the invitation!

Hope you enjoyed!