SPICE-SPIN+X Seminars

June 12th 2024 | *Online, SPICE YouTube channel*

Playing with magnetism in 2D van der Waals materials via first principles

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2Dsmarties

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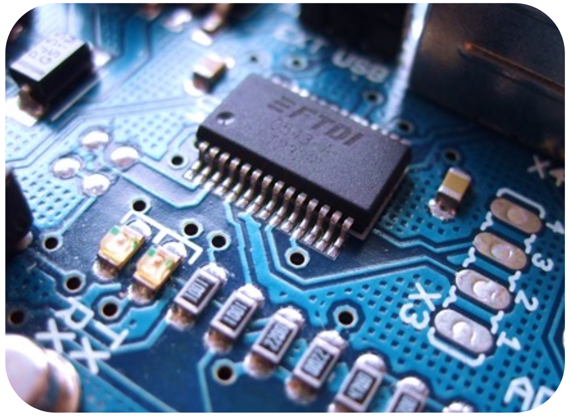
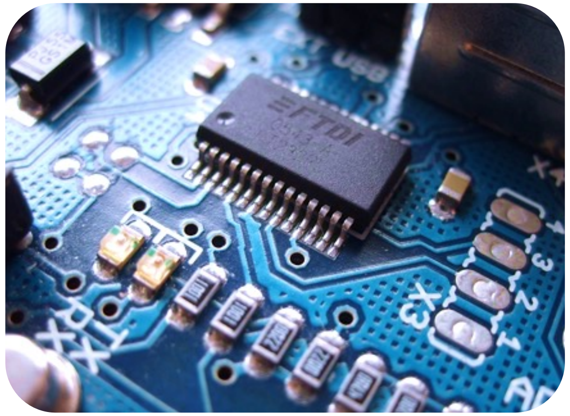
Gen—T

From electronics to magnonics



Electric current

From electronics to magnonics



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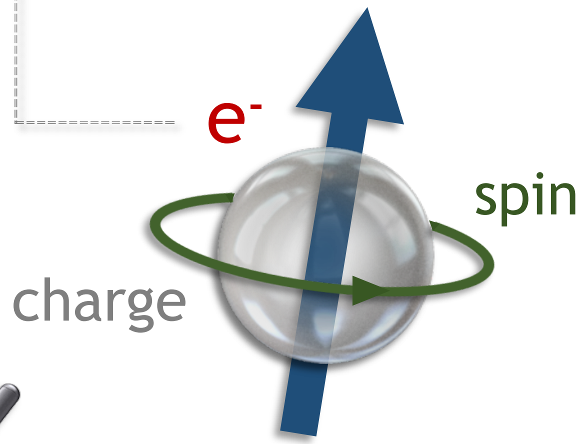
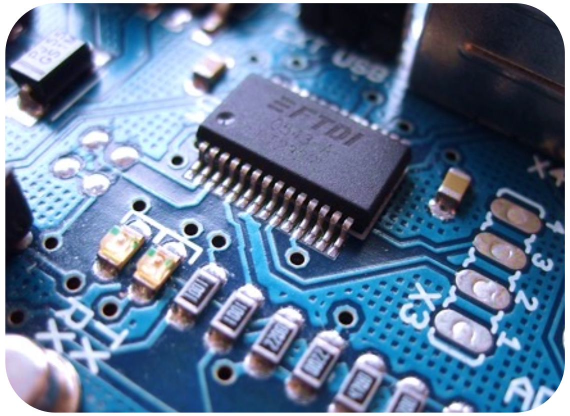


From electronics to magnonics



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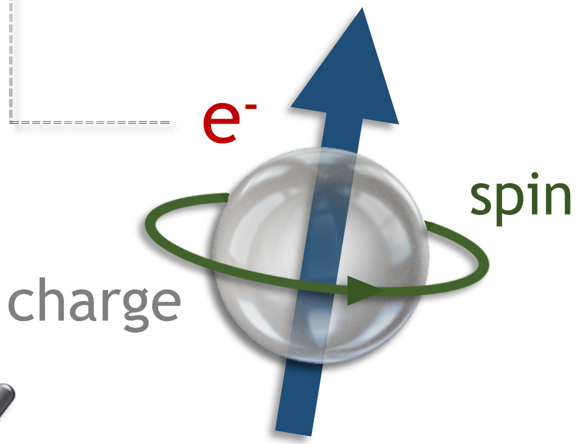

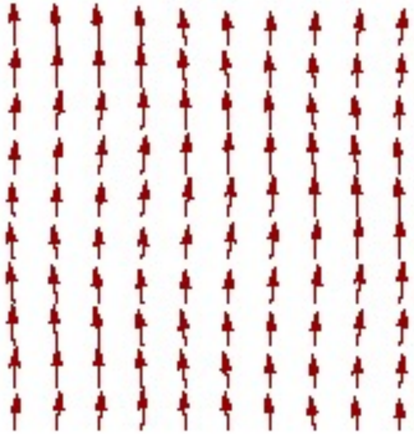

Spin waves

LOW-COST consumption

NANO-sized devices

WAVE-based computing

Challenges and opportunities

IOP Publishing
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Topical Review

The 2021 Magnonics Roadmap

Anjan Barman[1,51,*], Gianluca Gubbiotti[2,51,*], S Ladak[3], A O Adeyeye[4], M Krawczyk[5], J Gräfe[6], C Adelmann[7], S Cotofana[8], A Naeemi[9], V I Vasyuchka[10], B Hillebrands[10], S A Nikitov[11], H Yu[12], D Grundler[13], A V Sadovnikov[11,14], A A Grachev[11,14], S E Sheshukova[11,14], J-Y Duquesne[15], M Marangolo[15], G Csaba[16], W Porod[17], V E Demidov[18], S Urazhdin[19], S O Demokritov[18], E Albisetti[20], D Petti[20], R Bertacco[20], H Schultheiss[21,22], V V Kruglyak[23], V D Poimanov[24], S Sahoo[1], J Sinha[25], H Yang[26], M Münzenberg[27], T Moriyama[28,29], S Mizukami[29,30], P Landeros[31,32], R A Gallardo[31,32], G Carlotti[33,34], J-V Kim[35], R L Stamps[36], R E Camley[37], B Rana[38], Y Otani[38,39], W Yu[40], T Yu[41], G E W Bauer[30,42], C Back[43], G S Uhrig[44], O V Dobrovolskiy[45], B Budinska[45], H Qin[46], S van Dijken[46], A V Chumak[45], A Khitun[47], D E Nikonov[48], I A Young[48], B W Zingsem[49] and M Winklhofer[50]

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CrossMark

Abstract
Magnonics is a budding research field in nanomagnetism and nanoscience that addresses the use of spin waves (magnons) to transmit, store, and process information. The rapid advancements of this field during last one decade in terms of upsurge in research papers, review articles, citations, proposals of devices as well as introduction of new sub-topics prompted us to present the first roadmap on magnonics. This is a collection of 22 sections written by leading experts in this field who review and discuss the current status besides presenting their vision of future perspectives. Today, the principal challenges in applied magnonics are the excitation of sub-100 nm wavelength magnons, their manipulation on the nanoscale and the creation of sub-micrometre devices using low-Gilbert damping magnetic materials and its interconnections to standard electronics. To this end, magnonics offers lower energy consumption, easier integrability and compatibility with CMOS structure, reprogrammability, shorter wavelength, smaller device features, anisotropic properties, negative group velocity, non-reciprocity and efficient tunability by various external stimuli to name a few. Hence, despite being a young research field, magnonics has come a long way since its early inception. This roadmap asserts a milestone for future emerging research directions in magnonics, and hopefully, it will inspire a series of exciting new articles on the same topic in the coming years.

Keywords: magnonics, spin-waves, magnons, photons, magnetism

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Challenges and opportunities

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The 2024 Magnonics Roadmap

Benedetta Flebus[1], Dirk Grundler[2], Bivas Rana[3], Yoshichika Otani[4], Igor Barsukov[5], Anjan Barman[6], Gianluca Gubbiotti[7], Pedro Landeros[8], Johan Akerman[9], Ursula S Ebels[10], Philipp Pirro[11], V E Demidov[12], Katrin Schultheiss[13], Gyorgy Csaba[14], Qi Wang[15], Dmitri E. Nikonov[16], Florin Ciubotaru[17], Ping Che[18], Riccardo hertel[19], Teruo Ono[20], Dmytro Afanasiev[21], Johan H Mentink[22], Theo Rasing[23], Burkard Hillebrands[24], Silvia Viola Kusminskiy[25], Wei Zhang[26], Chunhui Rita Du[27], Aurore Finco[28], Toeno van der Sar[29], Yunqiu Kelly Luo[30], Yoichi Shiota[31], Joseph Sklenar[32], Tao Yu[33] and Jinwei Rao[34]

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DOI 10.1088/1361-648X/ad399c

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Abstract

Magnonics is a research field that has gained an increasing interest in both the fundamental and applied sciences in recent years. This field aims to explore and functionalize collective spin excitations in magnetically ordered materials for modern information technologies, sensing applications, and advanced computational schemes. Spin waves, also known as magnons, carry spin angular momenta that allow for the transmission, storage, and processing of information without moving charges. In integrated circuits, magnons enable on-chip data processing at ultrahigh frequencies without the Joule heating, which currently limits clock frequencies in conventional data processors to a few GHz. Recent developments in the field indicate that functional magnonic building blocks for in-memory computation, neural networks, and Ising machines are within reach. At the same time, the miniaturization of magnonic ... as the synergy of materials science, electrical engineering, and

Challenges and opportunities

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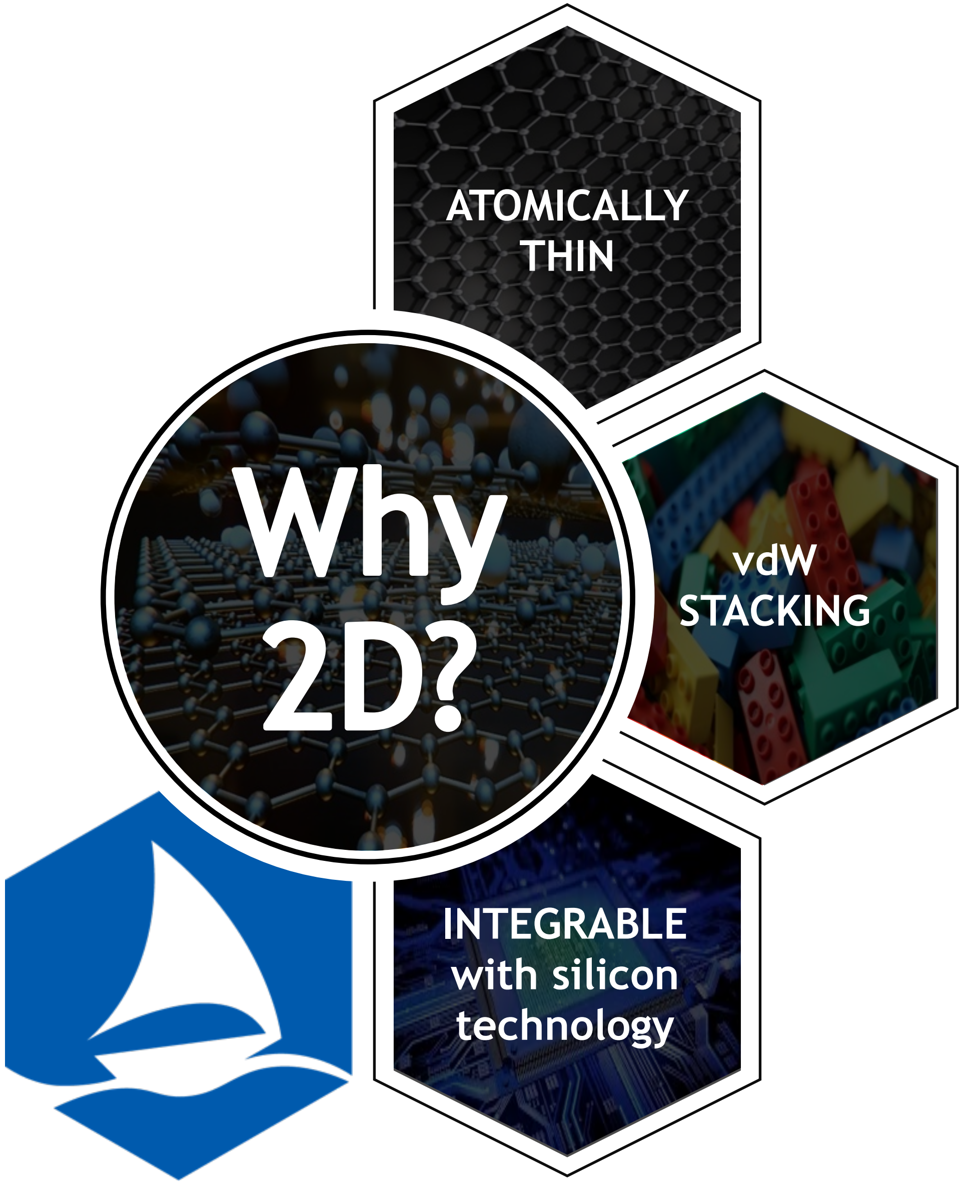
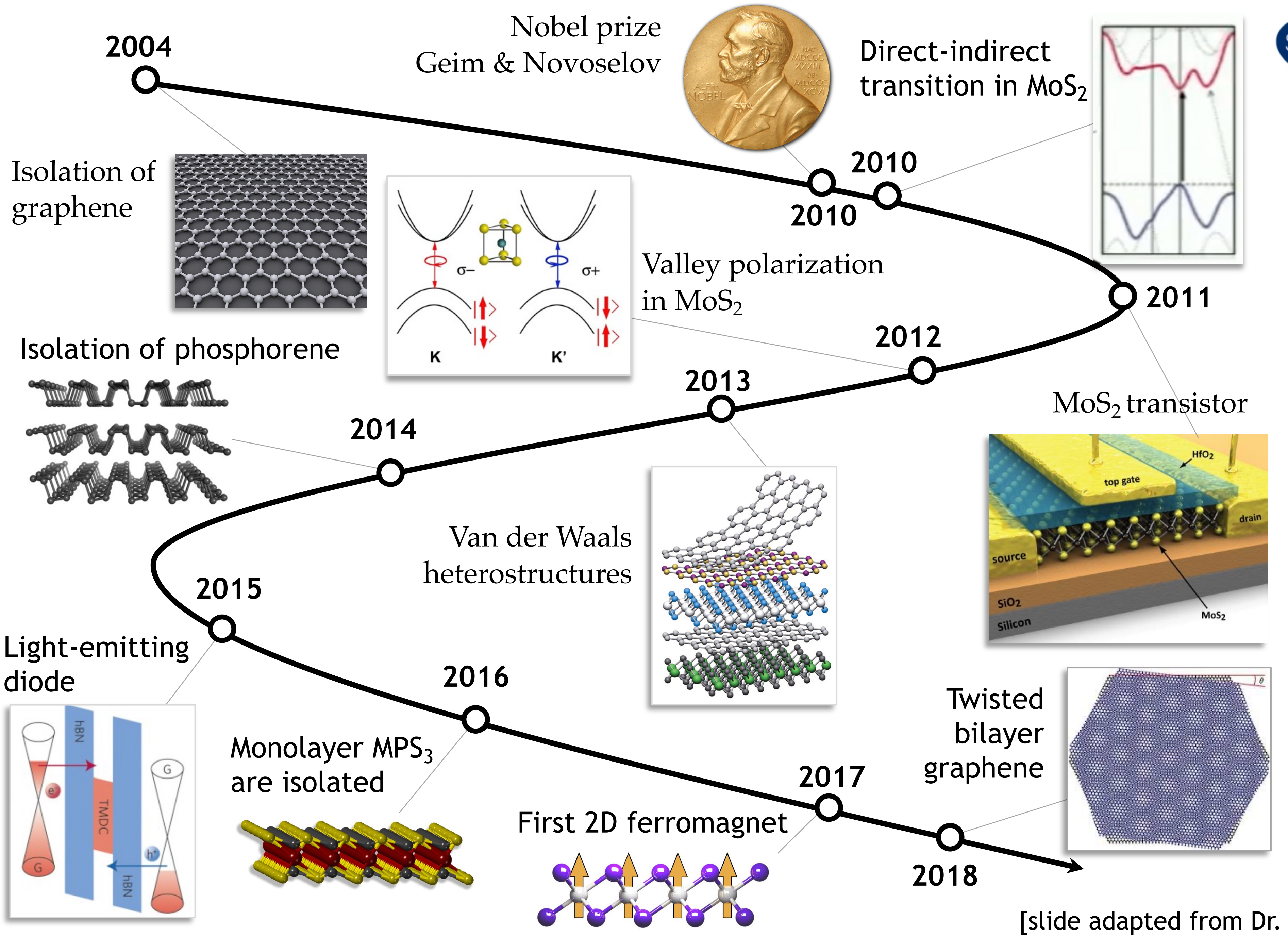
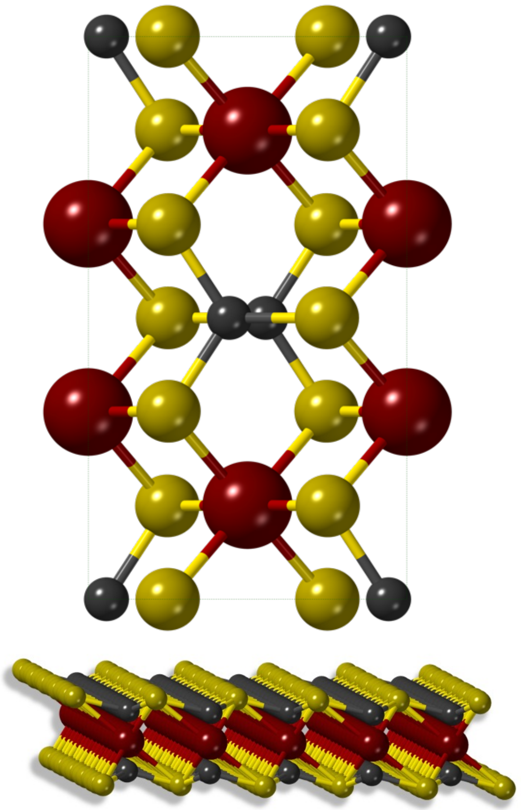




[slide adapted from Dr. Thygessen, DTU]

2D van der Waals magnetic materials

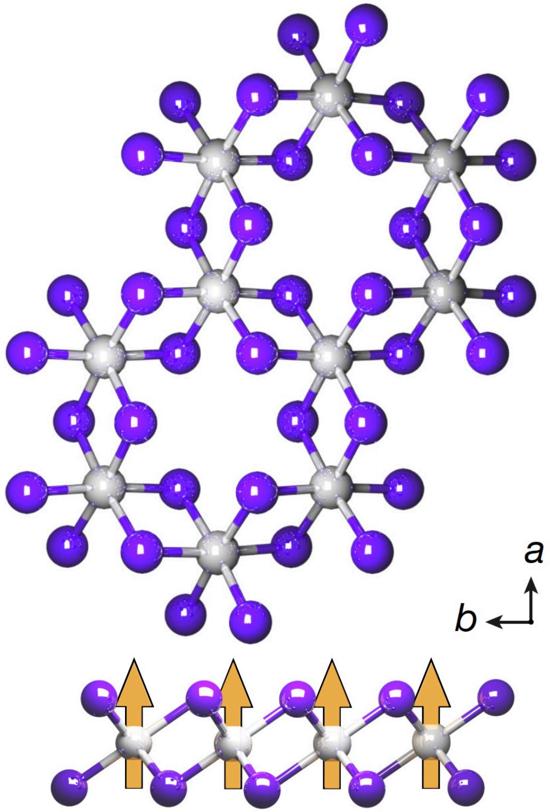
MPS_3



T_N = 70 - 150 K

Lee *et al.*, *Nano Lett.* 2016, **16(12)**, 7433–7438

CrI_3



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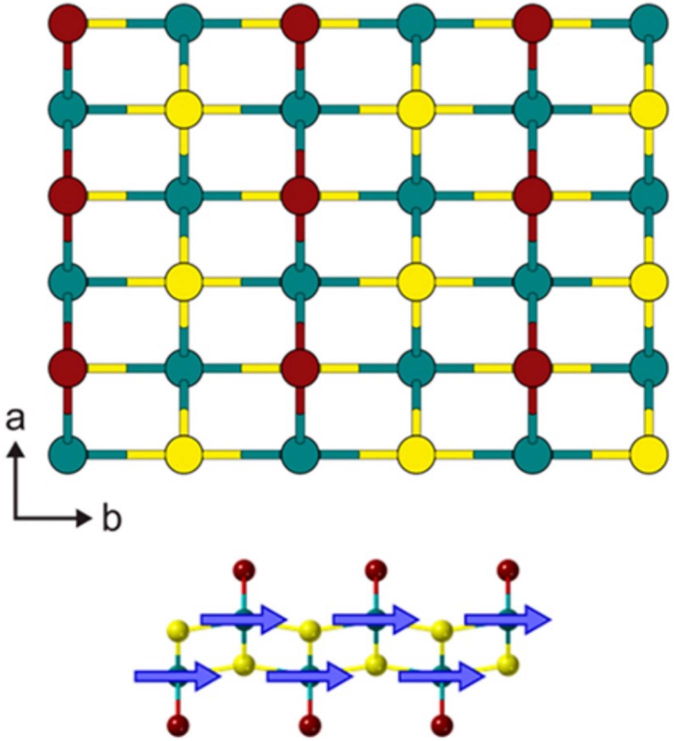
CrSBr



