NEW DEVELOPMENTS ON CHROMIUM TRIHALIDES 2D FERROMAGNETS

Dr. David Soriano
Radboud University of Nijmegen

2D van der Waals Spin Systems (4 – 7 August 2020)

SP/CE
SPIN PHENOMENA
INTERDISCIPLINARY CENTER

Outline

- What we already know about CrX_3
- Coulomb interactions in CrI_3
- Magnetic polarons in bilayer CrI_3
- Take-home message

What we already know about CrX_3

- **Magnetism in Bulk**

Physics. — "*Further experiments with liquid helium. Z. Magnetic researches.* XXVIII. *Magnetisation of anhydrous* $CrCl_3$, $CoCl_2$ *and* $NiCl_2$ *at very low temperatures.*" By H. R. WOLTJER and H. KAMERLINGH ONNES.

(Communicated at the meeting of May 30, 1925).

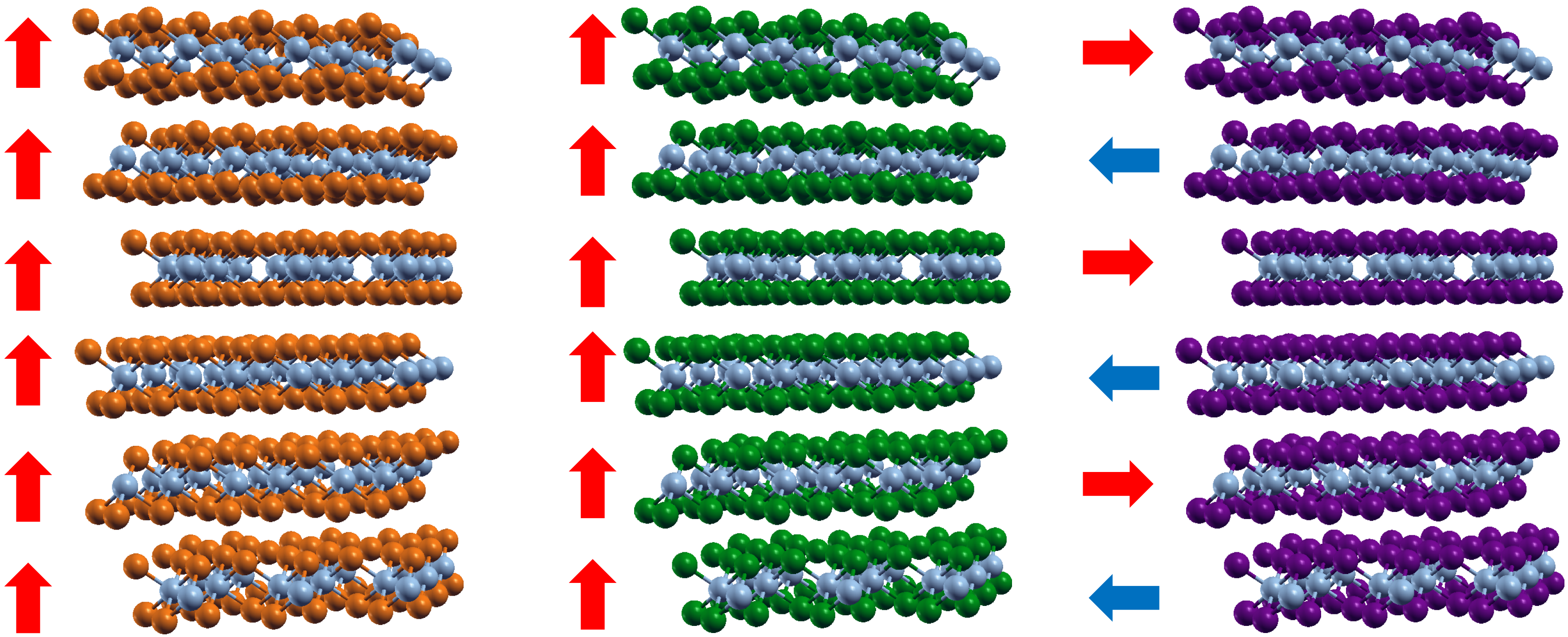
§ 1. *Introduction.* In the preceding communication the magnetic anomalies shown by anhydrous $CrCl_3$, $CoCl_2$ and $NiCl_2$ at the temperatures of liquid hydrogen have been pointed out. It seemed to be important to extend this research to the very low temperatures obtainable with liquid helium. The following questions came to the foreground: as regards $CrCl_3$ whether the initial susceptibility would increase with decreasing temperature as strongly as in the liquid hydrogen region and whether in strong fields saturation phenomena would appear; for the other substances whether the decrease of the susceptibility with decreasing temperature that seemed to be indicated by the measurements in liquid hydrogen would be continued.

H. Kamerlingh Onnes, *Leiden Comm.*, **167b**, 614 (1925)

What we already know about CrX_3

➢ **Magnetism in Bulk**

CrI_3 | $CrBr_3$ | $CrCl_3$



Dillon et al., *Journal of Applied Physics*, **36**, 1259 – 1260 (1965)
Tsubokawa, *Journal of the Physical Society of Japan*, **15**, 1664 – 1668 (1960)
Cable et al., *Journal of Physics and Chemistry of Solids*, **19**, 29 – 34 (1961)

What we already know about CrX_3

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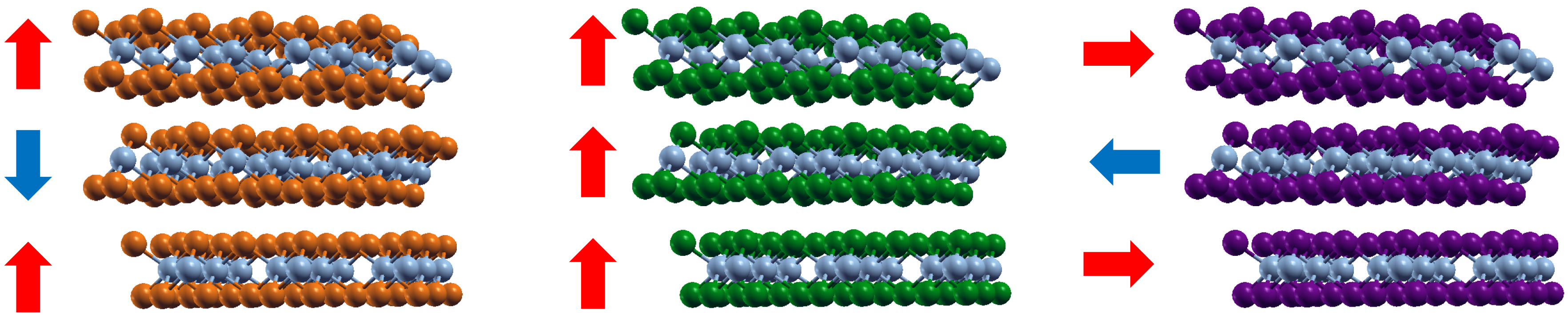
- **Magnetism in thin samples**

CrI_3 $CrBr_3$ $CrCl_3$



Huang et al., *Nature*, **546**, 270 – 273 (2017)
Klein, **DS**, *et al.*, *Science*, **360**, 1218 – 1222 (2018)
Song et al., *Science*, **360**, 1214 – 1218 (2018)
Klein et al., *Nature Physics*, **15**, 1255 – 1260 (2019)
Kim et al., *PNAS*, **116**, 11131 – 11136 (2019)

What we already know about CrX_3

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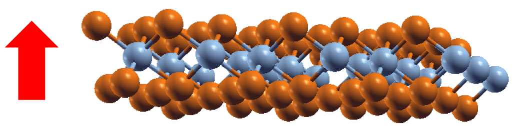
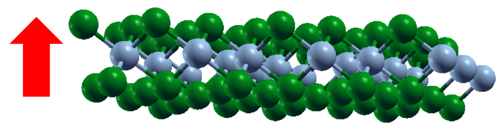
- **Magnetism in monolayer**

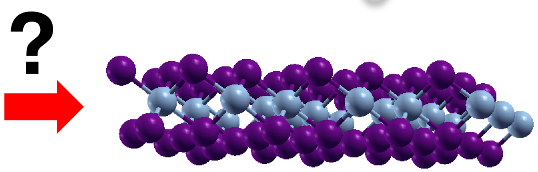
CrI_3



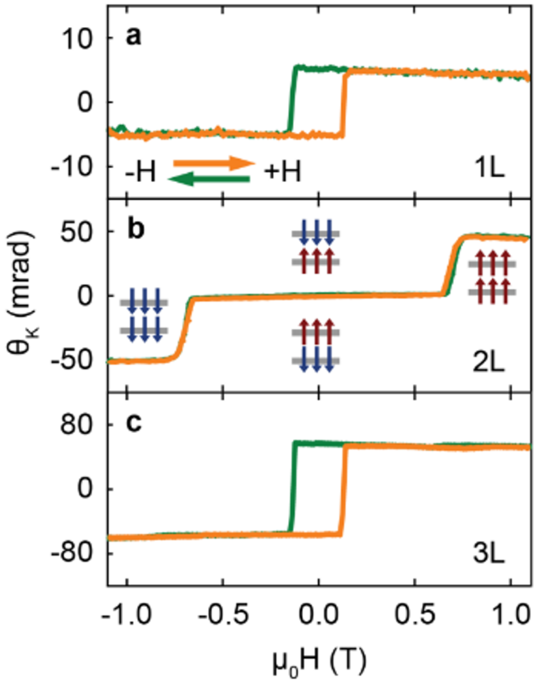
$CrBr_3$

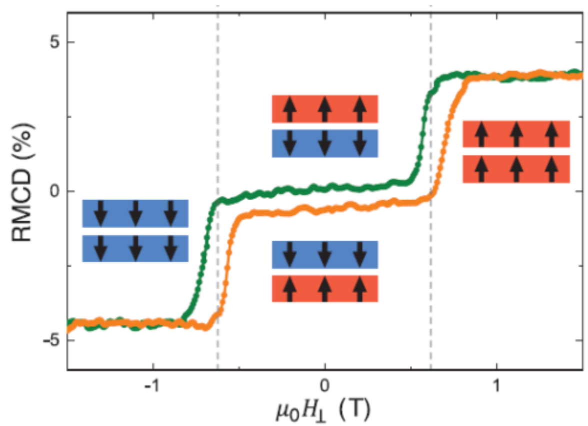


$CrCl_3$



Bedoya-Pinto et al., *arXiv:***2006.07605**
(Talk Thursday 9:00)





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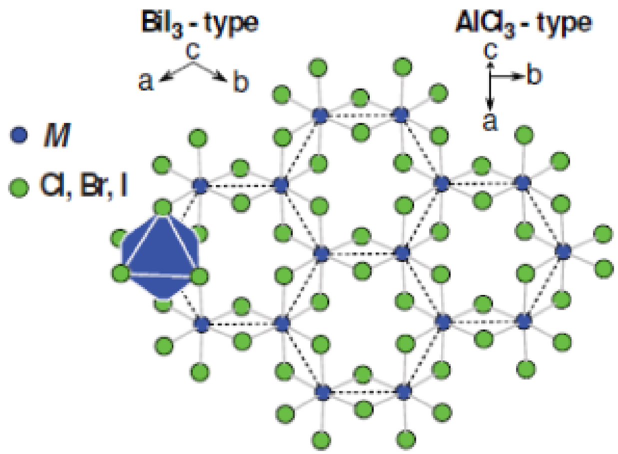
Kim et al., *PNAS*, **116**, 11131 – 11136 (2019)

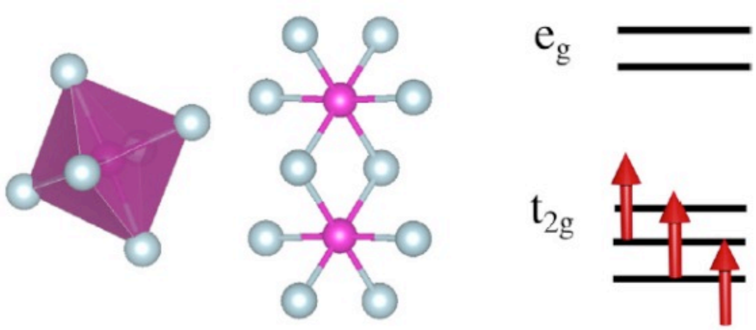
What we already know about CrX_3

- **Magnetism at the microscopic level: DFT**



Octahedral ligand field: **Crystal field splitting**

Cr:**[Ar]**$3d^5 4s^1$ → Cr^{3+}:**[Ar]**$3d^3$ **(m = 3 μ_B)**



Lado and Fernández-Rossier, *2D Materials* **4** 035002 (2017)
McGuire, *Crystals*, **7**, 121 (2017)
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

What we already know about CrX_3

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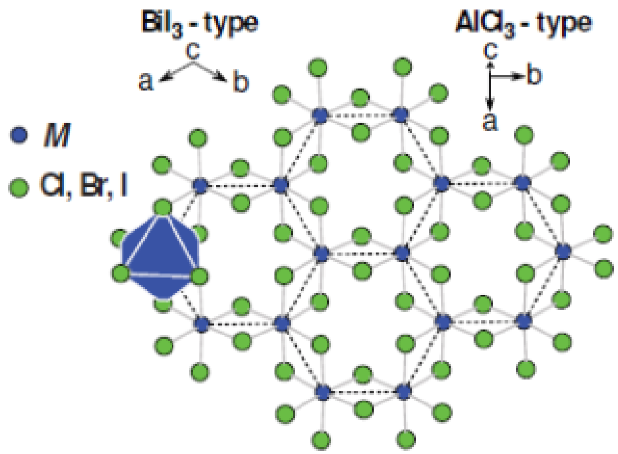

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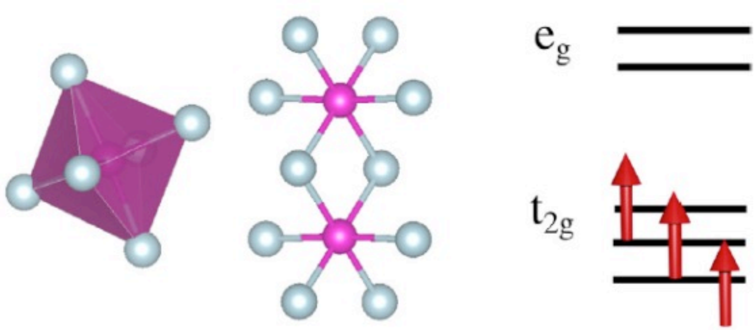

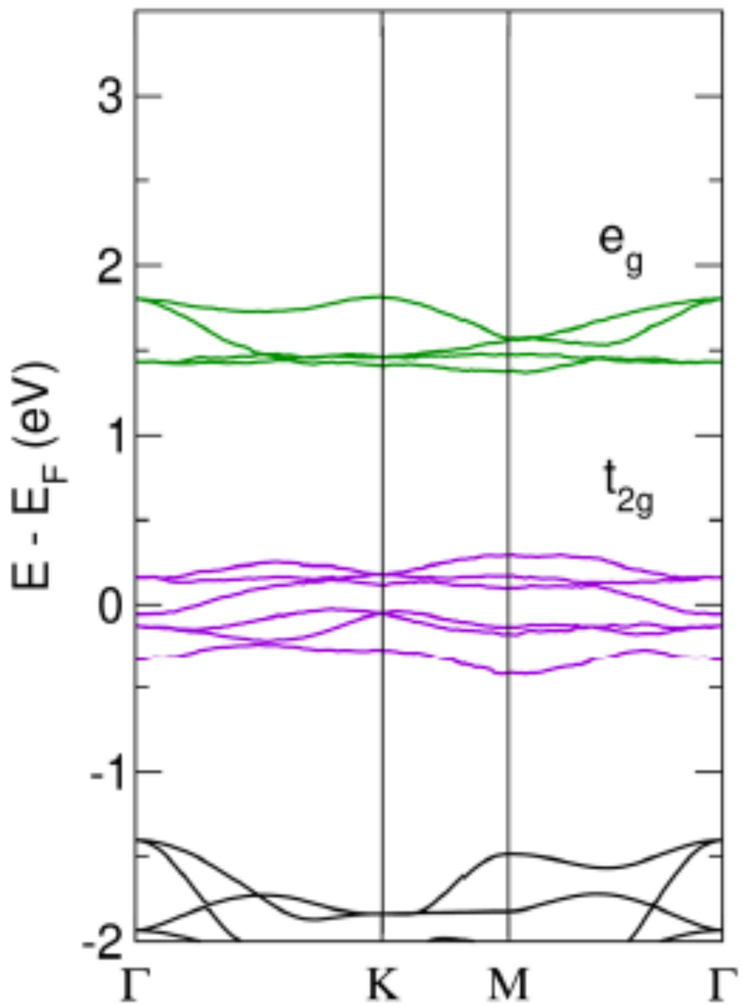

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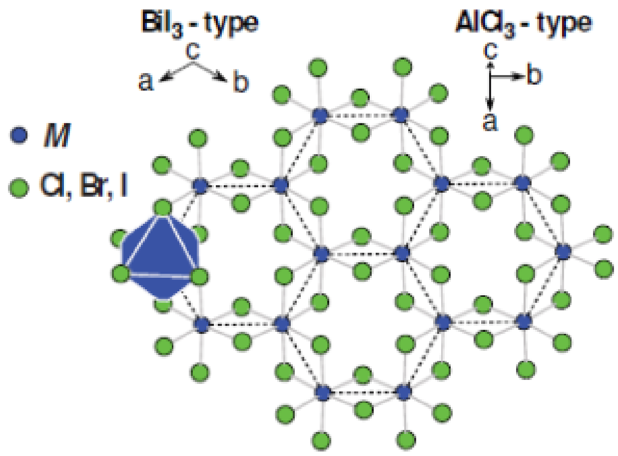
What we already know about CrX_3

