

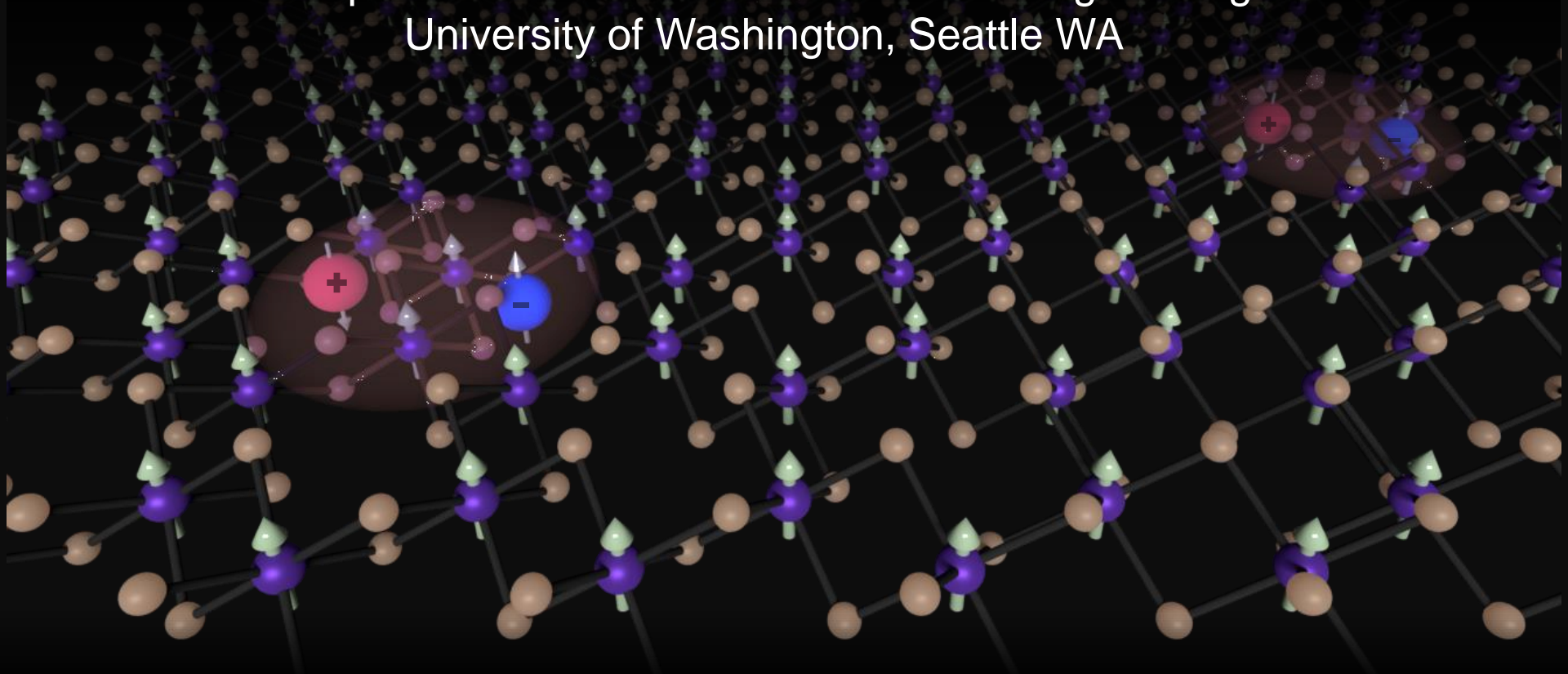
2D Magnets and Heterostructures

Xiaodong Xu

Department of Physics

Department of Materials Science and Engineering

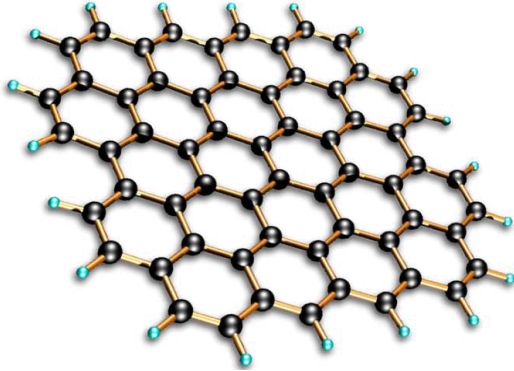
University of Washington, Seattle WA



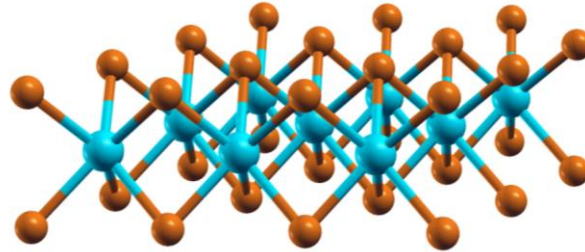
290K Condensed Seminar, UC Berkeley April 2018

New 2D Electronic Systems

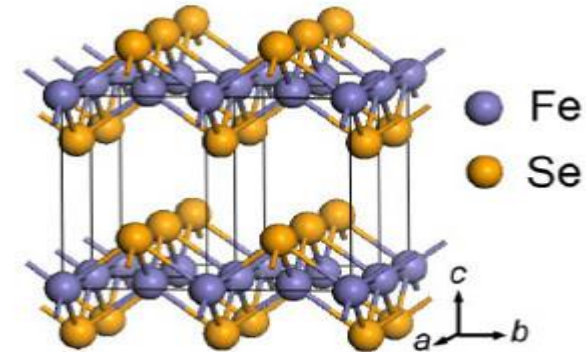
Graphene



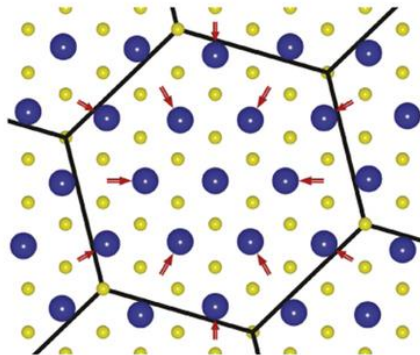
2D semiconductor (MX_2)



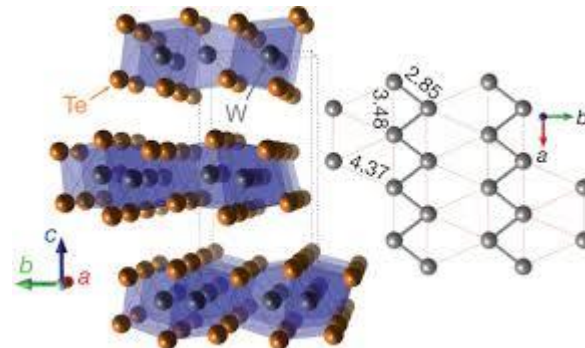
2D Superconductor
(FeSe, NbSe₂)



Tunable Strongly-
correlated system (TaS_2 ,
 TiSe_2)



Quantum Spin Hall
Insulator (WTe_2)



Two-Dimensional Magnets

Digital Data Storage



Google data center



History of 2D Magnetic Materials

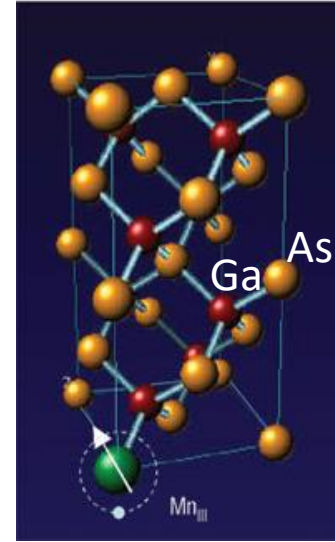
I. There was no monolayer semiconductor/isulator with *intrinsic* magnetism.

II. Monolayer metals with intrinsic magnetism indeed exist

Magnetic properties hinges on substrate properties and interface quality.

(monolayer Co/Cu, Ni/Cu)

Isolated 2D magnets did not exist



Dilute magnetic semiconductor quantum well

Emergence of Van der Waals Magnets

McGuire et al. Coupling of Crystal Structure and Magnetism in the Layered, Ferromagnetic Insulator CrI₃. *Chemistry of Materials* **27**, 612-620.(2015).

Zhang et al., Robust intrinsic ferromagnetism and half semiconductivity in stable two-dimensional single-layer chromium trihalides. *Journal of Materials Chemistry C* **3**, 12457-12468 (2015).

Li et al., CrXTe₃ (X = Si, Ge) nanosheets: two dimensional intrinsic ferromagnetic semiconductors. *Journal of Materials Chemistry C* **2**, 7071-7076.(2014).

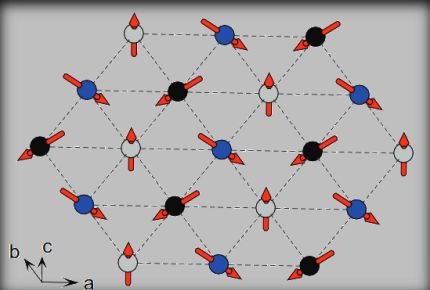
Lin et al., Ultrathin nanosheets of CrSiTe₃: a semiconducting two-dimensional ferromagnetic material. *Journal of Materials Chemistry C* **4**, 315-322.(2016).

Sivadas et al., D. Magnetic ground state of semiconducting transition-metal trichalcogenide monolayers. *Physical Review B* **91**, 235425.(2015).

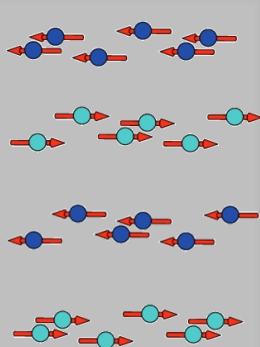
Wang et al, Raman spectroscopy of atomically thin two-dimensional magnetic iron phosphorus trisulfide (FePS₃) crystals. *2D Materials* **3**, 031009.(2016).

Layered Transition Metal Halides

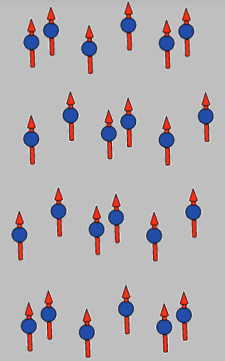
VCl_2, VBr_2



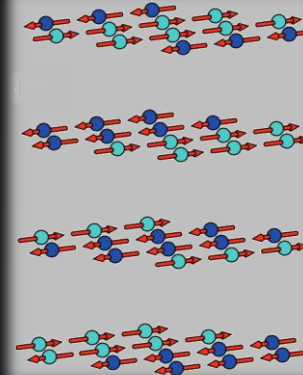
$CrCl_3$



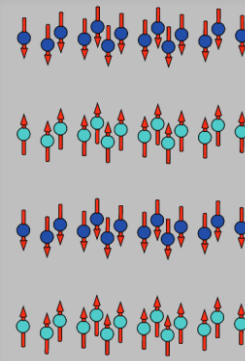
$CrBr_3, CrI_3$



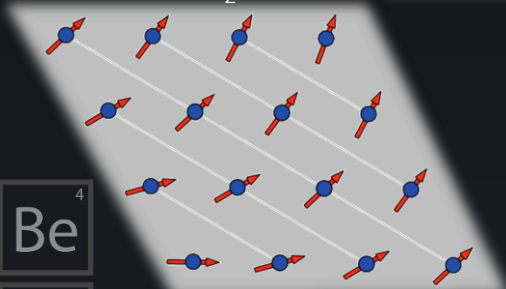
$MnBr_2$



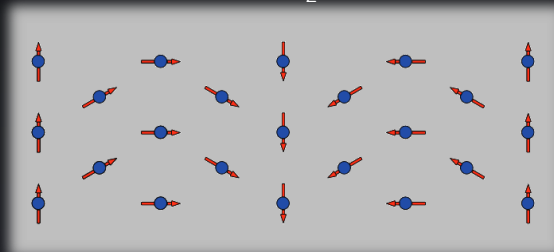
$FeCl_2, FeBr_2$



$NiBr_2$

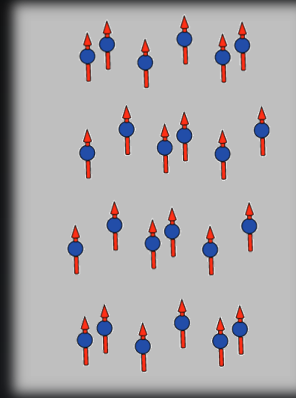


CoI_2



1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg	4	5	6	7	8	9	10	13 Al	14 Si	15 P	16 S	17 Cl	18 Ar			
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	26 Co	27 Ni	28 Cu	29 Zn	30 Ga	31 Ge	32 As	33 Se	34 Br	35 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	44 Rh	45 Pd	46 Ag	47 Cd	48 In	49 Sn	50 Sb	51 Te	52 I	53 Xe
55 Cs	56 Ba	57-71 La-Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	76 Ir	77 Pt	78 Au	79 Hg	80 Tl	81 Pb	82 Bi	83 Po	84 At	85 Rn
87 Li	88 Ra	89-103 Ac-Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Fl	115 Uup	116 Lv	117 Uus	118 Uuo

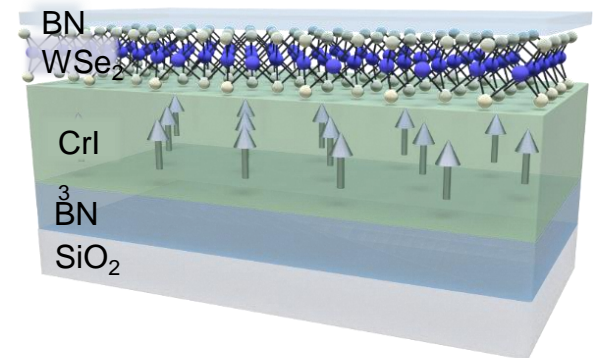
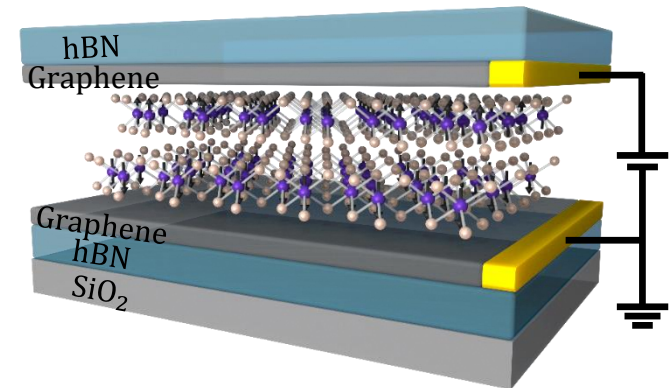
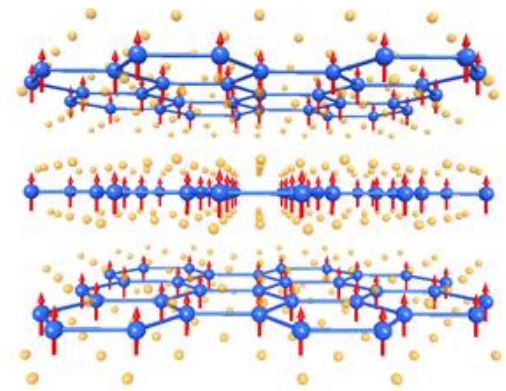
Layered Transition Metal Halides



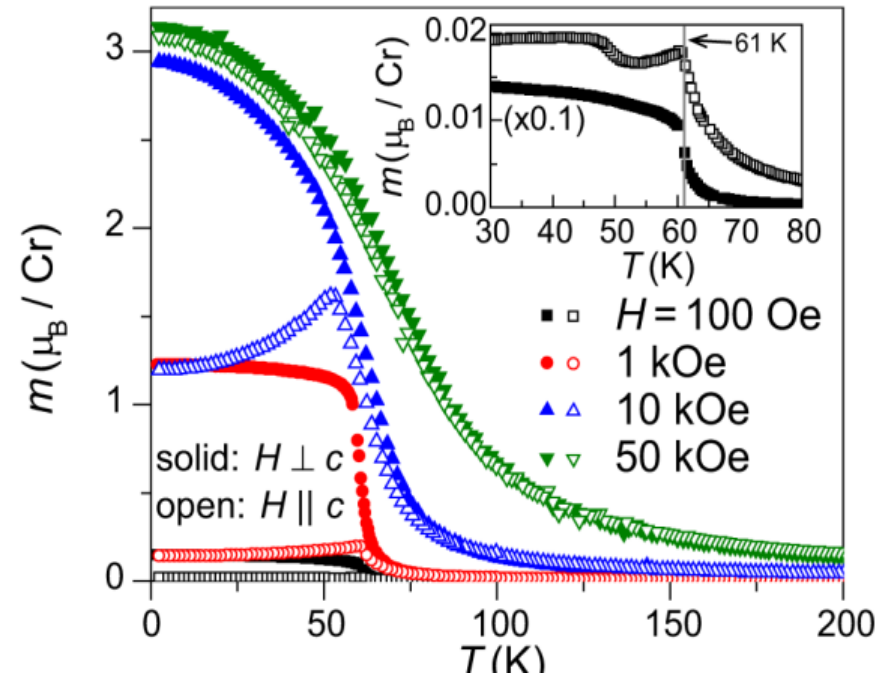
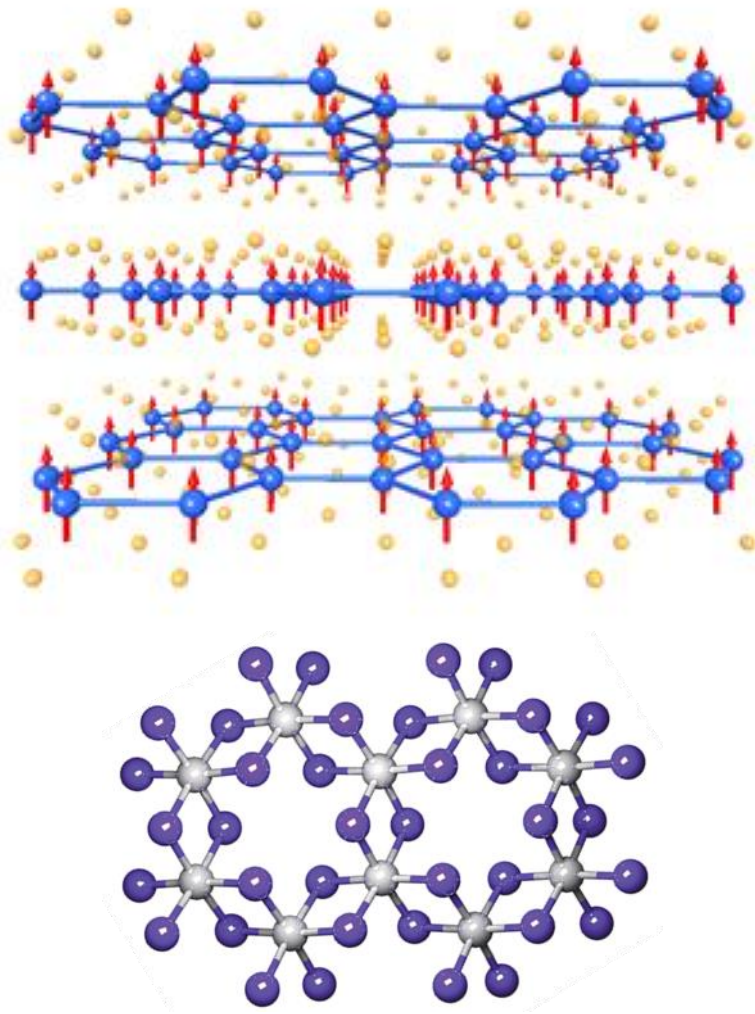
1 H																	2 He
3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
		4	5	6	7	8	9	10									
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
55 Cs	56 Ba	57-71 La-Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
87 Li	88 Ra	89-103 Ac-Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Cn	113 Uut	114 Fl	115 Uup	116 Lv	117 Uus	118 Uuo

Outline

- van der Waals ferromagnet CrI_3
 - Layer dependent ferromagnetism down to monolayer limit
- Giant tunneling magneto resistance of spin filter van der Waals heterostructure
- van der Waals ferromagnetic heterostructures ($\text{WSe}_2/\text{CrI}_3$)
 - Ferromagnetic control of spin/valley pseudospin in non-magnetic monolayer semiconductor WSe_2

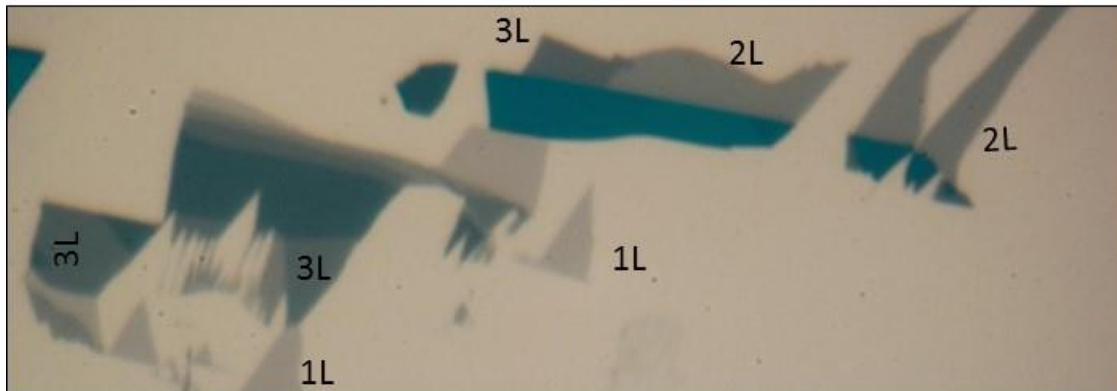


Basics of Bulk Chromium(III) iodide (CrI_3) Crystal



McGuire, M. A., Dixit, H., Cooper, V. R. & Sales, B. C. Coupling of Crystal Structure and Magnetism in the Layered, Ferromagnetic Insulator CrI_3 . *Chem. Mater.* **27**, 612–620 (2015).