T_C = 146 K

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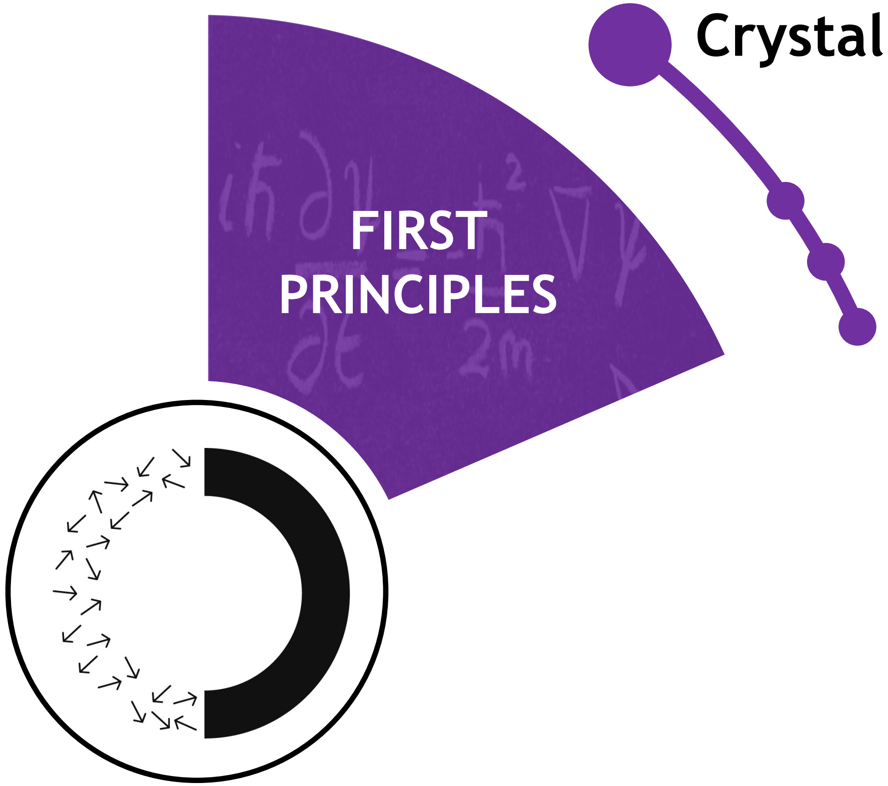
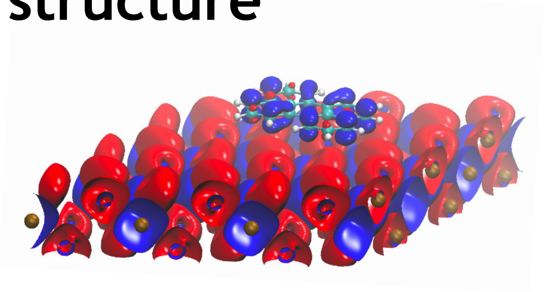
Computational and theoretical framework



Crystal structure



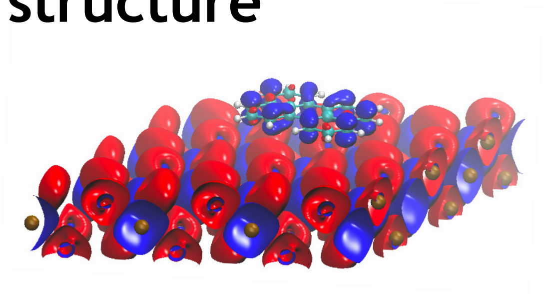
Perylene@CrI_3 FM monolayer

Computational and theoretical framework

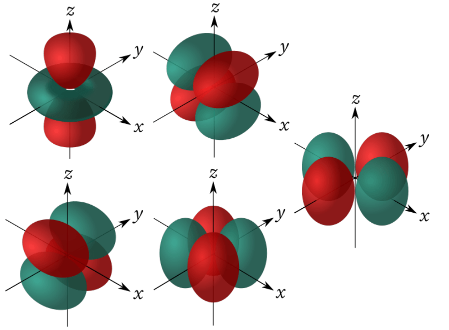
FIRST PRINCIPLES

TIGHT BINDING

Crystal structure

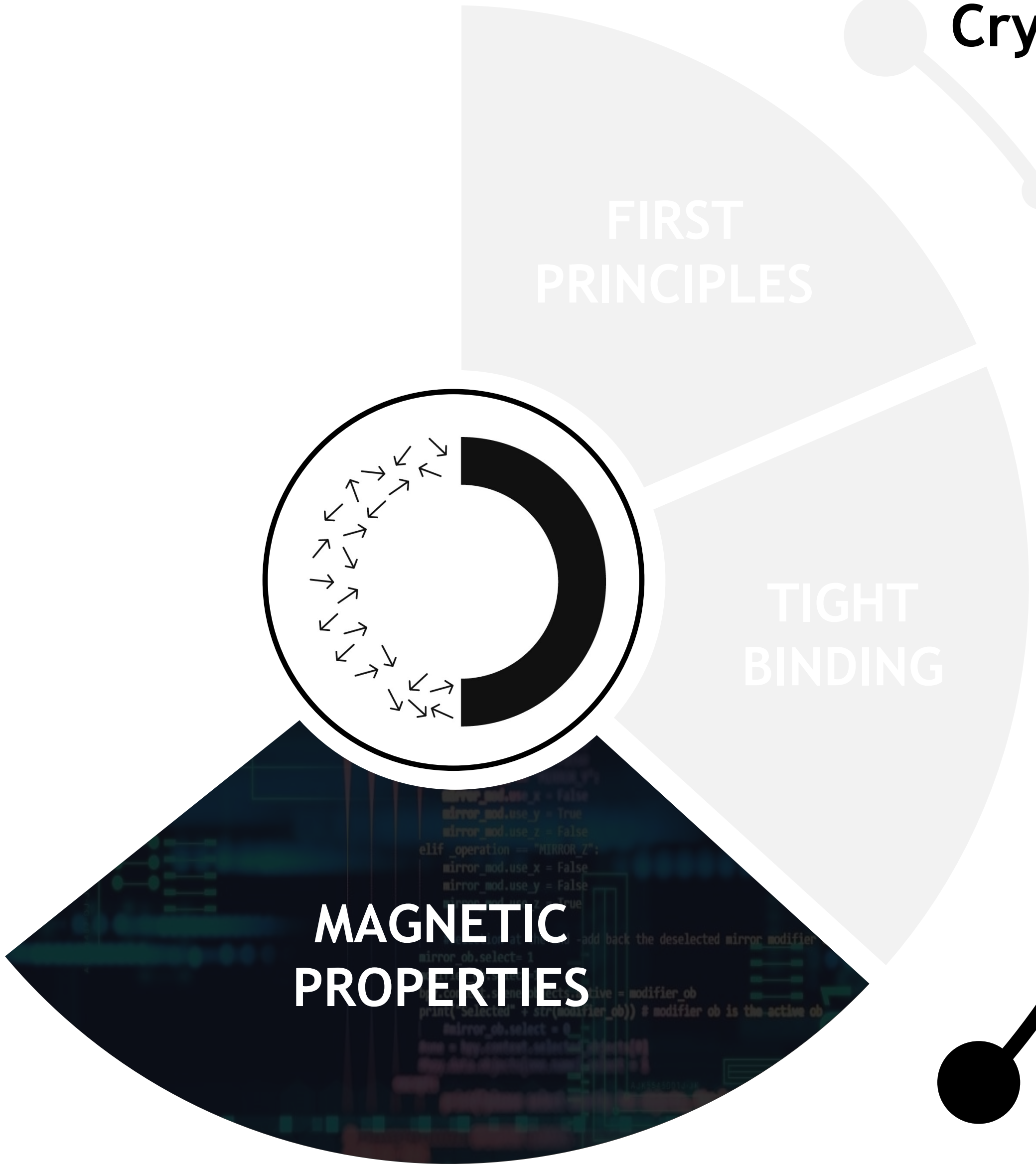


Wannierization



Maximally localized Wannier functions
d-orbitals

Computational and theoretical framework



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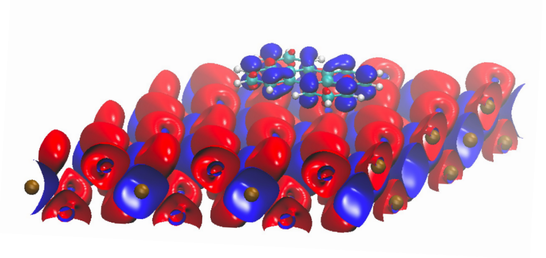

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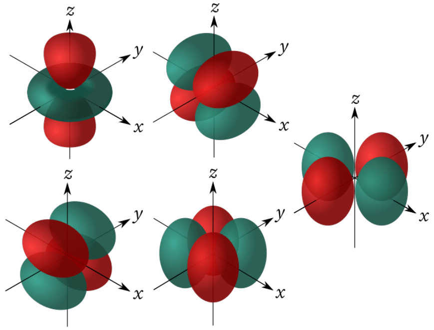
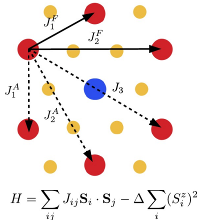
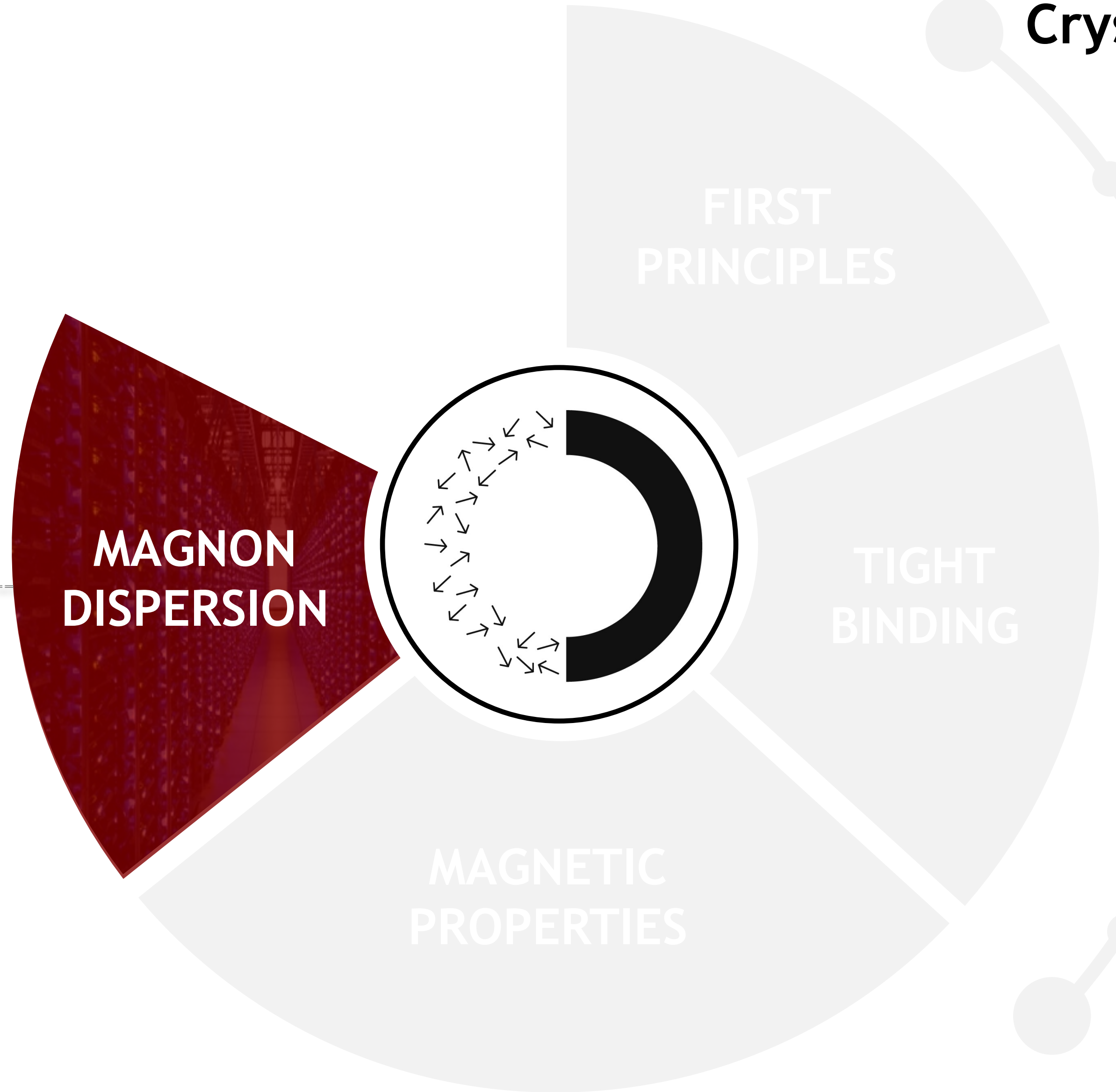


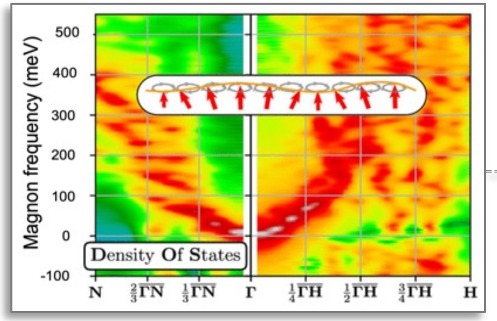
TB2J



Spin Hamiltonian

Computational and theoretical framework





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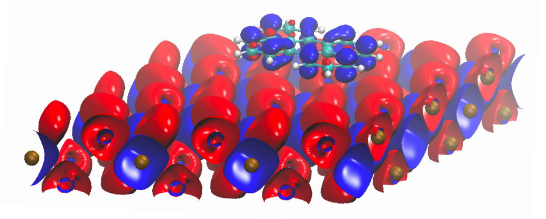

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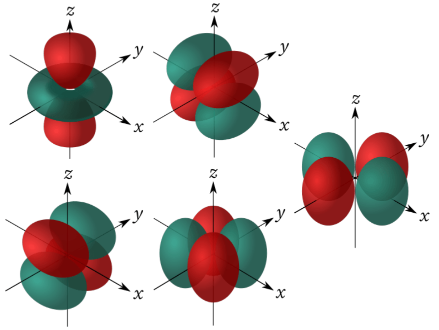

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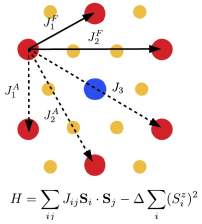
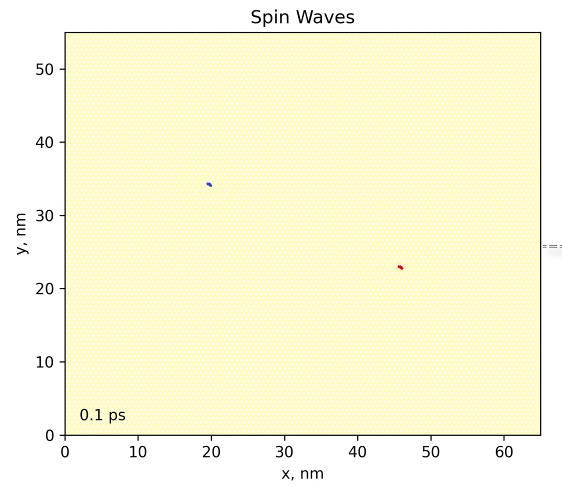
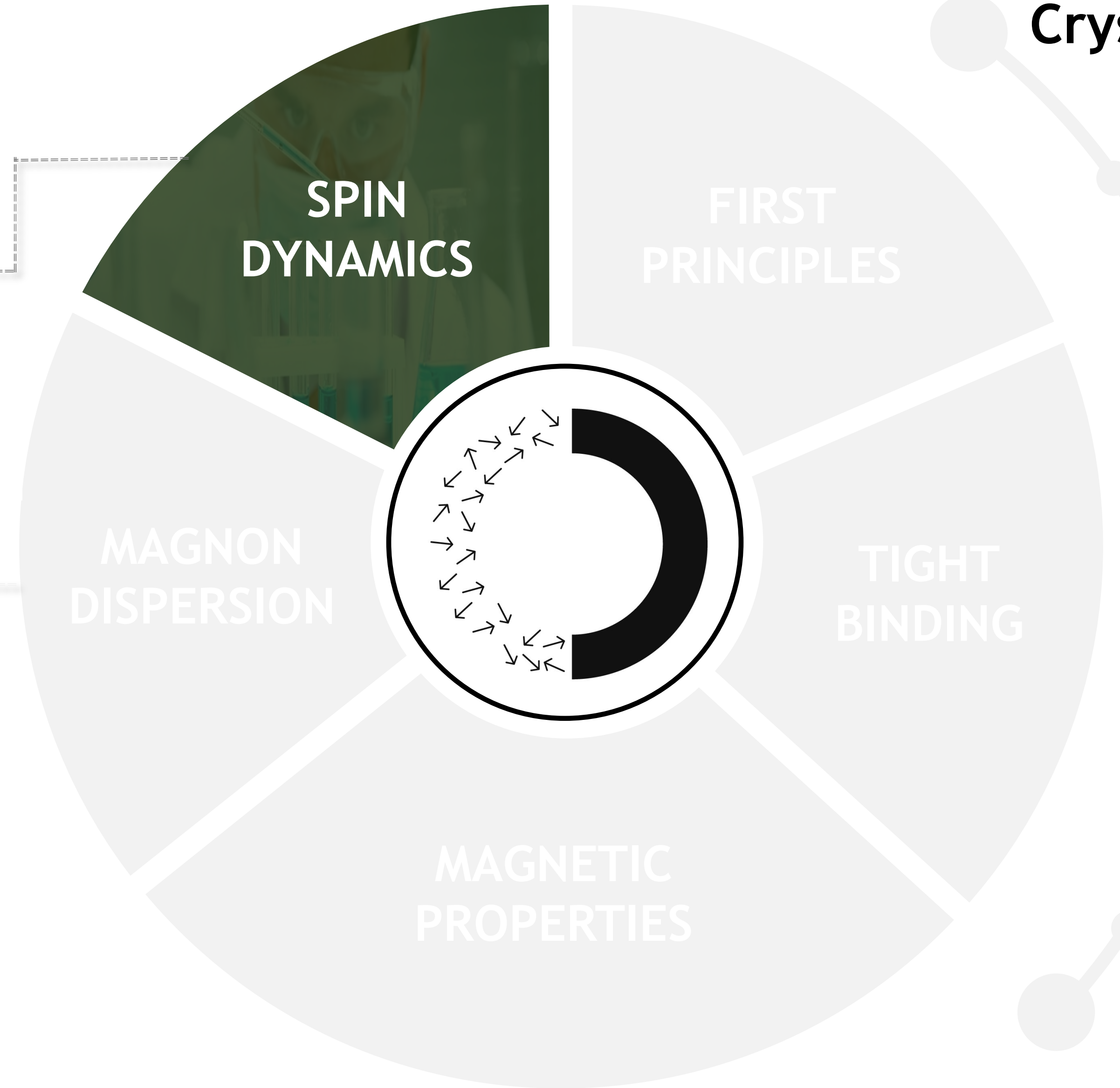
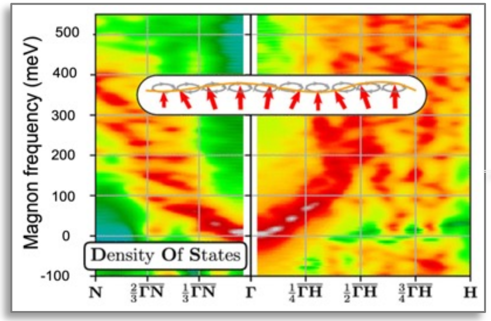