- **Magnetism at the microscopic level: DFT+U+J**



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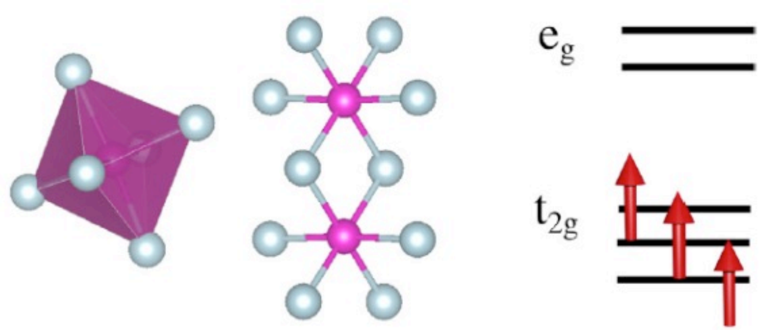

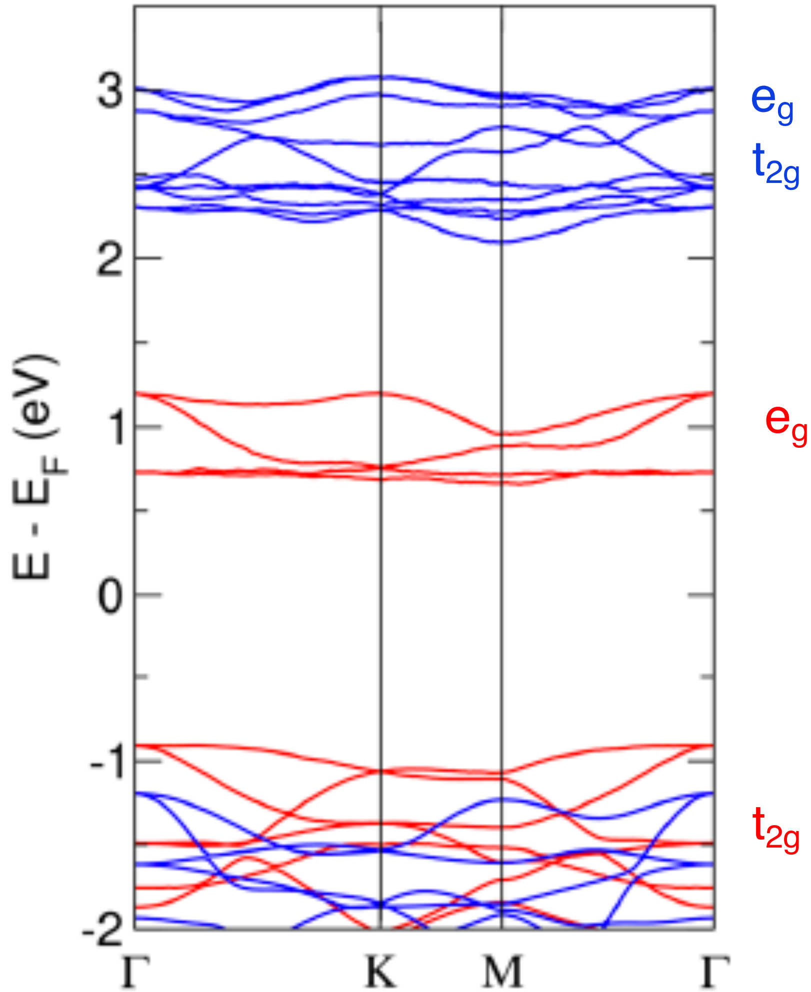

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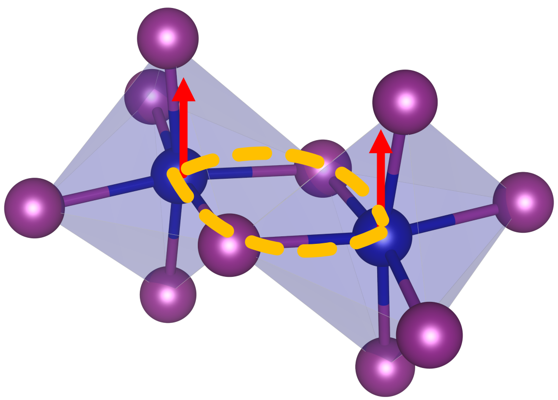
What we already know about CrX_3

- **Magnetism at the microscopic level: Intralayer Exchange**

Superexchange (Goodenough-Kanamori)



	d_{Cr-Cr}(Å)	α(deg)	$T_{C/N}$(K)	Type of anisotropy
CrI_3	4.026	97.5	45 [4] (68) [53]	Easy axis (z)
$CrBr_3$	3.722	94.9	27 [54] (37) [55]	Easy axis (z)
$CrCl_3$†	3.491	95.5	10* [48] 17** [56] (17) [57]	Easy plane (xy)

Kashin et al., *2D Materials*, **7**, 025036 (2020)

DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

What we already know about CrX_3

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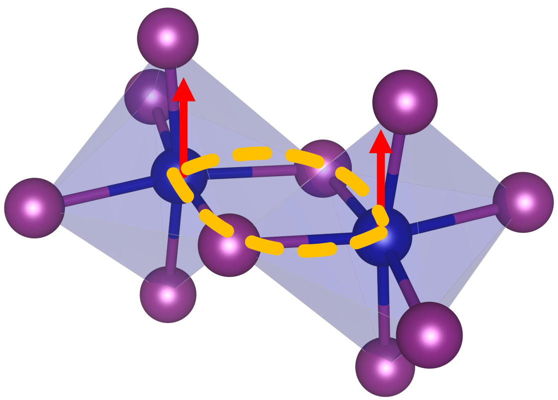

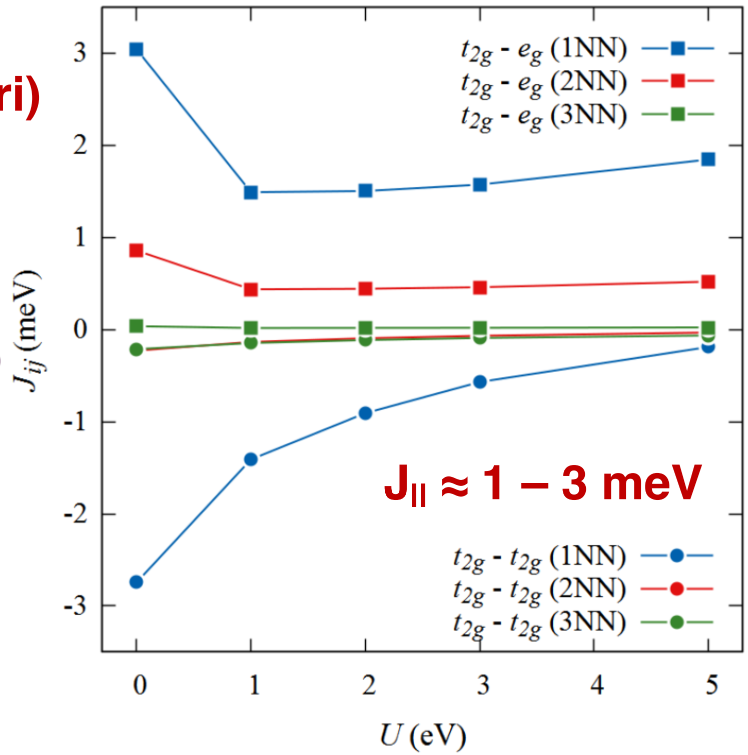

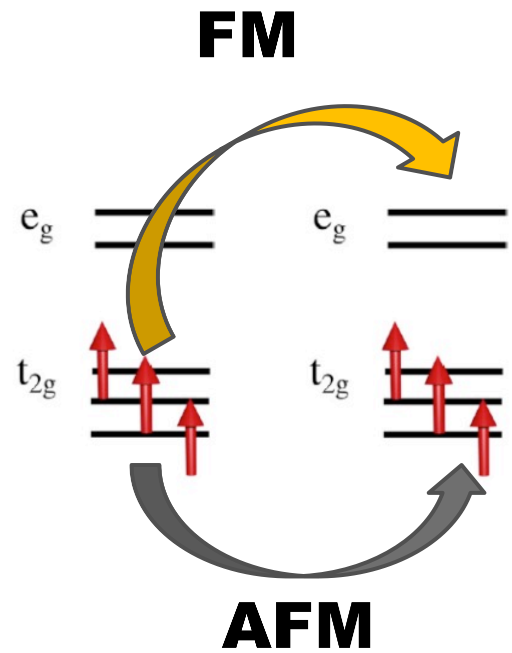

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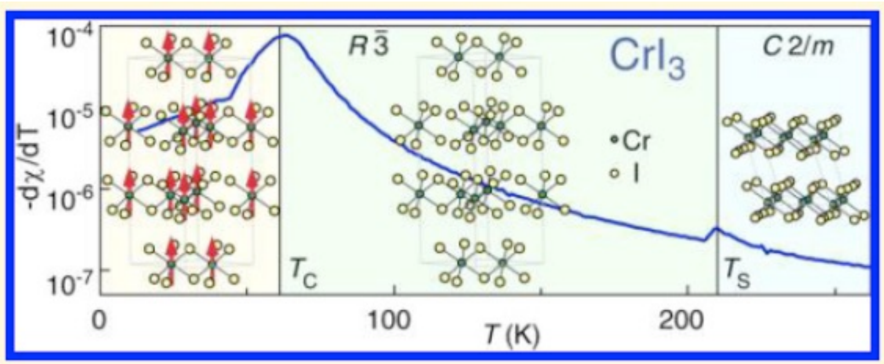
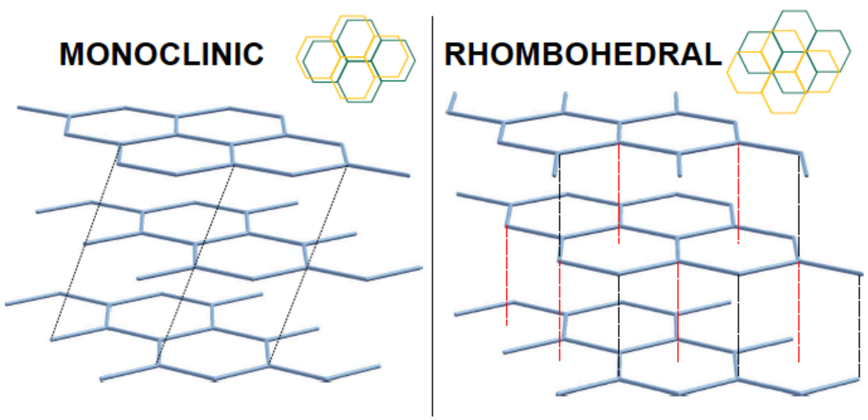
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

What we already know about CrX_3

➢ **Magnetism at the microscopic level: Interlayer Exchange**

T_S = 210 K (CrI_3); 420 K ($CrBr_3$); 240 K ($CrCl_3$)





McGuire et al., *Chem. Mater.*, **27**, 612 – 620 (2015)
Sivadas et al., *Nano Letters*, **18**, 7658 – 7664 (2018)
DS, Cardoso and Fernández-Rossier, Sol. Stat. Comm., **299**, 113662 (2019)
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

What we already know about CrX_3

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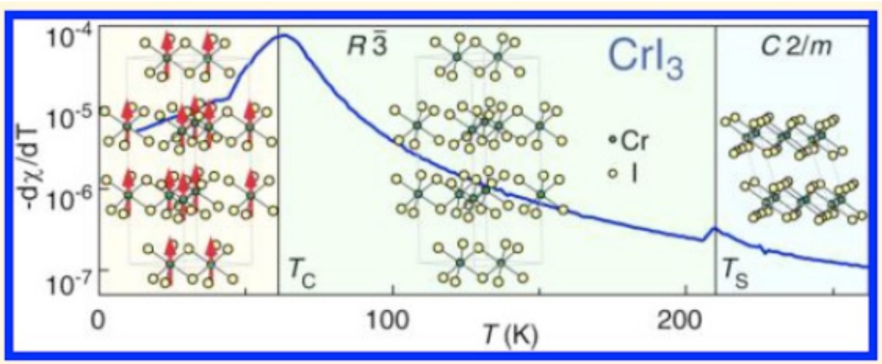
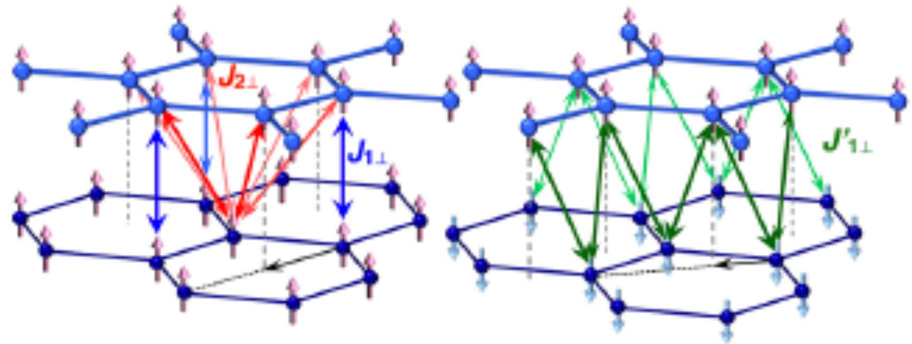
➢ **Magnetism at the microscopic level: Interlayer Exchange**

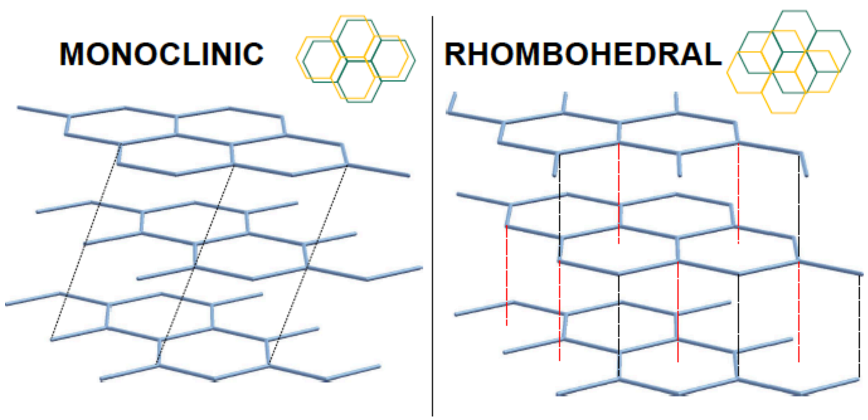
T_S = 210 K (CrI_3); 420 K ($CrBr_3$); 240 K ($CrCl_3$)