Exfoliation of Atomically-Thin CrI₃



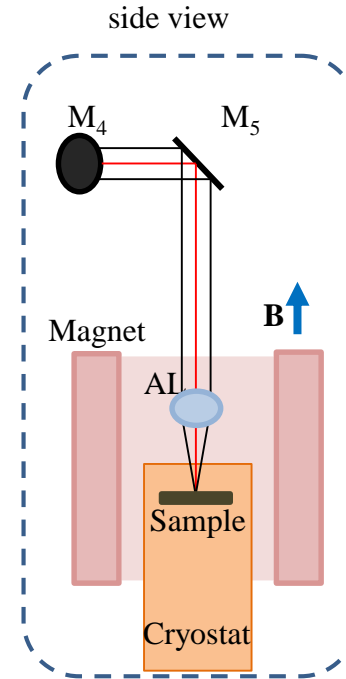
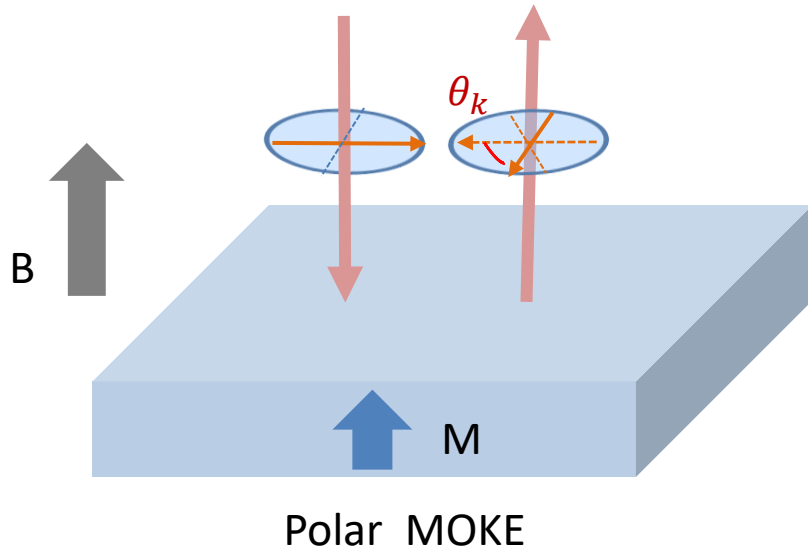
Dahlia Klein



Efren Navarro-Moratalla



Magneto Optical Kerr Effect (MOKE)

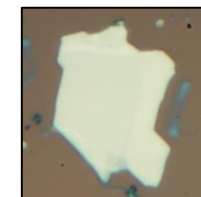
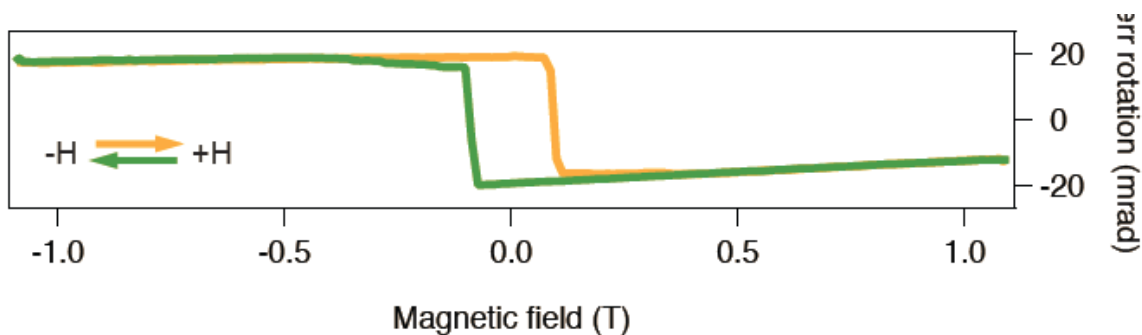


Bevin Huang



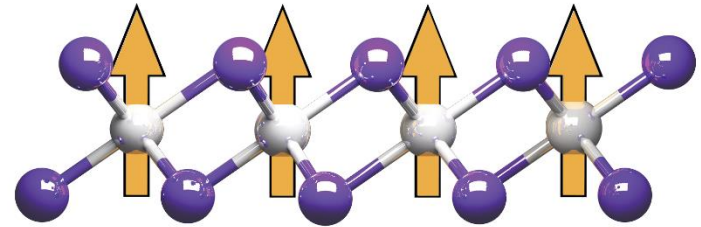
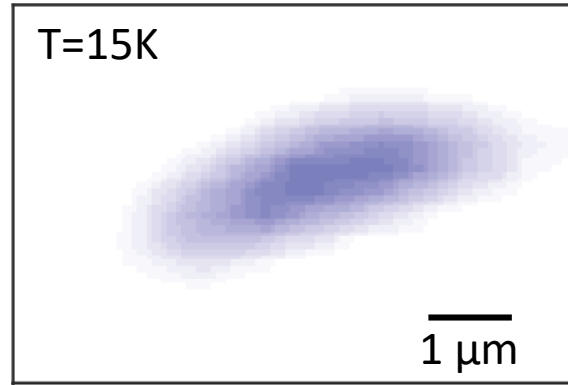
Gen Clark

MOKE of thin bulk crystal

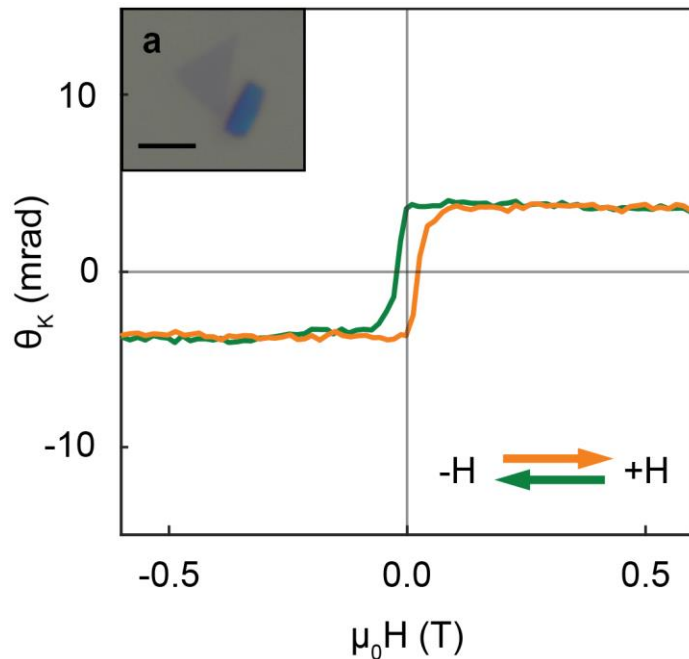


Demonstration of Monolayer Ising Ferromagnet

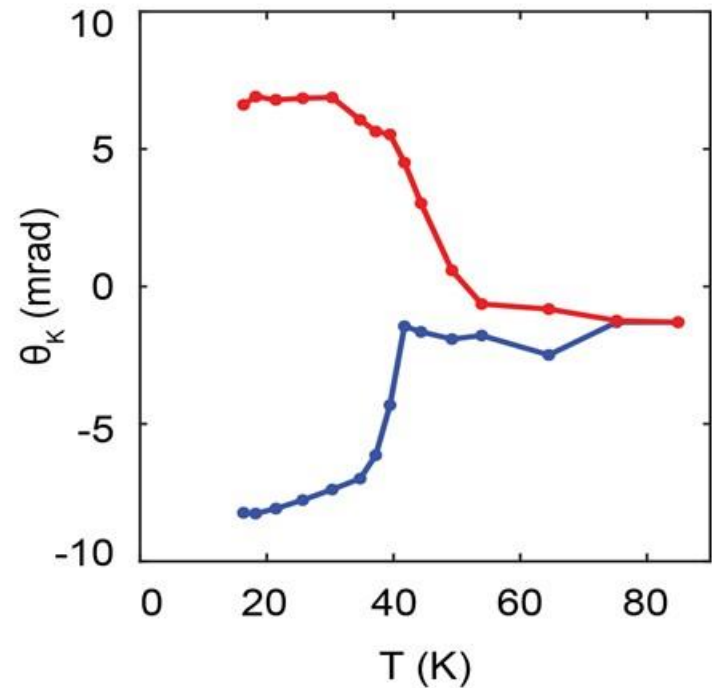
Kerr rotation intensity map at H=0



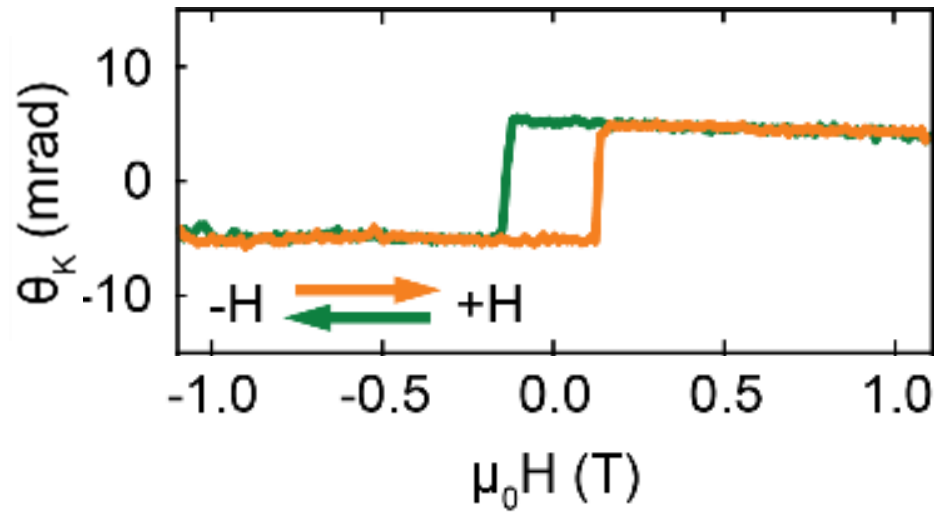
Monolayer MOKE



Temperature Dependent MOKE

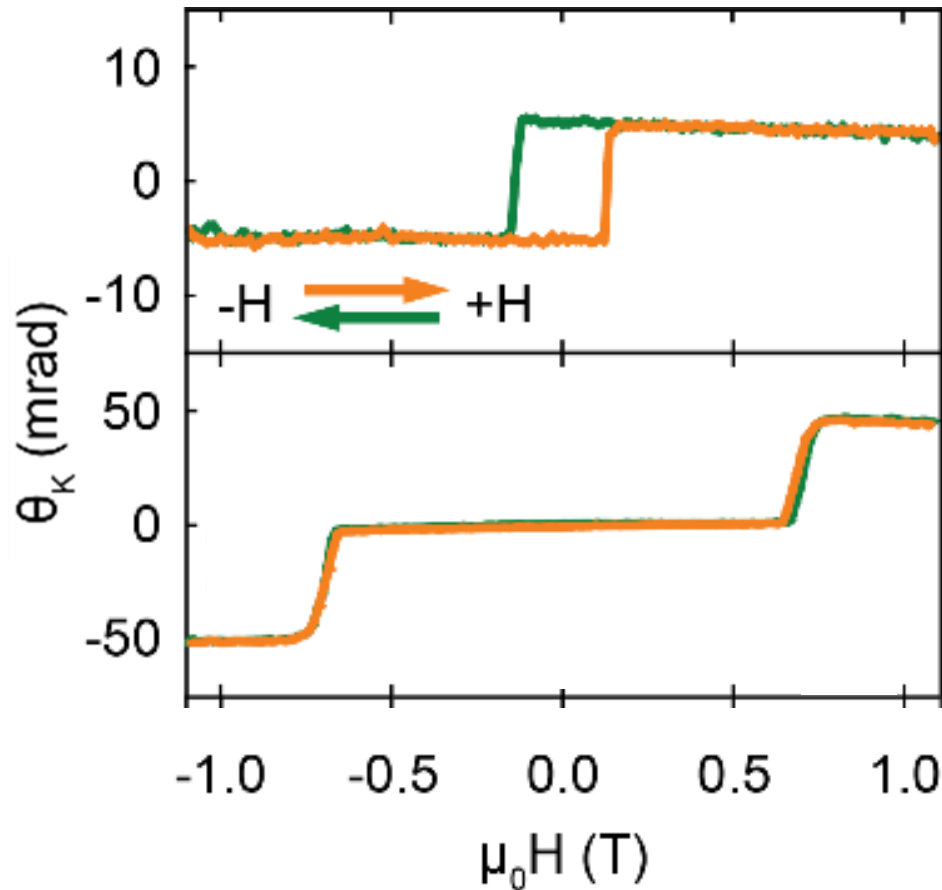


Layer Thickness Dependent Magnetic Phases



Monolayer

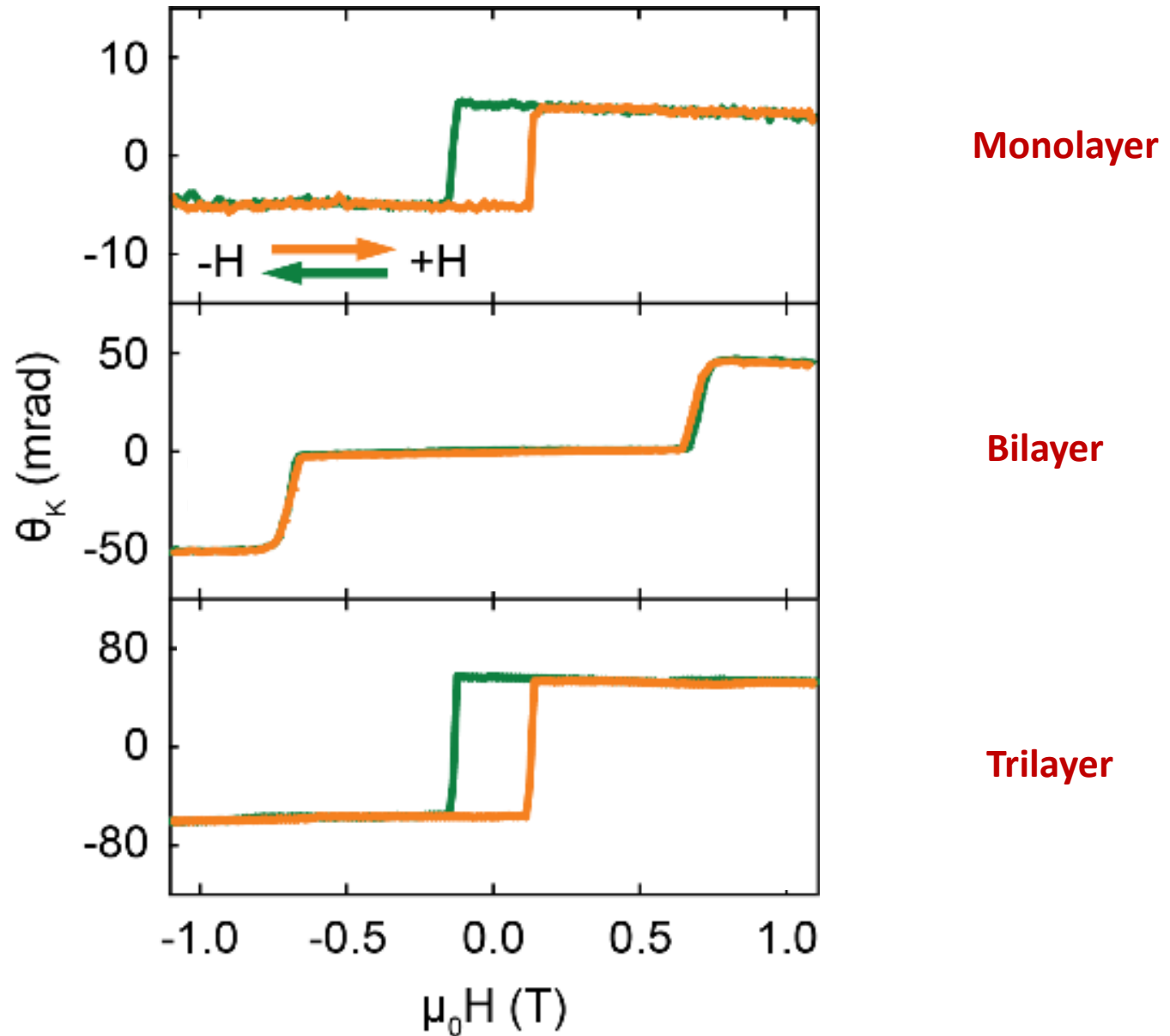
Layer Thickness Dependent Magnetic Phases