Spin Hamiltonian

Computational and theoretical framework







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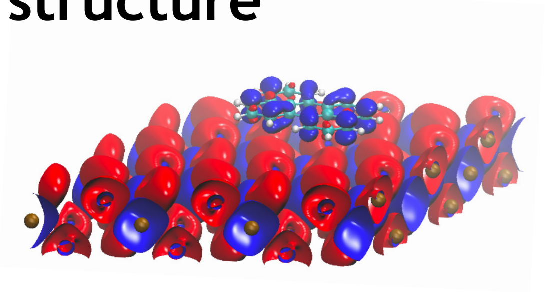

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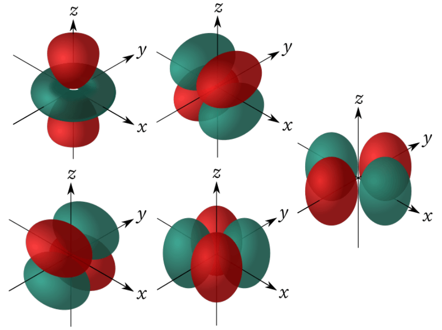
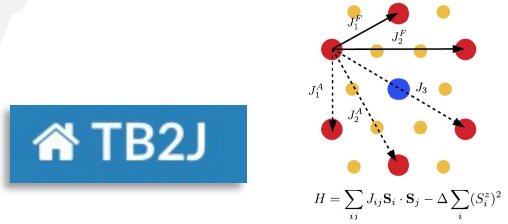
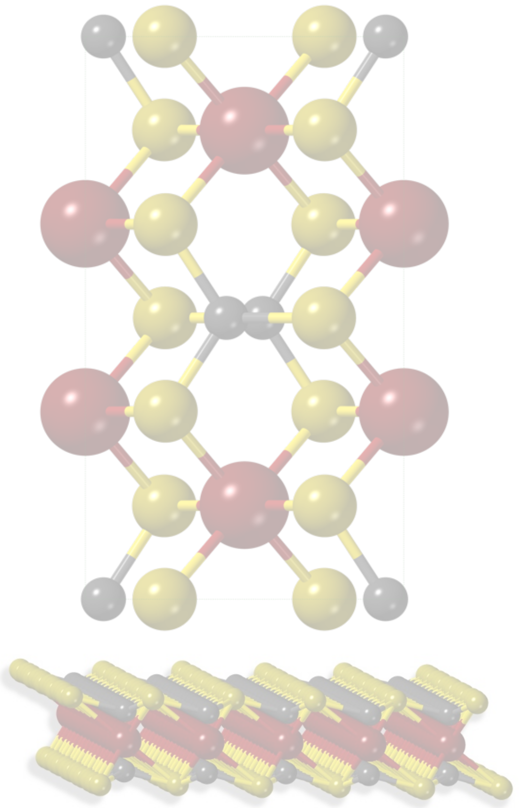




Spin Hamiltonian

2D van der Waals magnetic materials

MPS_3



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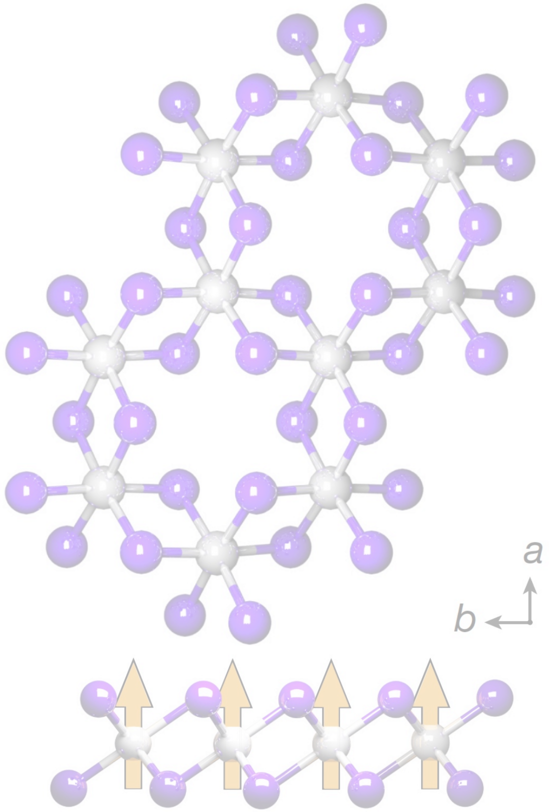

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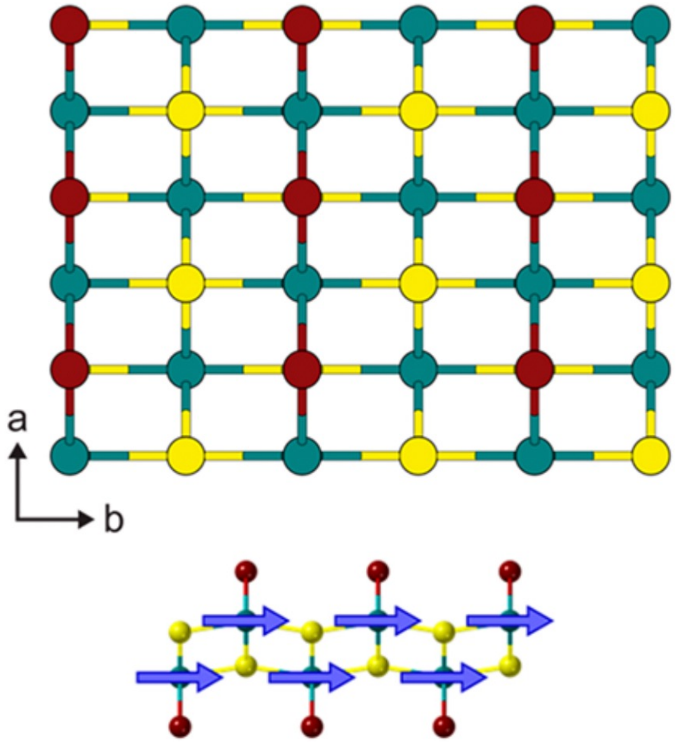

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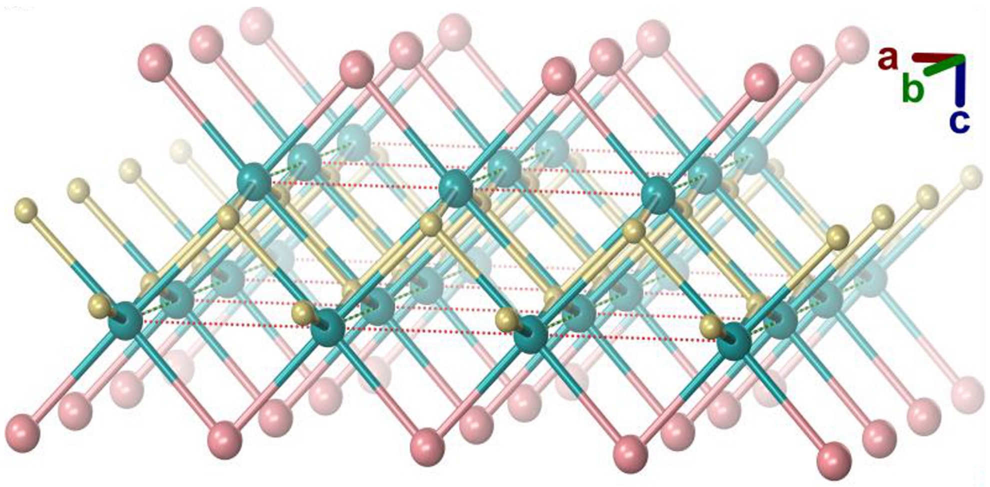
CrSBr: electronic structure



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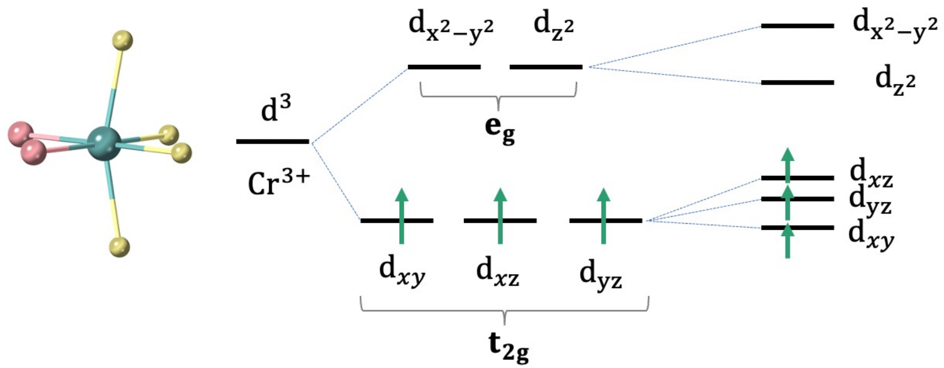
Rizzo *et al.*, *Adv. Mater.* **2022**, 2201000



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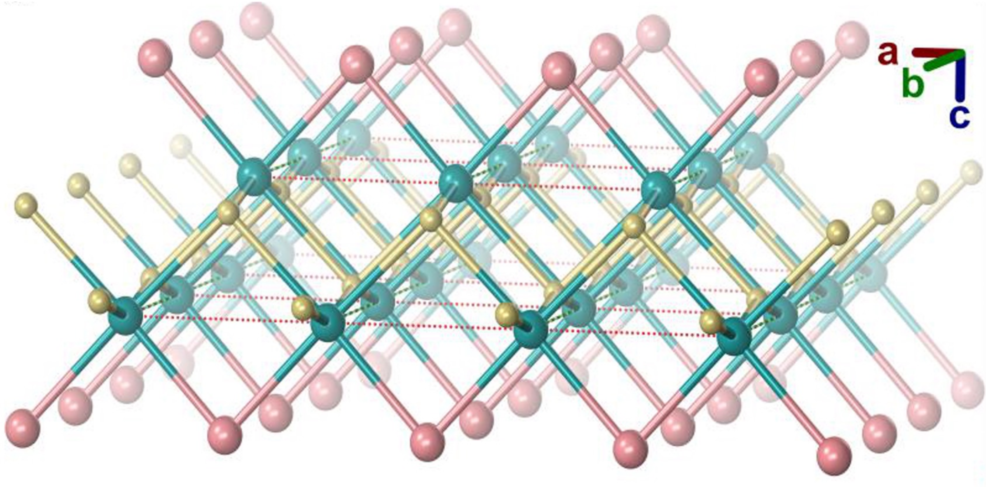

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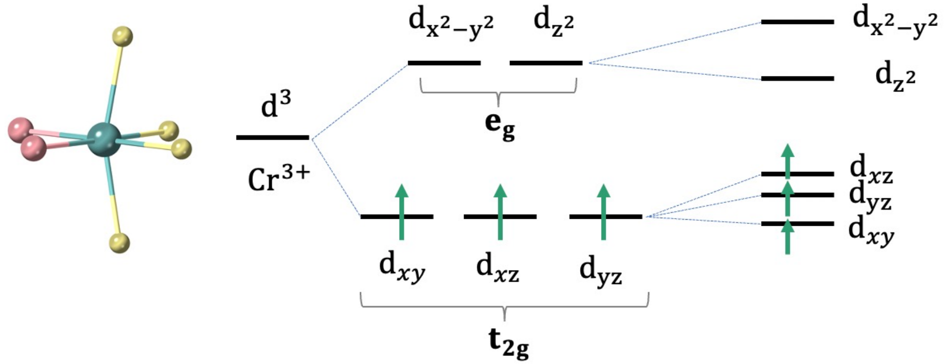
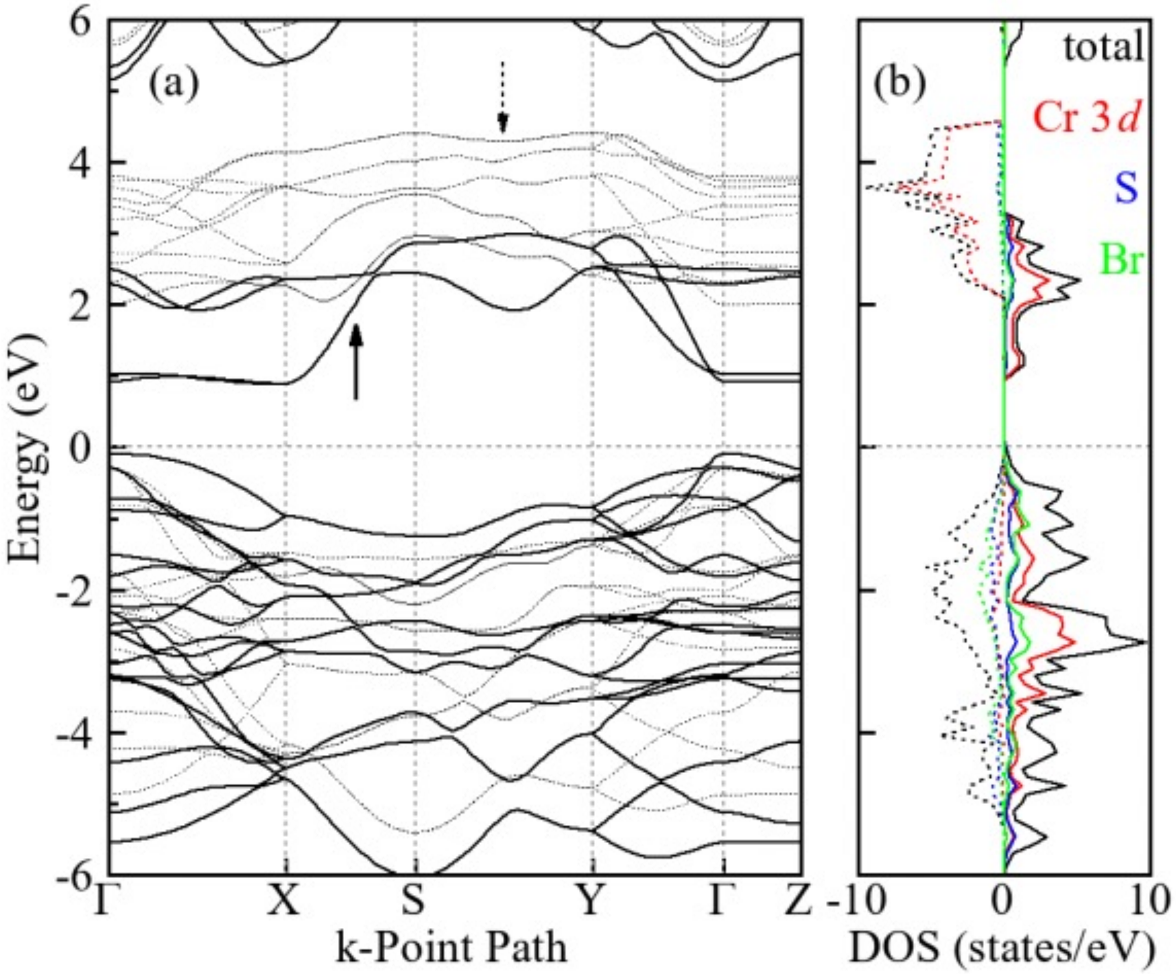
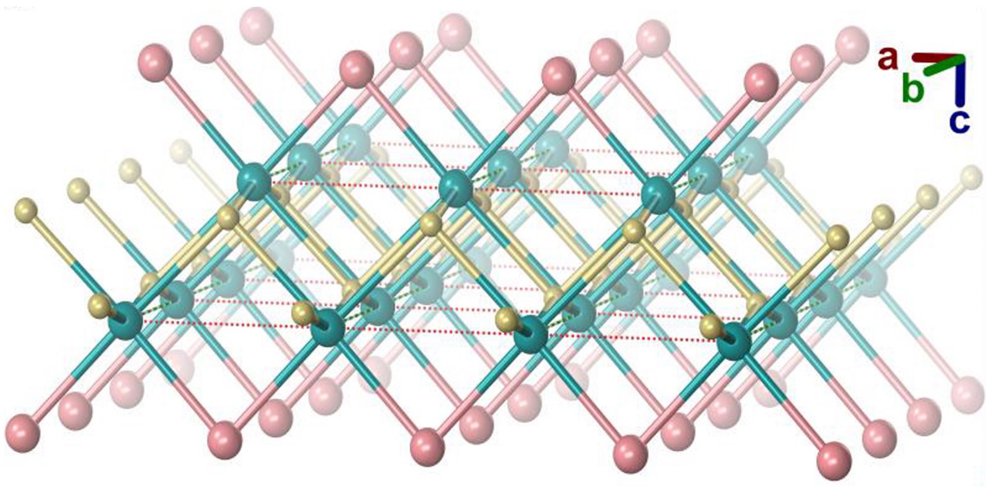


Electronic structure (DFT+U)



CrSBr: electronic structure



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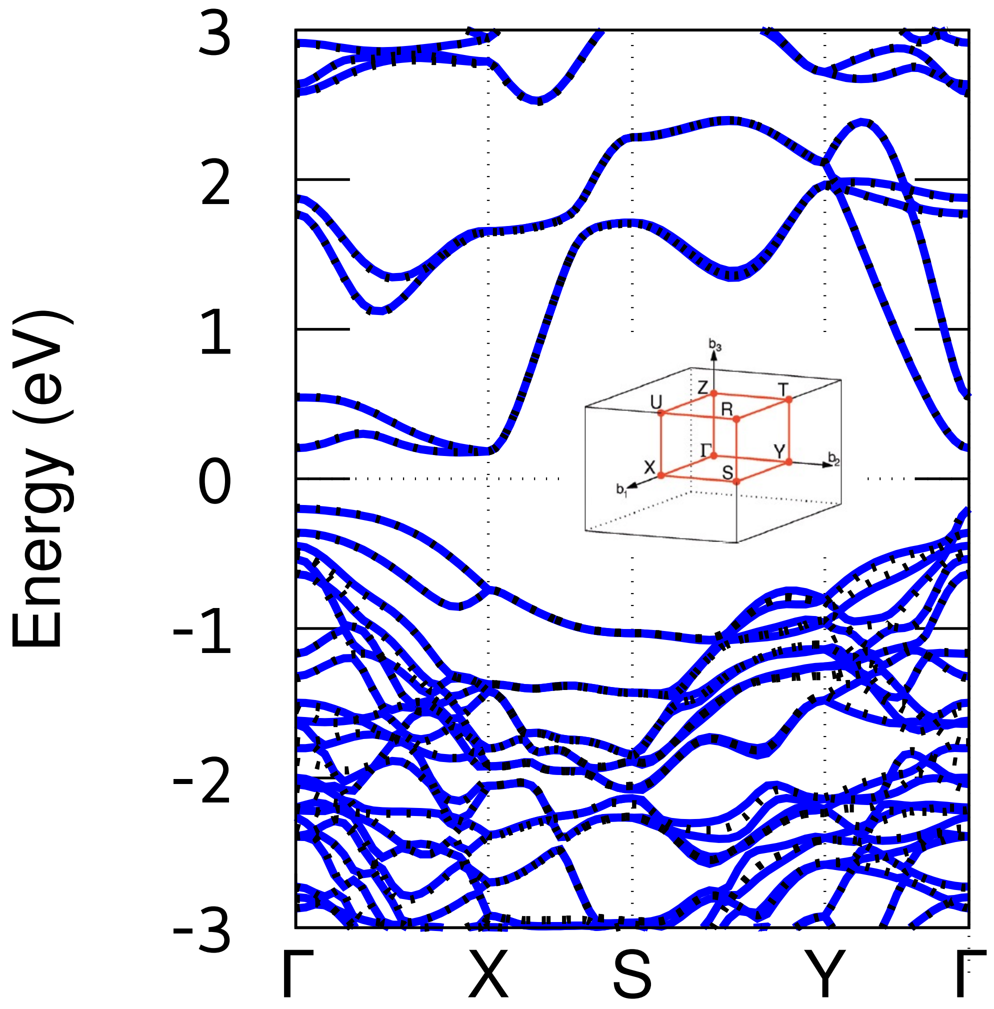
WANNIER90