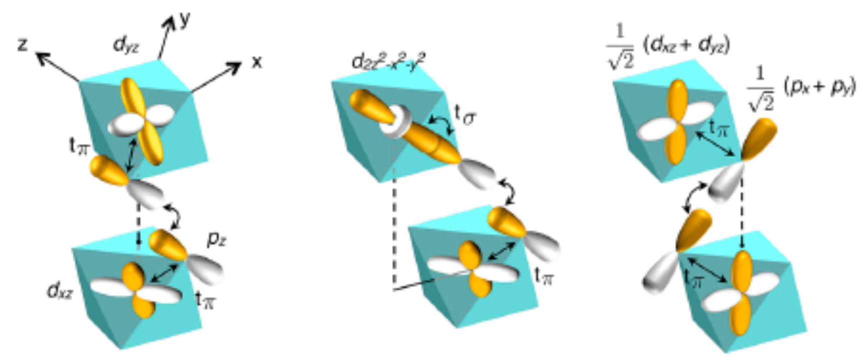
Super-superexchange





$J_{1\perp}$ $J_{2\perp}$ $J'_{1\perp}$





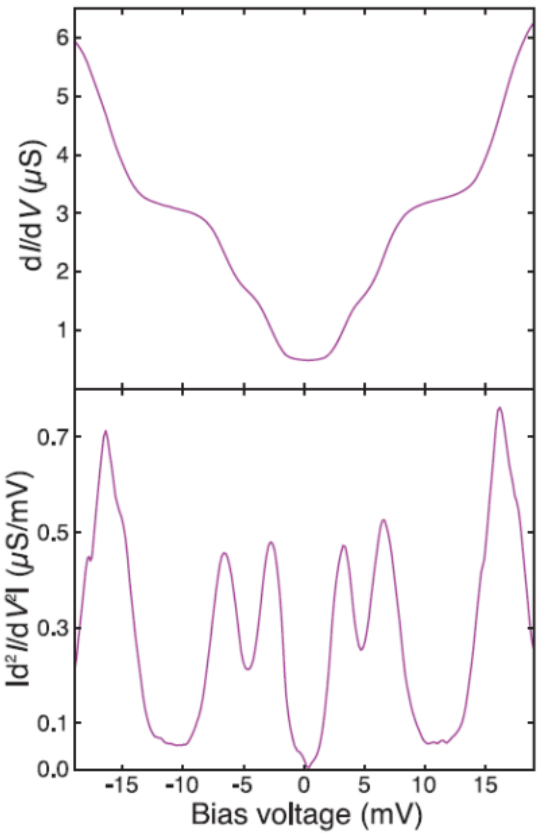
$J_\perp \approx 0.04$ meV

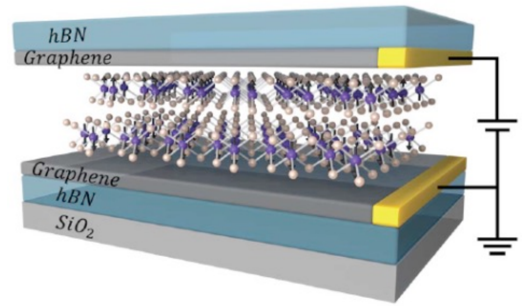
McGuire et al., *Chem. Mater.*, **27**, 612 – 620 (2015)
Sivadas et al., *Nano Letters*, **18**, 7658 – 7664 (2018)
DS, Cardoso and Fernández-Rossier, Sol. Stat. Comm., **299**, 113662 (2019)
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

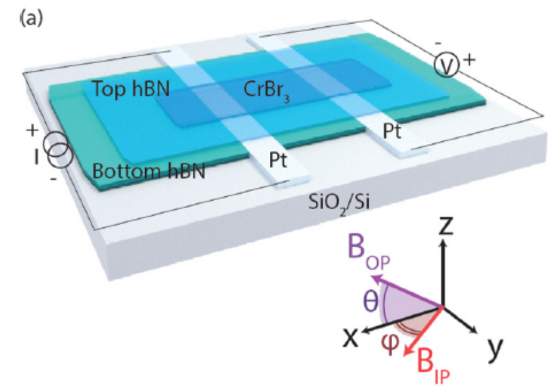
What we already know about CrX_3

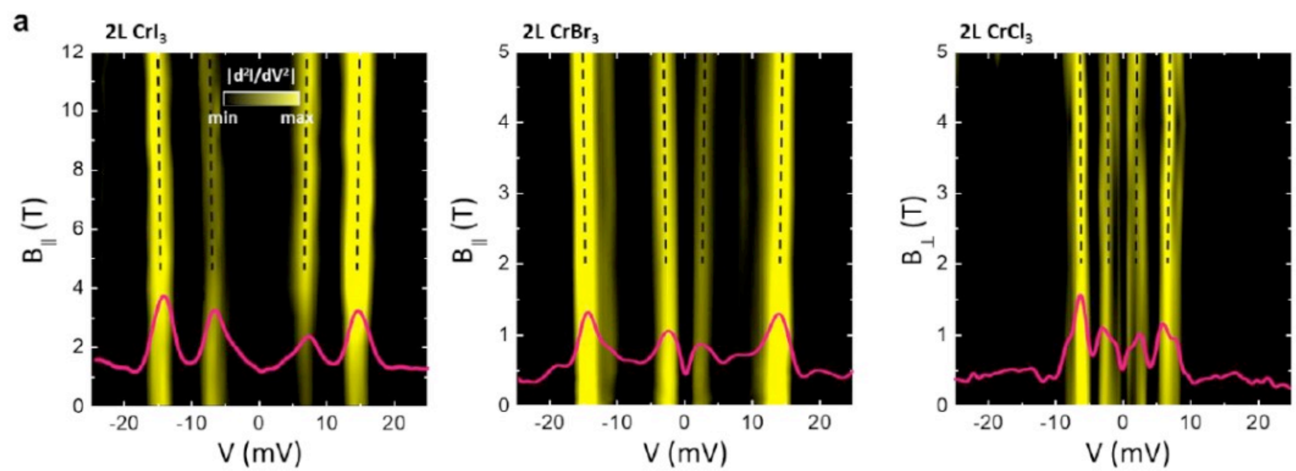
- **Magnetism at the microscopic level: Spin-Waves**

Inelastic Electron Tunneling Spectroscopy (IETS)









Klein, **DS**, *et al.*, *Science*, **360**, 1218 – 1222 (2018)
Kim et al., *PNAS*, **116**, 11131 – 11136 (2019)
Liu et al., *Phys. Rev. B*, **101**, 205407 (2020)

What we already know about CrX_3

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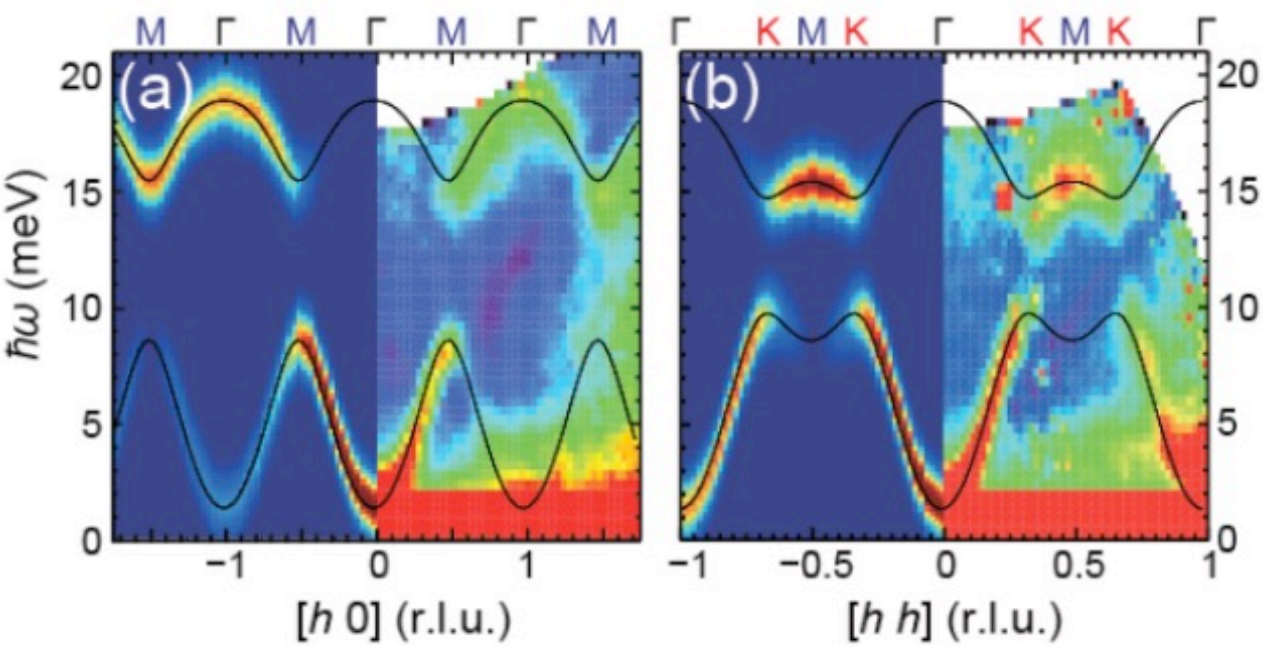
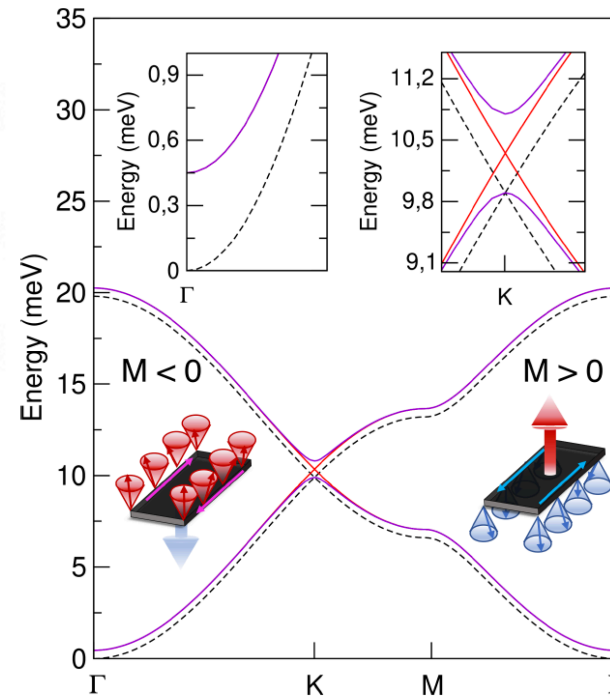
- **Magnetism at the microscopic level: Spin-Waves**

Inelastic Neutron Scattering (INS)





$\Delta_\Gamma = 2AS + 3SJ_z$
$\Delta_K \propto$ **K or D'**

A: Single-ion Anisotropy
J_z: Anisotropic Exchange
K: Kitaev interaction
D': 2nd Neighbor DMI

Chen et al., *Phys. Rev. X*, **8**, 041028 (2018)
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

What we already know about CrX_3

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➢ **Magnetism at the microscopic level: Anisotropy**

	$B^c_{\parallel}$(T)	$B^c_{\perp}$(T)	MAE(meV)	J_z (meV)	J_1 (meV)	J_2 (meV)	J_3 (meV)	K|D' (meV)	A (meV)
CrI_3	$6.5^{E\,31,54}$[FL] $3.8^{E\,12}$[BL]	$2.0^{E\,54}$[FL] $1.8^{E\,12,29}$[FL] $0.6^{E\,12}$[BL] $0.85^{E\,29}$[BL]	$0.65^{T\,43}$[ML] $0.68^{T\,45}$[ML]	$2.38^{E\,54}$[FL] $0.27^{E\,35}$[Bulk] $0.022^{T\,64}$[ML] $0.09^{T\,43}$[ML]	$2.29^{E\,54}$[FL] $2.01^{E\,34}$[Bulk] $0.20^{E\,35}$[Bulk] $3.24^{T\,64}$[ML] $2.2^{T\,43}$[ML] $1.44^{T\,67}$[ML] $1.0^{T\,46}$[ML] $2.86^{T\,45}$[ML] $2.29^{T\,68}$[ML]	$0.16^{E\,34}$[Bulk] $0.56^{T\,64}$[ML] $0.4^{T\,46}$[ML] $0.63^{T\,45}$[ML]	$-0.1^{E\,34}$[Bulk] $0.001^{T\,64}$[ML] $-0.15^{T\,45}$[ML]	$5.2^{E\,35}$[Bulk] $0.08^{T\,66}$[ML] $0.85^{T\,68}$[ML] $0.3^{E\,34}$[Bulk]	$0.22^{E\,34}$[FL] $0.056^{T\,64}$[ML] $0.1^{E\,67}$[ML]
$CrBr_3$	$0.4^{E\,54}$[FL]	$<0.01^{E\,54}$[FL]	$0.18^{T\,45}$[ML]	$1.58^{E\,54}$[FL]	$1.56^{E\,54}$[FL] $2.6^{T\,45}$[ML]	$0.38^{T\,45}$[ML]	$-0.15^{T\,45}$[ML]		
$CrCl_3$	$2.0^{E\,54}$[FL]	$2.4^{E\,54}$[FL] $0.85^{E\,49}$[BL] $1.6^{E\,49}$[FL]	$0.03^{T\,45}$[ML]	$0.91^{E\,54}$[FL]	$0.92^{E\,54}$[FL] $1.92^{T\,45}$[ML]	$0.23^{T\,45}$[ML]	$-0.13^{T\,45}$[ML]		

Still an open issue!

See recent preprint by Ke and Katsnelson **(arXiv:2007.14518)**
(Poster by Liqin Ke)

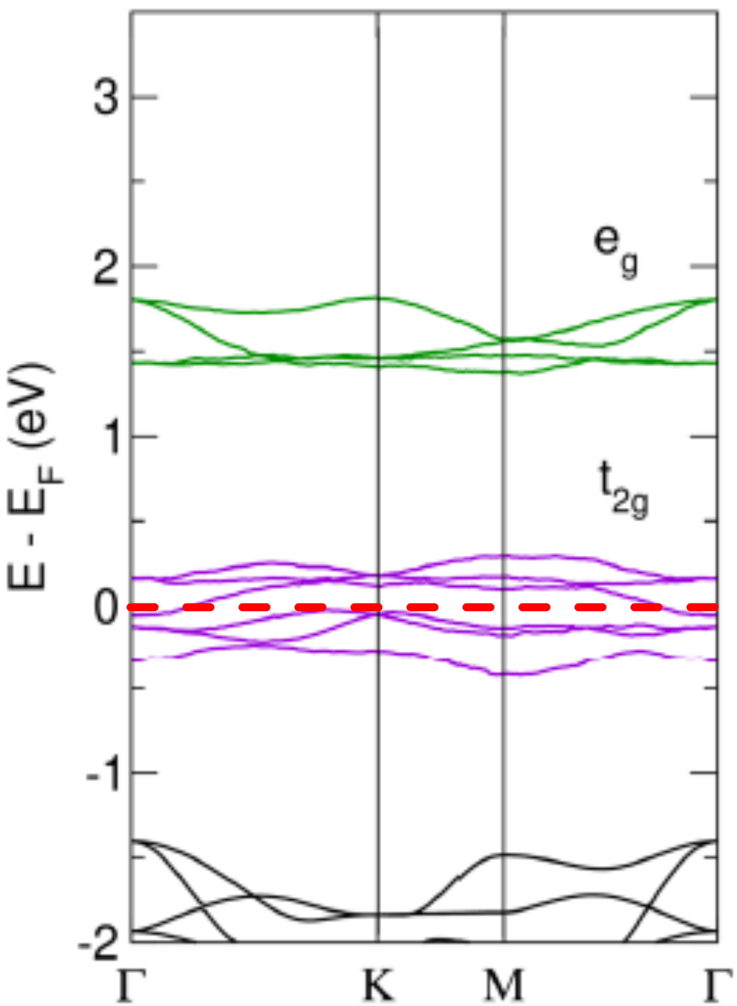
DS, Katsnelson and Fernández-Rossier, *Nano Letters (*Review) **10.1021/acs.nanolett.0c02381**