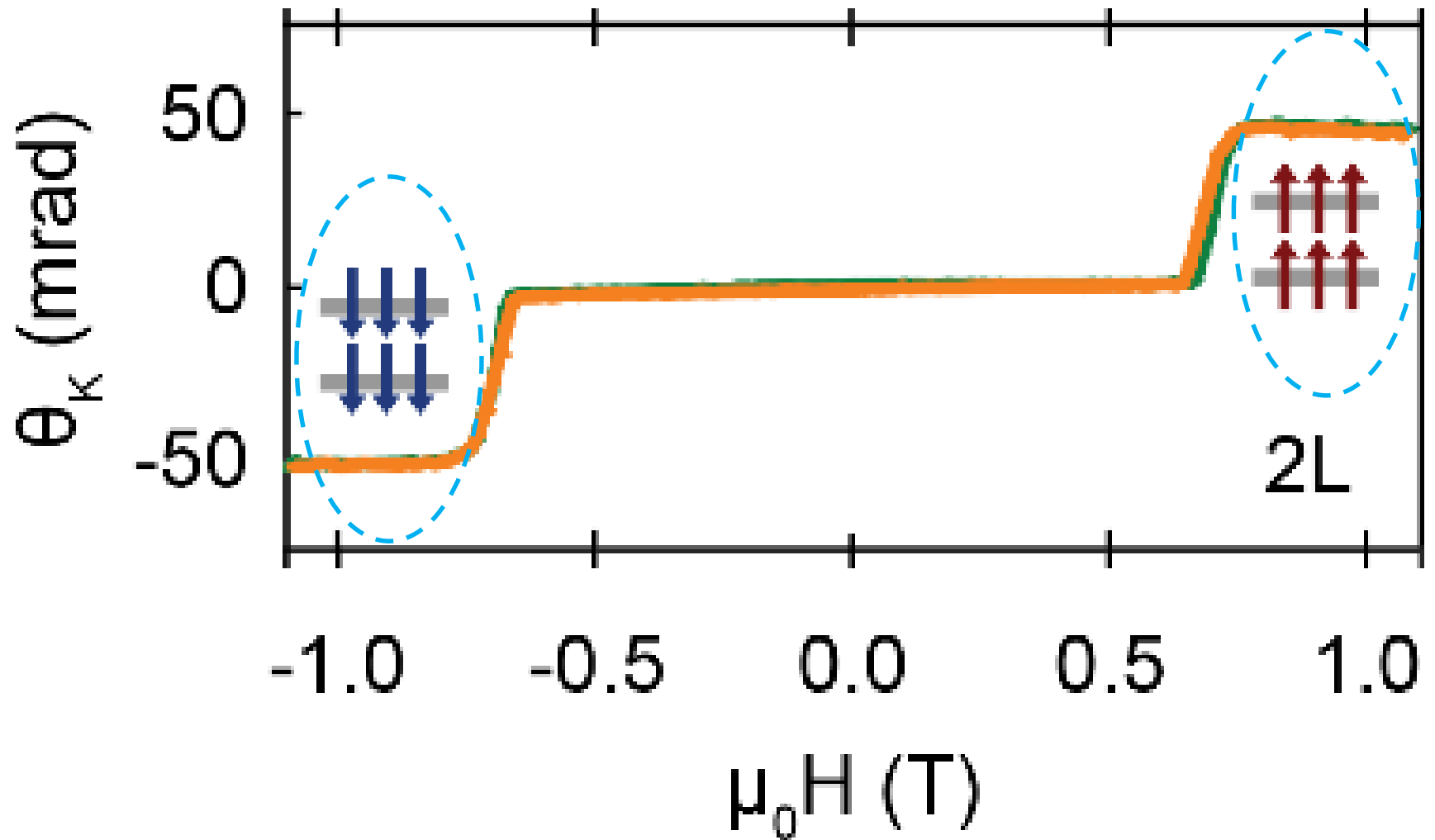
Monolayer

Bilayer

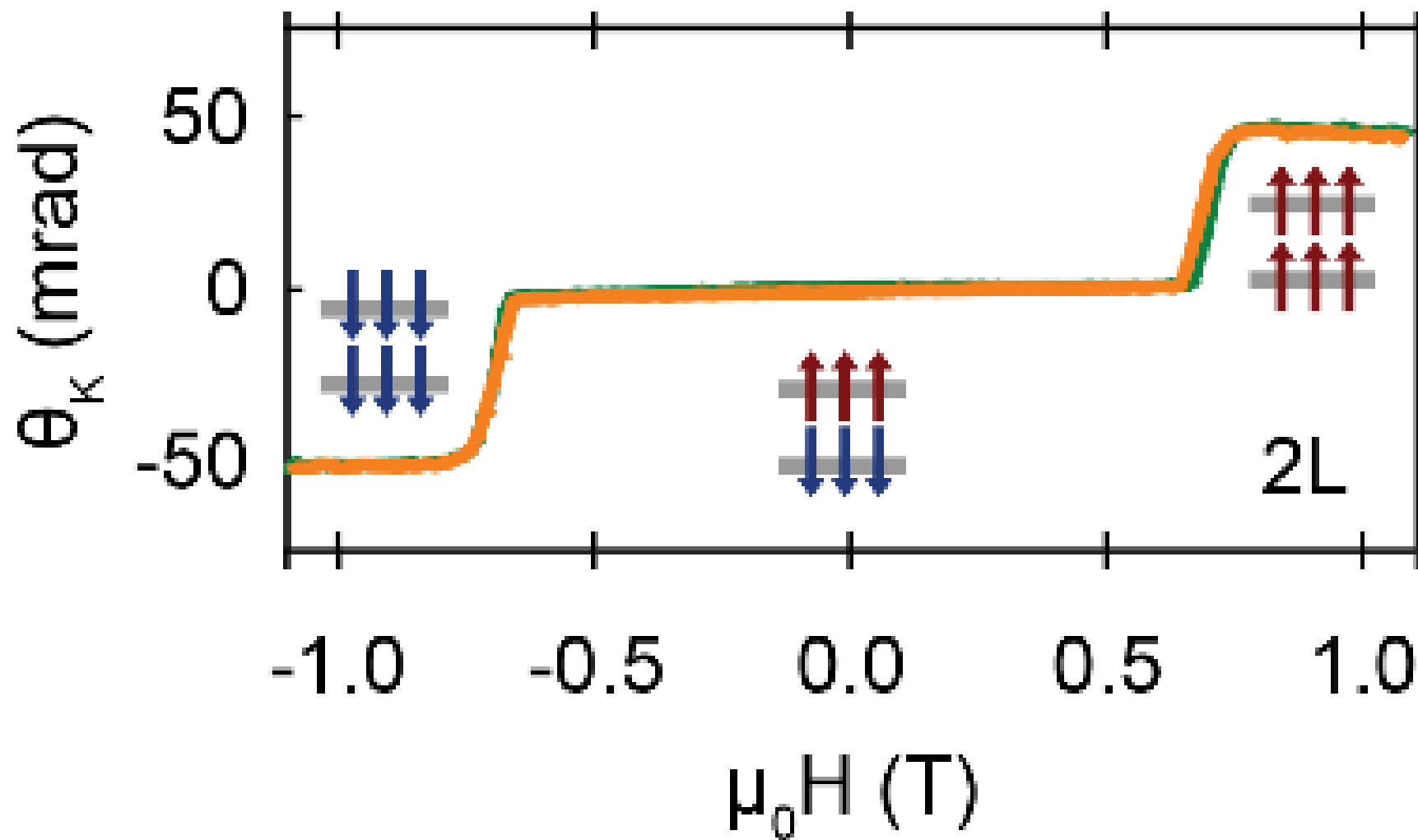
Layer Thickness Dependent Magnetic Phases



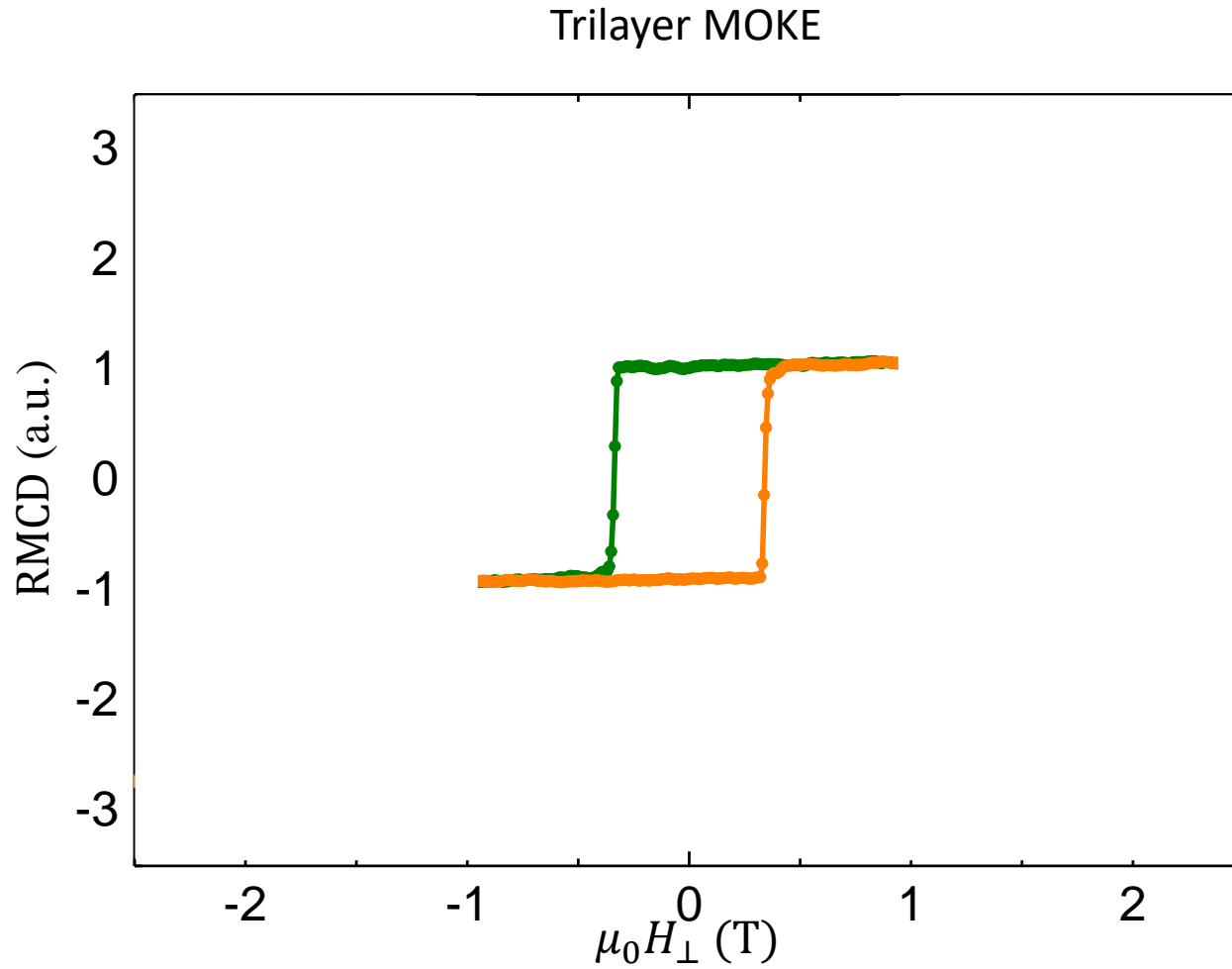
What is the magnetic ground state in bilayer?



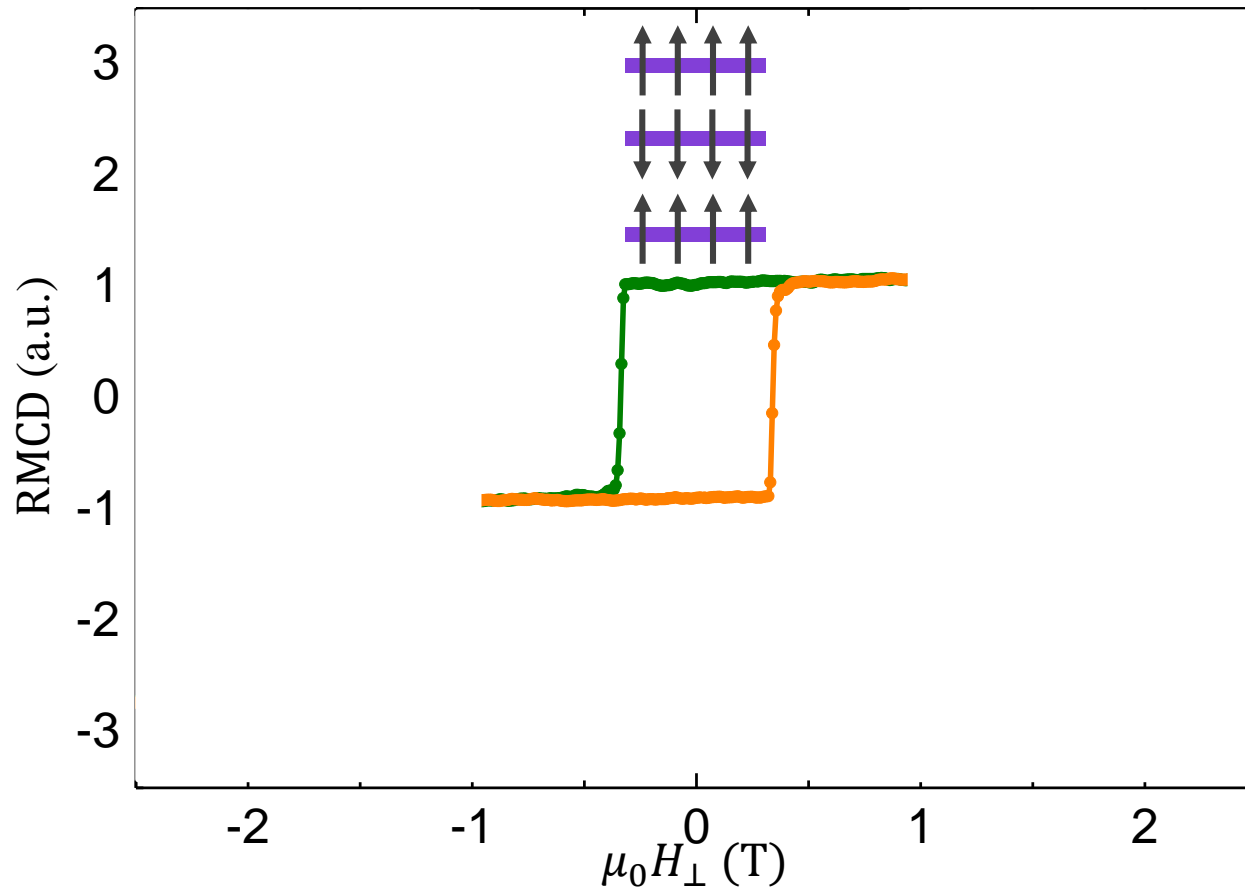
Anti-Ferromagnetic Interlayer Coupling in Bilayer



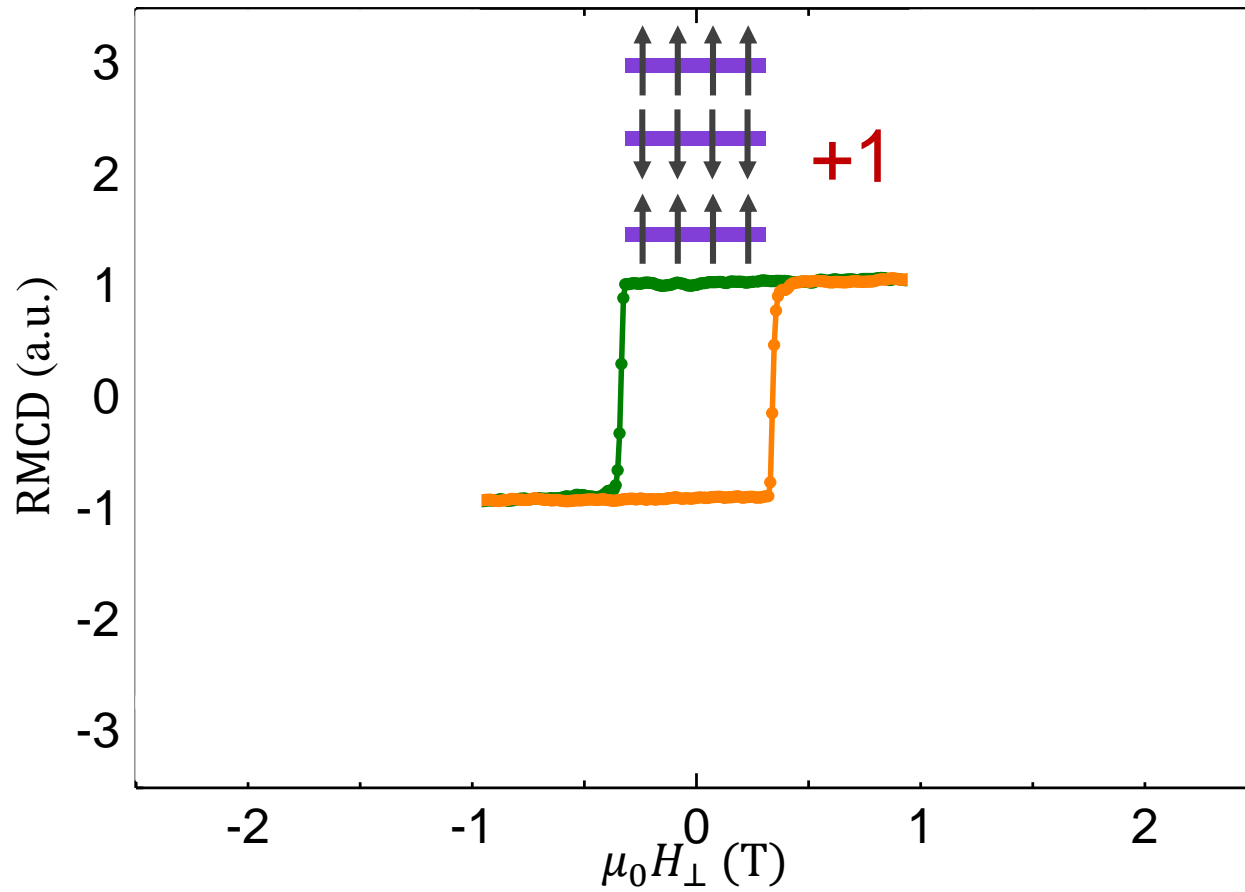
Trilayer – Interlayer Antiferromagnetic Coupling



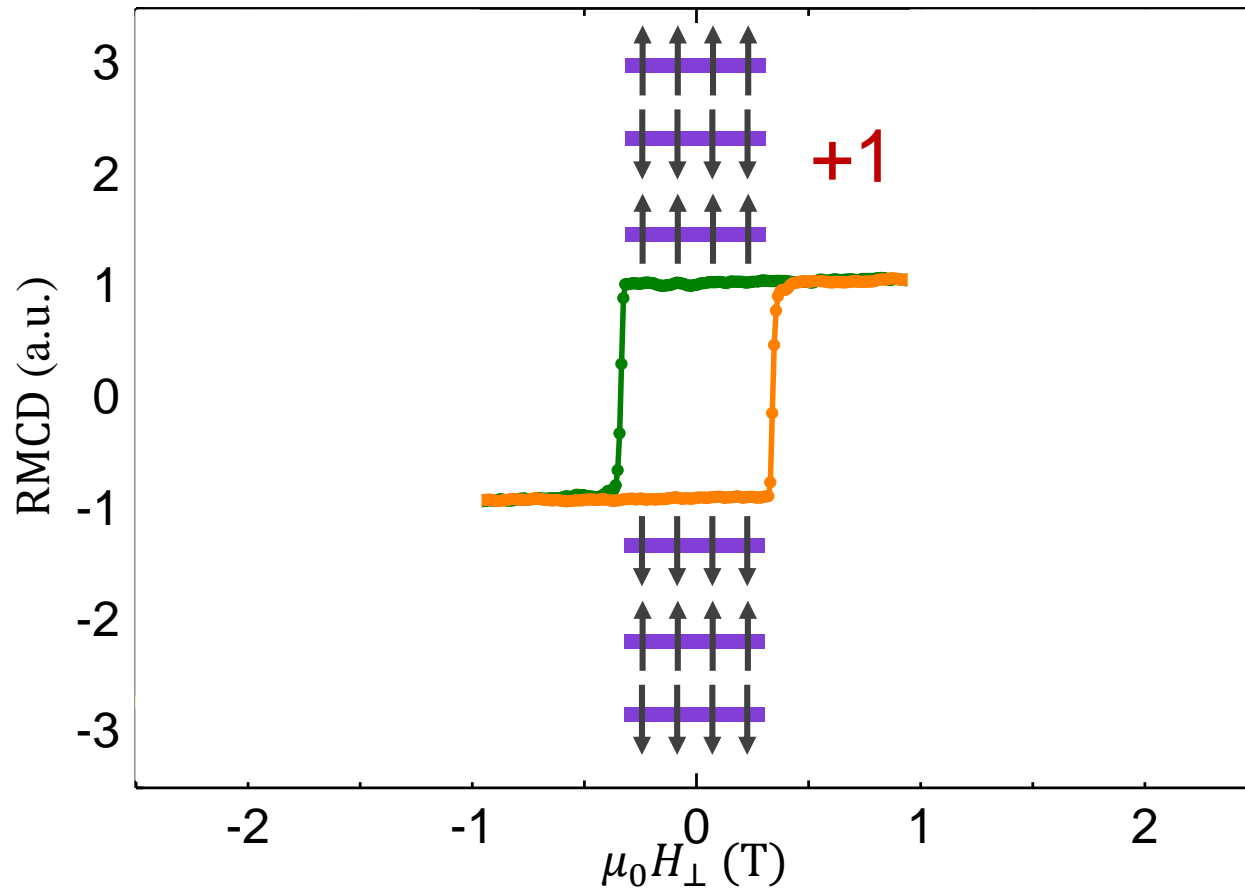
Trilayer – Interlayer Antiferromagnetic Coupling