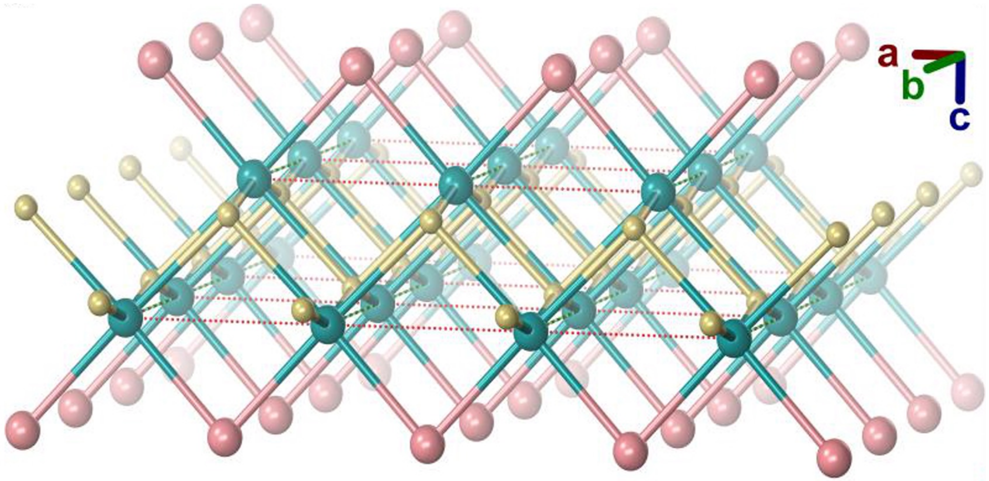
$$|\mathbf{R}n\rangle = \frac{V}{(2\pi)^3}\int_{\mathrm{BZ}} d\mathbf{k} e^{-i\mathbf{k}\cdot\mathbf{R}} \sum_{m=1}^{J} U_{mn}^{(\mathbf{k})} |\psi_{m\mathbf{k}}\rangle$$

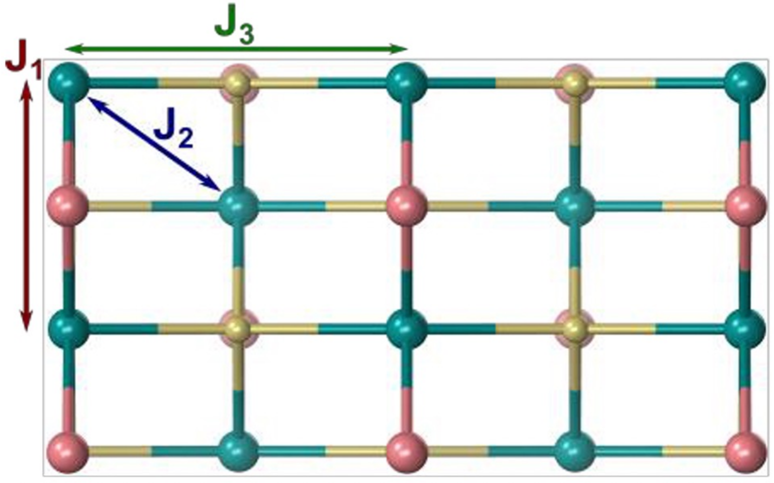
- **Tight-binding model** with *ab initio* parameters
- **Selected projectors:** Cr (d), S (p) and Br (p) orbitals
- Minimization of spread for the **Wannier functions**

DFT+U+SOC / TBH



CrSBr: magnetic interactions





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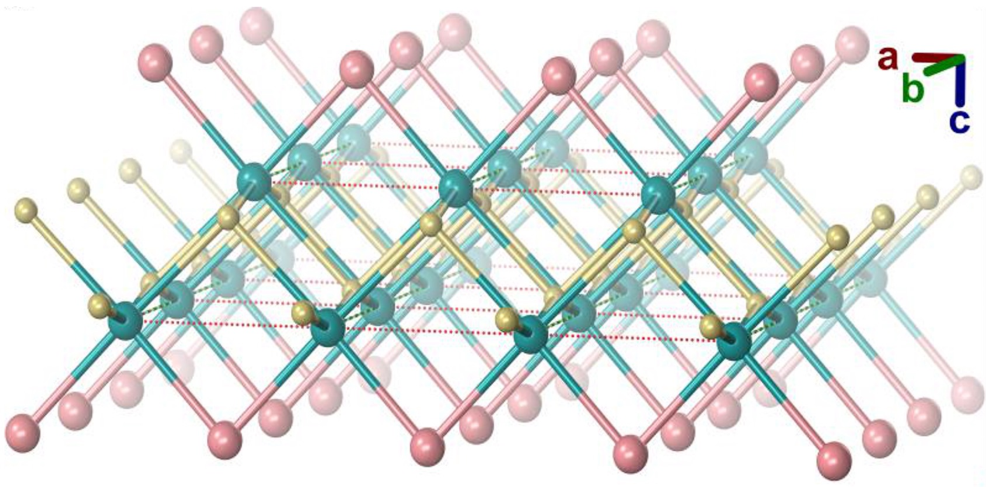
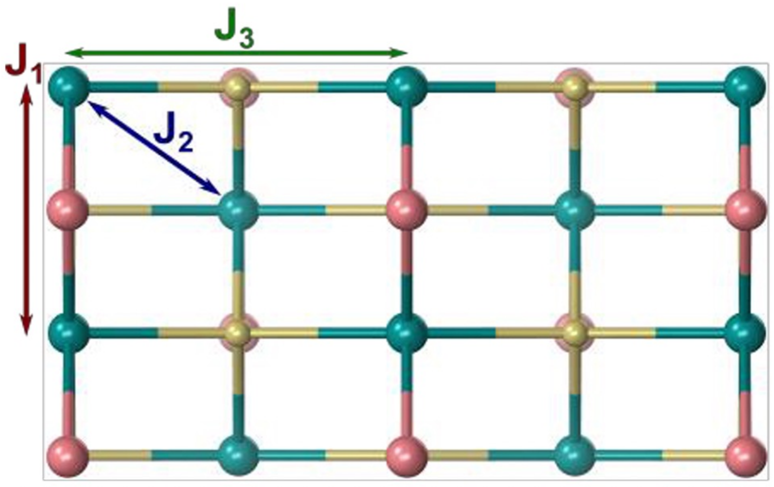
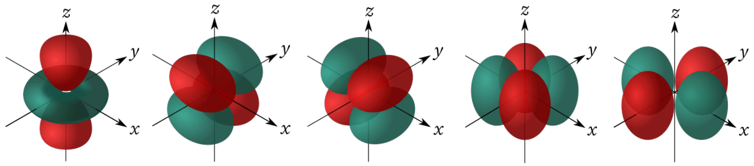






- Three Wannier hamiltonians that include SOC. Spin pointing x, y and z directions.

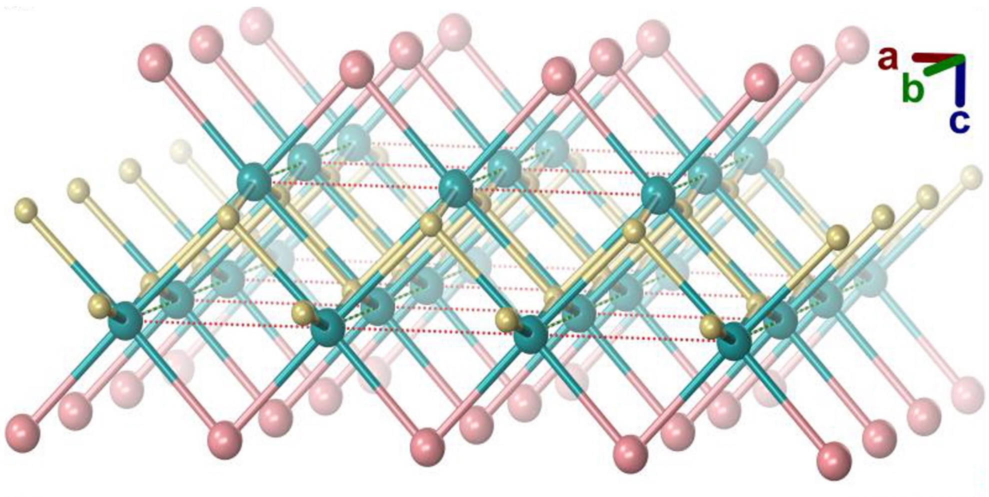
- Green's function method with local rigid spin rotation as a perturbation

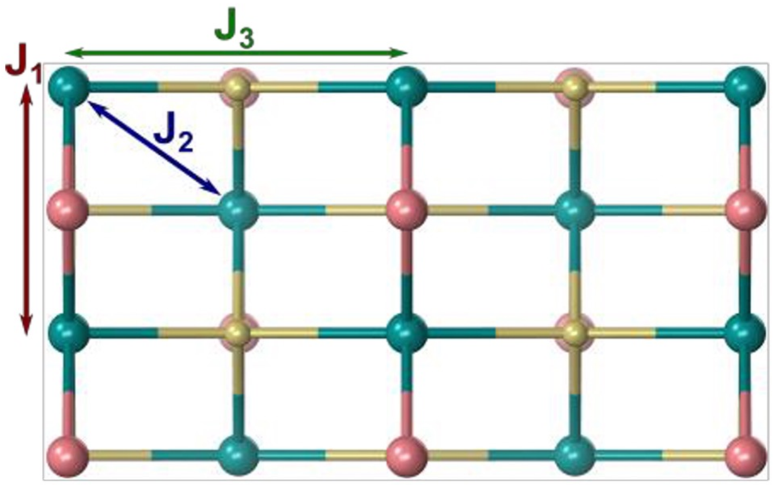
$$\hat{H} = \sum_{n=1}^{3}\left(-J_n \sum_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j - J_n^z \sum_{ij} \hat{S}_i^z \hat{S}_j^z\right) - D \sum_i (\hat{S}_i^z)^2$$

TB2J code : https://github.com/mailhexu/TB2J

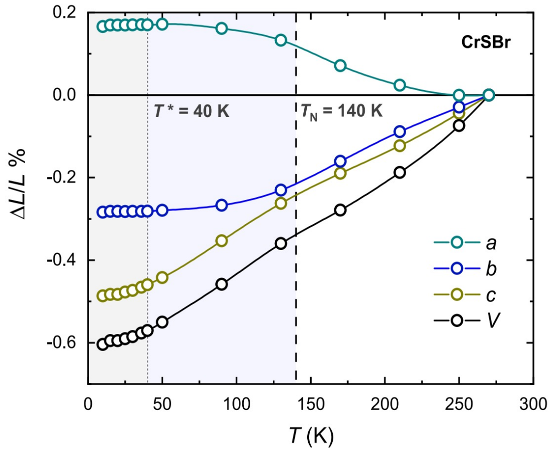
U, eV	1	2	3	4	5	6
J_1, meV	2.67	3.11	3.54	3.93	4.30	4.51
J_2, meV	3.48	3.29	3.08	2.87	2.63	2.43
J_3, meV	6.38	5.36	4.15	2.82	1.21	-0.64

CrSBr: magnetic interactions



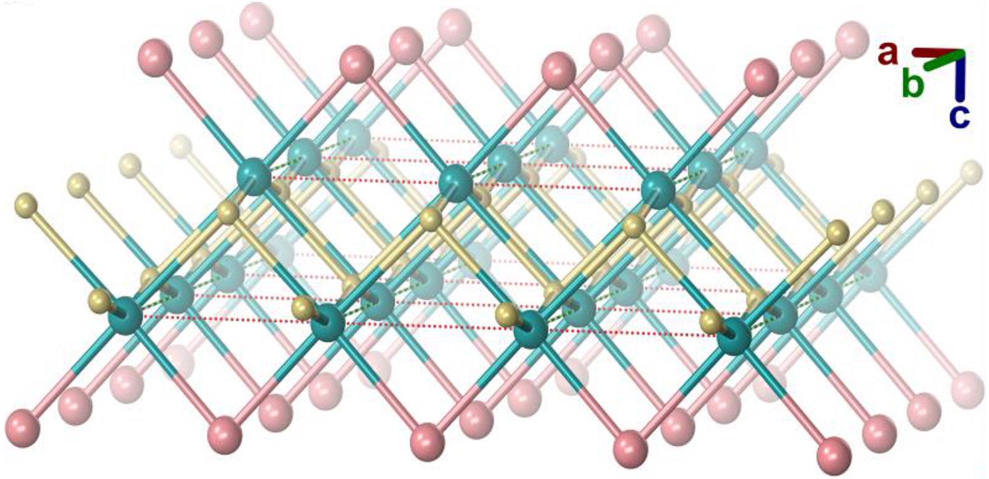


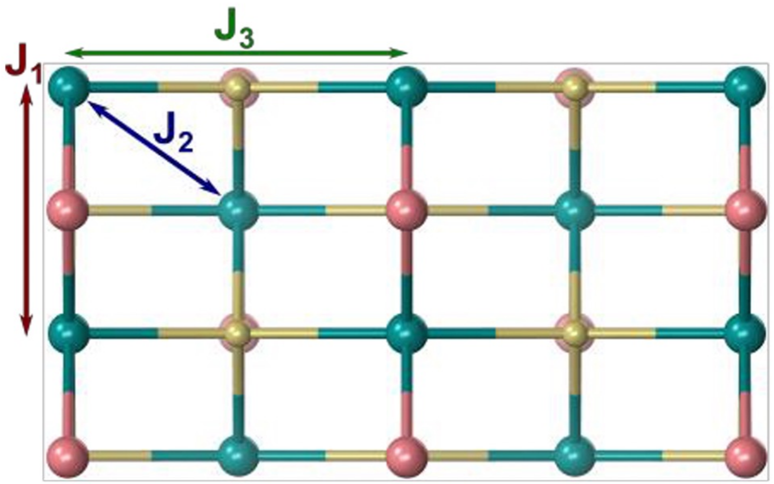
Synchrotron X-ray diffraction (250-10K)
López-Paz *et al. Nat. Commun.* 2022, **13**, 4745



Boix-Constant, Mañas-Valero,* Ruiz, Rybakov, Konieczny, Pillet, Baldoví and Coronado* *Adv. Mater*. 2022, **34**, 2204940

CrSBr: magnetic interactions





Synchrotron X-ray diffraction (250-10K)
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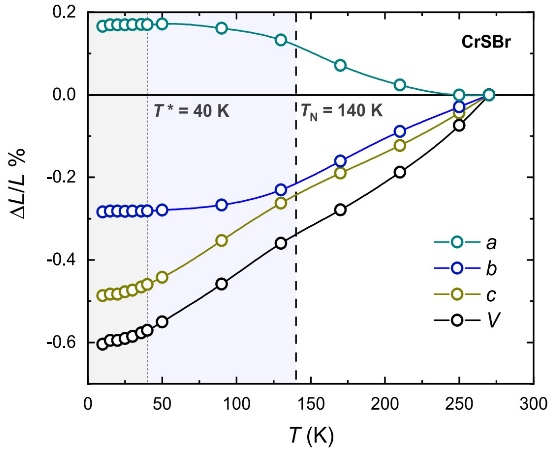
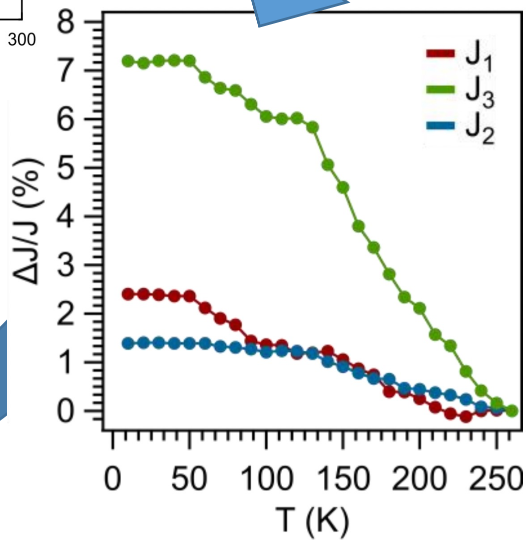
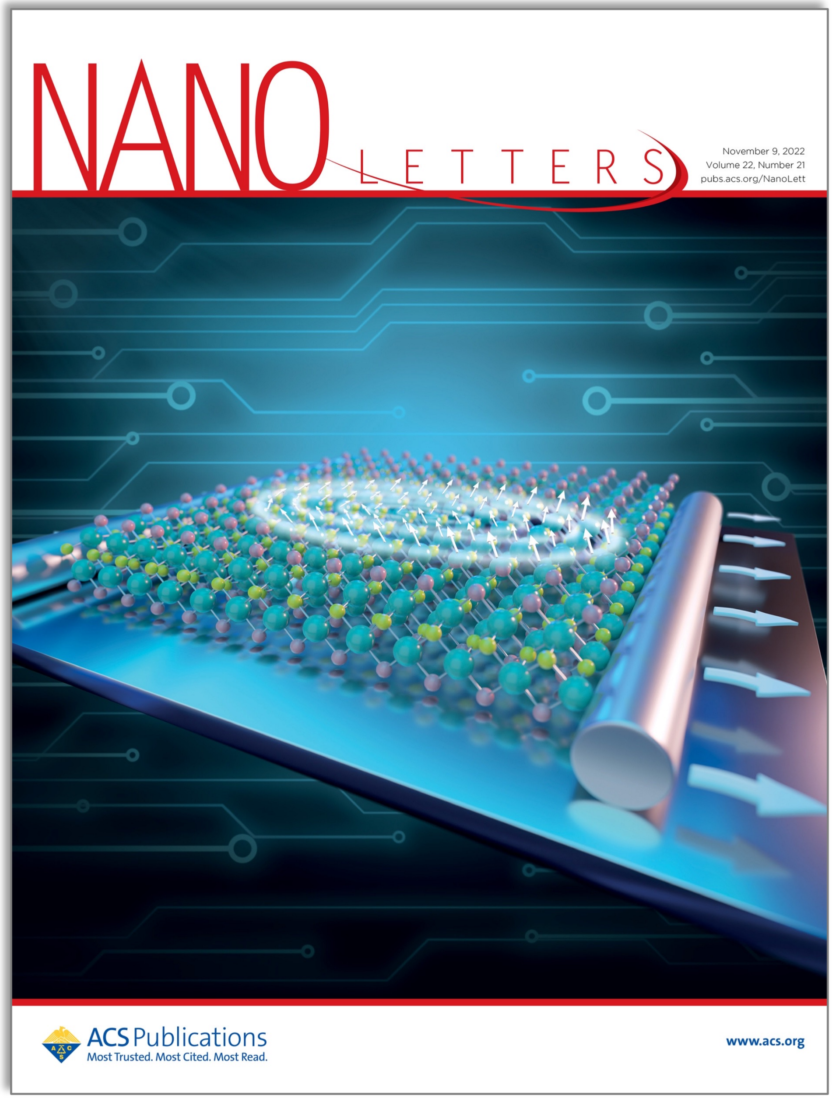


Alberto M. Ruiz



Boix-Constant, Mañas-Valero,* Ruiz, Rybakov, Konieczny, Pillet, Baldoví and Coronado* *Adv. Mater*. 2022, **34**, 2204940

Magnon straintronics in single-layer CrSBr



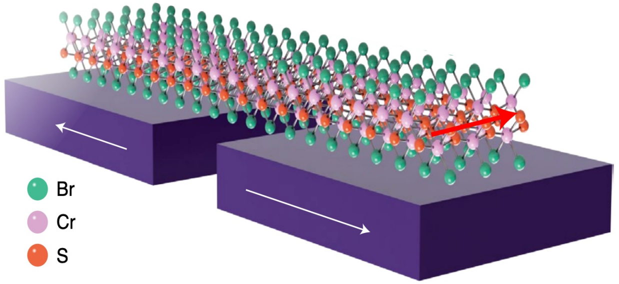
2D materials

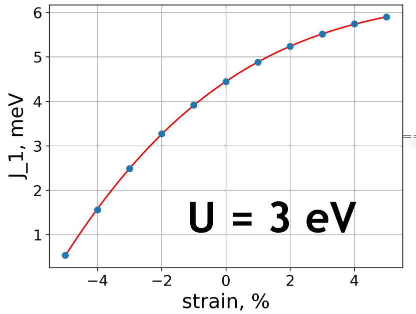
Outstanding deformation capacity

Can we use strain engineering to control spin waves propagation?