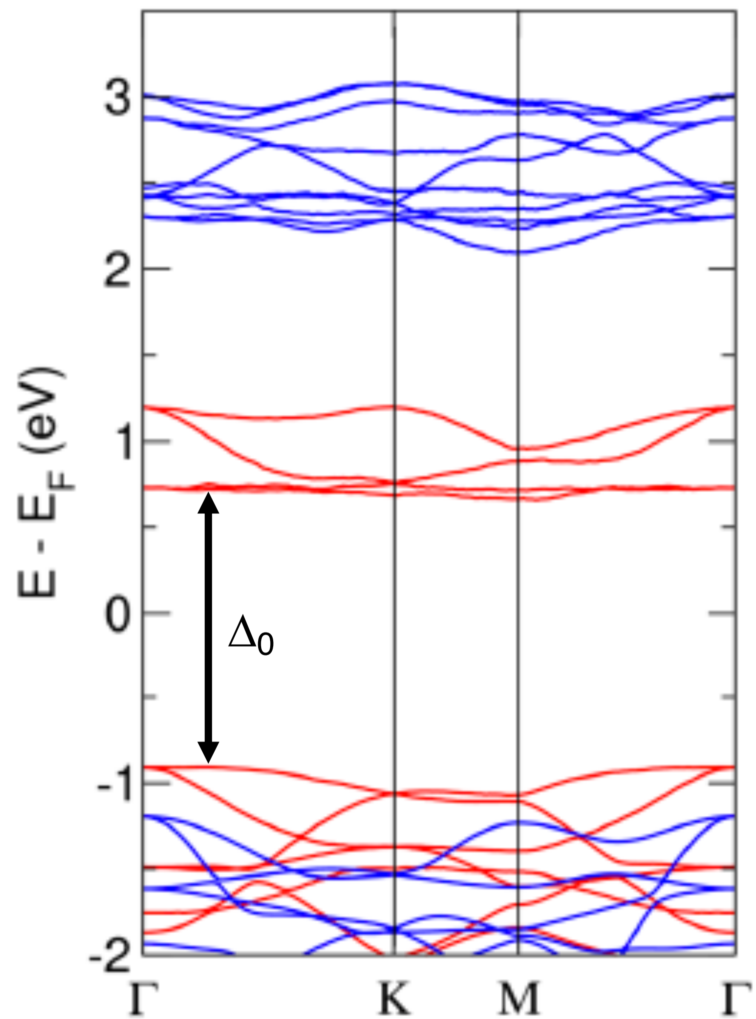
Coulomb Interactions in CrI_3

Coulomb interactions in CrI_3

- **Strongly correlated materials: LSDA+U+J**

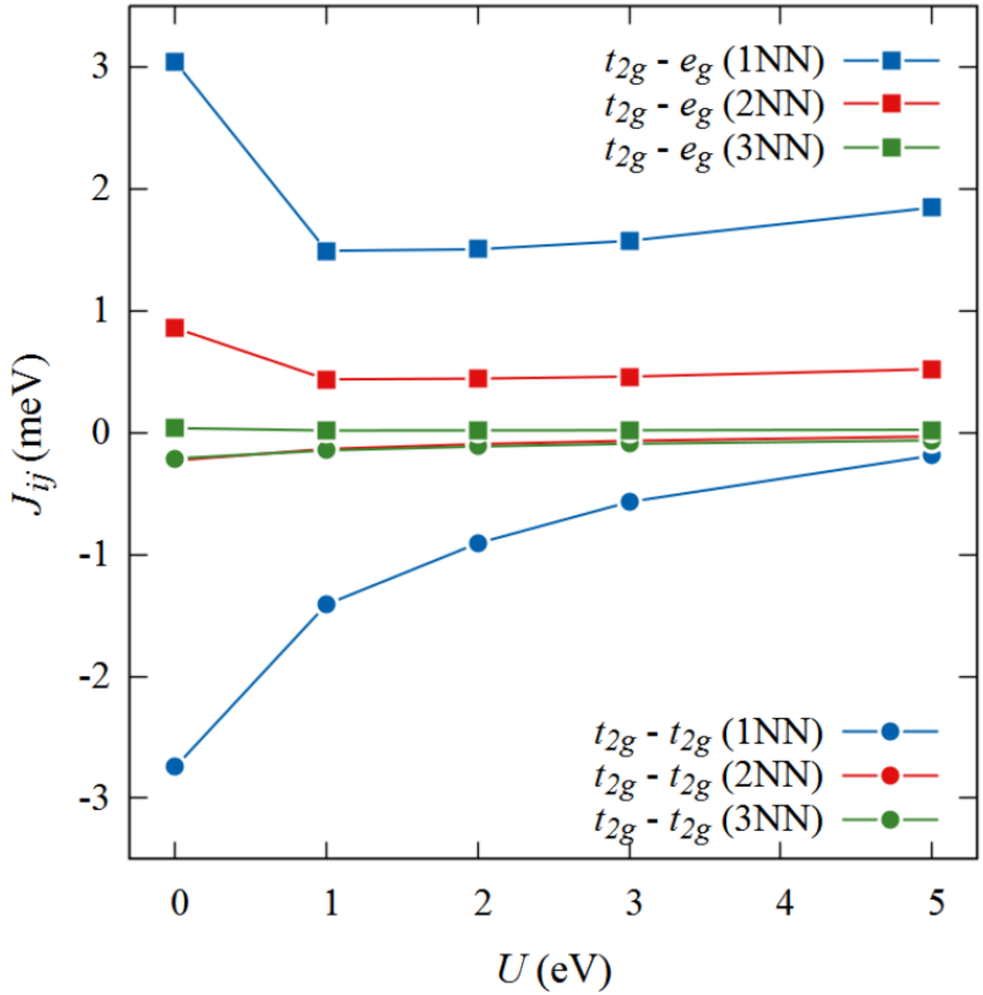
U = 3 eV; J = 0.5 eV





Coulomb interactions in CrI_3

- **Strongly correlated materials: J(U) dependence**



Kashin et al., *2D Materials*, **7**, 025036 (2020)

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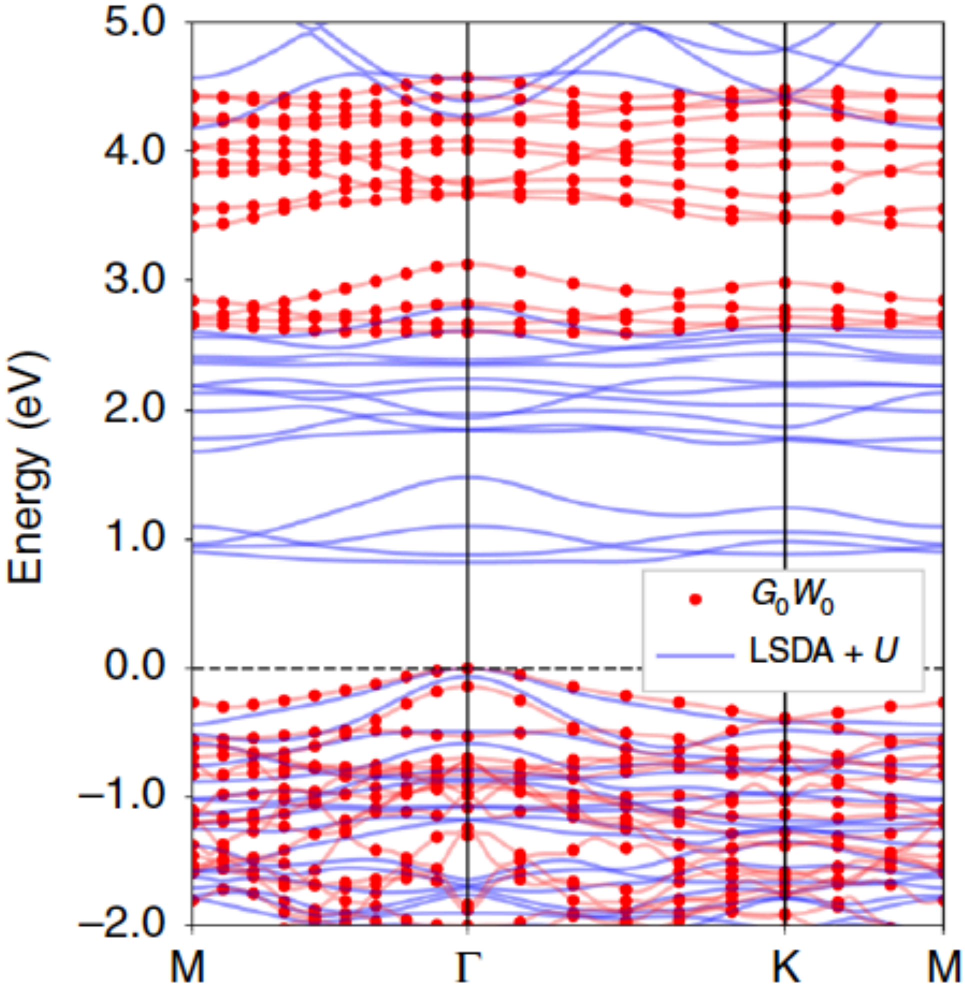
- **Strongly correlated materials: GW**

  Coulomb and dielectric screening effects are very important

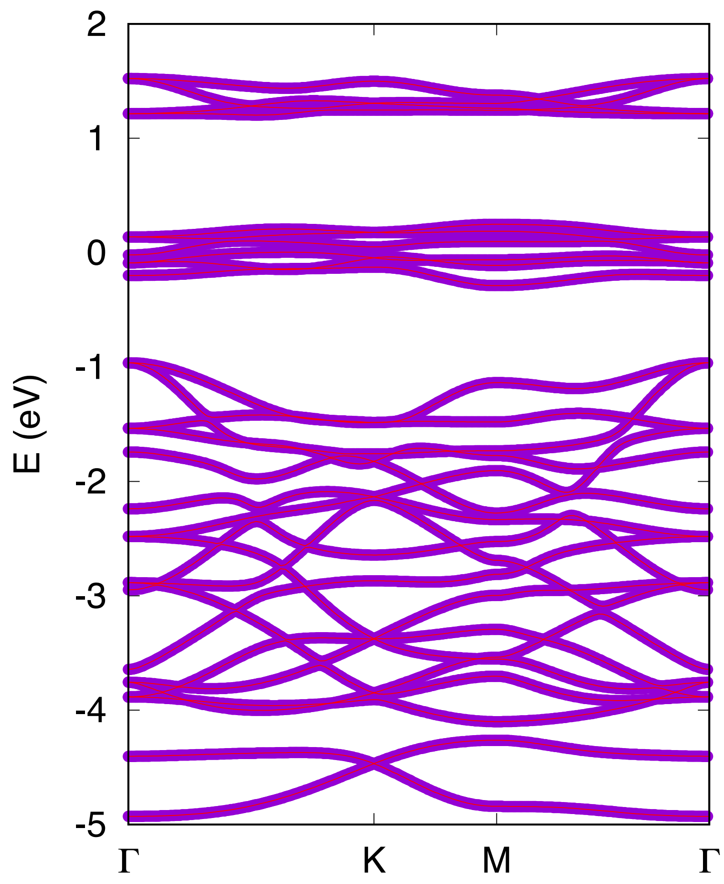


Wu et al., *Nature Communications*, **10**, 2371 (2019)

Coulomb interactions in CrI_3

➢ Wannier-Hartree-Fock Approach

Step 1. Calculate the spin-unpolarized Wannier Hamiltonian (H^W) from first-principles (LDA) projected on Cr *d*-orbitals and I *p*-orbital



Coulomb interactions in CrI_3

➢ Wannier-Hartree-Fock Approach

Step 1. Calculate the spin-unpolarized Wannier Hamiltonian (H^W) from first-principles (LDA) projected on Cr *d*-orbitals and I *p*-orbital

Step 2. Introduce Coulomb interactions using Hartree-Fock (H^{HF}) in the mean-field approximation, and calculate the electronic structure self-consistently

$$\begin{aligned} H^{HF} &= U \sum_{m} \hat{n}_{m\uparrow}\hat{n}_{m\downarrow} + U' \sum_{m \neq m'} \hat{n}_{m\uparrow}\hat{n}_{m'\downarrow} + \\ &+ (U' - J) \sum_{m<m',\sigma} \hat{n}_{m\sigma}\hat{n}_{m'\sigma} + \\ &- J \sum_{m \neq m'} d^{\dagger}_{m\uparrow} d_{m\downarrow} d^{\dagger}_{m'\downarrow} d_{m'\uparrow} \end{aligned}$$

Coulomb interactions in CrI_3

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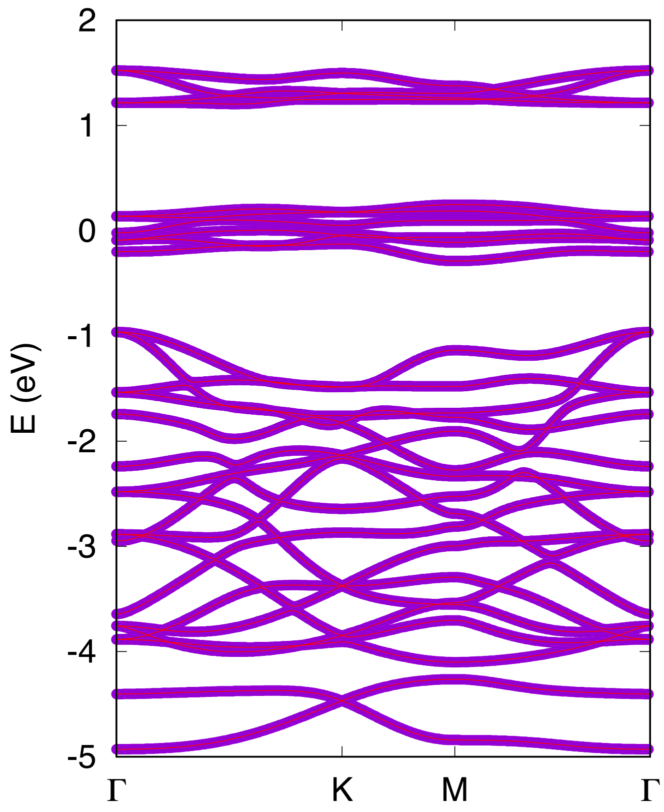

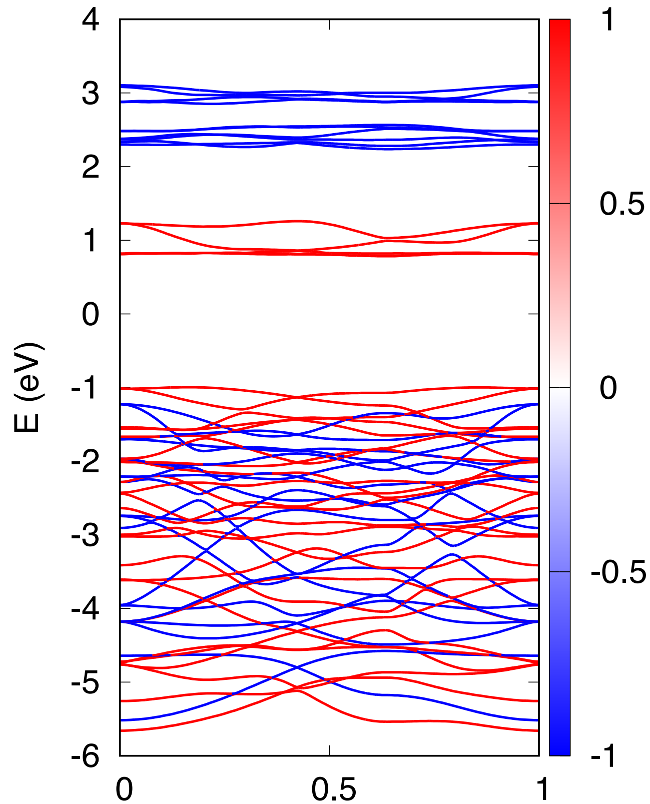

Coulomb interactions in CrI_3

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$$
\begin{aligned}
H^{HF} &= U\sum_{m}\hat{n}_{m\uparrow}\hat{n}_{m\downarrow} + U'\sum_{m\neq m'}\hat{n}_{m\uparrow}\hat{n}_{m'\downarrow} + \\
&+ (U'-J)\sum_{m<m',\sigma}\hat{n}_{m\sigma}\hat{n}_{m'\sigma} + \\
&- J\sum_{m\neq m'} d^{\dagger}_{m\uparrow}d_{m\downarrow}d^{\dagger}_{m'\downarrow}d_{m'\uparrow}
\end{aligned}
$$

Step 3. The orbital-dependent exchange interactions are calculated by means of the magnetic force theorem using the converged total Hamiltonian ($H = H^W + H^{HF}$)

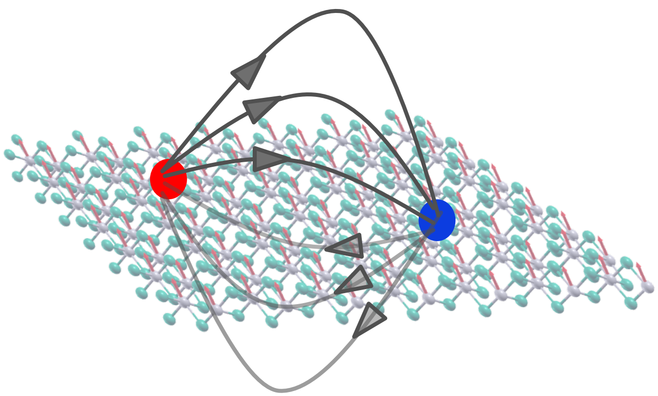
$$
J^{\alpha\beta}_{kl} = \frac{1}{4\pi}\int_{-\infty}^{E_F} d\epsilon \,\mathrm{Im}\left[\Delta_{\alpha\beta}G^{\beta\alpha\downarrow}_{kl}(\epsilon)\Delta_{\beta\alpha}G^{\alpha\beta\uparrow}_{kl}(\epsilon)\right]
$$