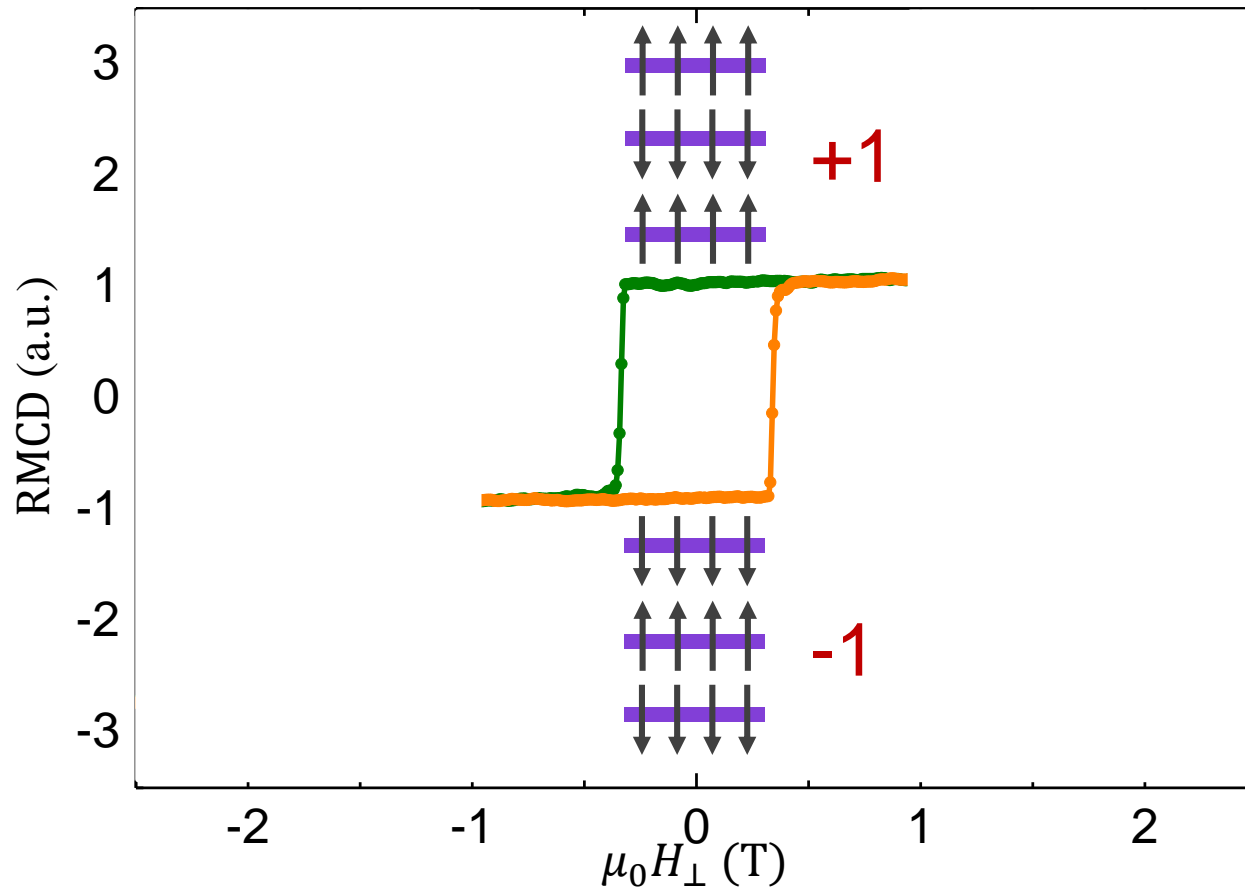
Trilayer – Interlayer Antiferromagnetic Coupling



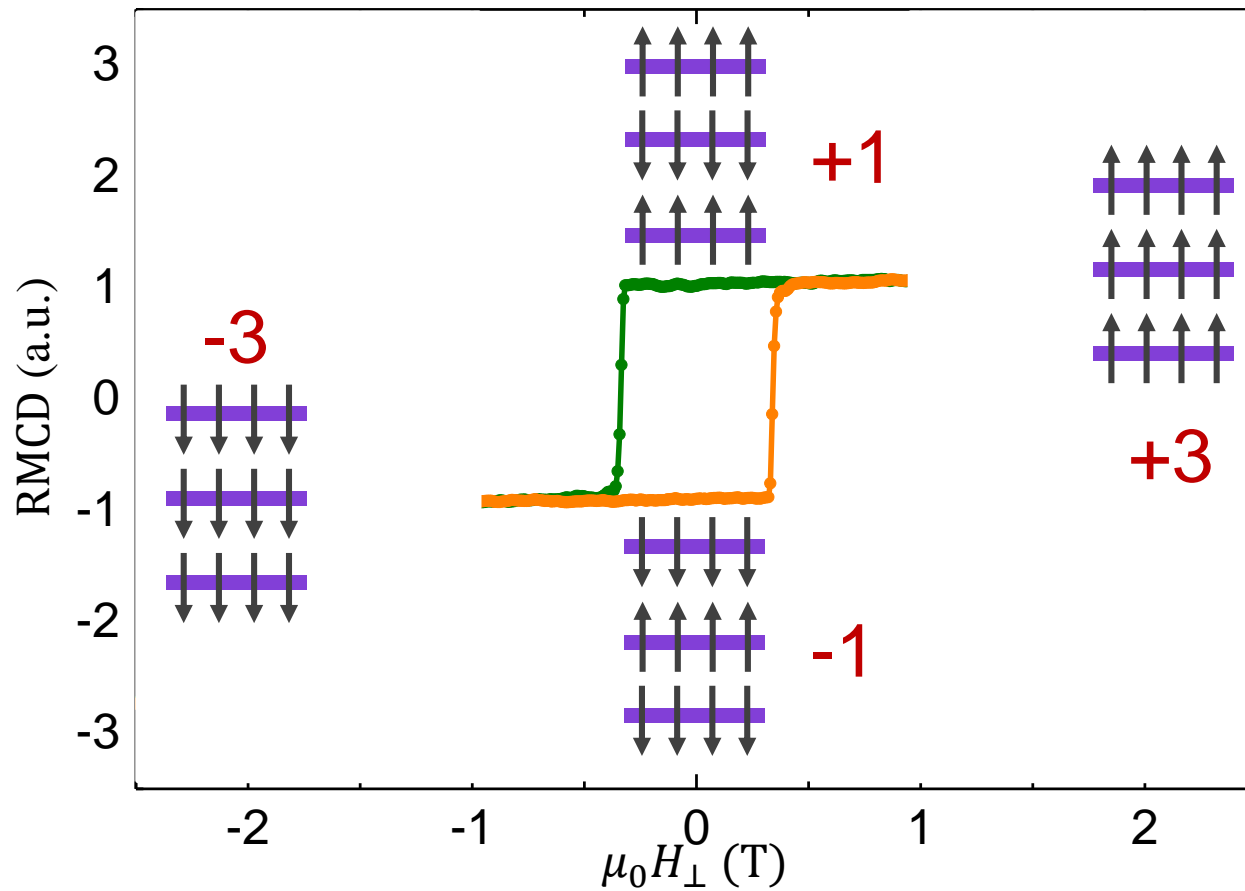
Trilayer – Interlayer Antiferromagnetic Coupling



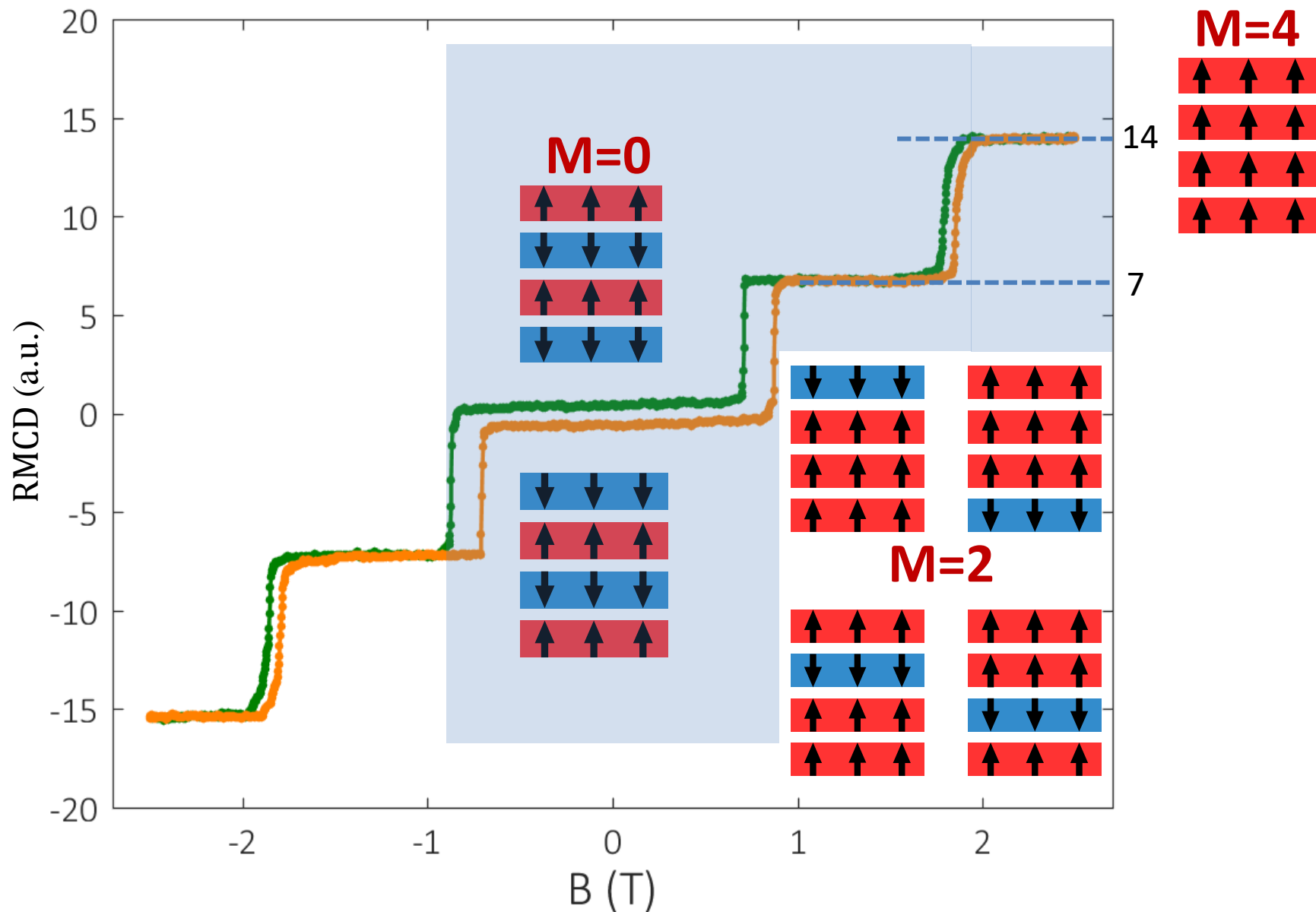
Trilayer – Interlayer Antiferromagnetic Coupling



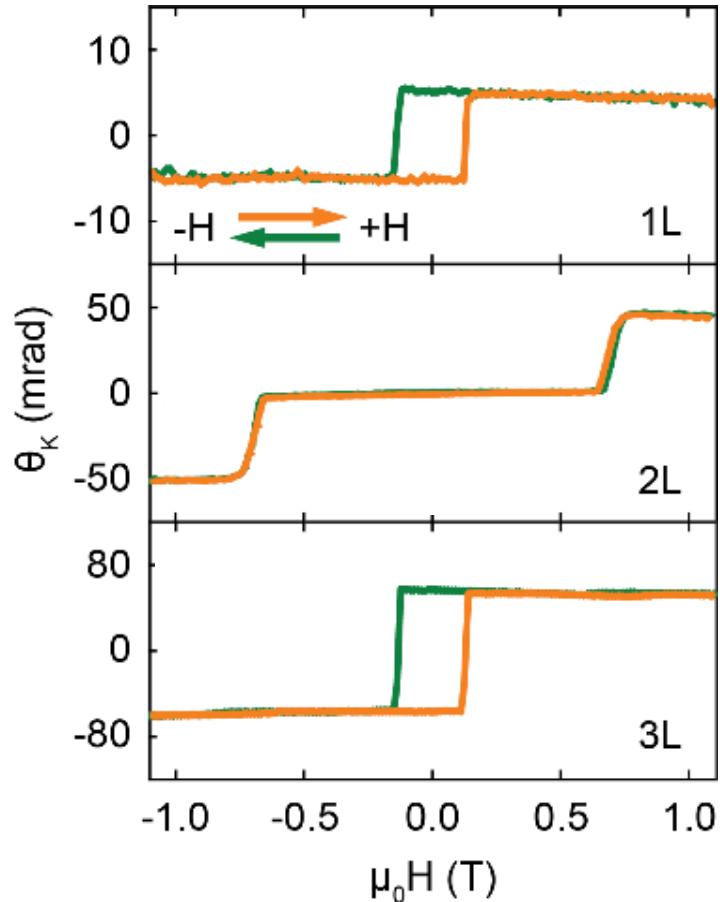
Observation of Magnetic Devil's Staircase



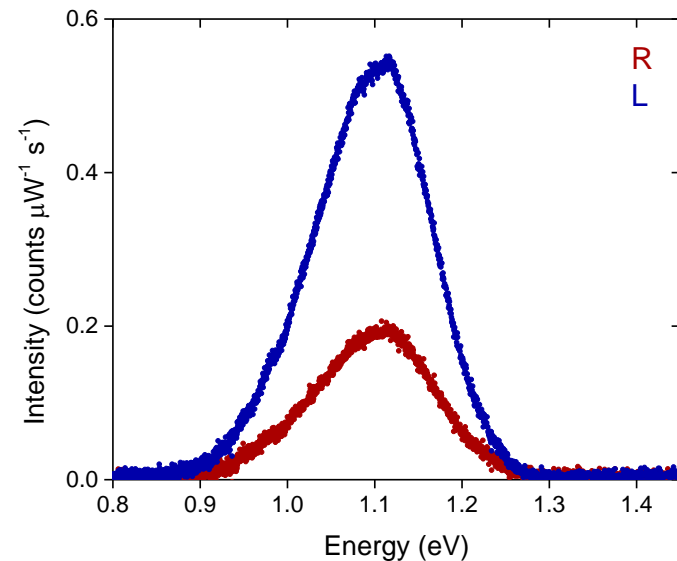
Four Layer – Interlayer Antiferromagnetic Coupling



Short Summary



- **Monolayer CrI₃ is an intrinsic ferromagnet.**
- **Atomically-thin CrI₃ is a layered antiferromagnet: Intralayer coupling is ferromagnetic while interlayer coupling is antiferromagnetic**
- **Spontaneously circularly polarized-photoluminescence in monolayer CrI₃ with magnetization determined helicity**



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

Kyle Seyler, Xu et al, *Nature Physics* **14**, 277 (2018);

CrI_3 : atomically-thin Insulator with **magnetic function**

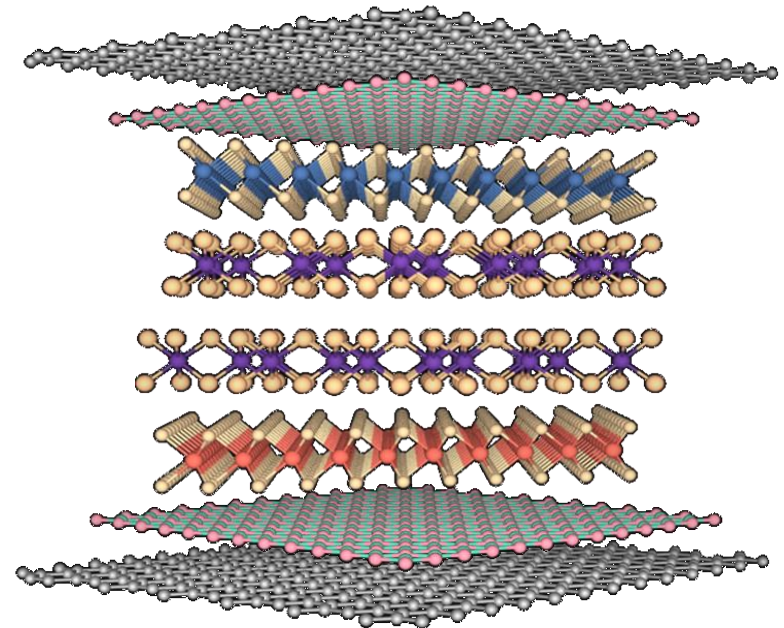
Hexagonal boron nitride (hBN):
atomically-thin insulator
atomically-smooth substrate,
tunnel barrier...

Boron nitride substrates for high-quality
graphene electronics, **Nature
nanotechnology** 5 (10), 722-726

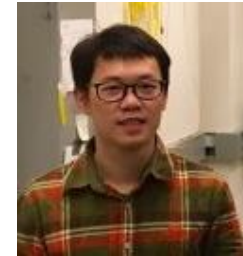
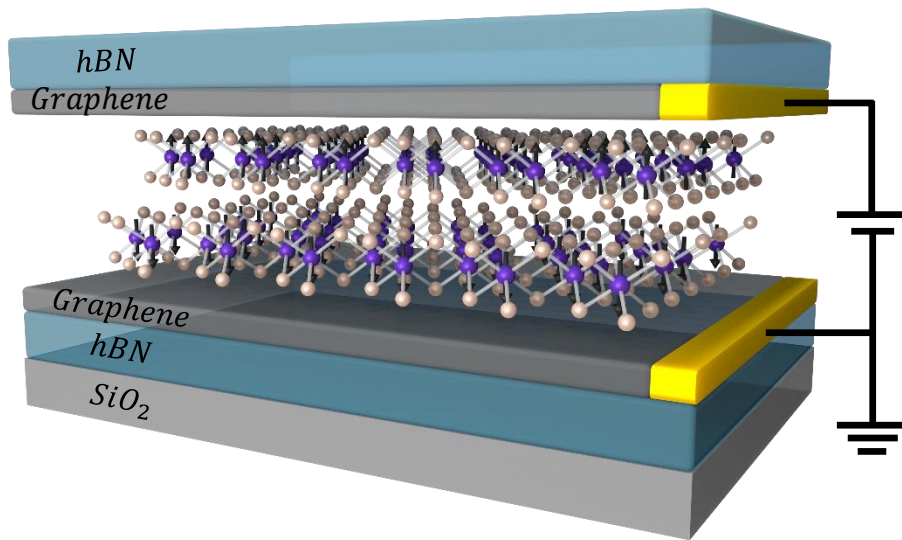
Electron tunneling through atomically flat and
ultrathin hexagonal boron nitride,
Applied physics letters 99 (24), 243114

Hofstadter's butterfly and the fractal quantum
Hall effect in moire superlattices, **Nature** 497
(7451), 598

CrI_3 : Key component for van
der Waals structures with
spintronic functionalities,
topological properties...