Esteras, Rybakov, Ruiz and Baldoví* *Nano Lett*. 2022, **22**, 8771–8778

High-density maps of magnetic exchange

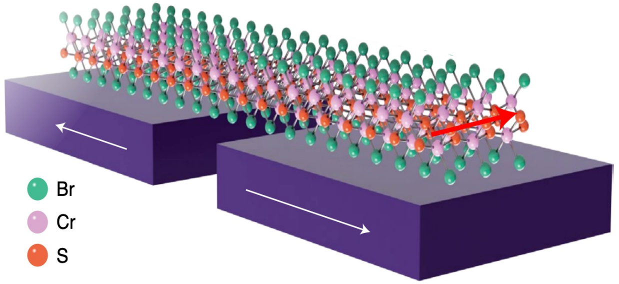


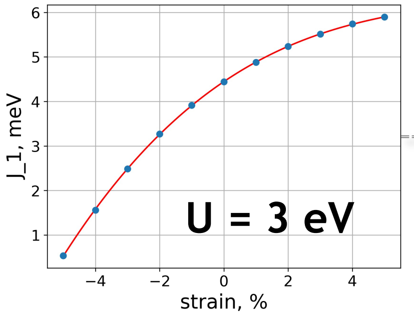


$$J = \sum_{i=0}^{3}\sum_{j=0}^{3} a_{ij} U^{i} \varepsilon^{j}$$

Dorye L. Esteras

High-density maps of magnetic exchange

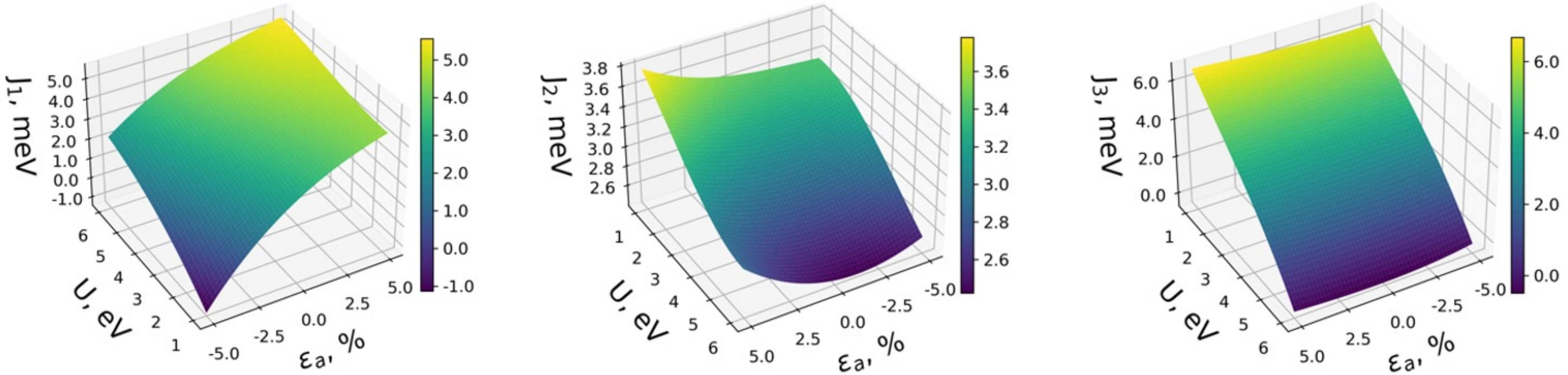




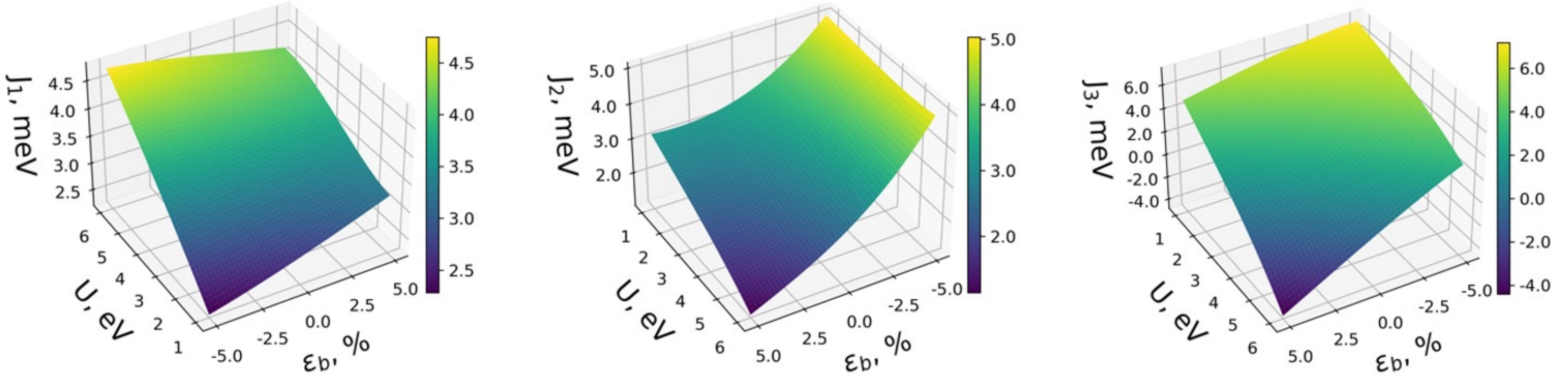
$$J = \sum_{i=0}^{3}\sum_{j=0}^{3} a_{ij} U^i \varepsilon^j$$

Dorye L. Esteras

- Uniaxial strain (a)

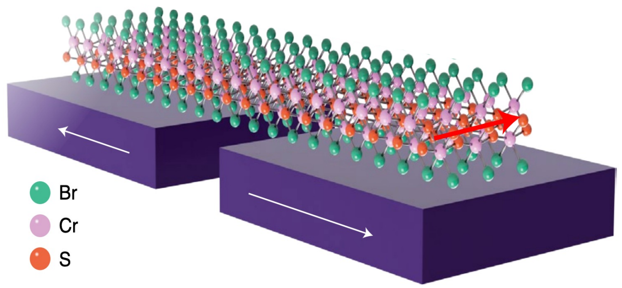


- Uniaxial strain (b)



Esteras, Rybakov, Ruiz and Baldoví* *Nano Lett*. 2022, **22**, 8771–8778

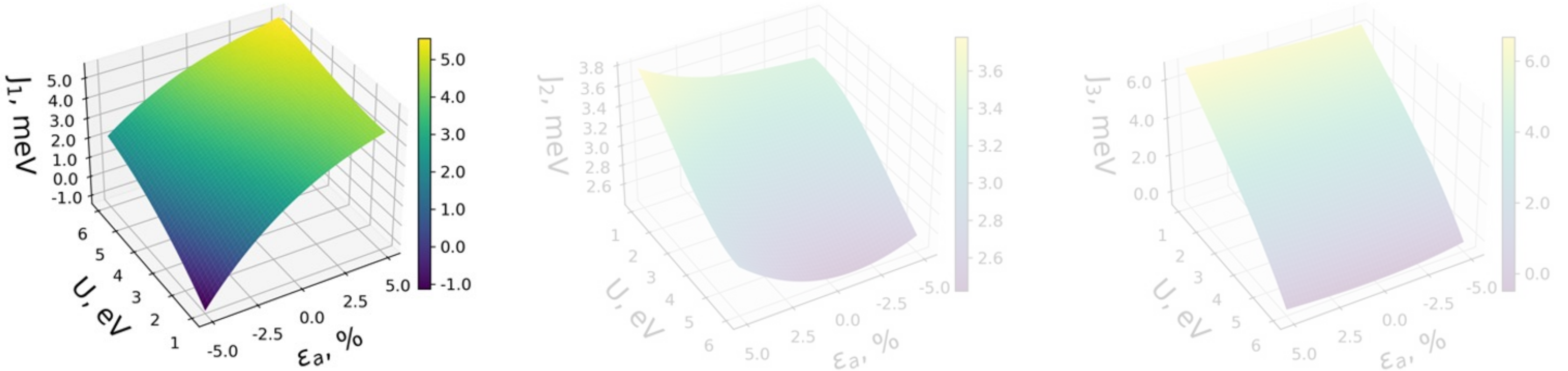
High-density maps of magnetic exchange



$$J = \sum_{i=0}^{3}\sum_{j=0}^{3} a_{ij} U^{i} \varepsilon^{j}$$

Dorye L. Esteras

- **Uniaxial strain (a)**

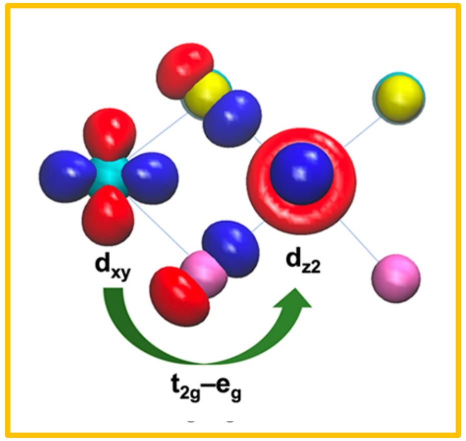


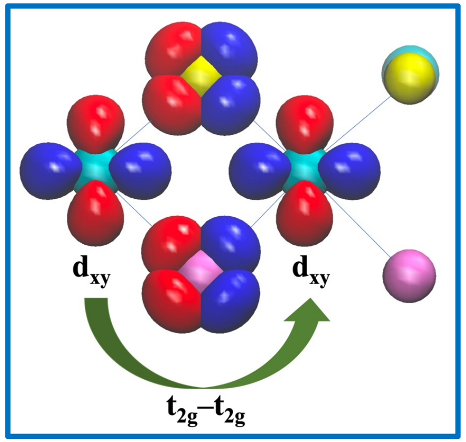
- **Uniaxial strain (b)**

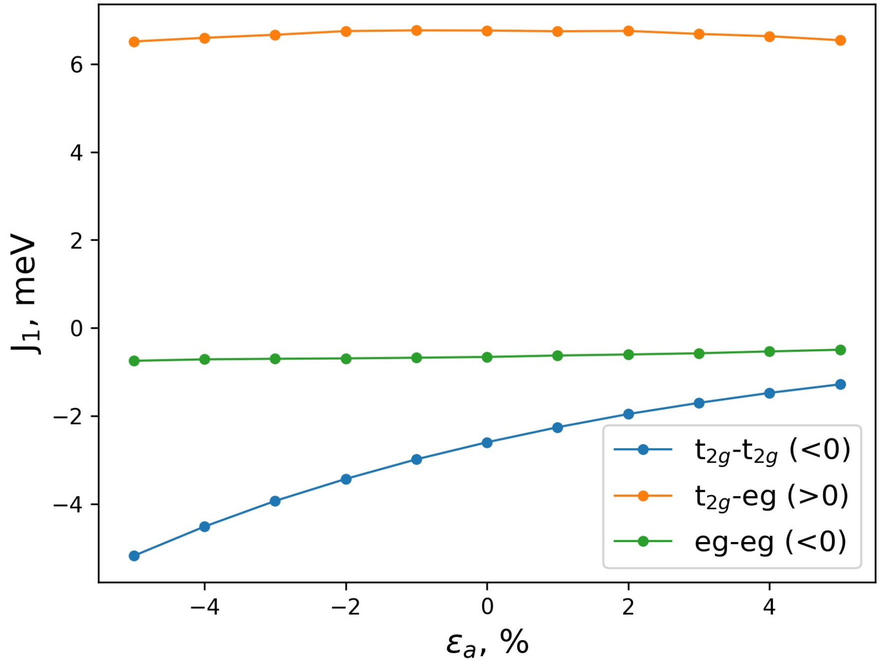
Esteras, Rybakov, Ruiz and Baldoví* *Nano Lett*. 2022, **22**, 8771–8778

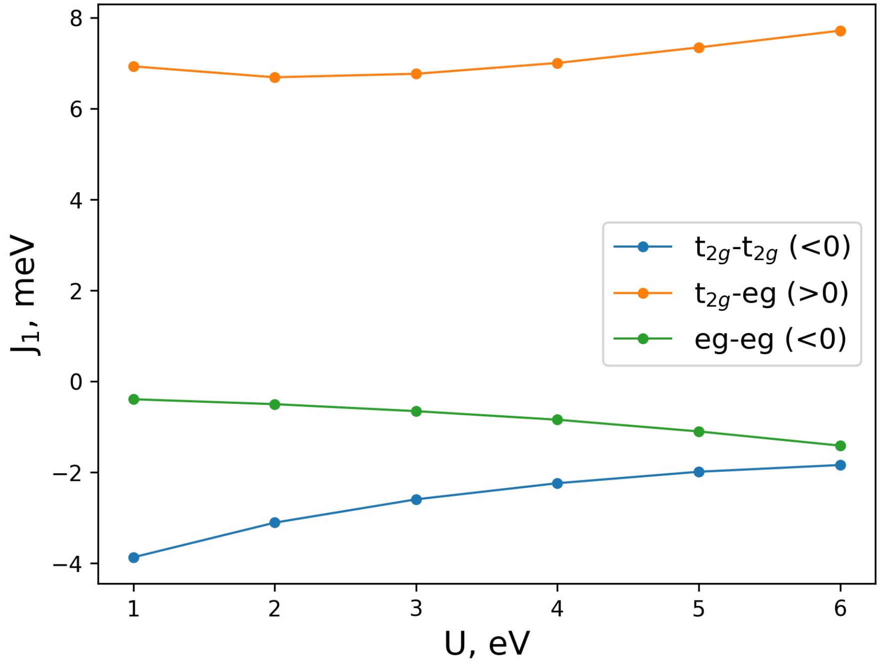
Orbital decomposed J_1 channels

- **Uniaxial strain (a)**



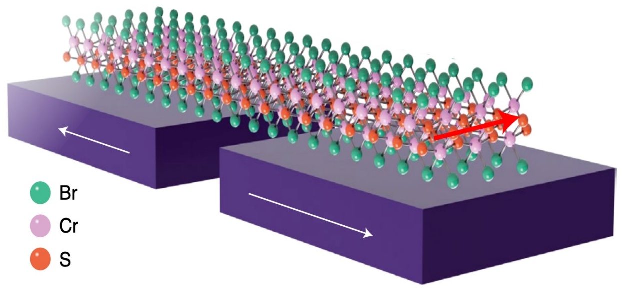






- Competing FM (t_{2g}-e_g) vs AFM (t_{2g}-t_{2g}) pathways
- Tensile strain and an enhancement of Coulomb interactions lead to a decrease of the AFM channel, thus enhancing J_1.

High-density maps of magnetic exchange

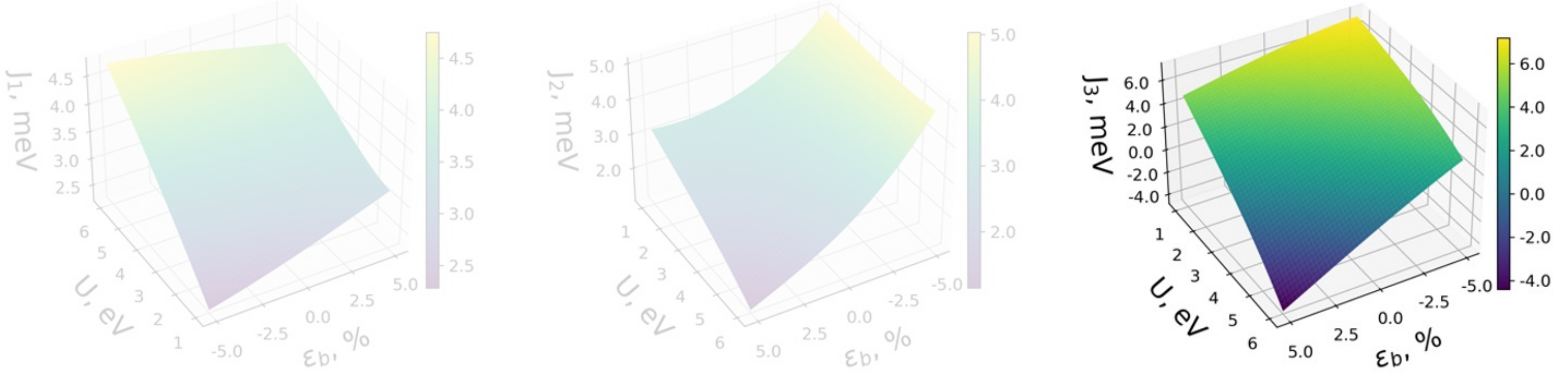


$$J = \sum_{i=0}^{3}\sum_{j=0}^{3} a_{ij}U^i\varepsilon^j$$

Dorye L. Esteras

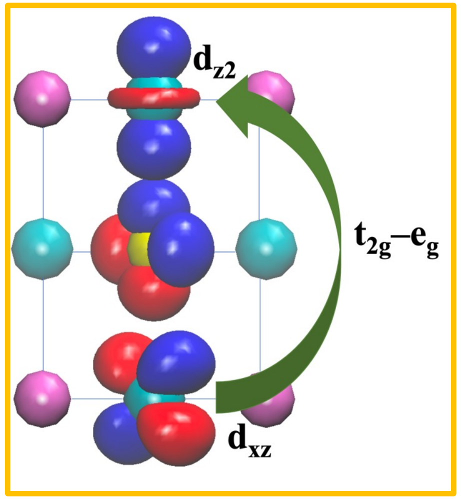
- Uniaxial strain (a)