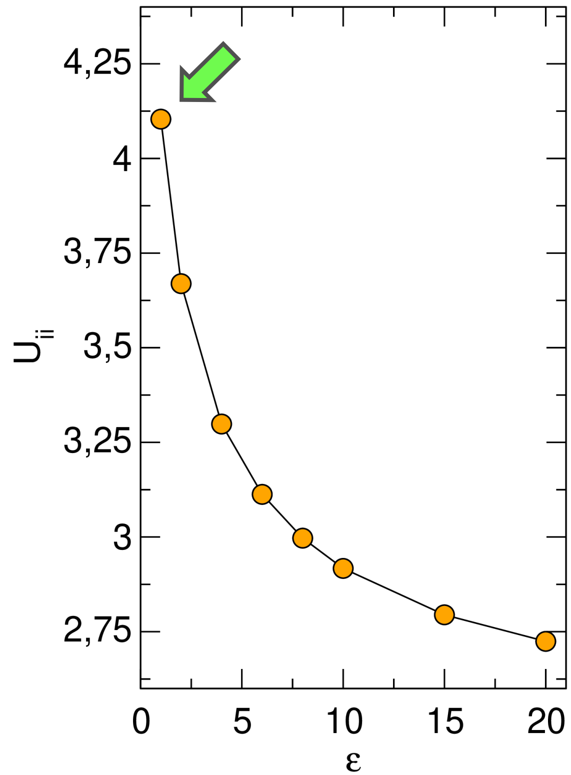
A. I. Liechtenstein *et al.*, *J. Magn. Magn. Mater.*, **67**, 65 (1987)

Coulomb interactions in CrI_3

➢ **Wannier-Hartree-Fock Approach:** **Substrate dielectric screening effects**

Van der Waals heterostructure: **hBN/CrI_3/hBN**





Rösner *et al.*, *Phys. Rev. B*, **92**, 085102 (2015)
Rösner *et al.*, *Nano Letters*, **16**, 2322 – 2327 (2016)
Florian et al., Nano Letters, **18**, 2725 – 2732 (2018)

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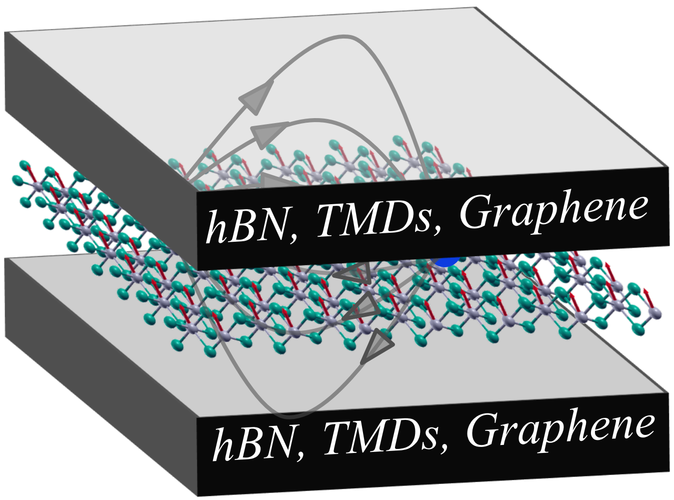
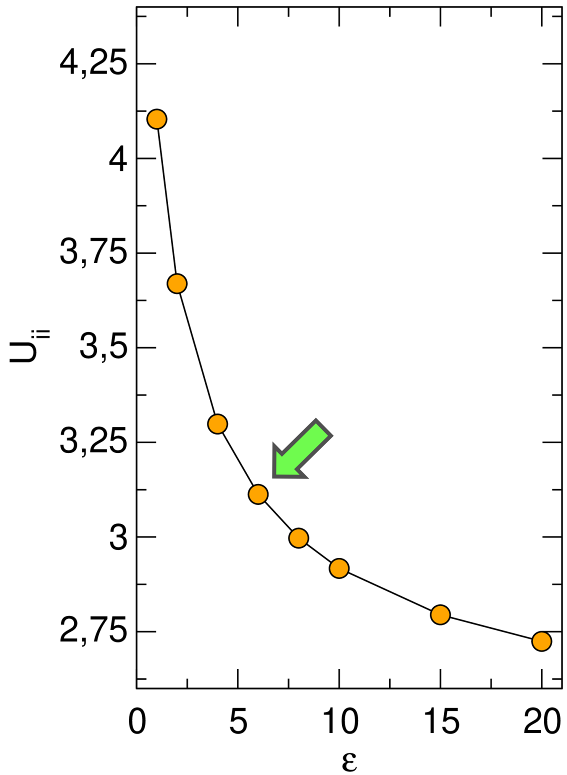


Dielectric environment

$$U_{ij}^{cRPA} = \varepsilon^{-1} v_{ij}$$



❑ **Impact on the quasi-particle/excitonic gap**

Rösner *et al.*, *Phys. Rev. B*, **92**, 085102 (2015)
Rösner *et al.*, *Nano Letters*, **16**, 2322 – 2327 (2016)
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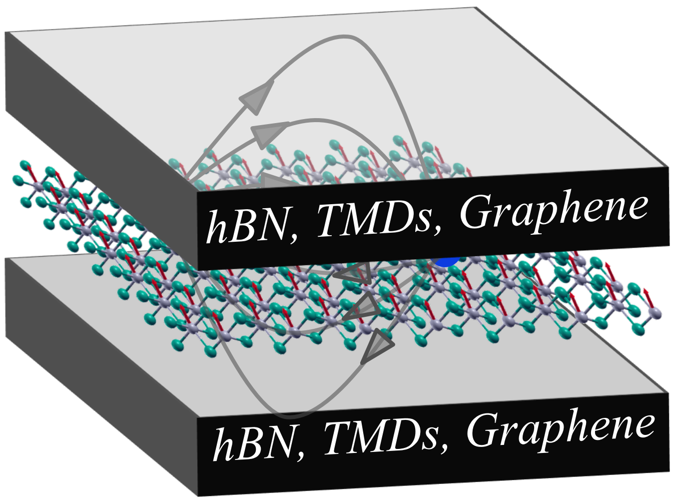

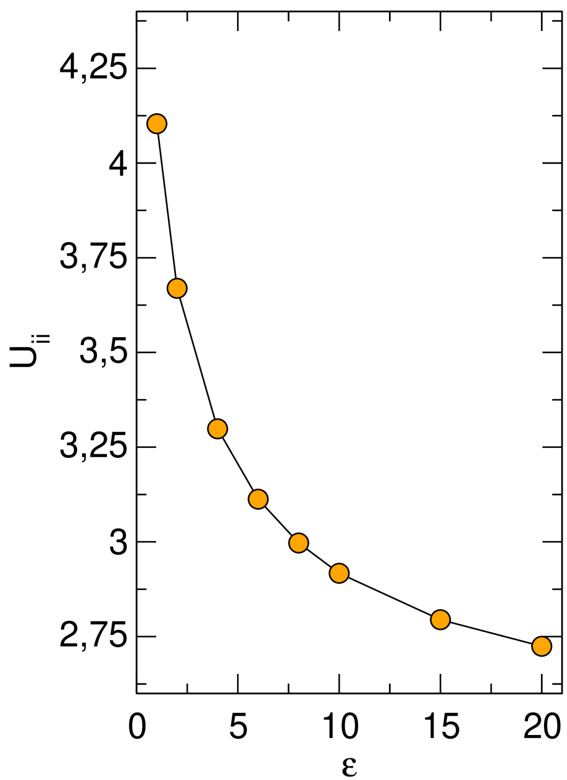

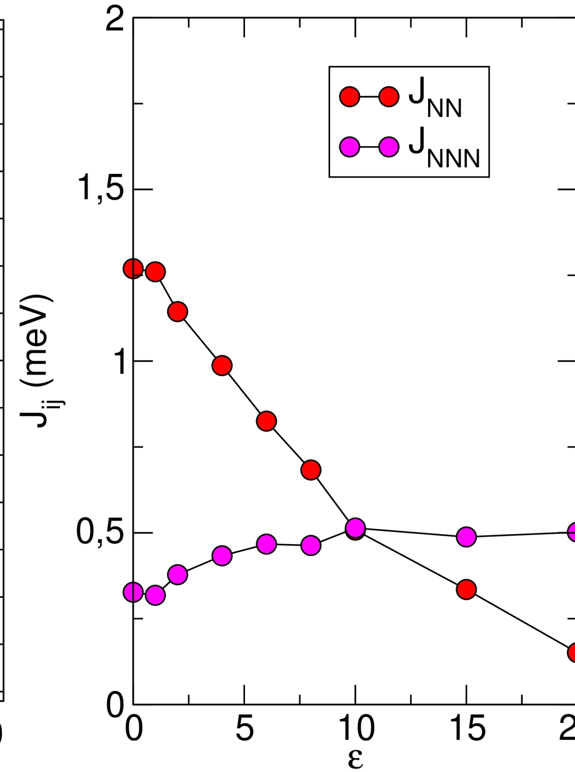

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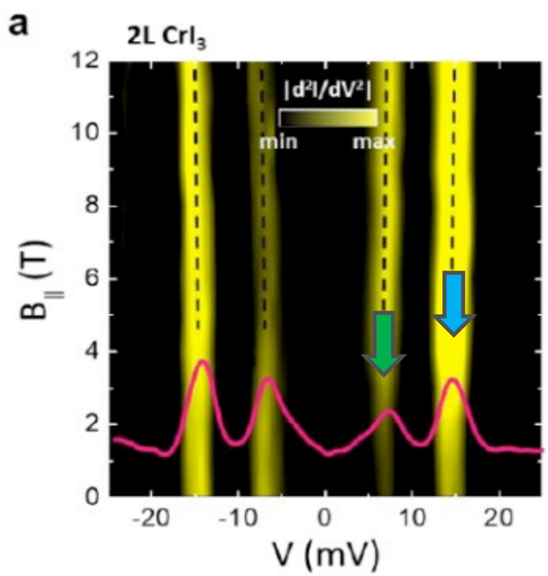
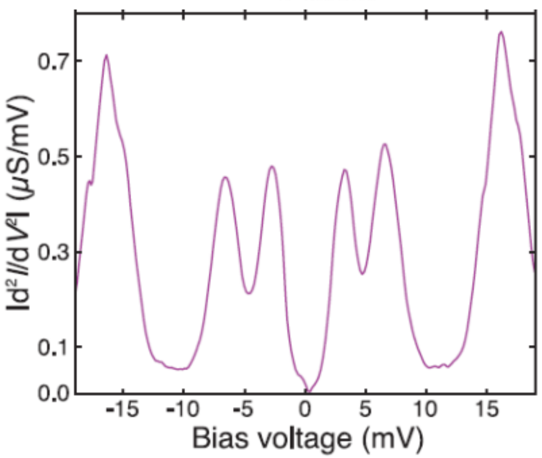
Florian et al., Nano Letters, **18**, 2725 – 2732 (2018)

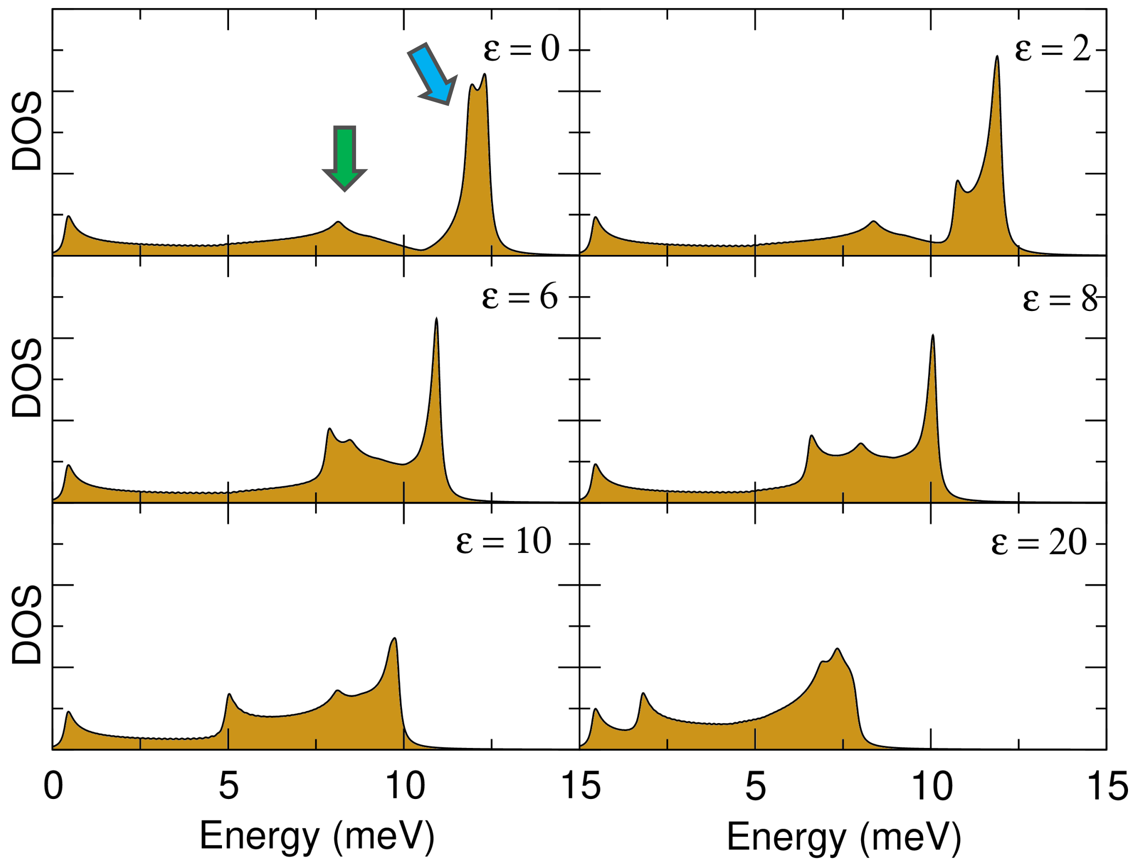
Coulomb interactions in CrI_3

➢ **Wannier-Hartree-Fock Approach: Spin-wave spectrum**

Impact of dielectric environment on the spin-wave dispersion







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Radboud University

- **Wannier-Hartree-Fock Approach: Conclussions**

Coulomb and substrate dielectric screening effects impact the magnon spectrum of Chromium Trihalides

We have developed a new computational tool to study Coulomb interactions in CrX_3 combining Wannier-HF and the magnetic force theorem (MFT)

Magnetic Polarons in Bilayer CrI_3

Magnetic polarons in bilayer CrI_3

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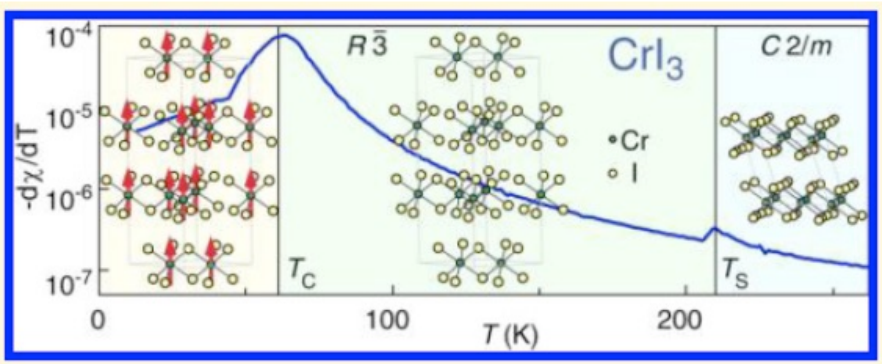
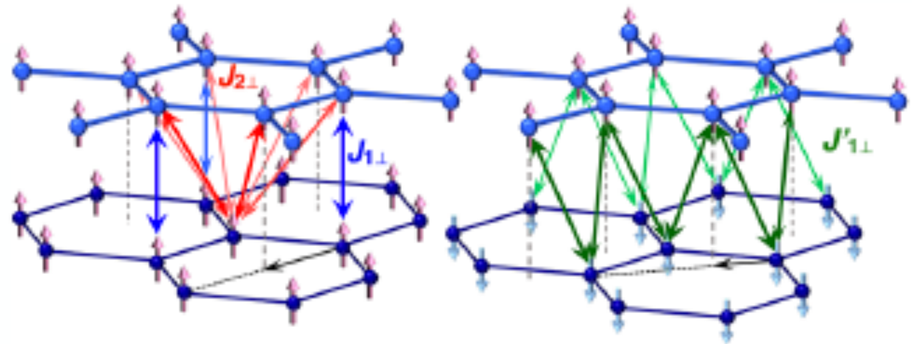
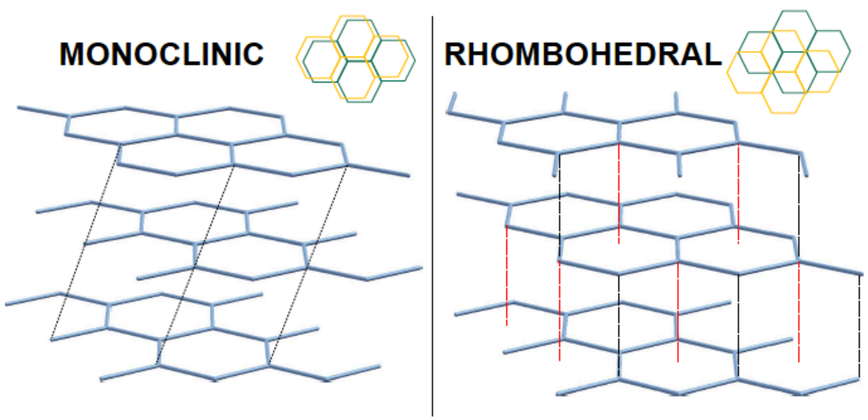
Radboud University