Giant Tunneling Magnetoresistance in Spin-Filter van der Waals Heterostructures



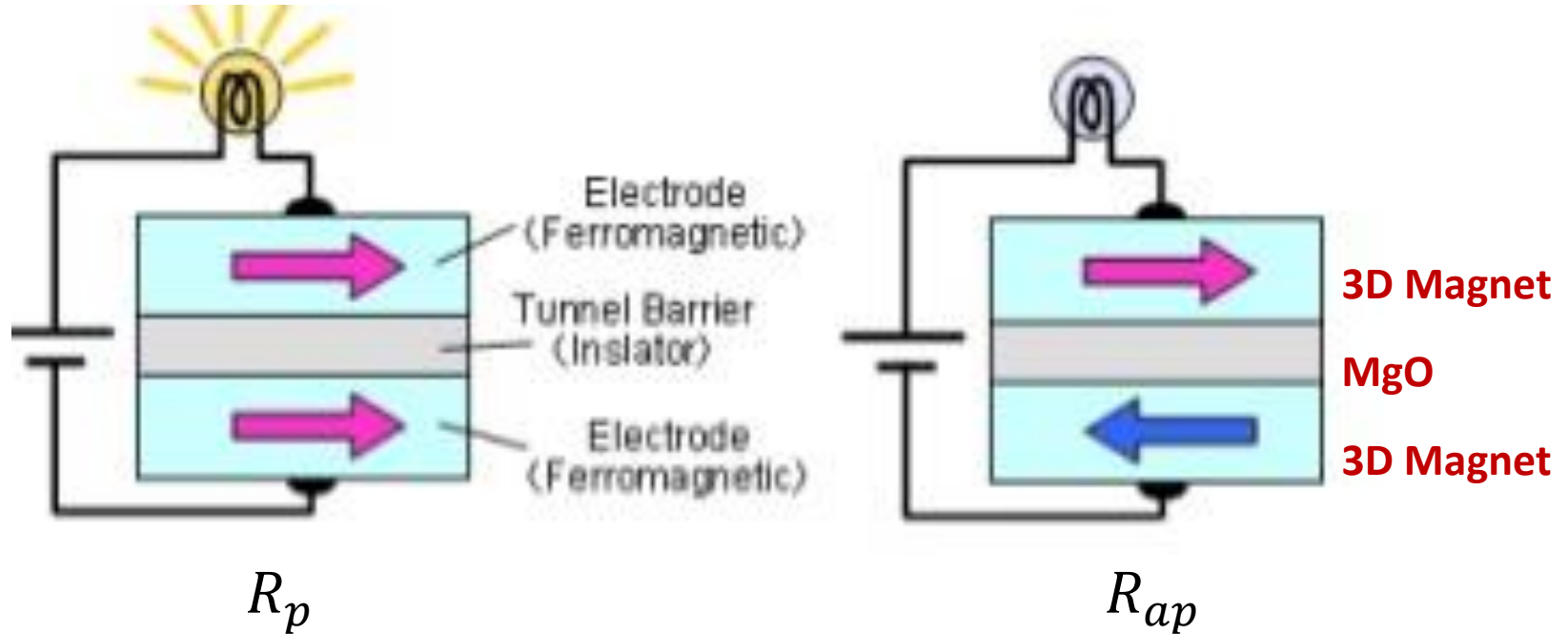
Tiancheng Song

Song, T., Xu, X *et al.* <http://arxiv.org/abs/1801.08679>

also see Klein, D. R. <http://arxiv.org/abs/1801.10075>;

Wang, Z. *et al.* <http://arxiv.org/abs/1801.08188>

Giant Magnetoresistance (GMR)



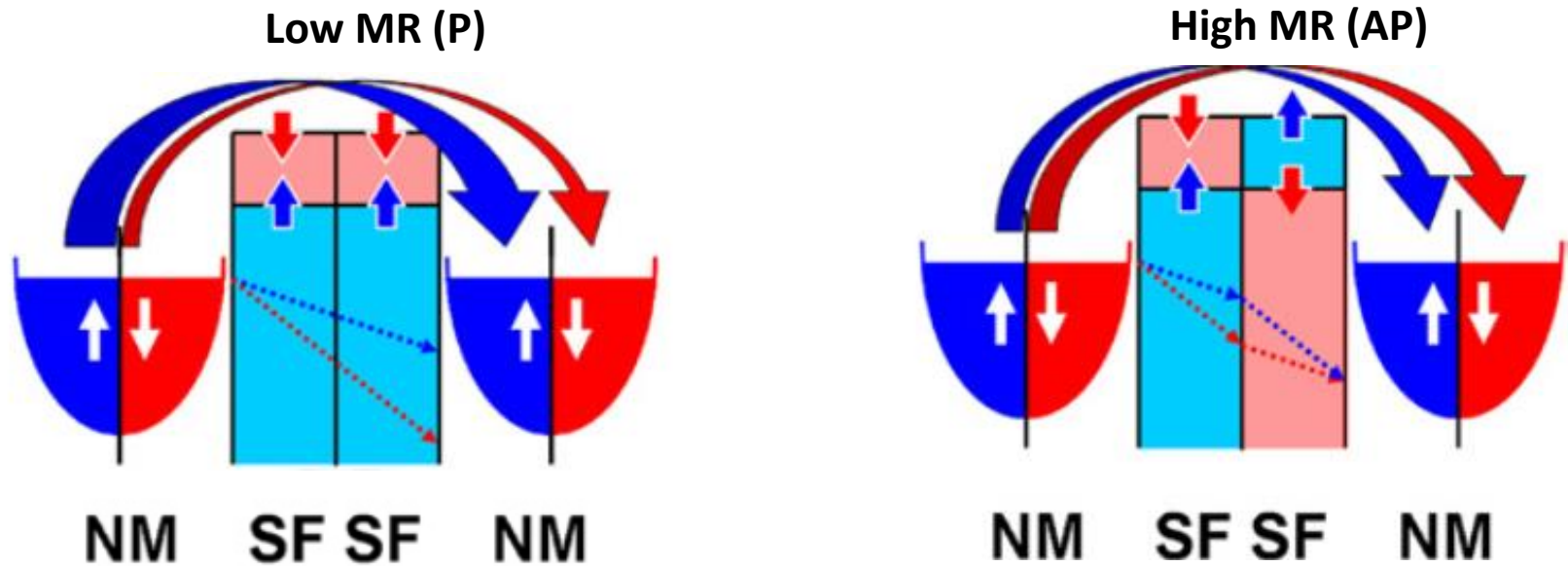
$$TMR = \frac{R_{ap} - R_p}{R_p}$$

S. Yuasa, T. Nagahama, A. Fukushima, Y. Suzuki and K. Ando, *Nature Materials*, vol. 3, pp. 868-871, 2004.

S. S. P. Parkin, K. Kaiser, A. Panchula, P. M. Rice, B. Hughes, M. Samant and S.-H. Yang, *Nature Materials*, vol. 3, pp. 862-867, 2004.

S. Ikeda, J. Hayakawa, Y. Ashizawa, Y. M. Lee, K. Miura, H. Hasegawa, M. Tsunoda, F. Matsukura and H. Ohno, *Appl. Phys. Lett.*, vol. 93, art. 082508, 2008.

Spin-Filter Magnetic Tunnel Junctions



D. C. Worledge and T. H. Geballe, J. Appl. Phys. 88, 5277 (2000).
Z. W. Xie and B. Z. Li, J. Appl. Phys. 93, 9111 (2003).
A. Saffarzadeh, J. Phys. Condens. Matter 15, 3041 (2003).

Double Spin-Filter Magnetic Tunnel Junctions

PRL 102, 076601 (2009)

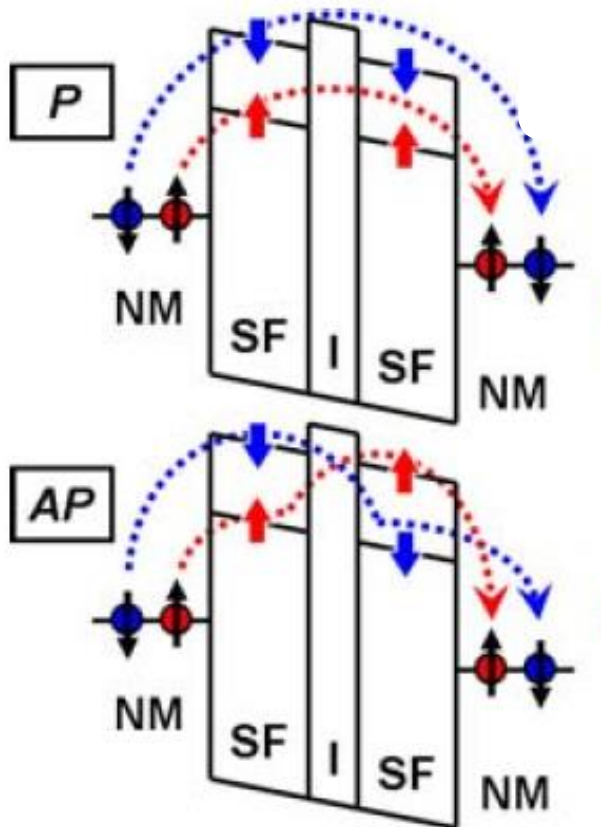
Selected for a *Viewpoint in Physics*
PHYSICAL REVIEW LETTERS

week ending
20 FEBRUARY 2009

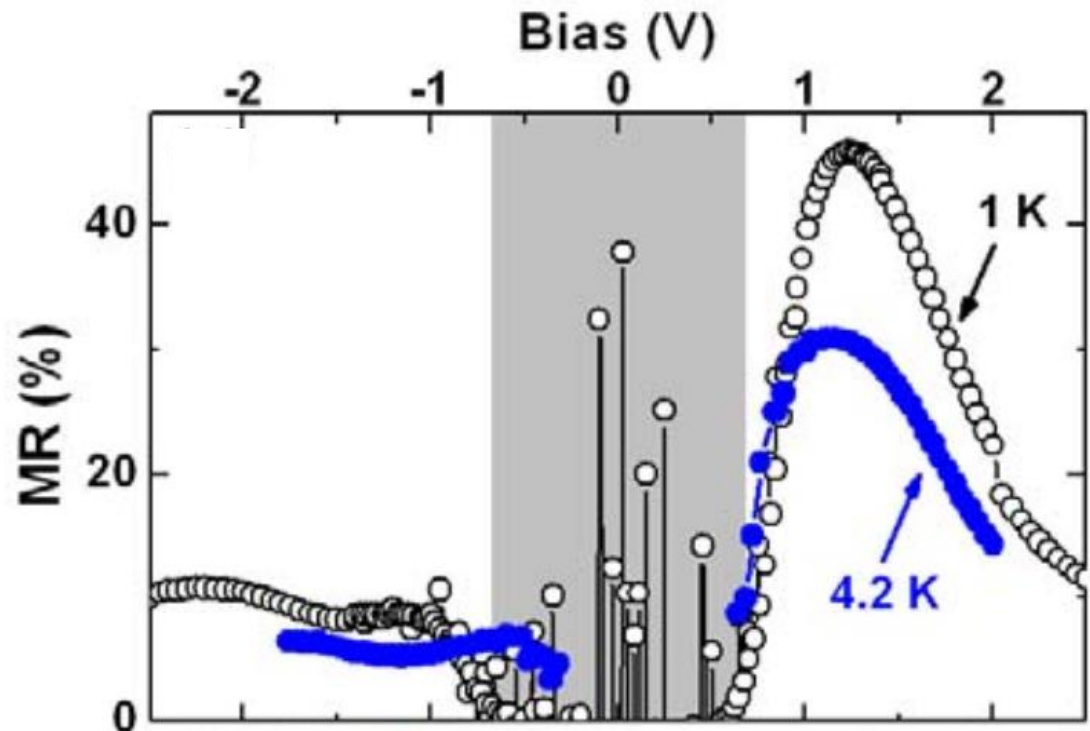


Magnetoresistance in Double Spin Filter Tunnel Junctions with Nonmagnetic Electrodes and its Unconventional Bias Dependence

Guo-Xing Miao,* Martina Müller, and Jagadeesh S. Moodera

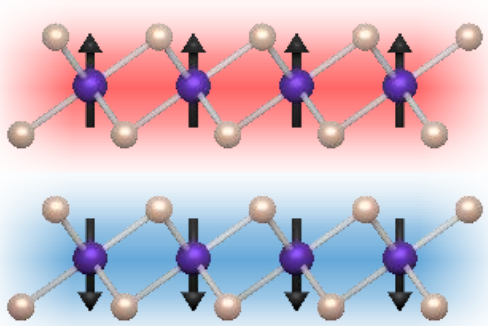


EuS/Al₂O₃/EuS

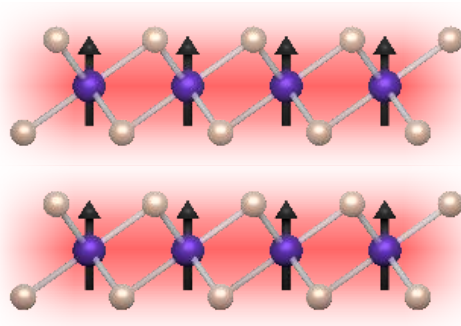


Bilayer CrI₃ - a Double Spin-Filter

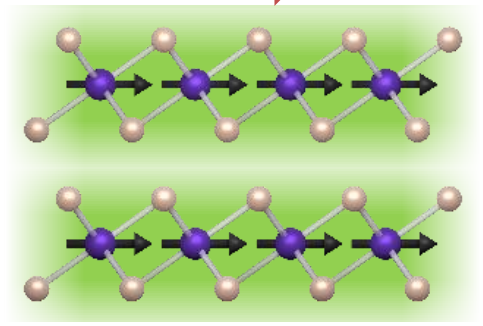
$$\mu_0 H_{\perp} = 0$$



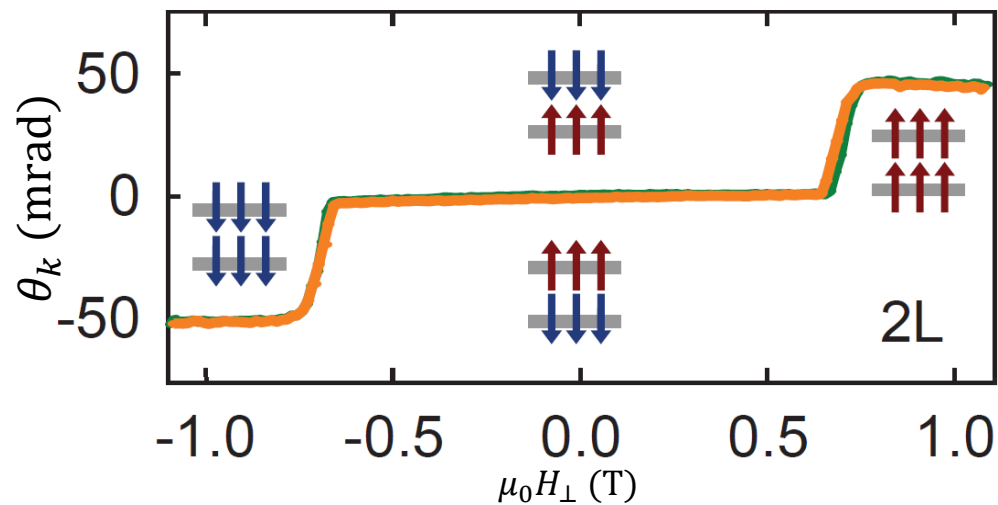
$$\mu_0 H_{\perp} > \mu_0 H_{\perp c}$$



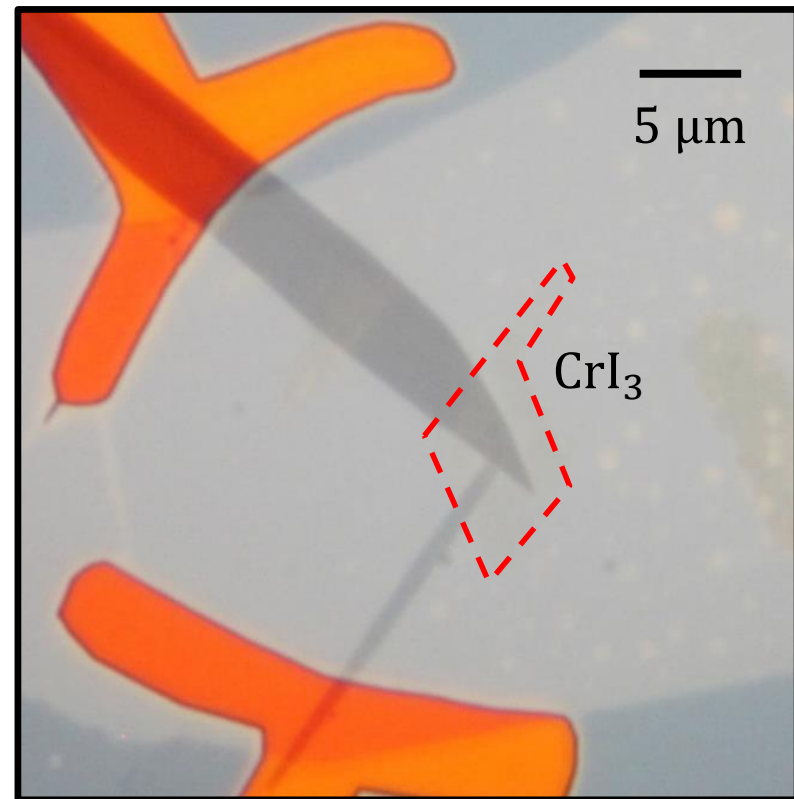
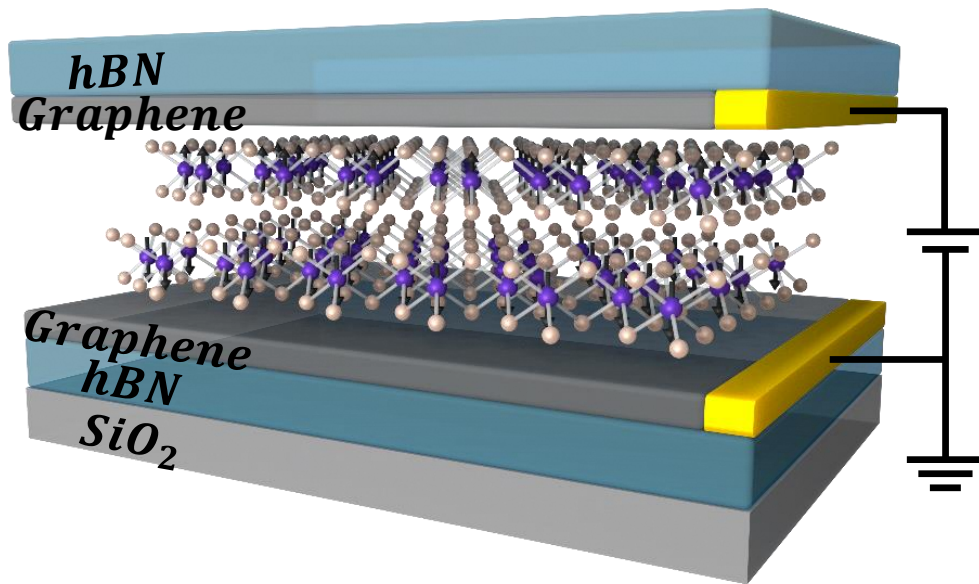
$$\mu_0 H_{\parallel} > \mu_0 H_{\parallel c}$$



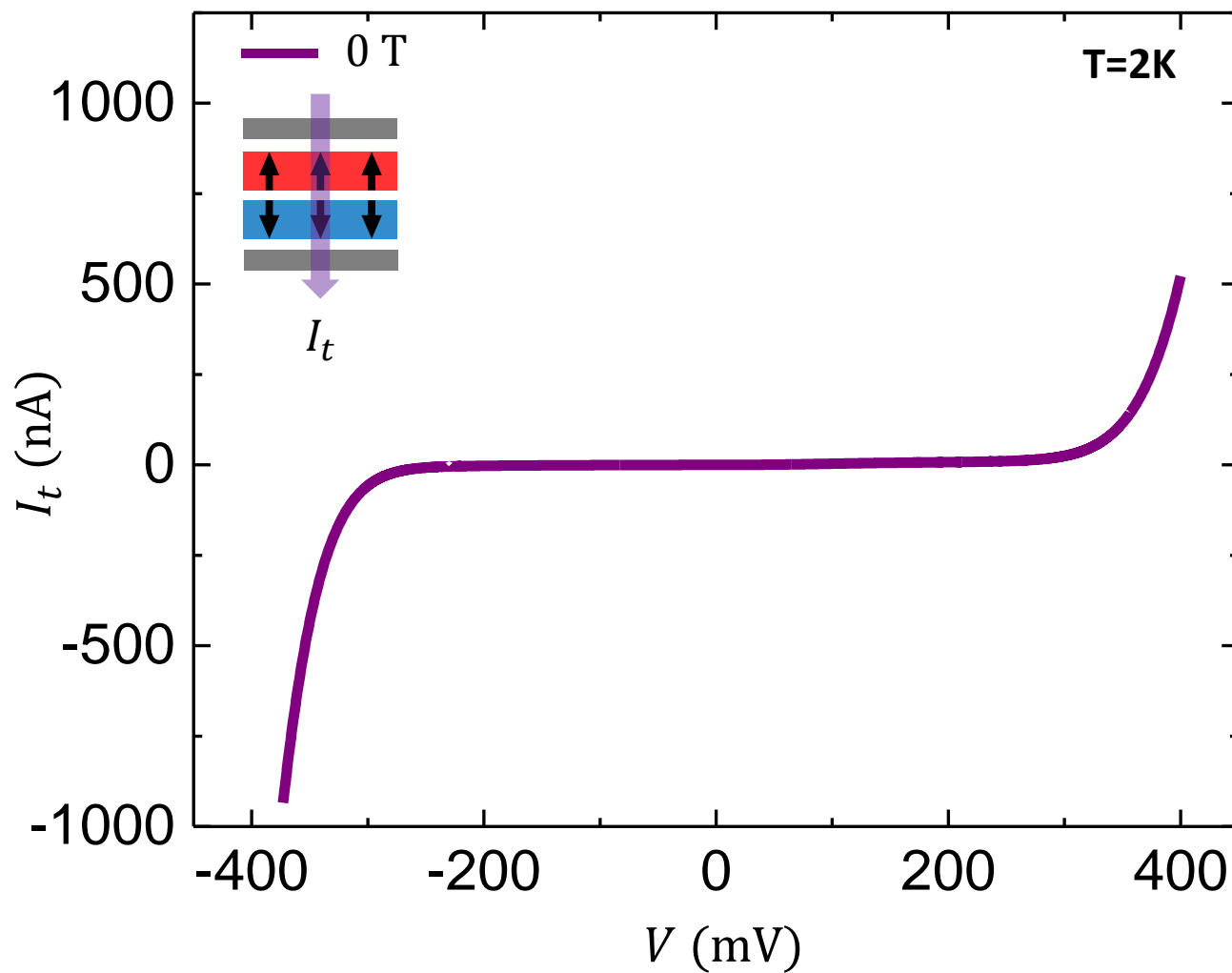
Magneto-optical Kerr effect (MOKE)