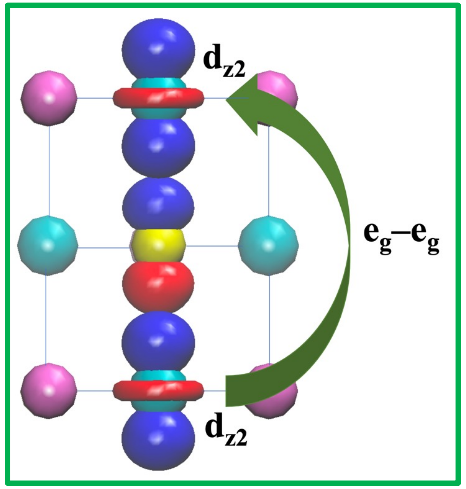
- **Uniaxial strain (b)**



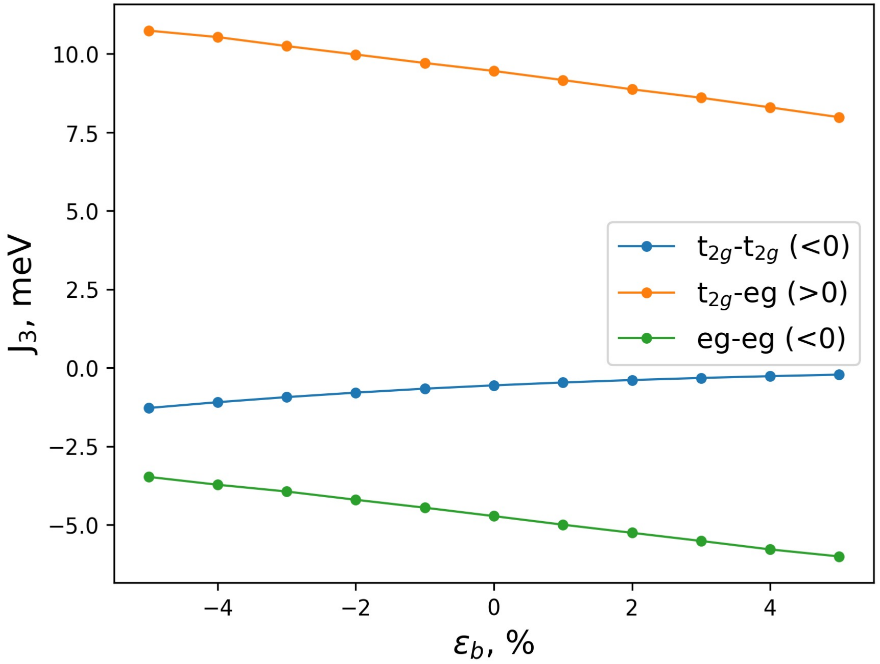
Esteras, Rybakov, Ruiz and Baldoví* *Nano Lett*. 2022, **22**, 8771–8778

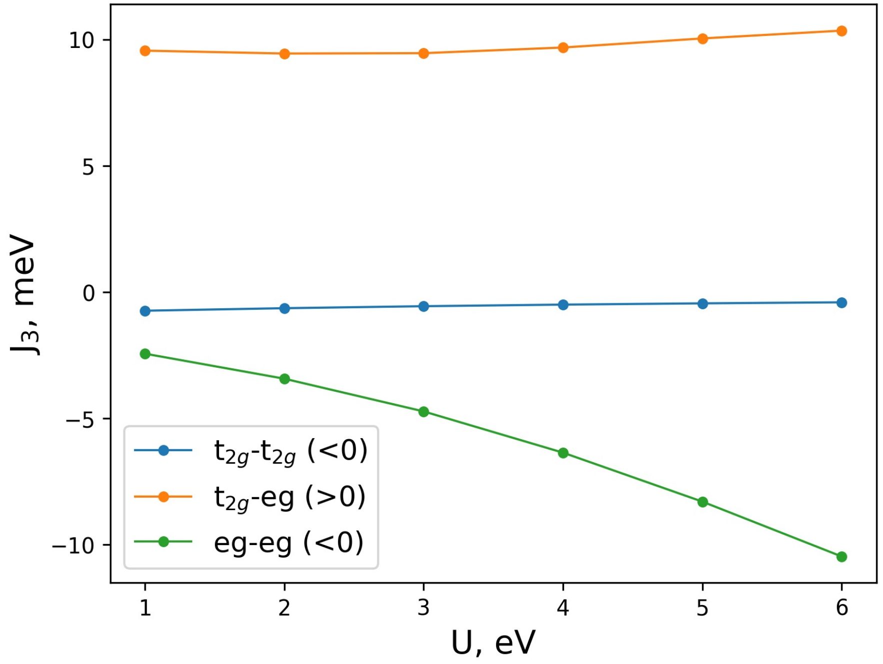
Orbital decomposed J_3 channels





- **Uniaxial strain (b)**

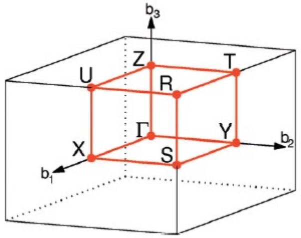




- Competing FM (t_{2g}-e_g) vs AFM (e_g-e_g) pathways
- **Compressive strain** (*b*) and **decrease of Hubbard U** lead to a decrease of the AFM channel, thus enhancing J_3.

Strain-engineering of magnon dispersion

- Holstein-Primakoff transformation in linear spin wave approximation



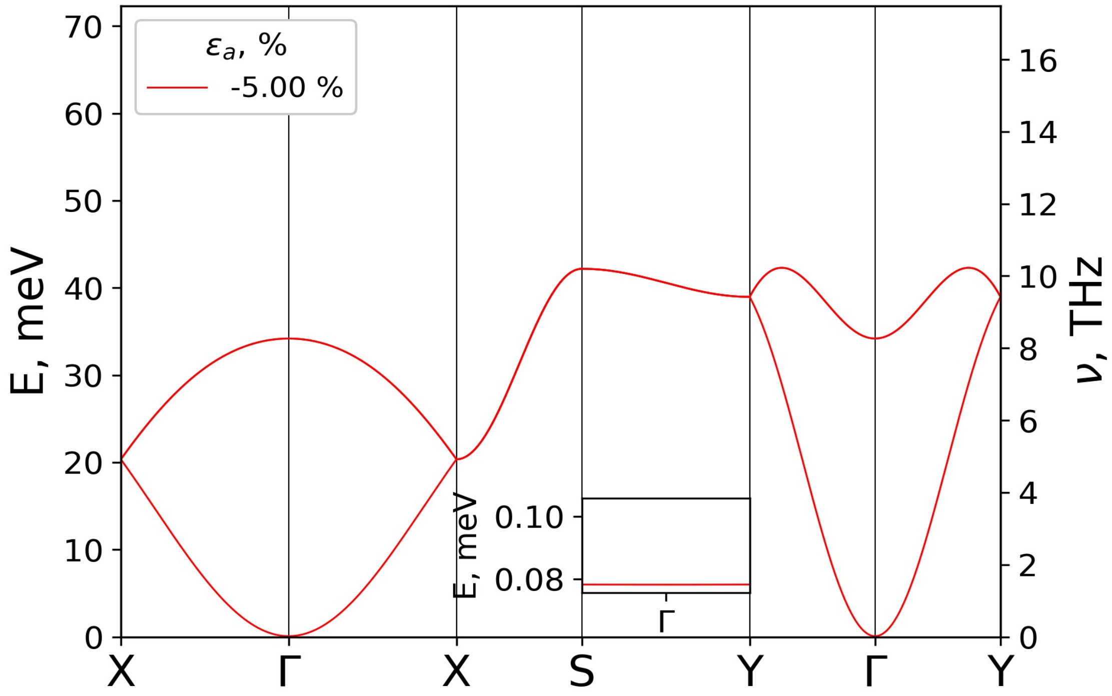
Andrey Rybakov

$$\hat{H} = \sum_{n=1}^{3}\left(-J_n \sum_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j - J_n^z \sum_{ij} \hat{S}_i^z \hat{S}_j^z\right) - D \sum_i (\hat{S}_i^z)^2$$

$$\hat{H}_{SW} = E_0 + \sum_k \omega_k \cdot \hat{a}_k^{+} \hat{a}_k$$

$$\omega_k = 2S \sum_{n=1}^{3} \left(J_n n_n (1 - \gamma_k^{(n)}) + J_n^z n_n\right) + 2SD$$

Uniaxial strain (a)



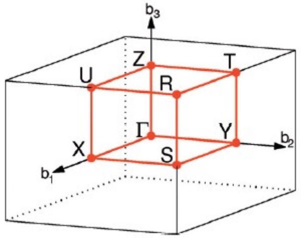
Strain-engineering of magnon dispersion

- Holstein-Primakoff transformation in linear spin wave approximation

$$\hat{H} = \sum_{n=1}^{3}\left(-J_n \sum_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j - J_n^z \sum_{ij} \hat{S}_i^z \hat{S}_j^z\right) - D \sum_i (\hat{S}_i^z)^2 \quad \Rightarrow$$

$$\hat{H}_{SW} = E_0 + \sum_k \omega_k \cdot \hat{a}_k^+ \hat{a}_k$$

$$\omega_k = 2S \sum_{n=1}^{3} \left(J_n n_n (1 - \gamma_k^{(n)}) + J_n^z n_n\right) + 2SD$$



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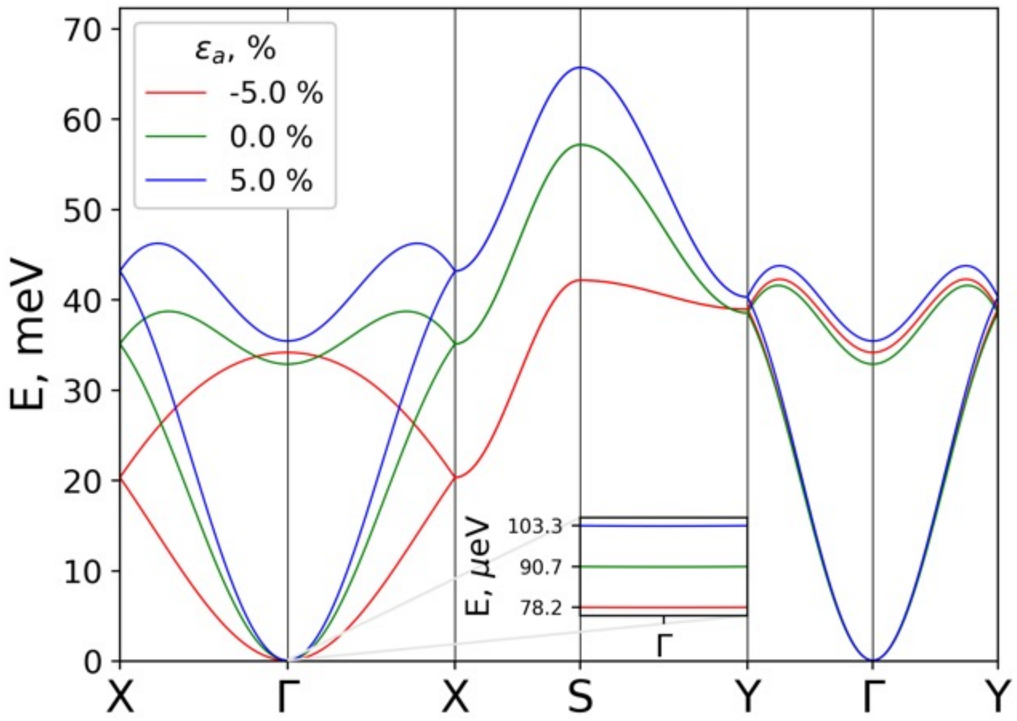
Uniaxial strain (a)

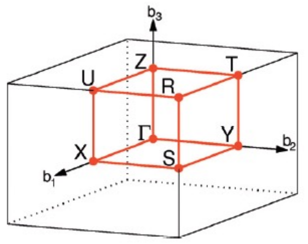


Strain-engineering of magnon dispersion

- Holstein-Primakoff transformation in linear spin wave approximation

$$\hat{H} = \sum_{n=1}^{3}\left(-J_n \sum_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j - J_n^z \sum_{ij} \hat{S}_i^z \hat{S}_j^z\right) - D \sum_i (\hat{S}_i^z)^2 \rightarrow \hat{H}_{SW} = E_0 + \sum_k \omega_k \cdot \hat{a}_k^+ \hat{a}_k$$

$$\omega_k = 2S \sum_{n=1}^{3} \left(J_n n_n (1 - \gamma_k^{(n)}) + J_n^z n_n\right) + 2SD$$



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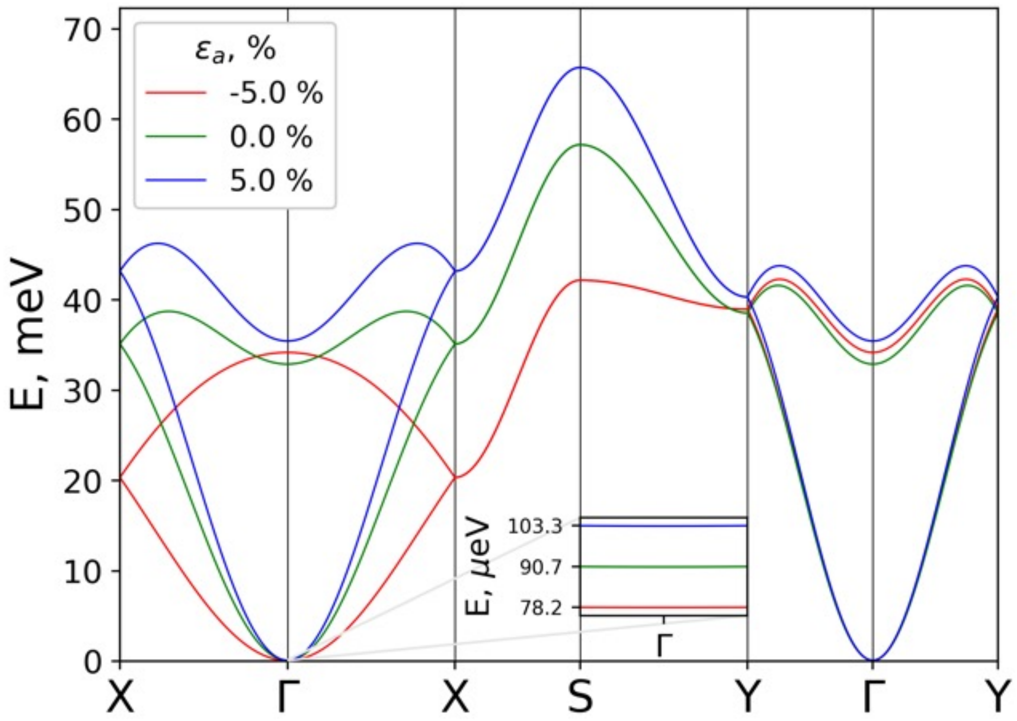
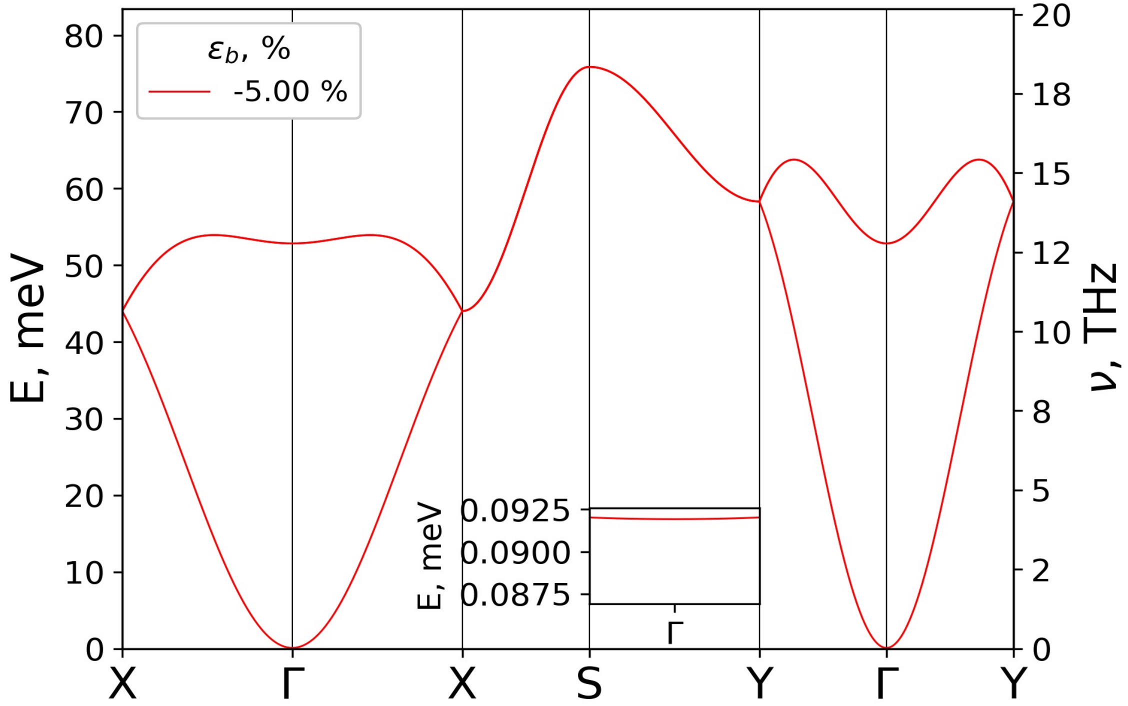
Uniaxial strain (a)



Uniaxial strain (b)

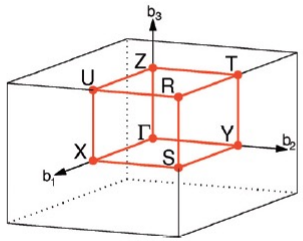


Strain-engineering of magnon dispersion

- Holstein-Primakoff transformation in linear spin wave approximation

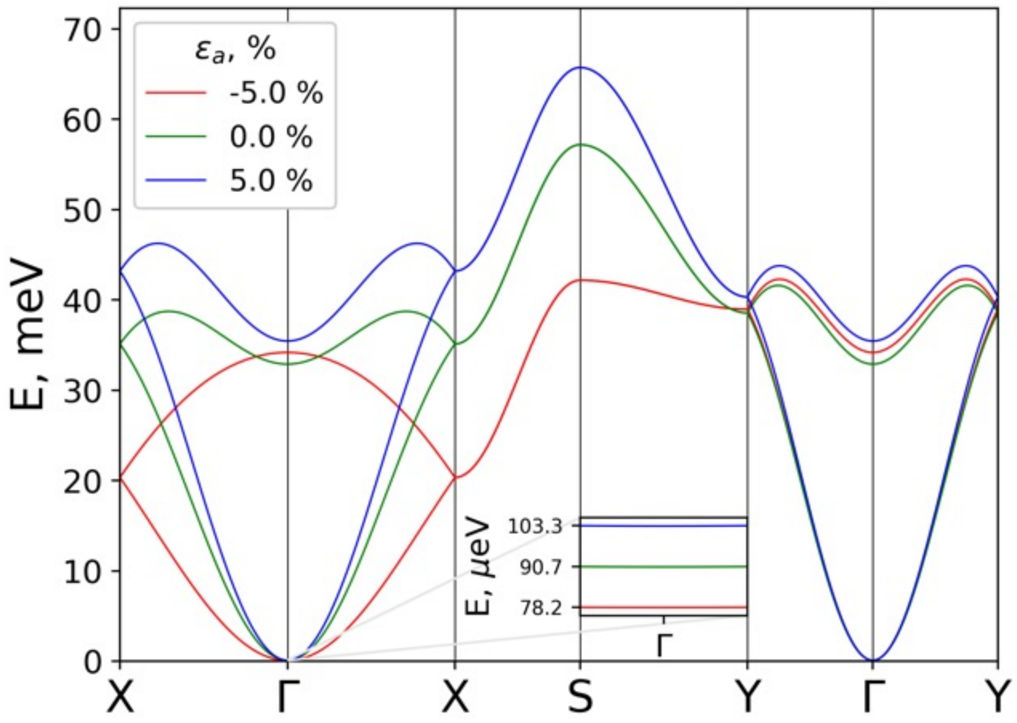
$$\hat{H} = \sum_{n=1}^{3}\left(-J_n \sum_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j - J_n^z \sum_{ij} \hat{S}_i^z \hat{S}_j^z\right) - D \sum_i (\hat{S}_i^z)^2 \quad \Rightarrow \quad \hat{H}_{SW} = E_0 + \sum_k \omega_k \cdot \hat{a}_k^{+} \hat{a}_k$$

$$\omega_k = 2S \sum_{n=1}^{3} \left(J_n n_n (1 - \gamma_k^{(n)}) + J_n^z n_n\right) + 2SD$$