➢ **Magnetism at the microscopic level: Interlayer Exchange**

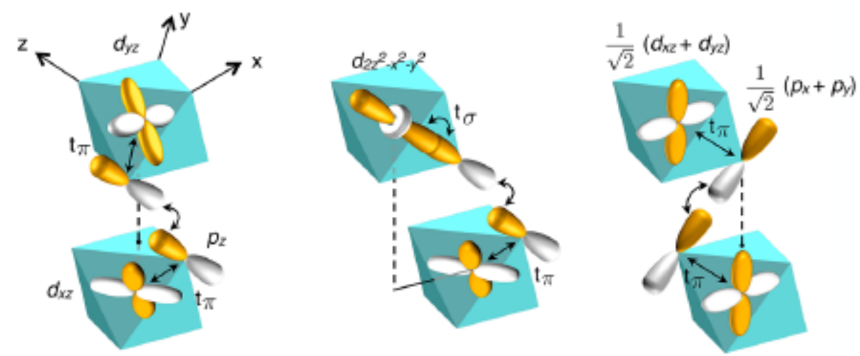
T_S = 210 K (CrI_3); 420 K ($CrBr_3$); 240 K ($CrCl_3$)







$J_{1\perp}$ $J_{2\perp}$ $J'_{1\perp}$

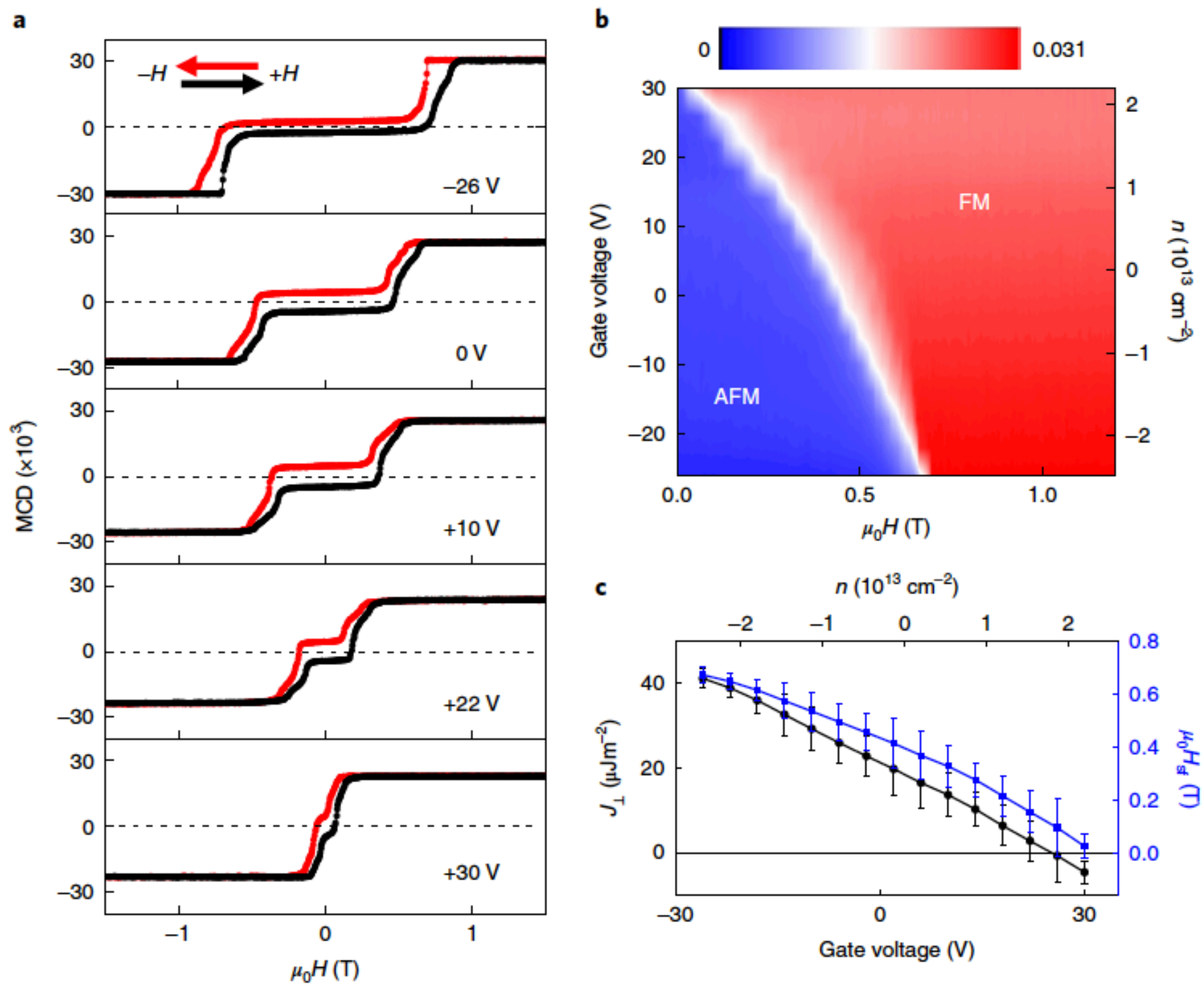


$J_\perp \approx 0.04$ meV

McGuire et al., *Chem. Mater.*, **27**, 612 – 620 (2015)
Sivadas et al., *Nano Letters*, **18**, 7658 – 7664 (2018)
DS, Cardoso and Fernández-Rossier, Sol. Stat. Comm., **299**, 113662 (2019)
DS, Katsnelson and Fernández-Rossier, *Nano Letters* (Mini-Review - Accepted)

Magnetic polarons in bilayer CrI_3

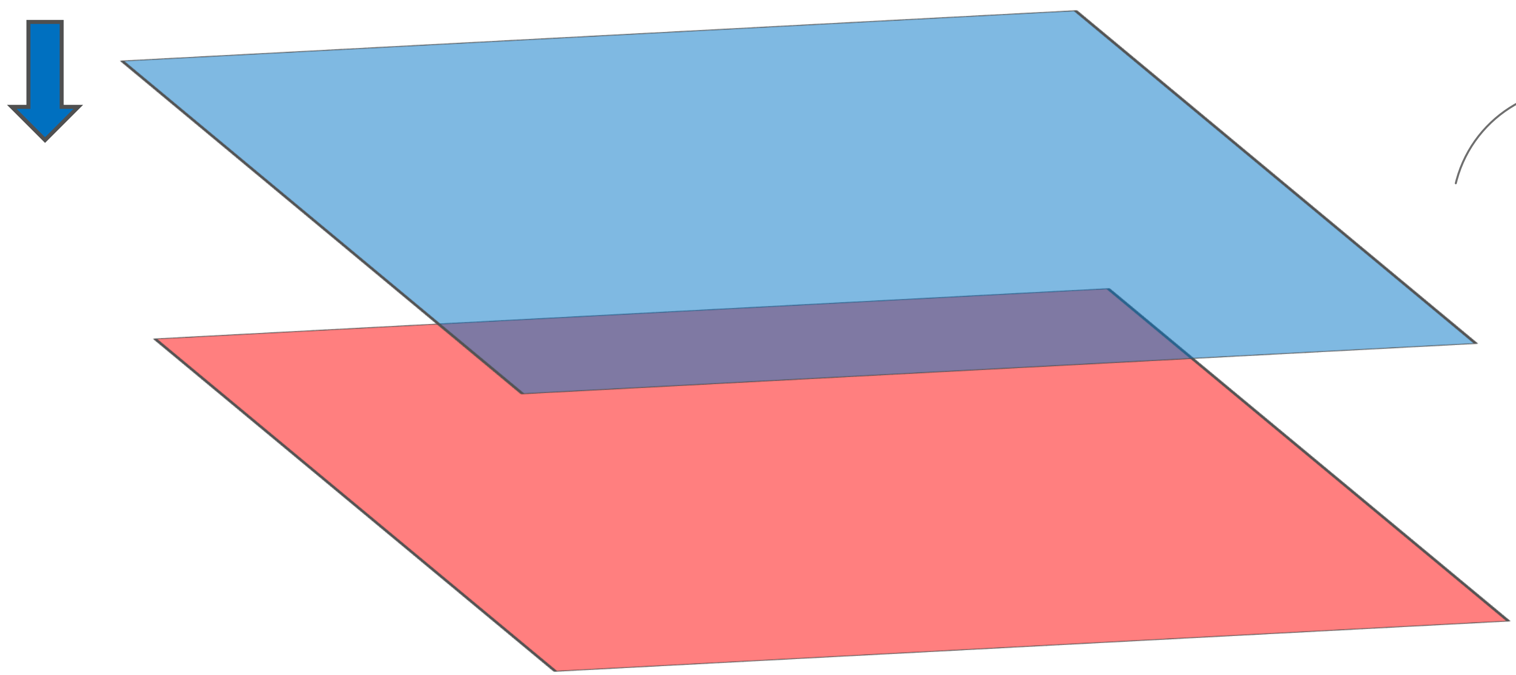
- **Electic field control of interlayer magnetism**

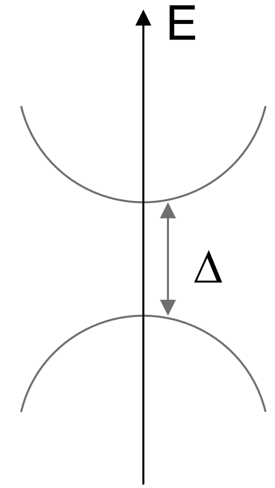


Jiang et al., *Nature Nanotechnology*, **13**, 549 – 553 (2018)
Jiang et al., *Nature Materials*, **17**, 406 – 410 (2018)
Huang et al., Nature Nanotechnology, **13**, 544 - 548 (2019)

Magnetic polarons in bilayer CrI_3

➢ **Magnetic polarons in layered semiconducting magnets**





Auslander and Katsnelson, *J. Magn. Magn. Mater.*, **24**, 117 (1981)
Vischer, *Phys. Rev. B*, **10**, 943 (1974)
Mott, *Metal-Insulator Transitions*, Taylor & Francis, London (1974)

Magnetic polarons in bilayer CrI_3

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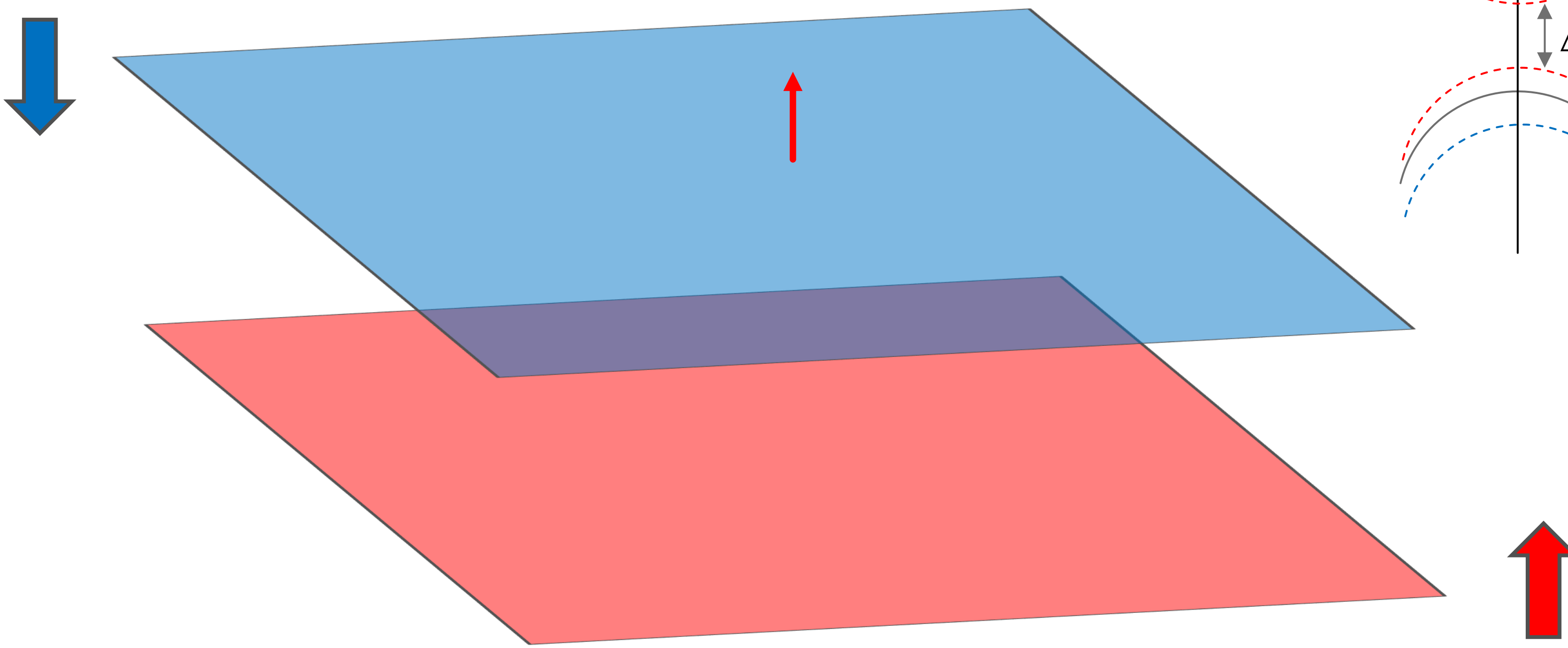

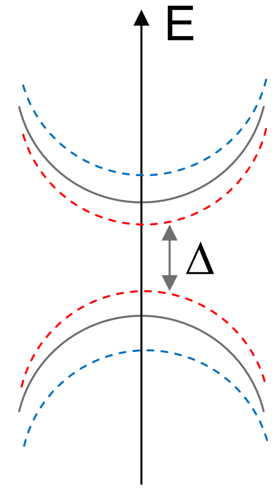

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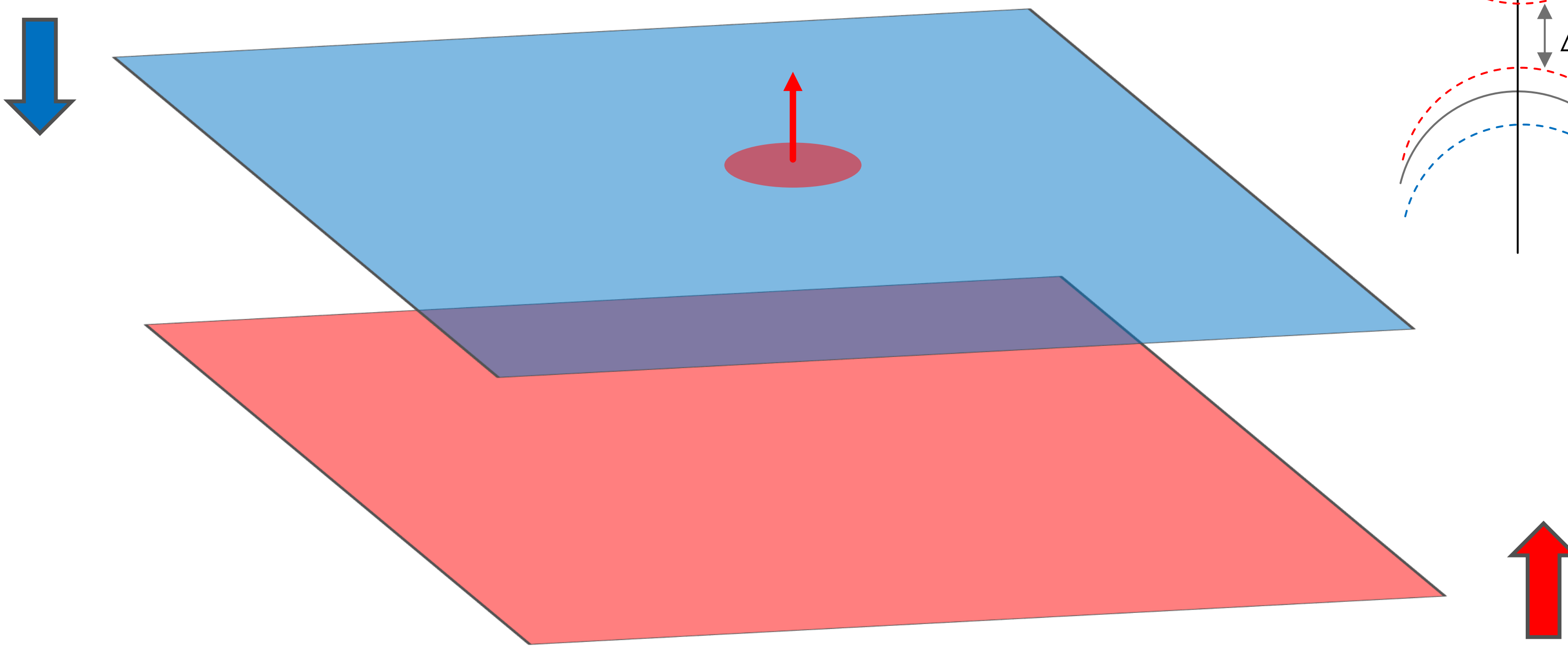