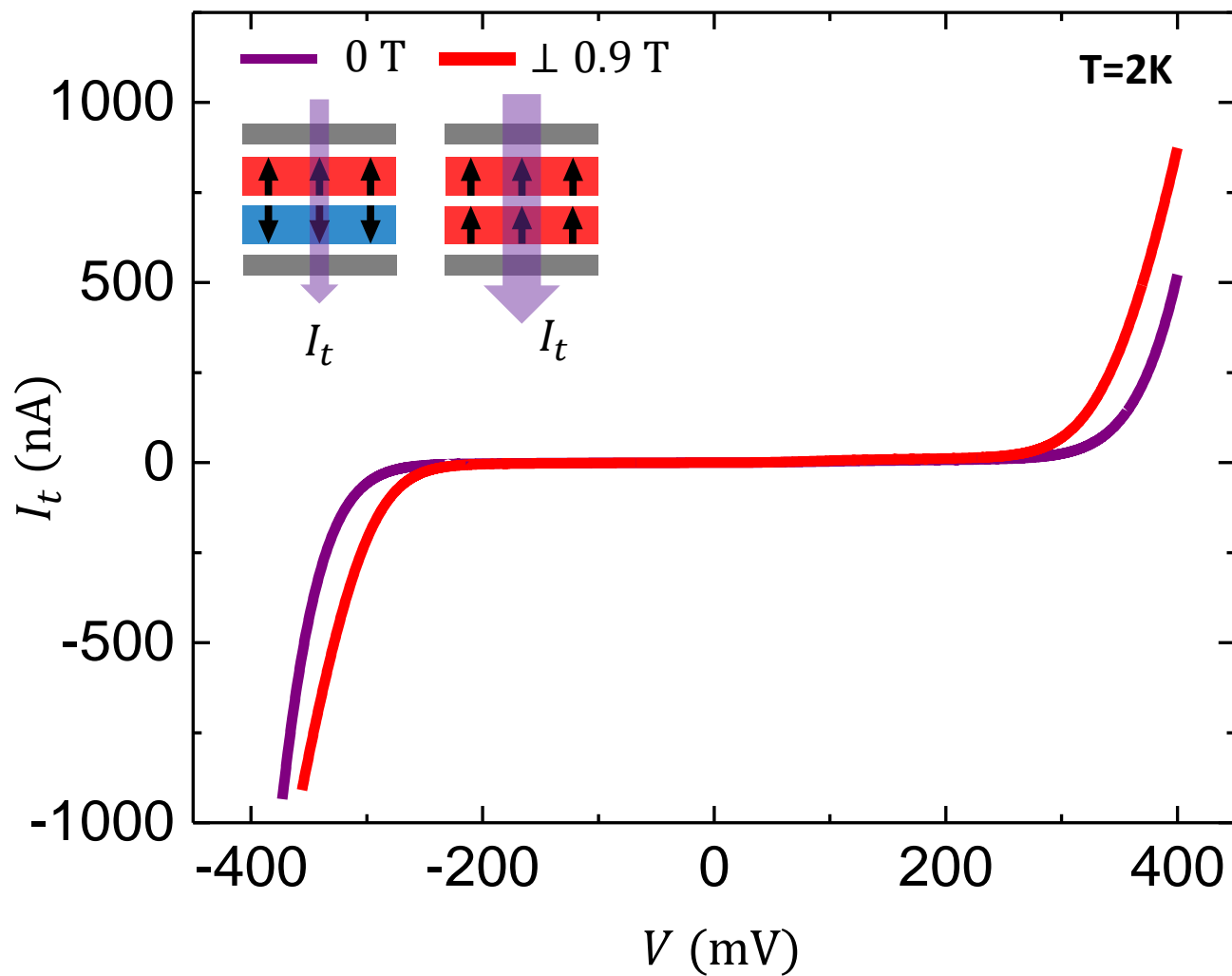
Van der Waals Double Spin-Filter MTJs



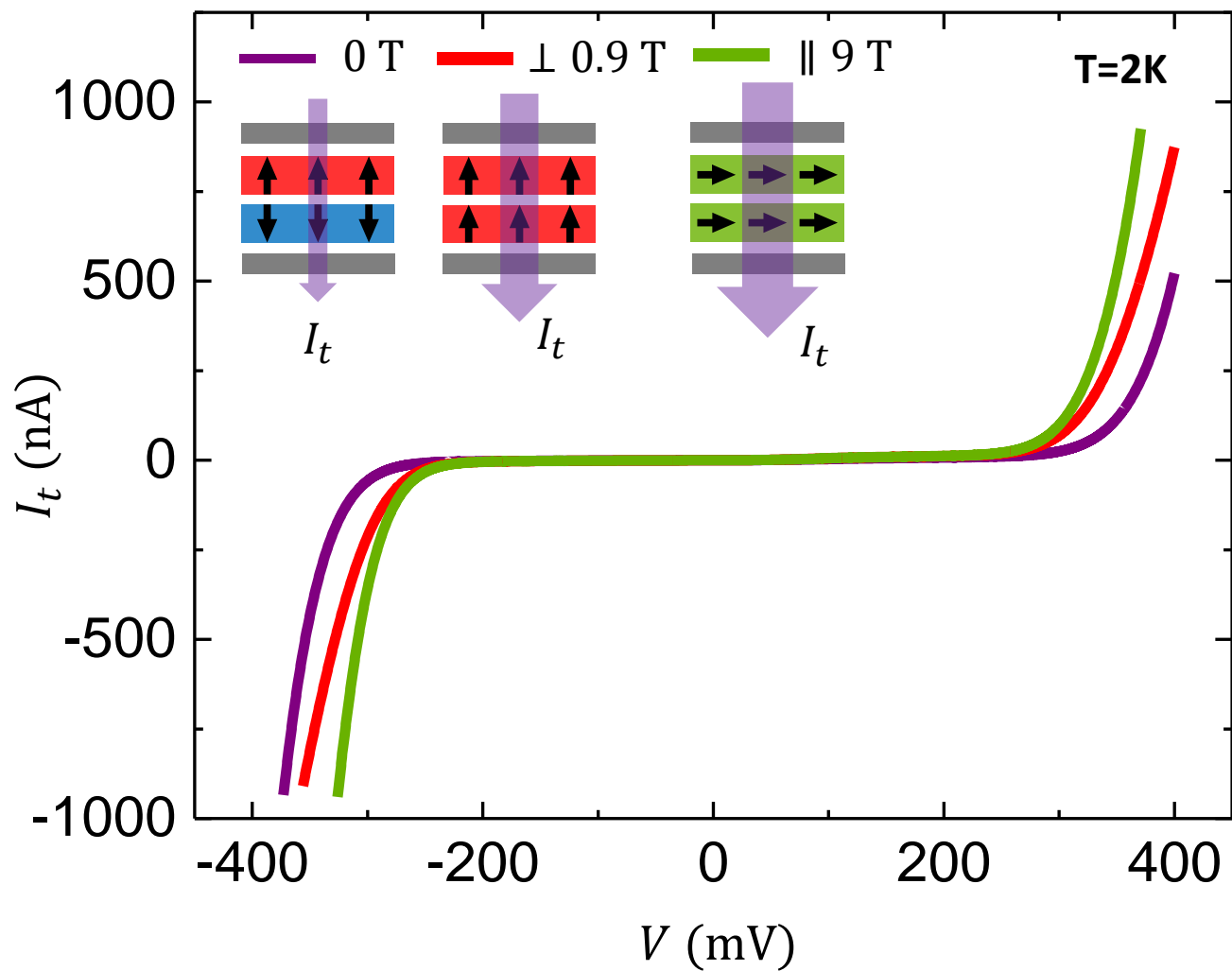
Tunneling current at selected magnetic field



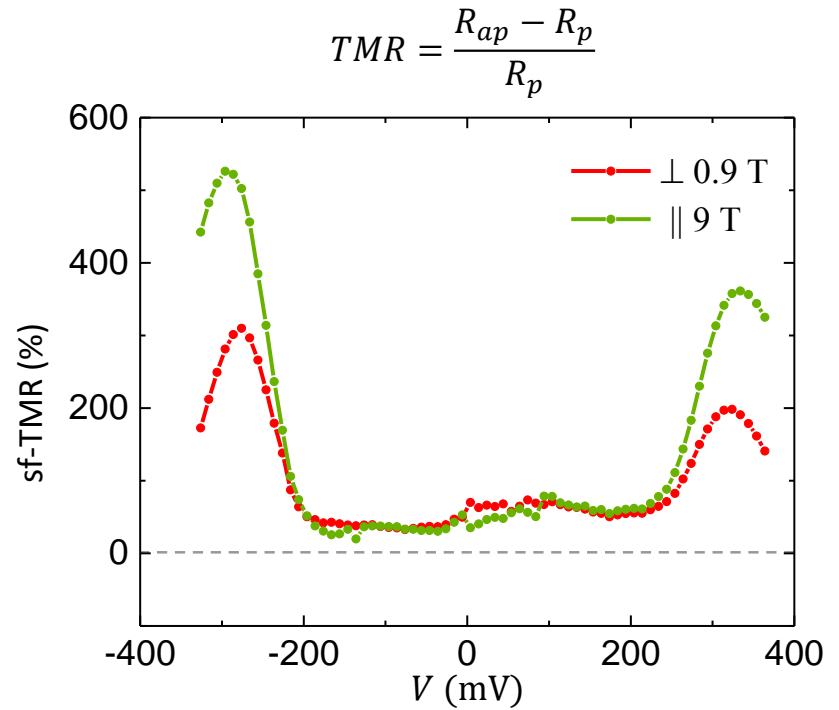
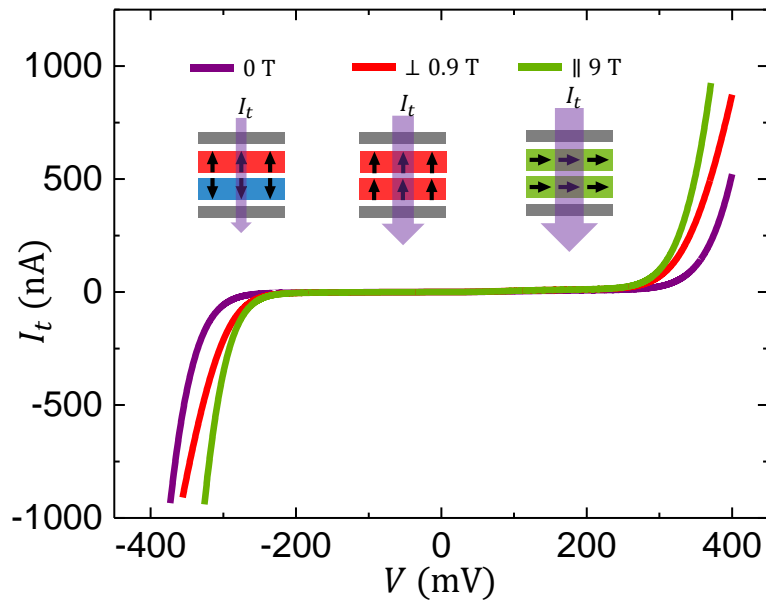
Tunneling current at selected magnetic field



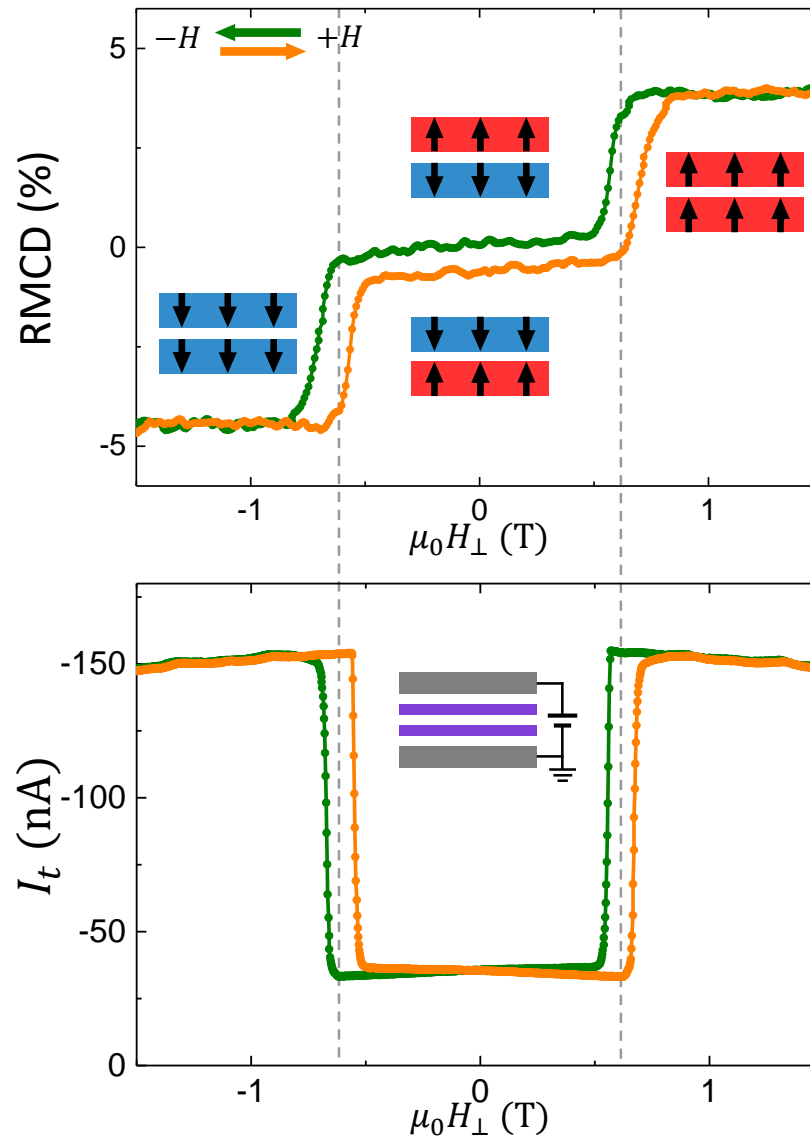
Tunneling current at selected magnetic field