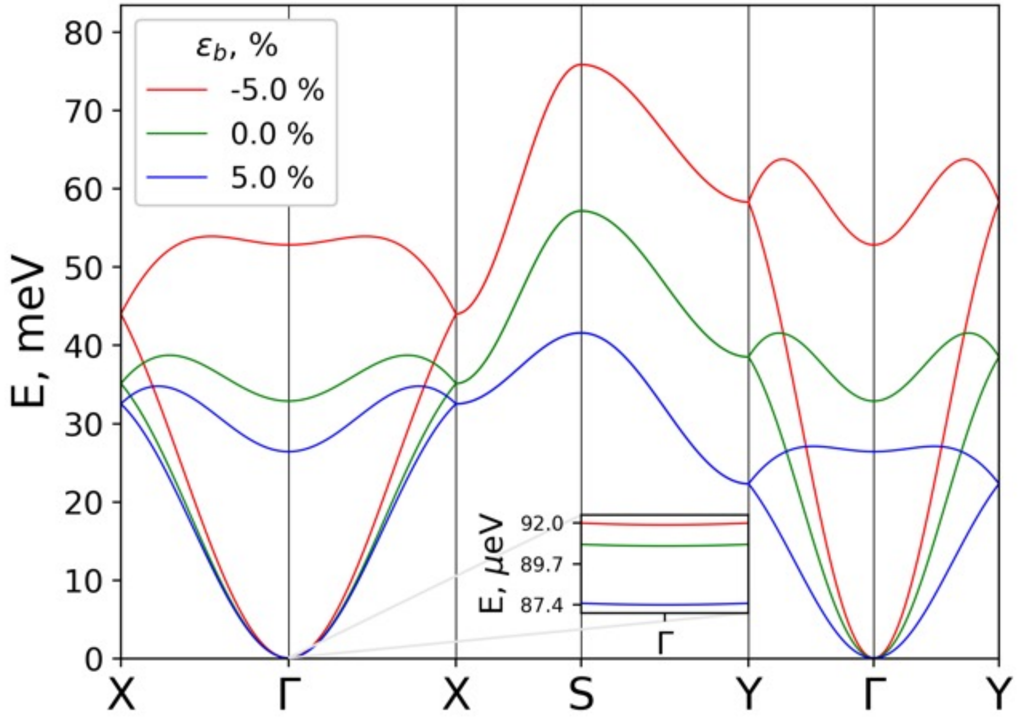


Andrey Rybakov

Uniaxial strain (a)



Uniaxial strain (b)



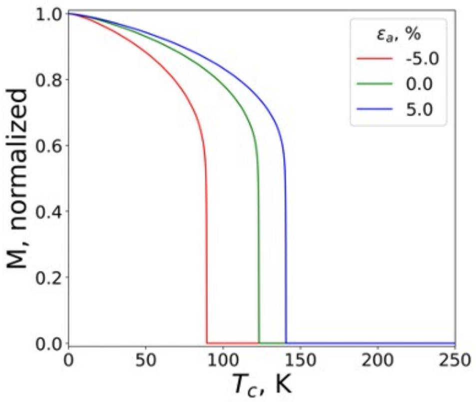
Strain-engineering of Curie temperature

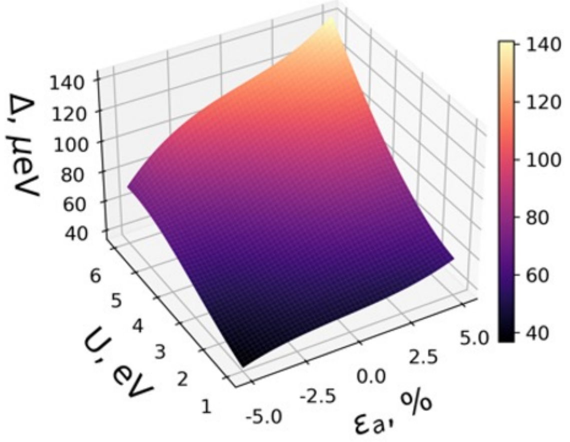
- From **magnon dispersion relation** through renormalized spin-wave theory

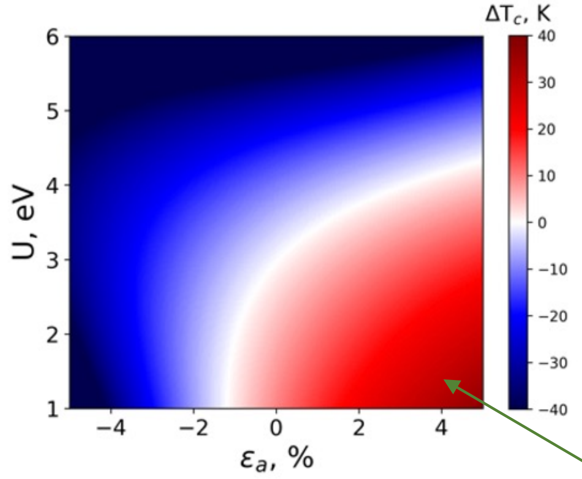
$$M = S - \frac{1}{2(2\pi)^2}\int_{BZ}\frac{d^2\vec{k}}{e^{\beta M E(\vec{k})/S}-1}$$

Lado *et al.*, *2D Materials* 2017, **4(3)**, 03002

- **Uniaxial strain (a)**







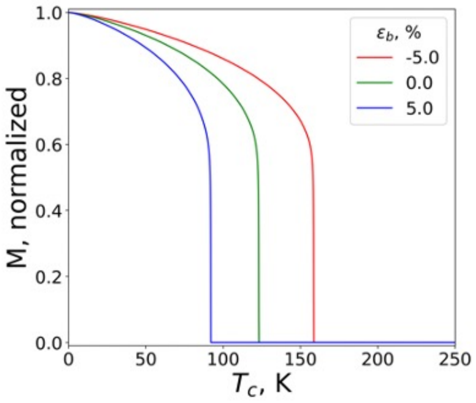
Fight t_{2g}-t_{2g} (J_1)

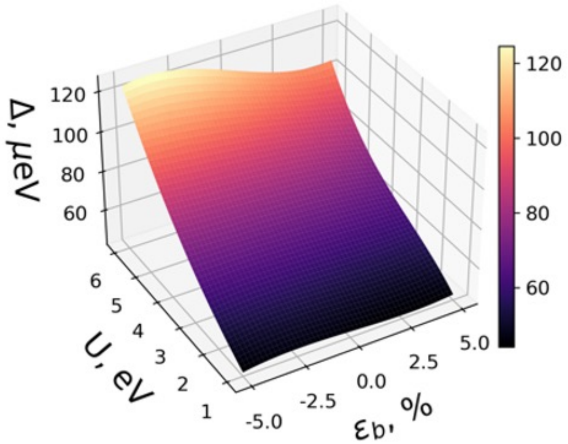
T_C (calc) = 122 K

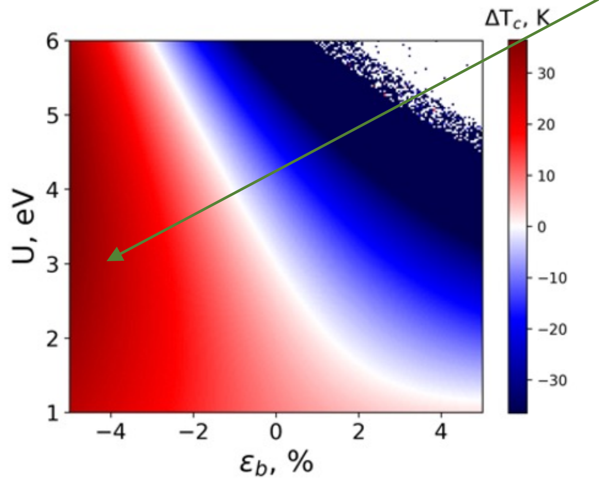
30% enhancement!

Fight e_g-e_g (J_3)

- **Uniaxial strain (b)**





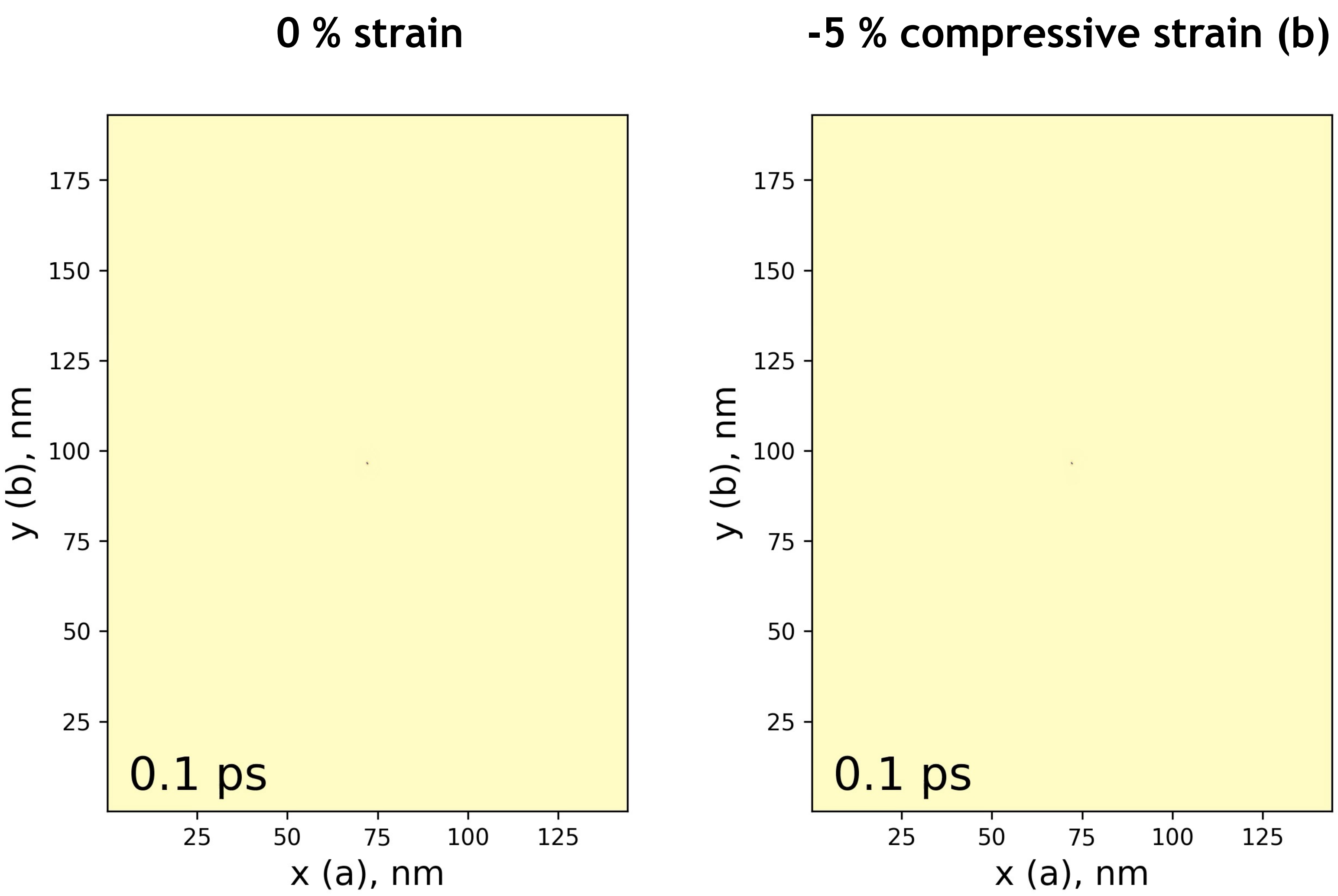


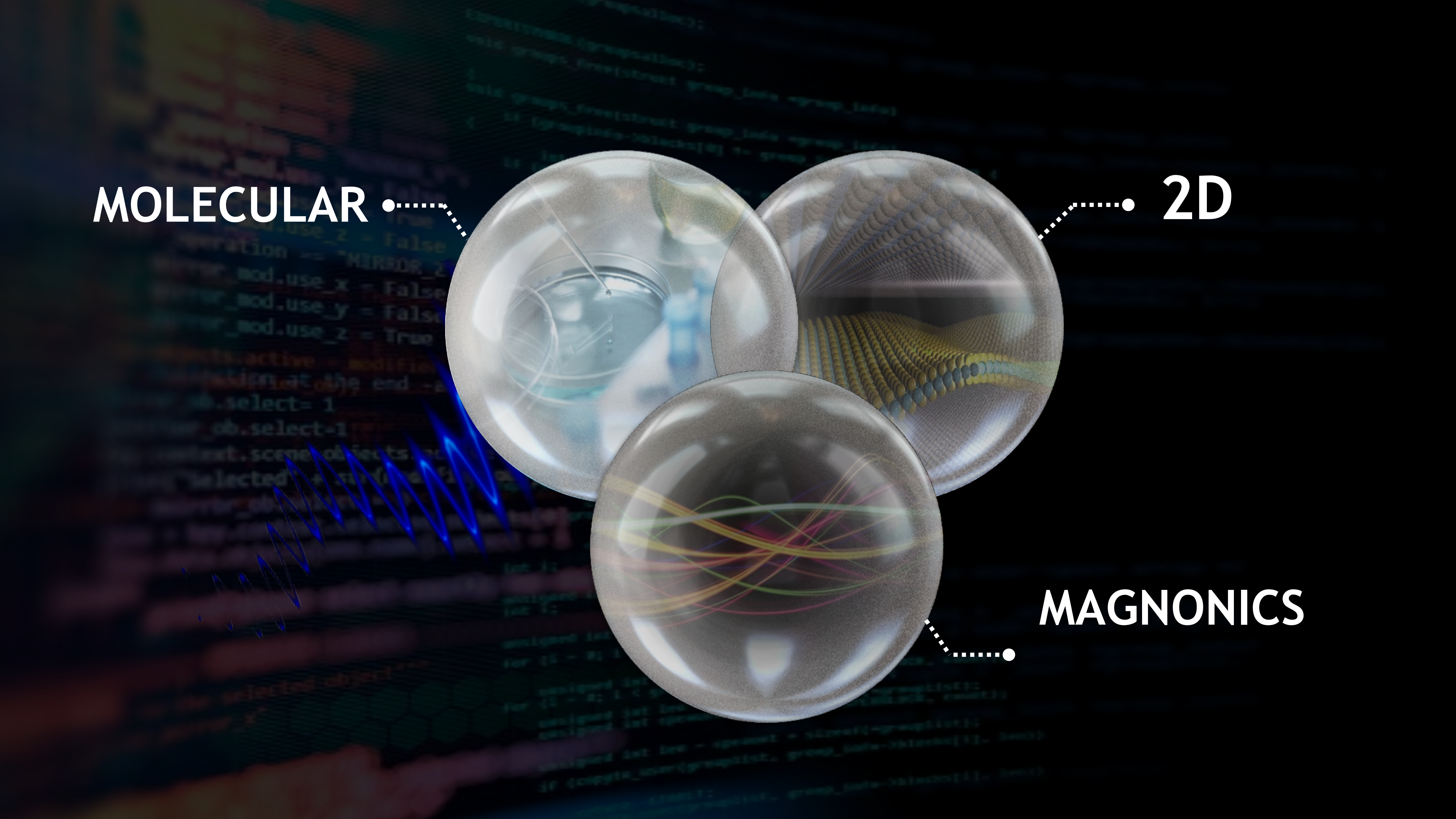
Atomistic spin dynamics simulations

- Real-time real-space SWs propagation (LLG)

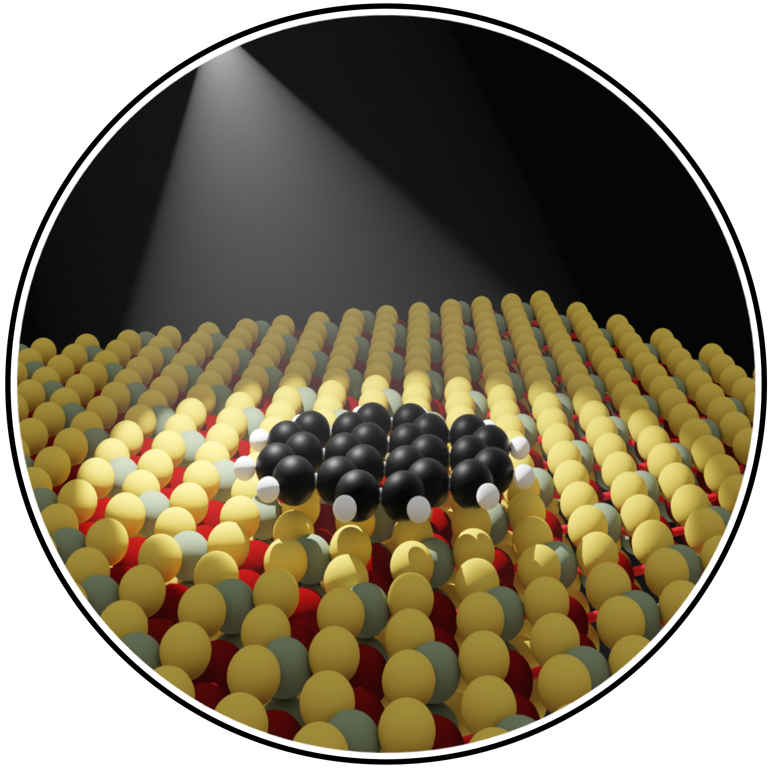
$$\frac{d\vec{m}}{dt} = -\gamma\mu_0\vec{m} \times \vec{H} + \alpha\vec{m} \times \frac{d\vec{m}}{dt}$$

- Gilbert damping: 0.01 (from CrI_3 literature)
- Ultrashort oscillating magnetic field (1 ps) to generate a SW.
- Tensile strain *a* reaches group velocity v_a = $4.2 \cdot 10^3$ m/s vs compressive strain *b* reaches group velocity v_b = $7.5 \cdot 10^3$ m/s