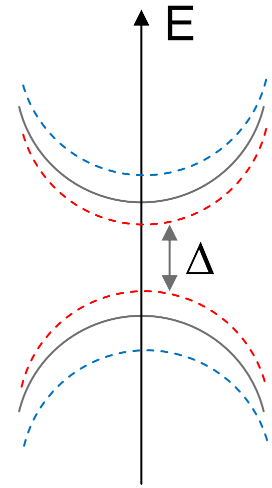

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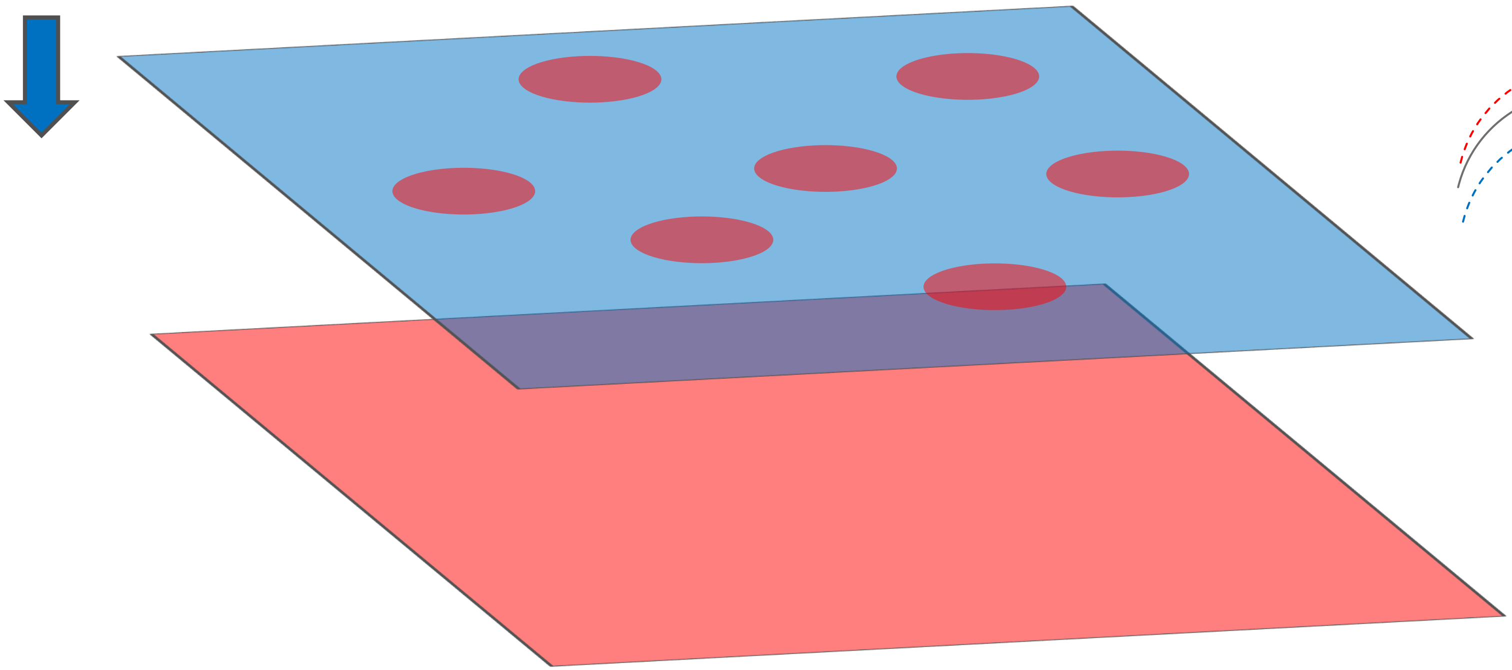

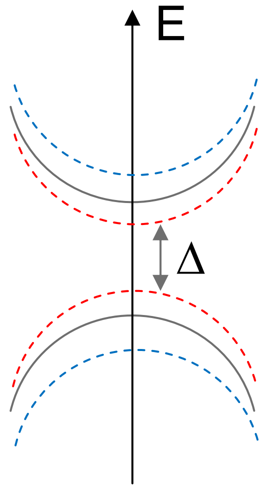

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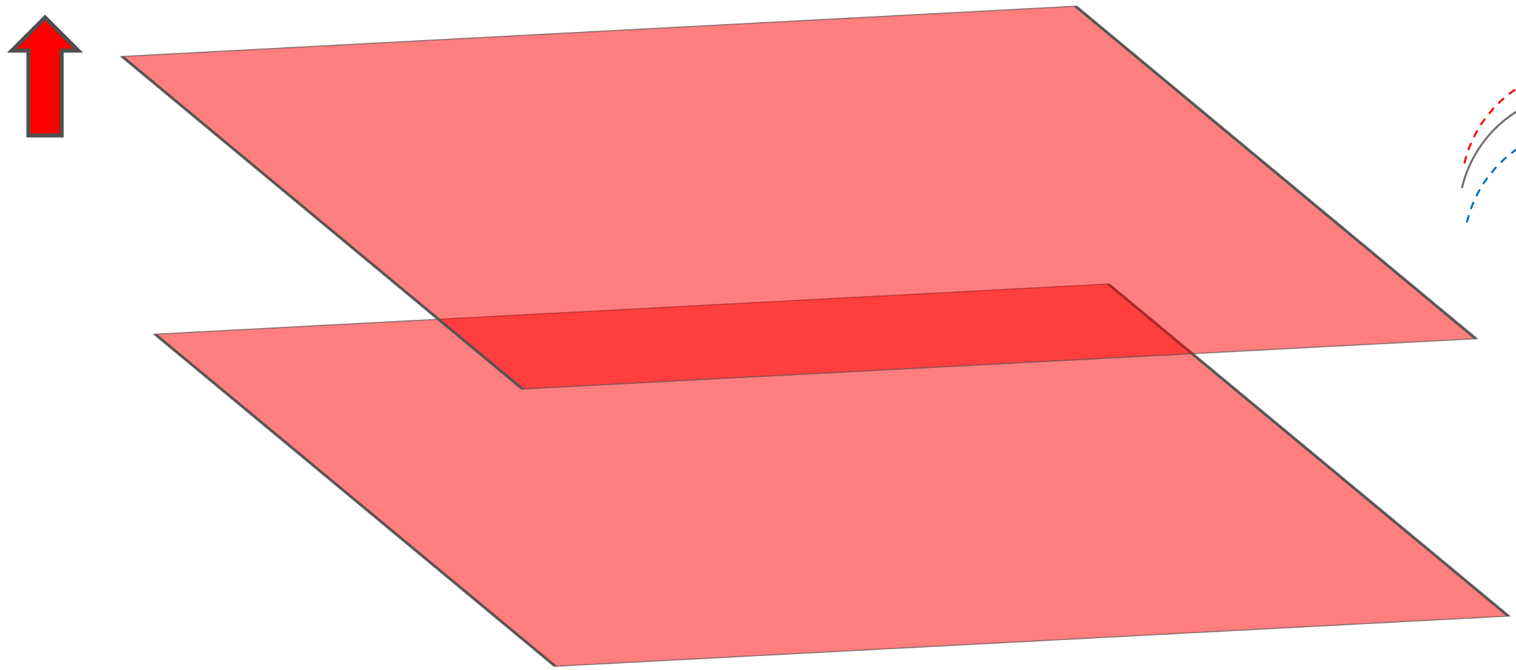

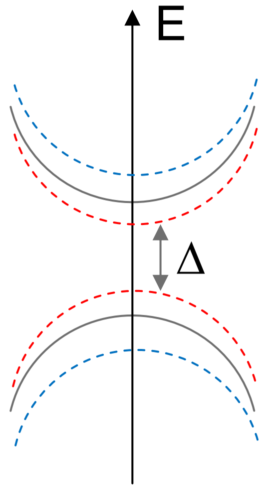

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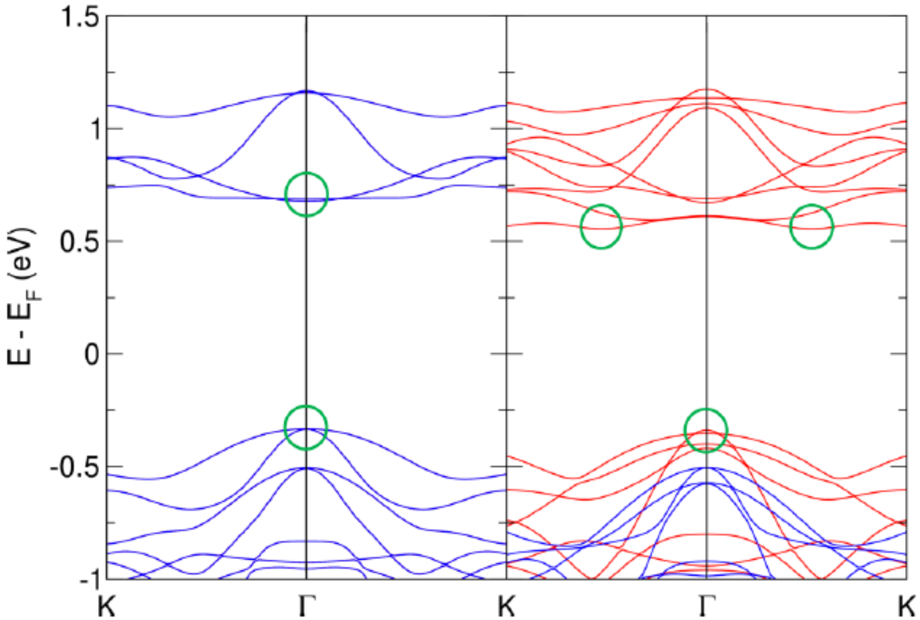
Mott, *Metal-Insulator Transitions*, Taylor & Francis, London (1974)

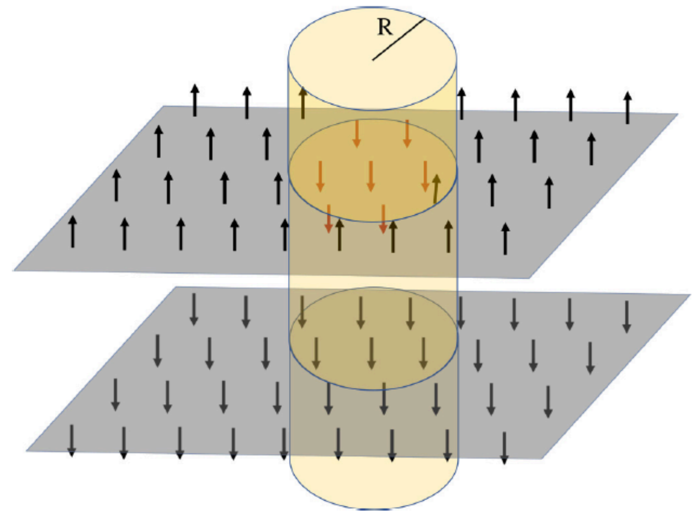
- **Magnetic polarons in bilayer CrI_3**



	VBMax (eV)	CBMin (eV)	m_h^*/m_e	m_e^*/m_e
AFM	−0.333	0.678	0.15	0.18
FM	−0.339	0.555	0.02	0.11

Formation energy of a magnetic polaron

$$\mathcal{E}(R) = -\Delta + \frac{\hbar^2 z_0^2}{2m^* R^2} + J\frac{\pi R^2}{S_0}$$

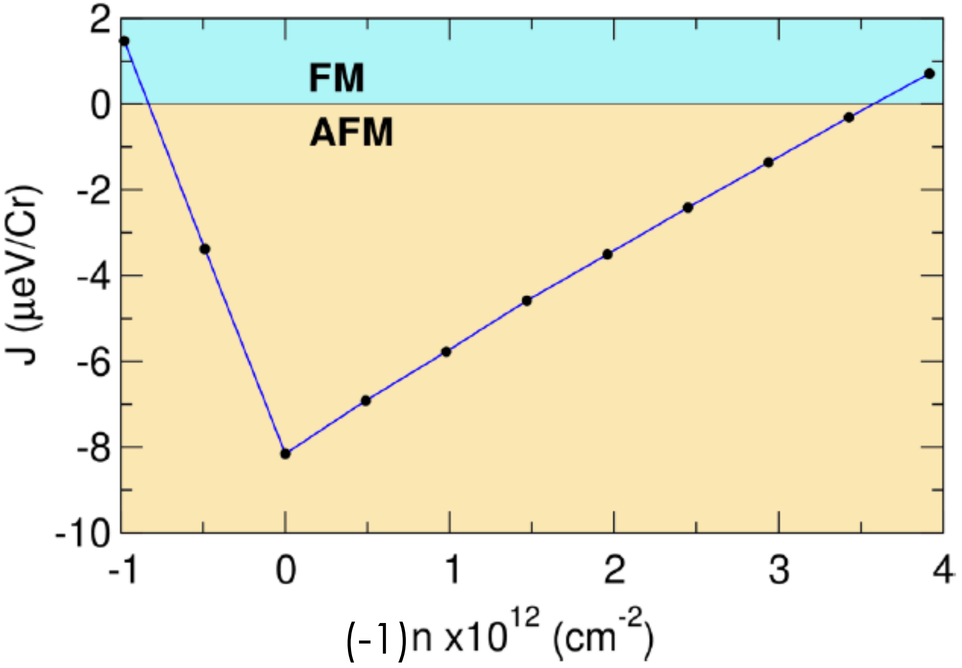


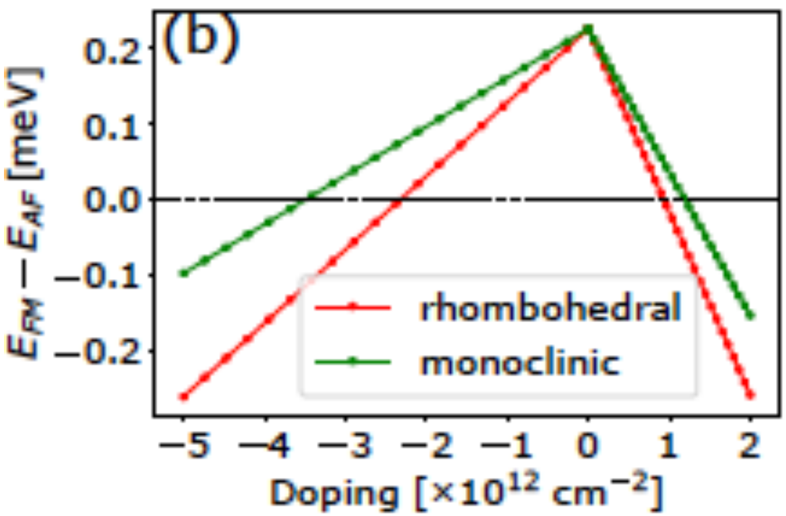
$$\mathcal{E}_e = -91.3 \text{ meV and } \mathcal{E}_h = 67.9 \text{ meV}$$

DS and Katsnelson, *Phys. Rev. B*, **101**, 041402(R) (2020)

➢ Magnetic polarons in bilayer CrI_3

DFT calculations





DS and Katsnelson, *Phys. Rev. B*, **101**, 041402(R) (2020)
Lei et al., **arXiv:1902.06418**