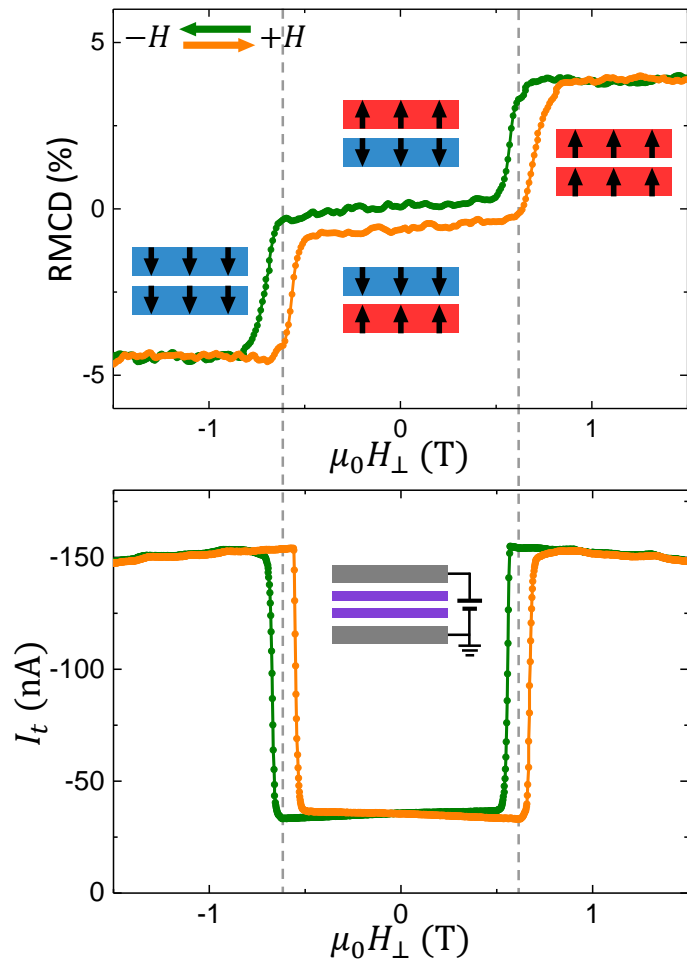
Giant Spin-Filter TMR Effect



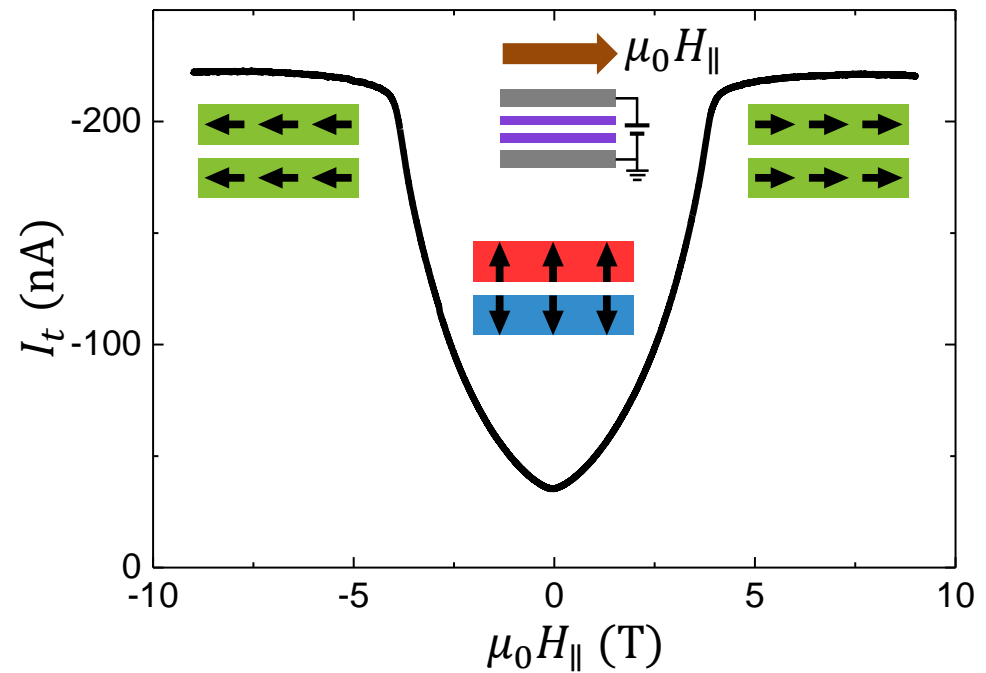
Giant Spin-Filter TMR Effect



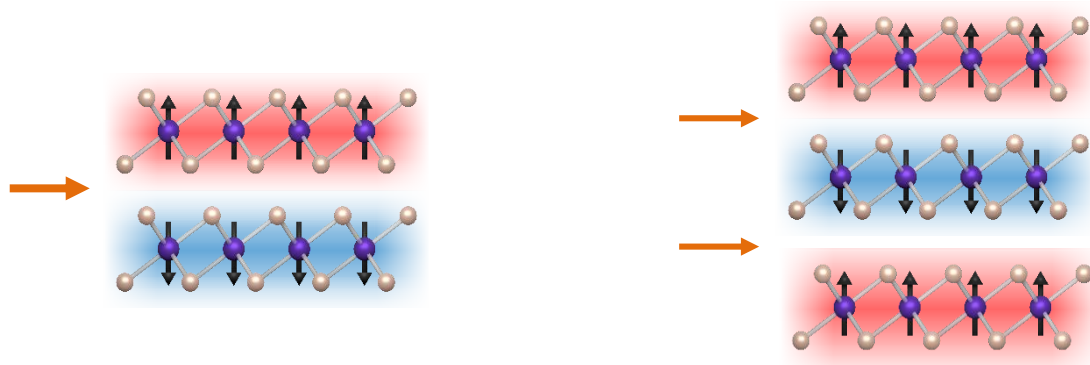
Measuring Magnetic Anisotropy Field



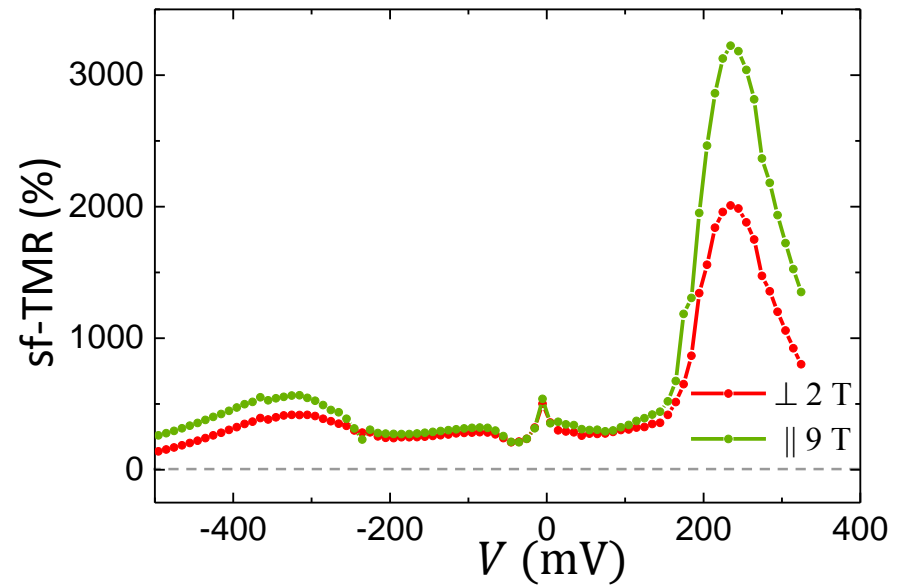
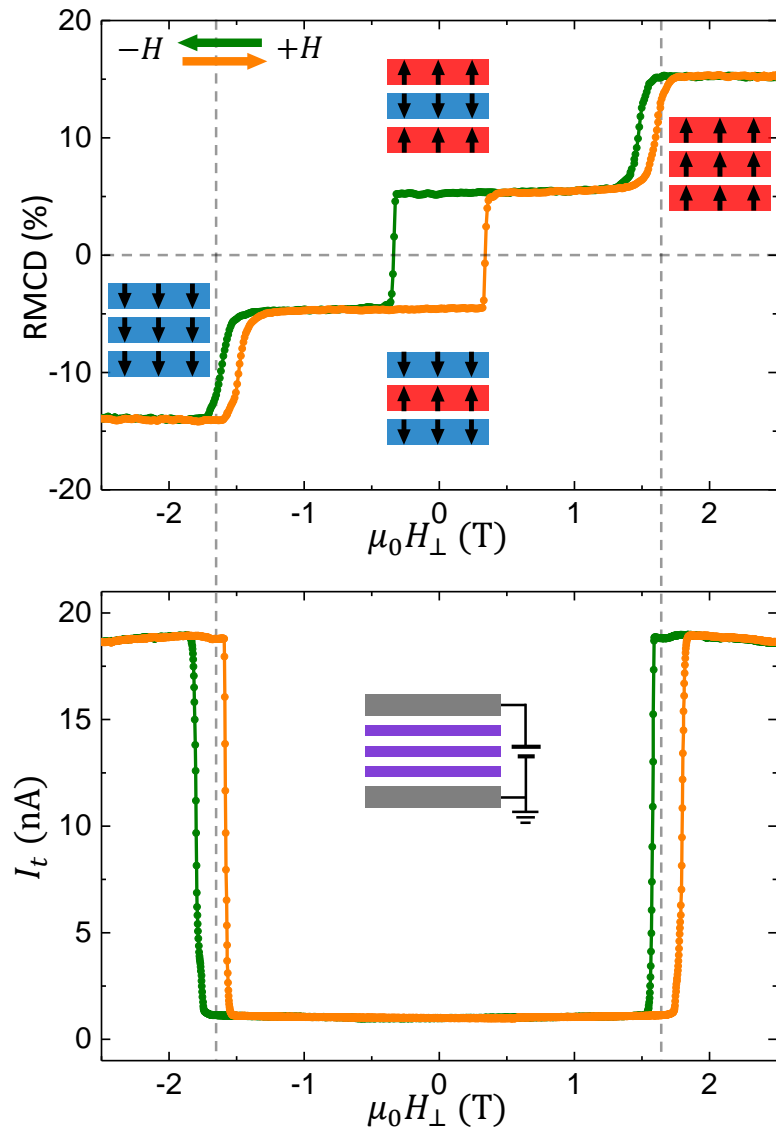
- **Out-of-plane magnetic anisotropy**
- **Spin-canting effect**
- **3.8 T magnetic anisotropy field**



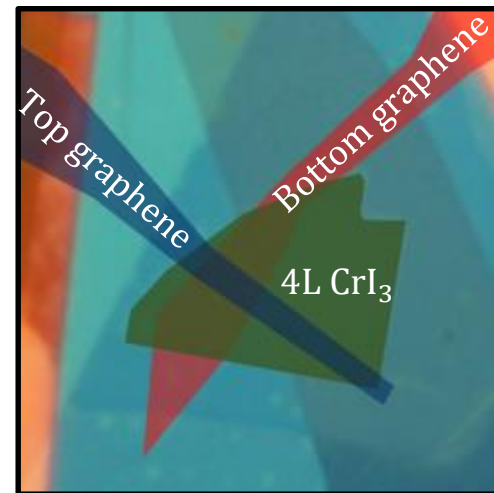
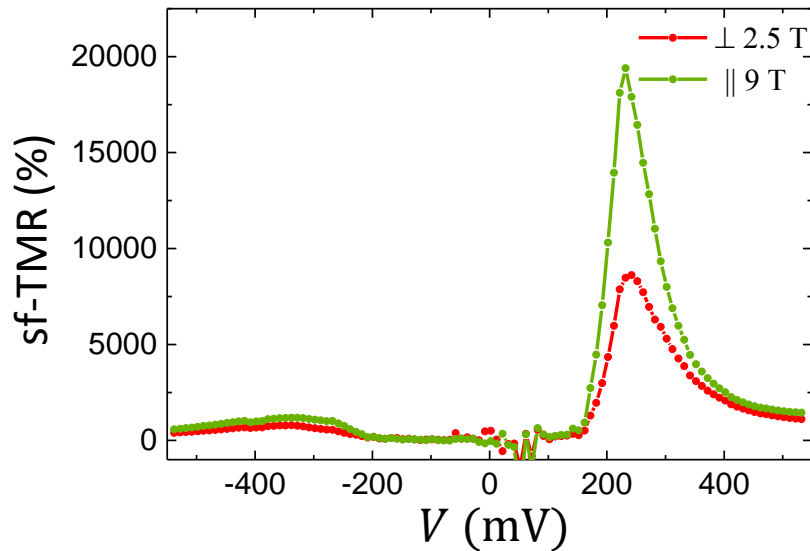
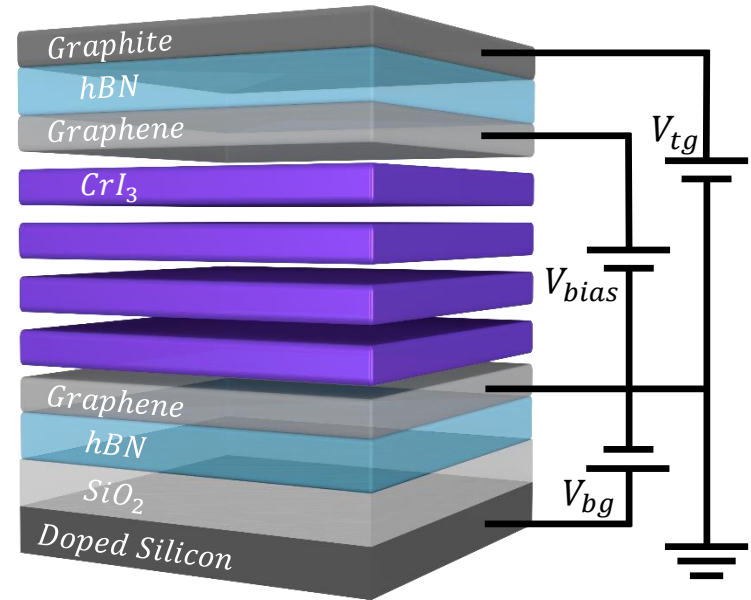
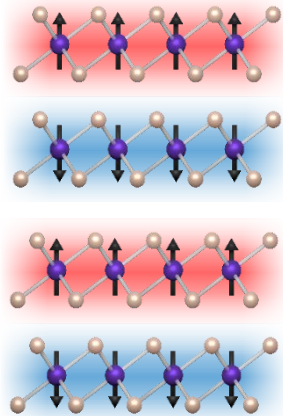
Can we further increase TMR?



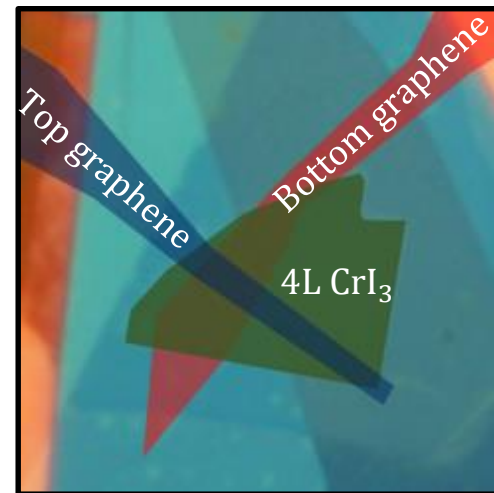
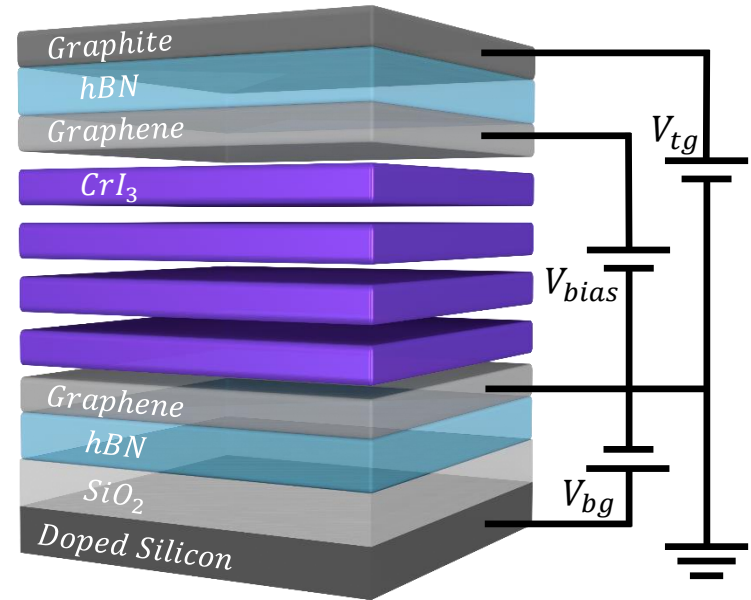
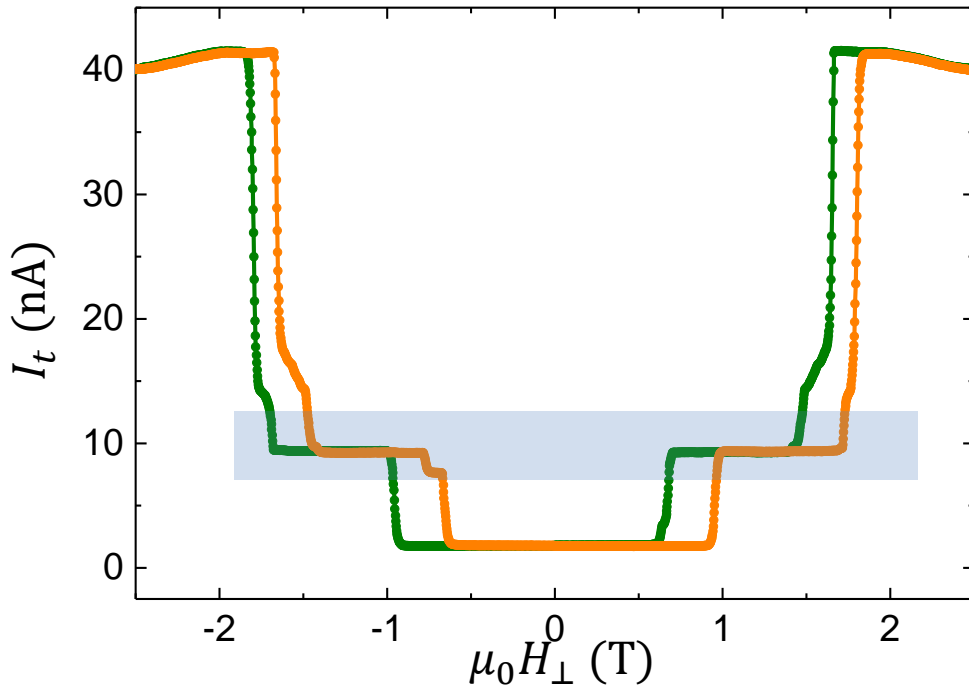
Trilayer CrI₃ MTJ



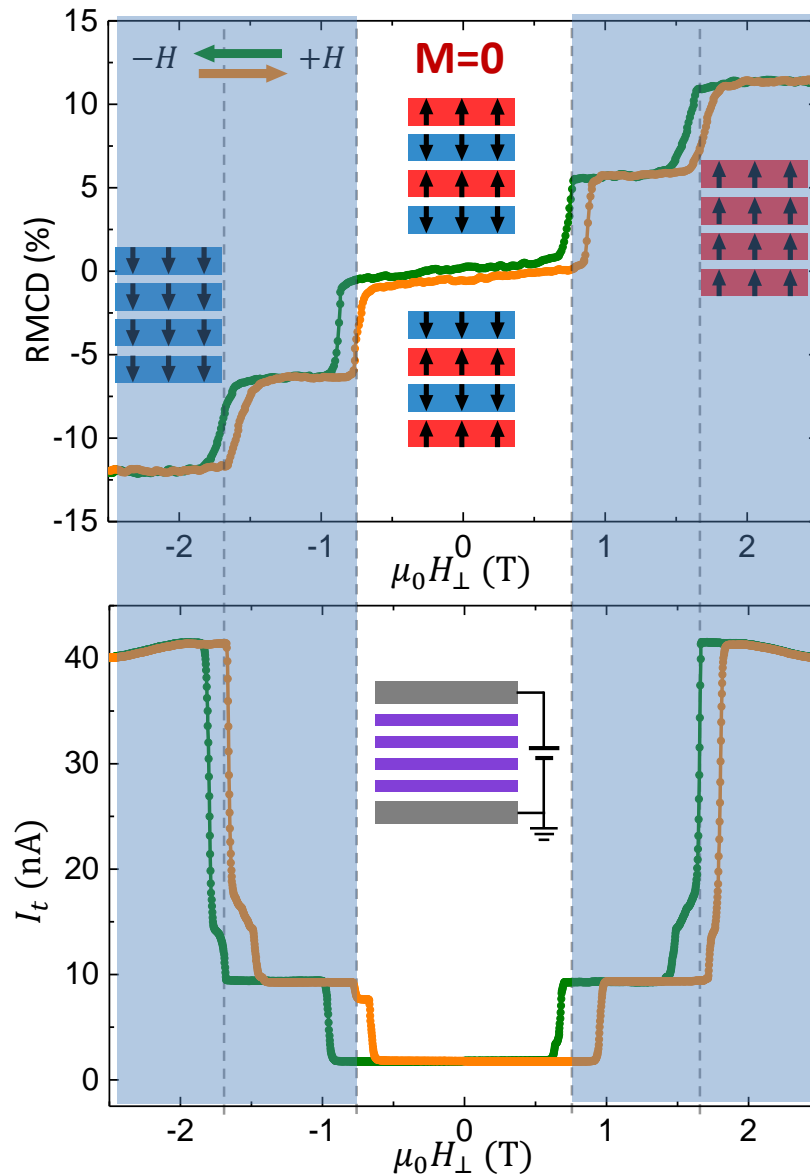
Giant Spin-Filter TMR in Four Layer CrI₃ MTJ