Magnetic polarons in bilayer CrI_3

➢ **Magnetic polarons in bilayer CrI_3: Conclussions**

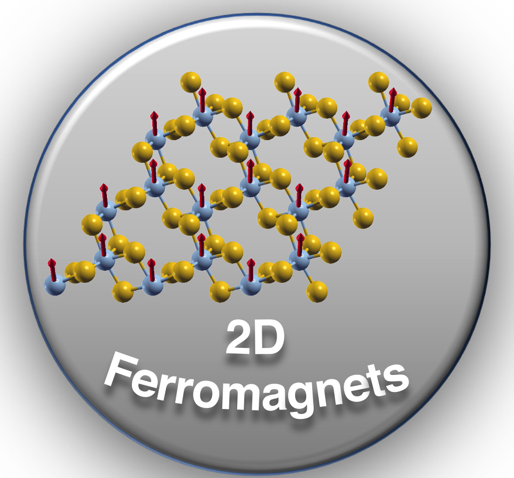
Electric field modulation of interlayer magnetism is driven by magnetic polarons in bilayer CrI_3

The formation of magnetic polarons is only allowed for electron doping which facilitates the antiferro-to-ferro phase transition

Still several open issues

- Origin of magnetic anisotropy (single-ion, Kitaev, 2nd neighbor DMI, etc.)
- Role of van der Waals interactions in interlayer exchange (different results with different functionals)
- Effect of substrates on the magnetic and electronic properties of CrX_3
- Moiré magnets (Hejazi et al., *PNAS*, **117**, 10721 – 10726 (2020))
- Magneto-optical properties

Take home message



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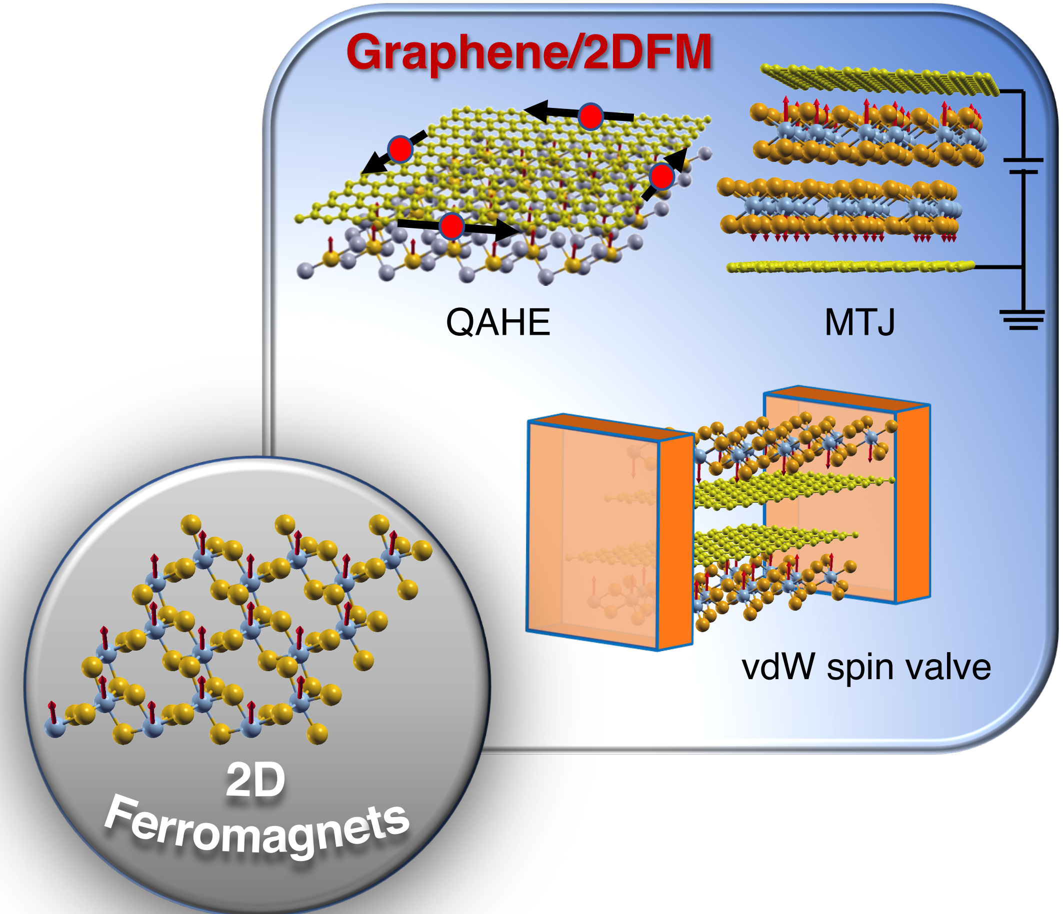
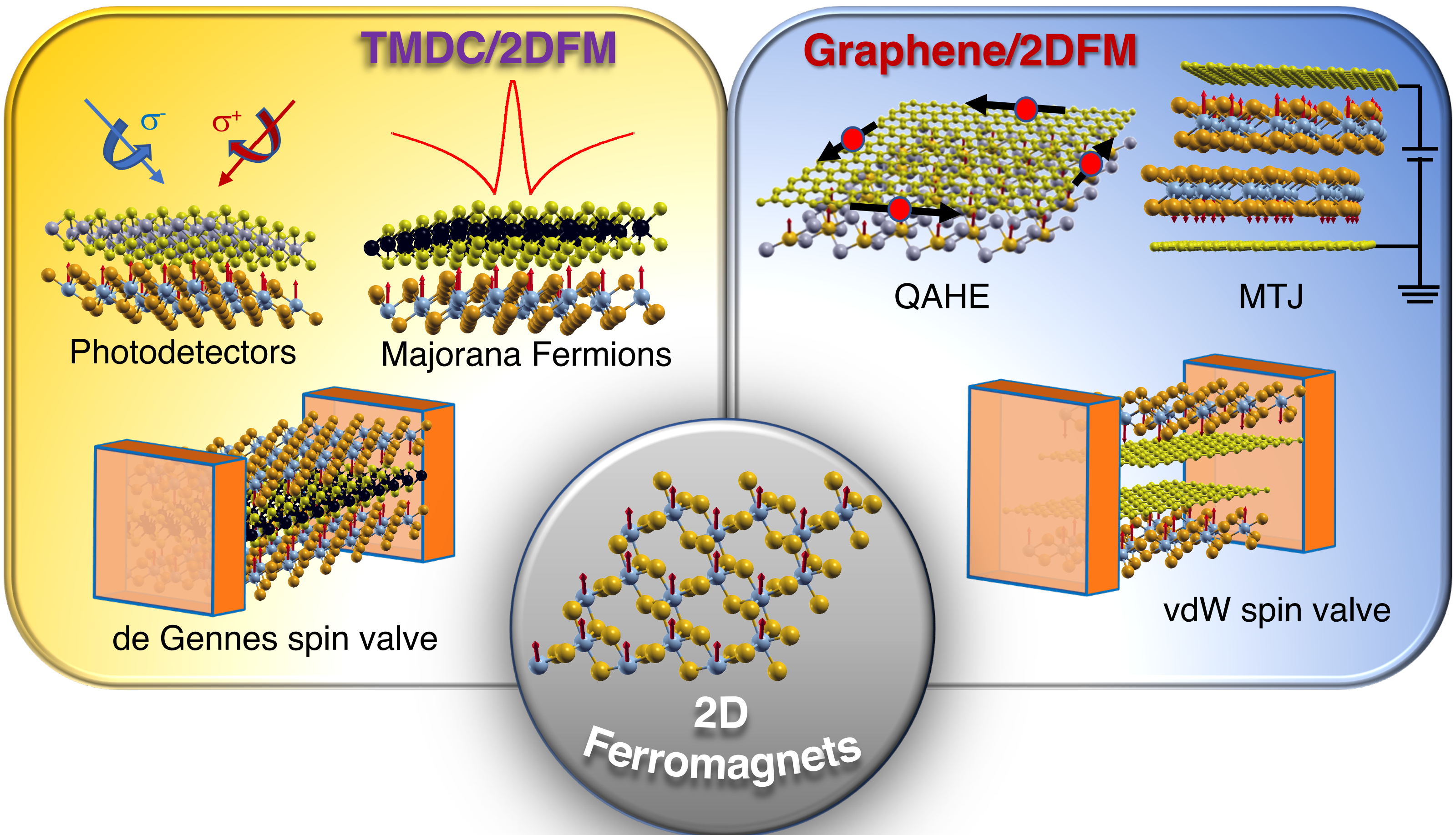


Take home message



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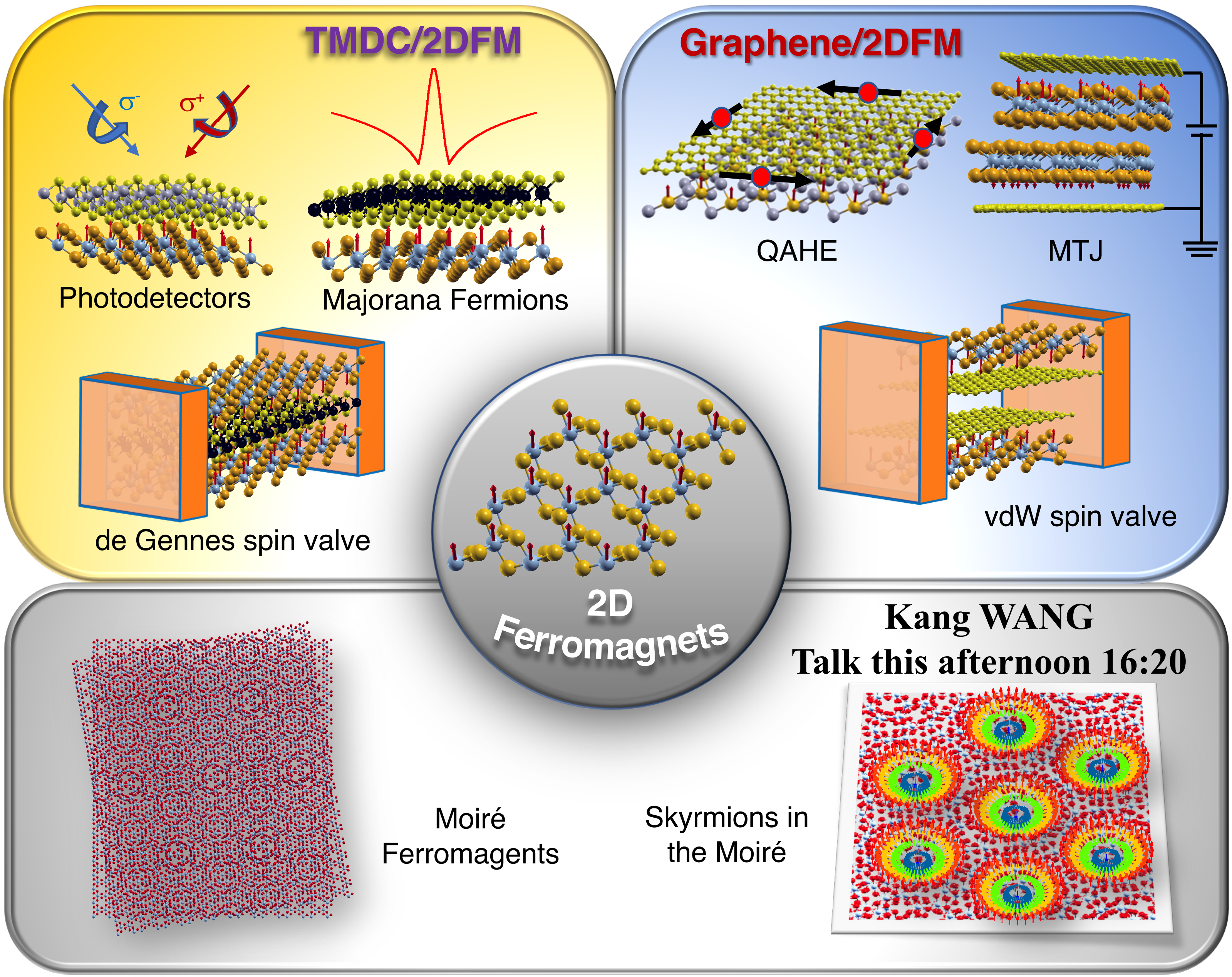
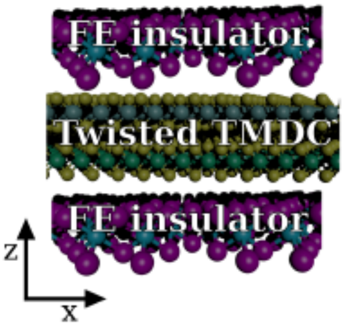
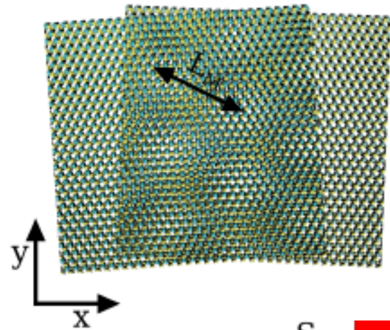


Take home message

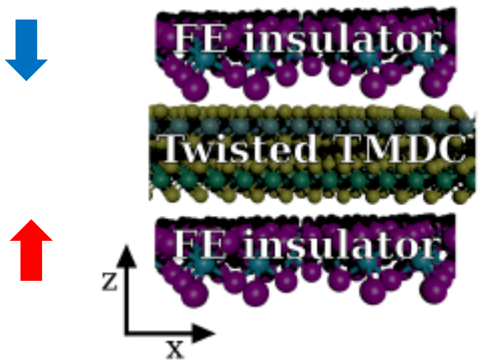
- ➢ **Exchange proximity effects in twisted TMDs**

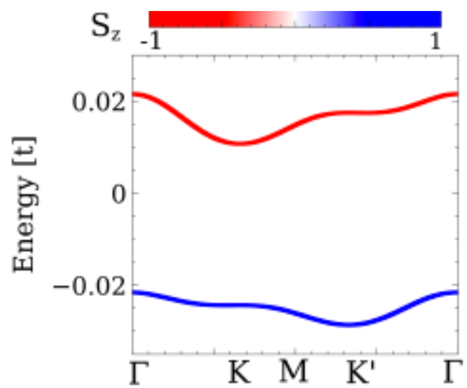


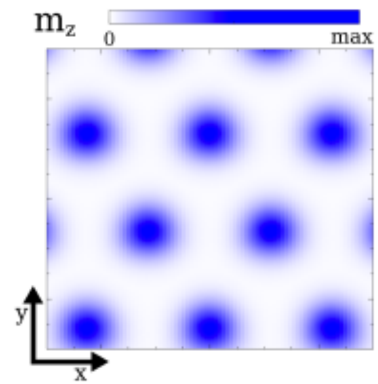


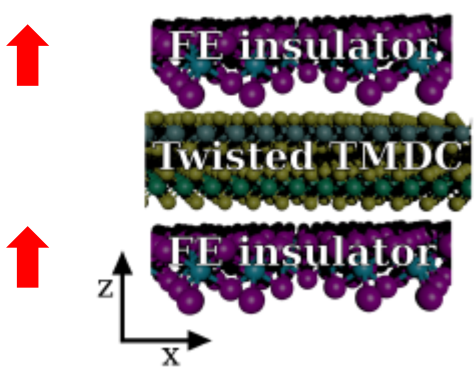
DS and Lado, J. *Phys. D: Appl. Phys.*
https://doi.org/10.1088/1361-6463/abaa15

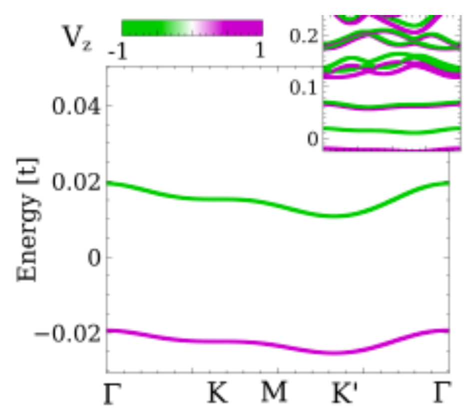
For the coffe break!

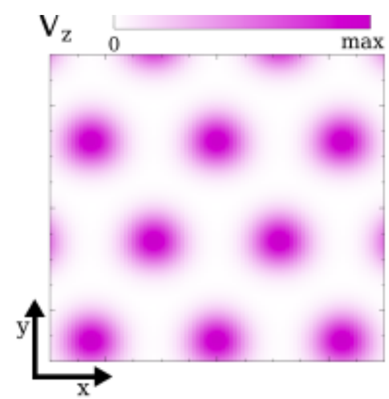












Acknowledgments

Institute for Molecules and Materials
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Dr. Malte Rösner

Dr. Alexander Rudenko

INL
INTERNATIONAL IBERIAN NANOTECHNOLOGY LABORATORY

Prof. Joaquín Fernández Rossier

A"
Aalto University

Dr. José Lado

Acknowledgments

Thank You!

MARIE CURIE ACTIONS

SURF SARA

Acknowledgments