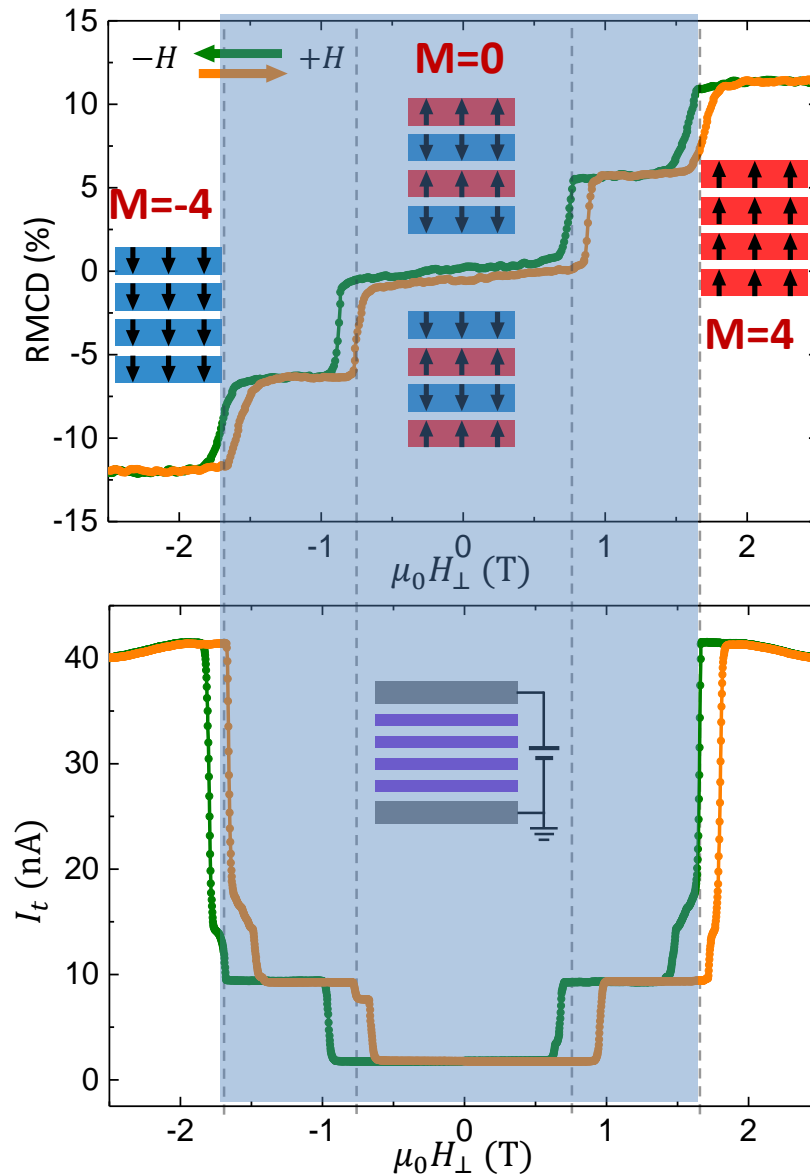
Multiple Magneto-Resistance States



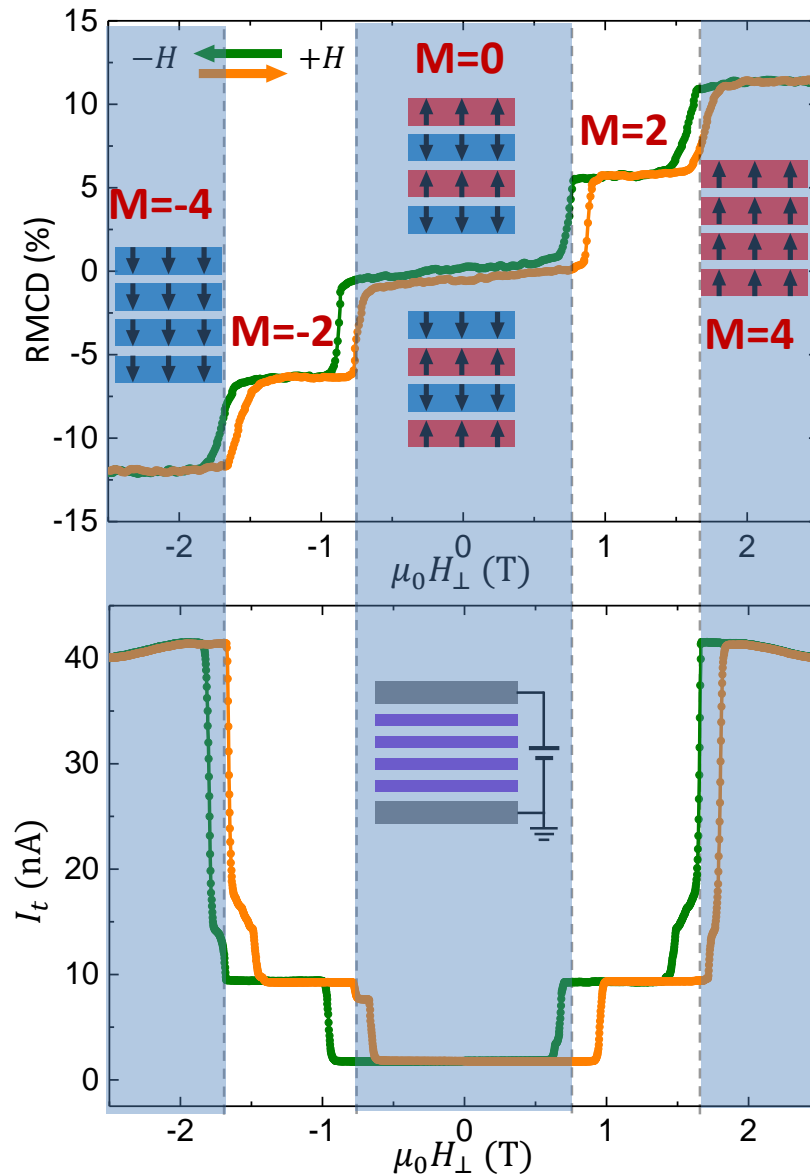
Multiple Magneto-Resistance States



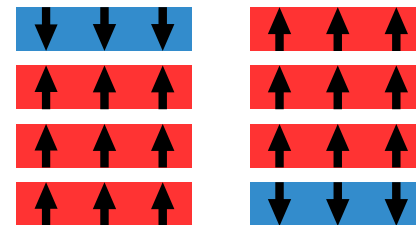
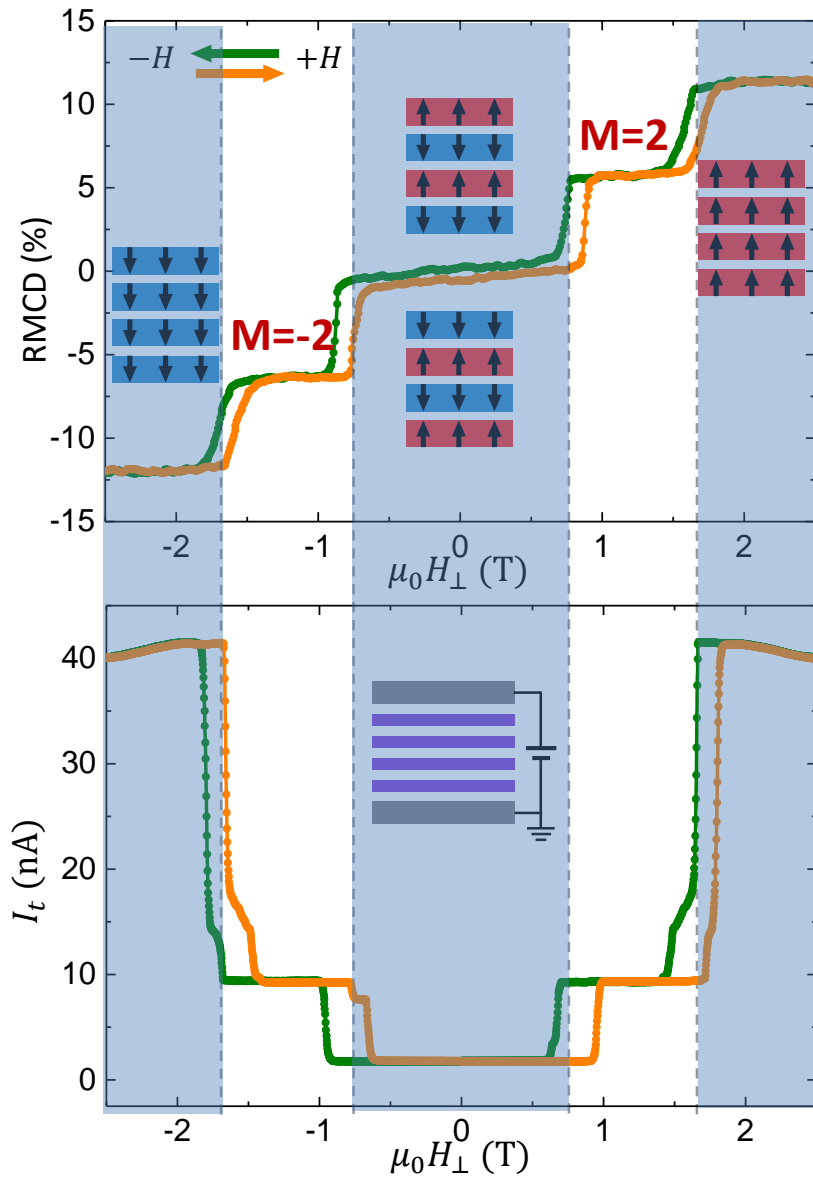
Multiple Magneto-Resistance States



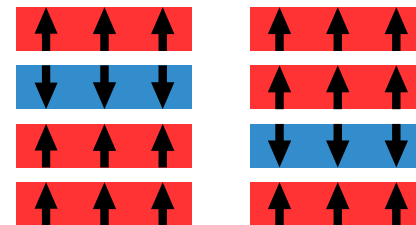
Multiple Magneto-Resistance States



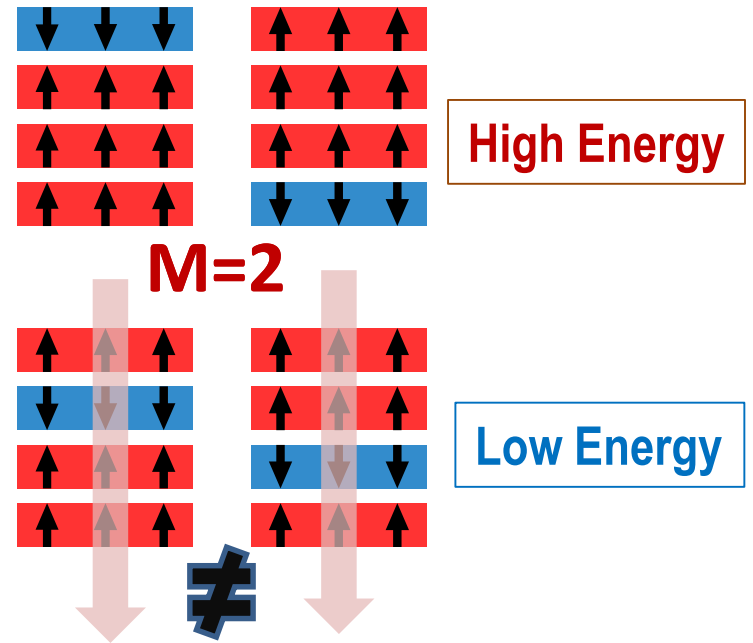
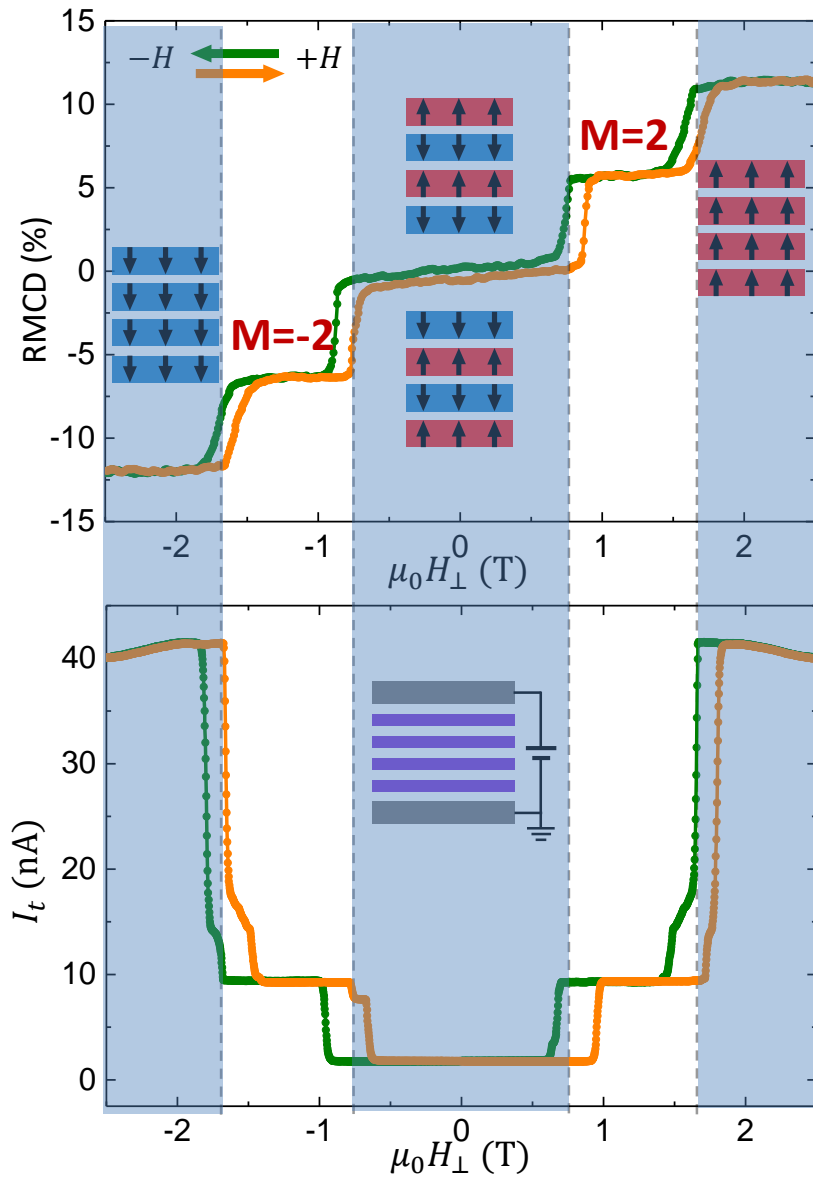
Multiple Magnetic-States



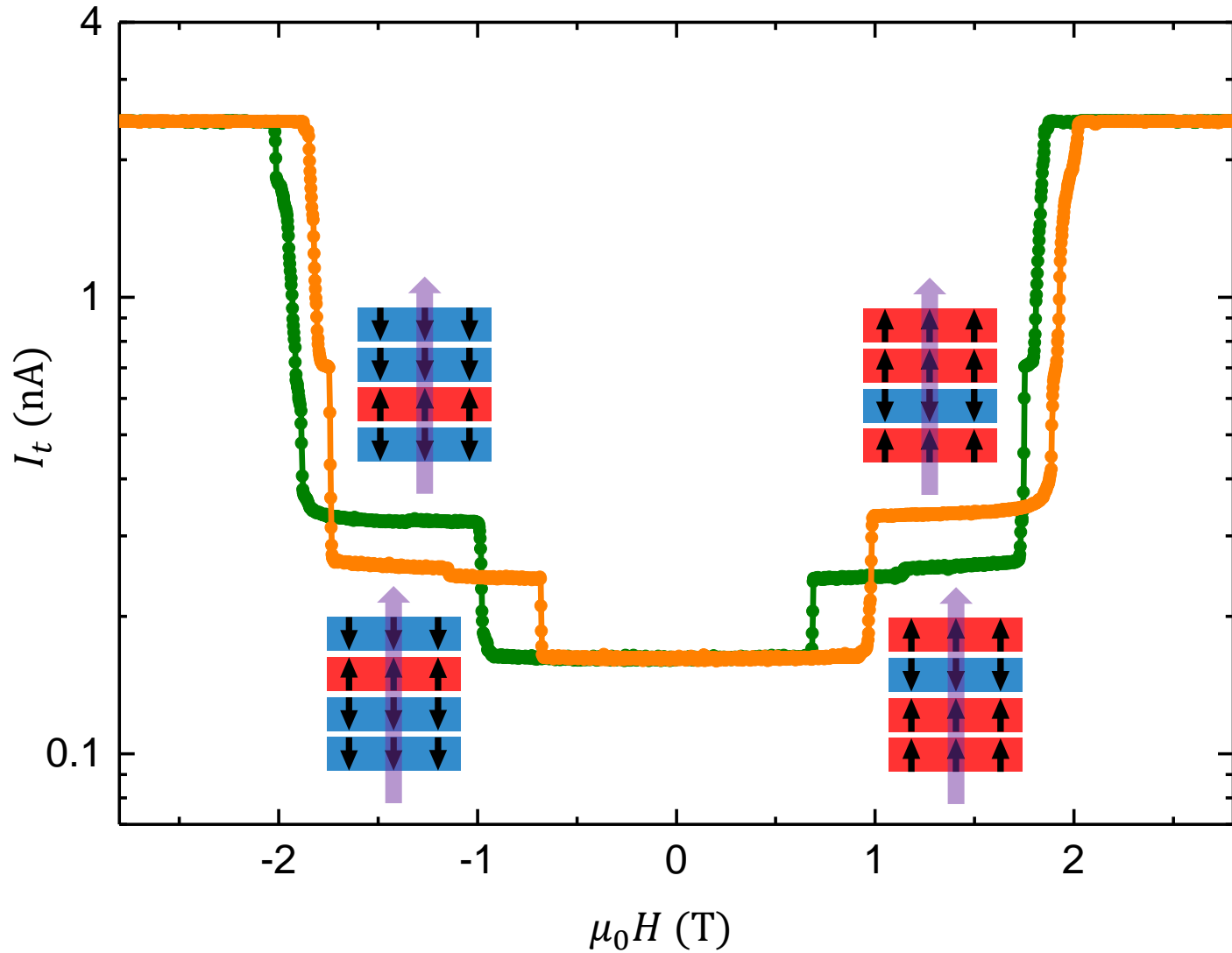
M=2



Multiple Magnetic-States

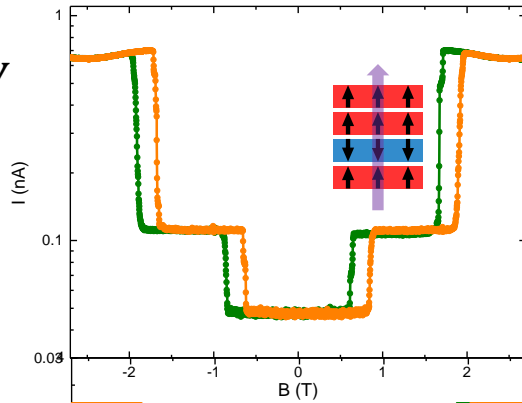


Multiple Magneto Resistance States

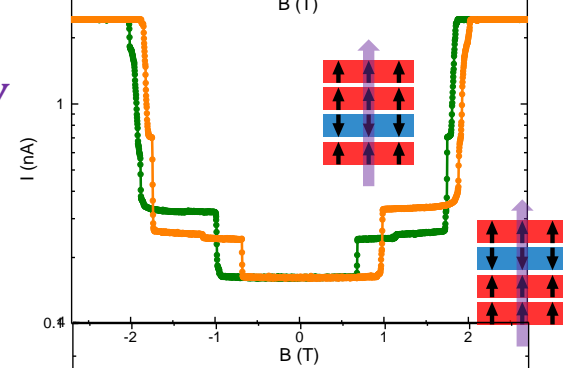


Electrical Switching of Magneto Resistance States

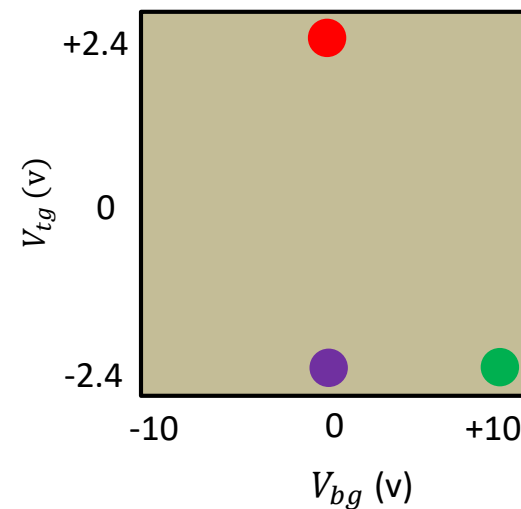
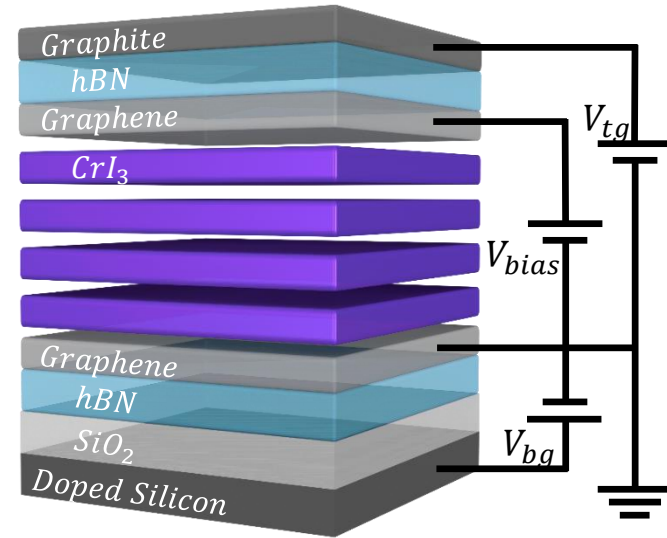
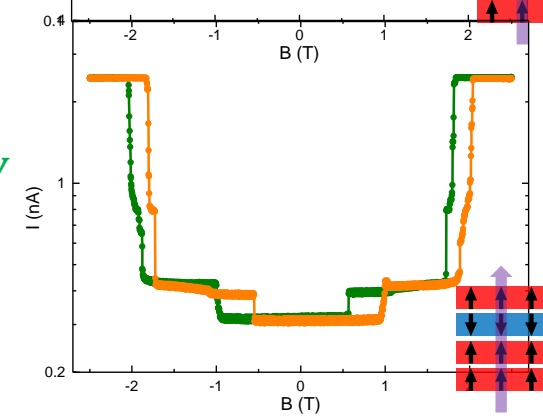
$V_{tg} = +2.4 \text{ V}$
 $V_{bg} = 0 \text{ V}$



$V_{tg} = -2.4 \text{ V}$
 $V_{bg} = 0 \text{ V}$



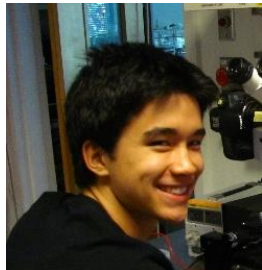
$V_{tg} = -2.4 \text{ V}$
 $V_{bg} = 10 \text{ V}$



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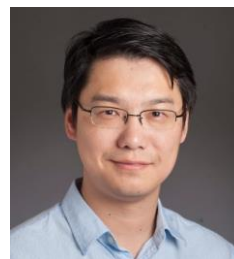


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Di Xiao



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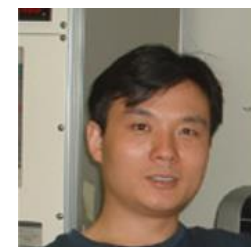


ONRL/UT

David Mandrus



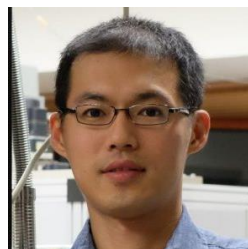
Jiaqiang Yan



David Cobden



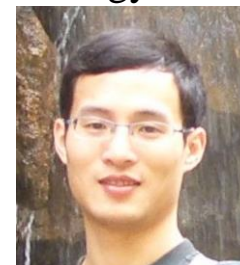
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