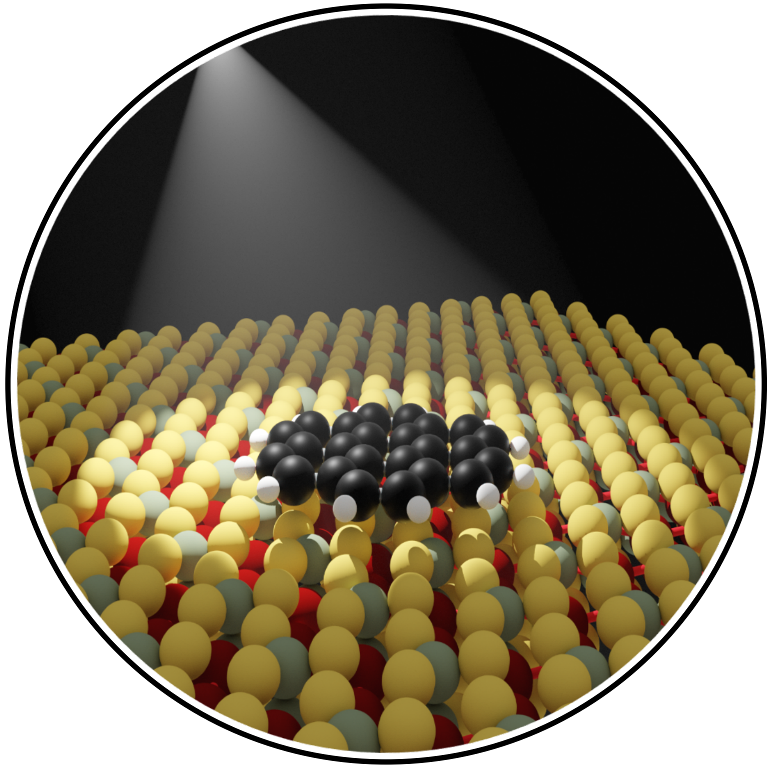


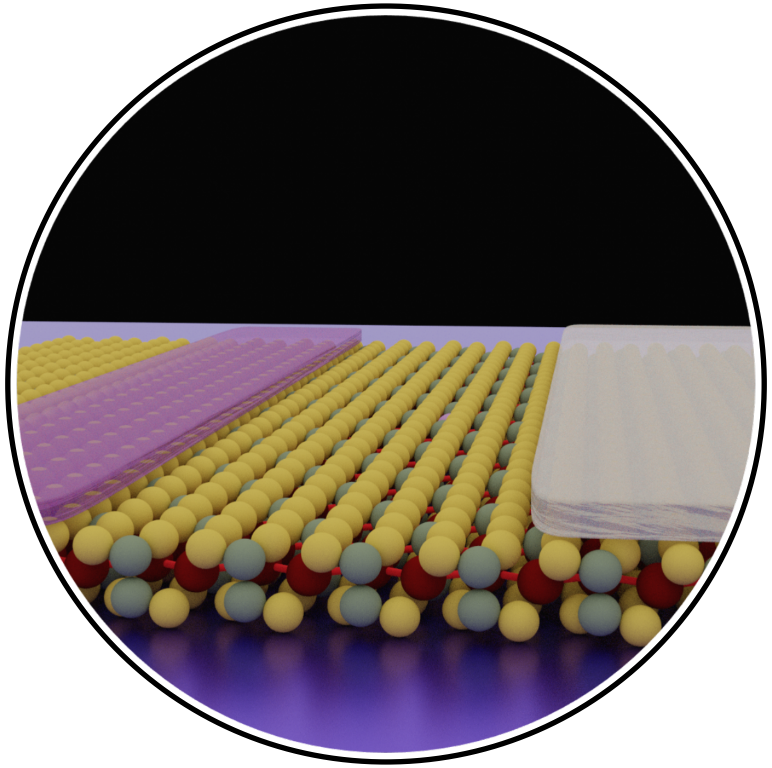

MOLECULAR
2D
MAGNONICS

Chemistry to excite,

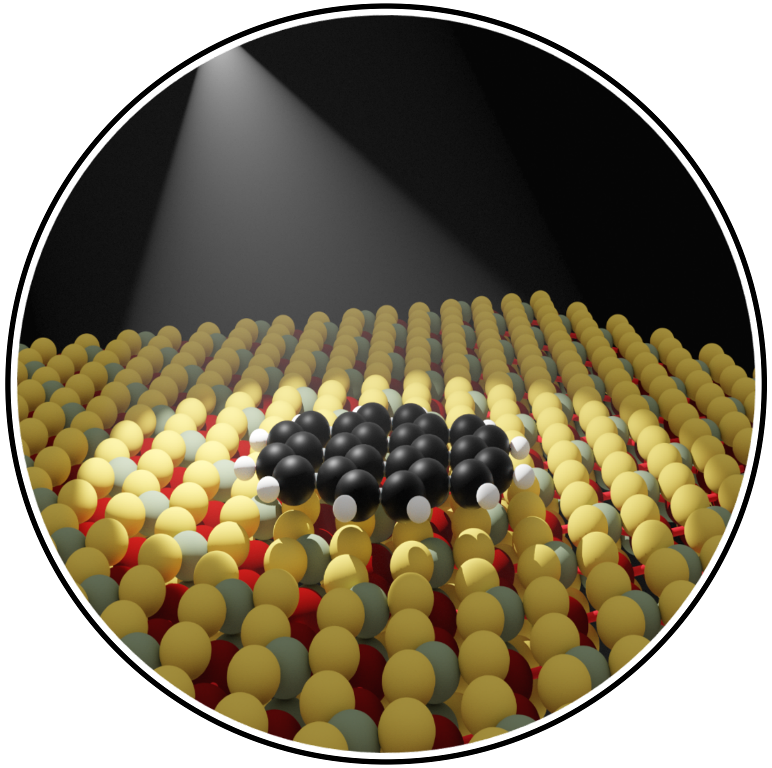
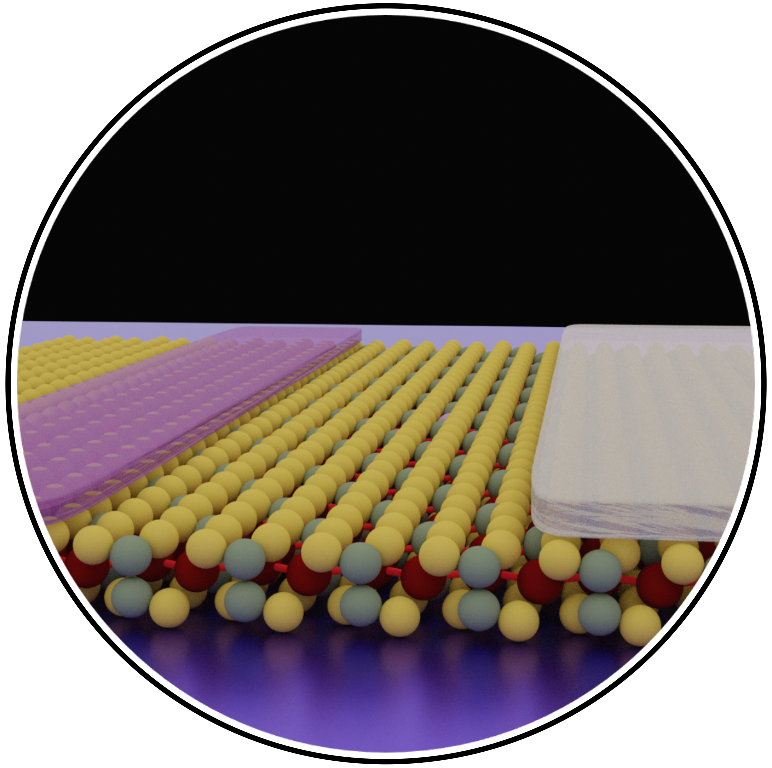
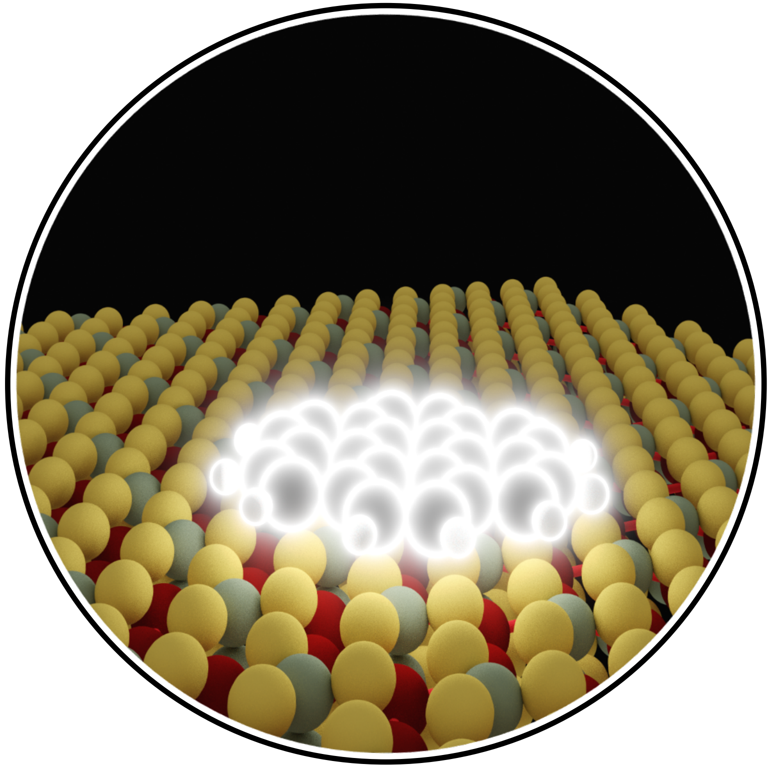


Chemistry to excite, modulate



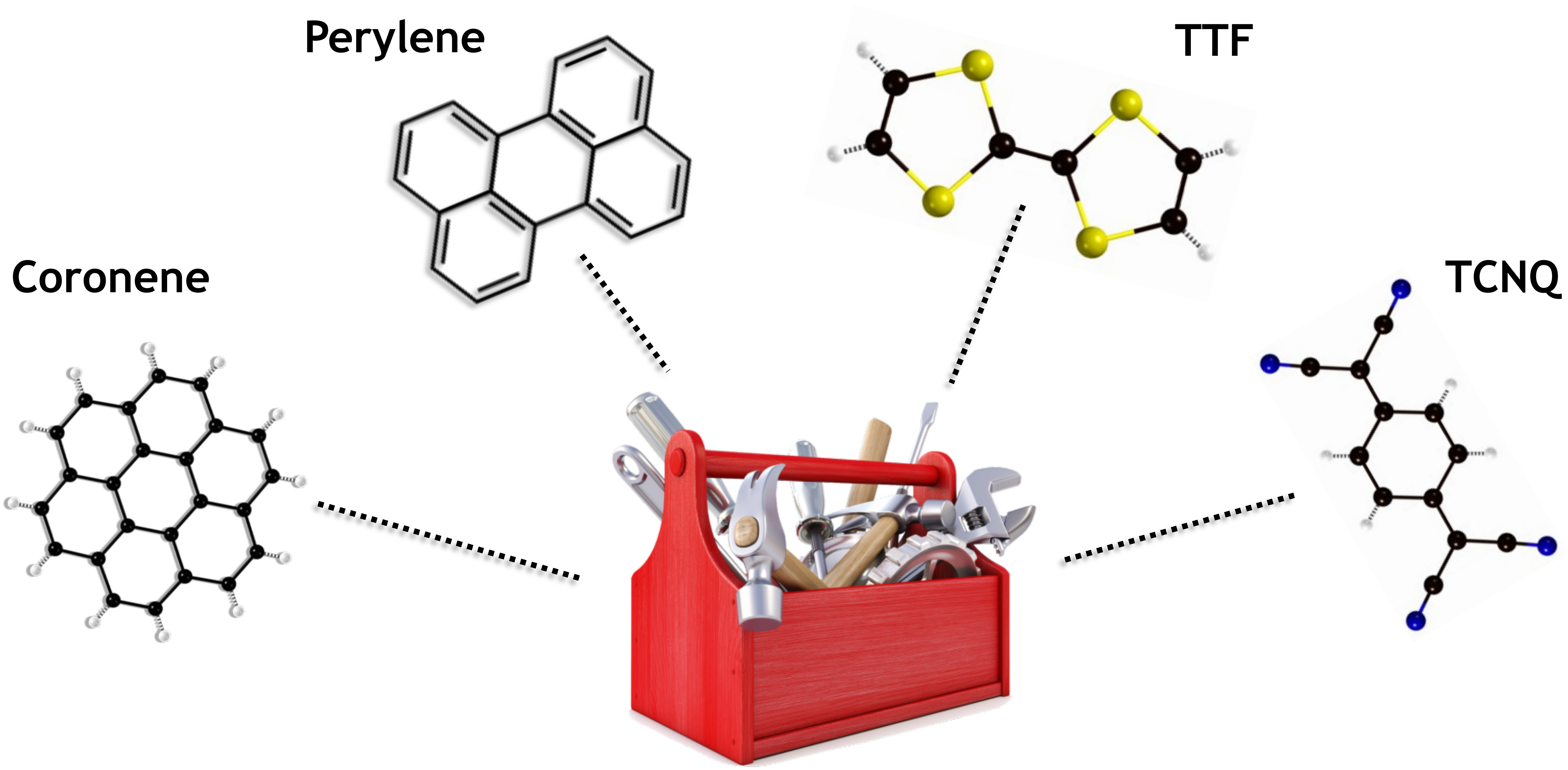


Chemistry to excite, modulate and detect

Spin waves in hybrid molecular/2D magnetic materials!

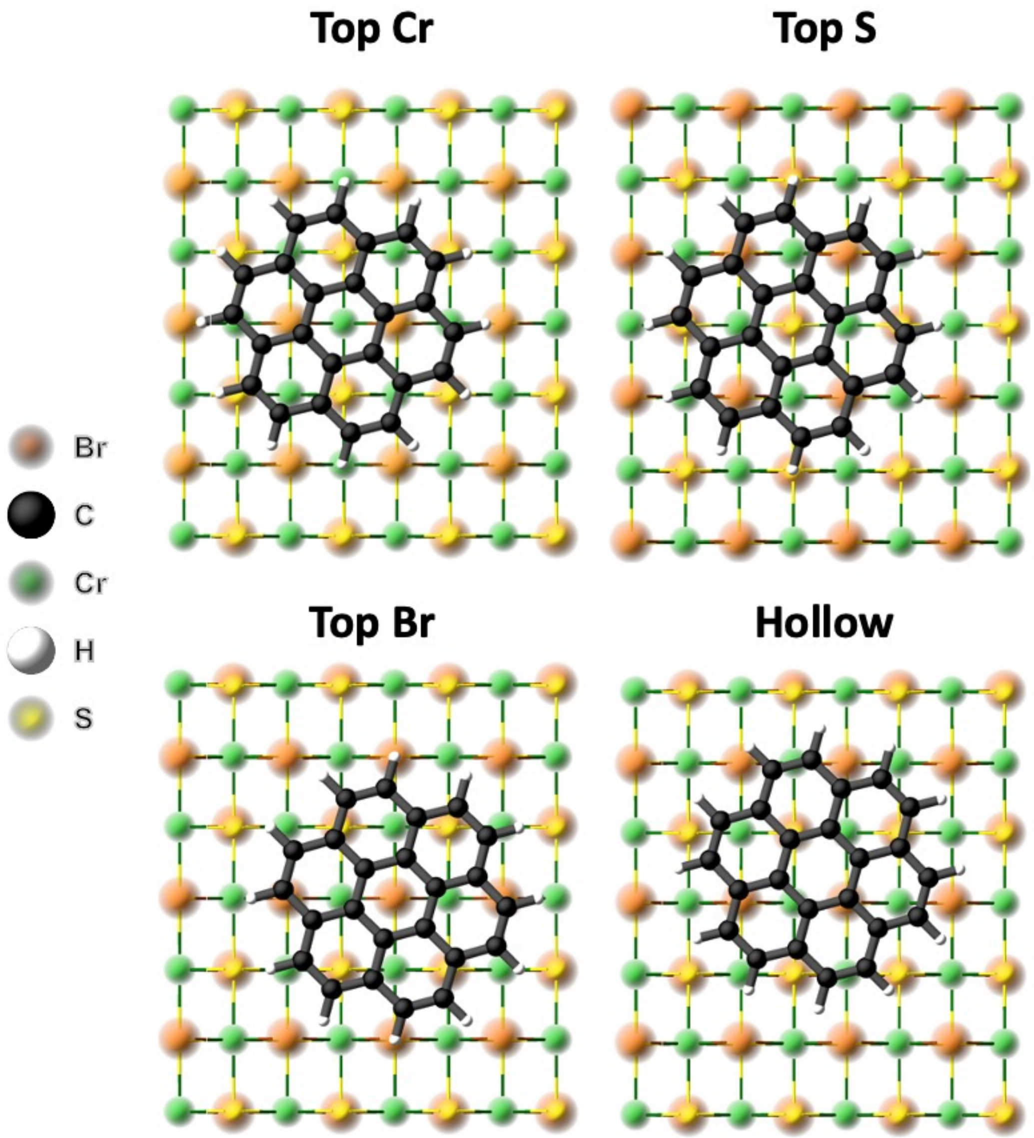
Organic molecules toolbox



DFT structural optimization (PBE + vdW)

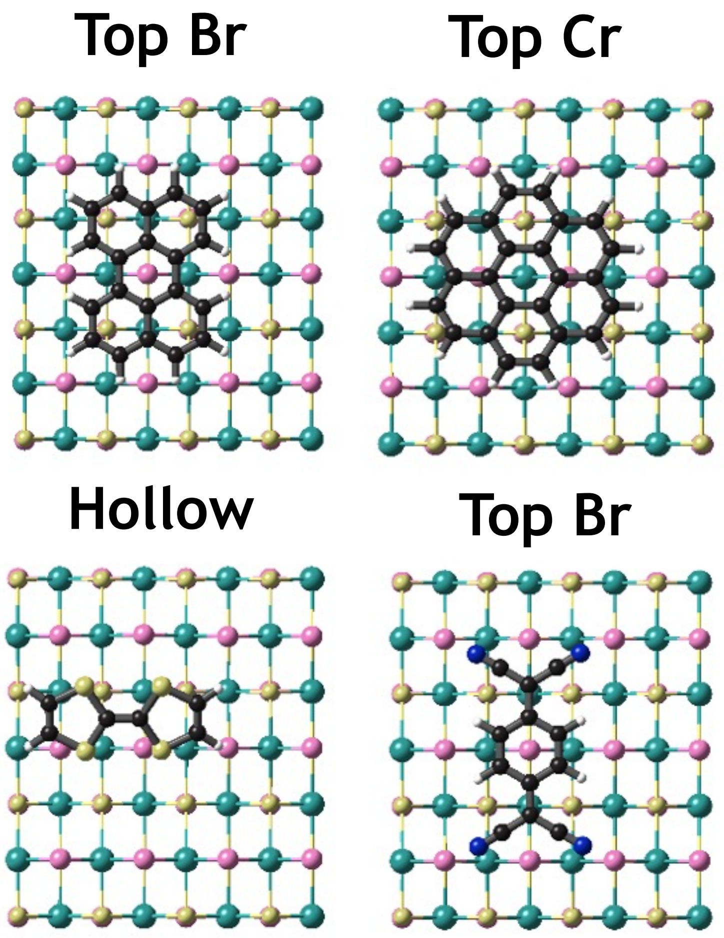
- Different adsorption sites
- Different orientation of the molecules (rotation ϕ axis)
- 4x4 supercells (~12 Å between molecules)
- Dispersion corrections
- Calculation of E_{ads}

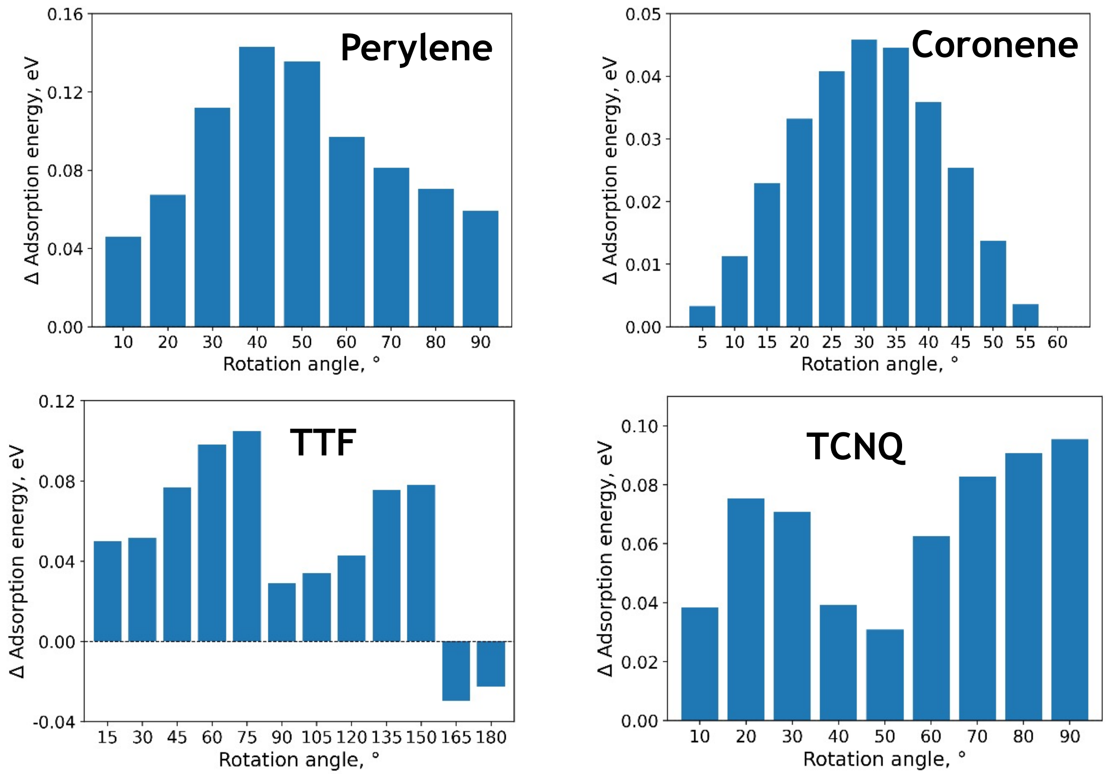
$$E_{ads} = E_{hybrid} - E_{2D} - E_{mol}$$



DFT structural optimization (PBE + vdW)

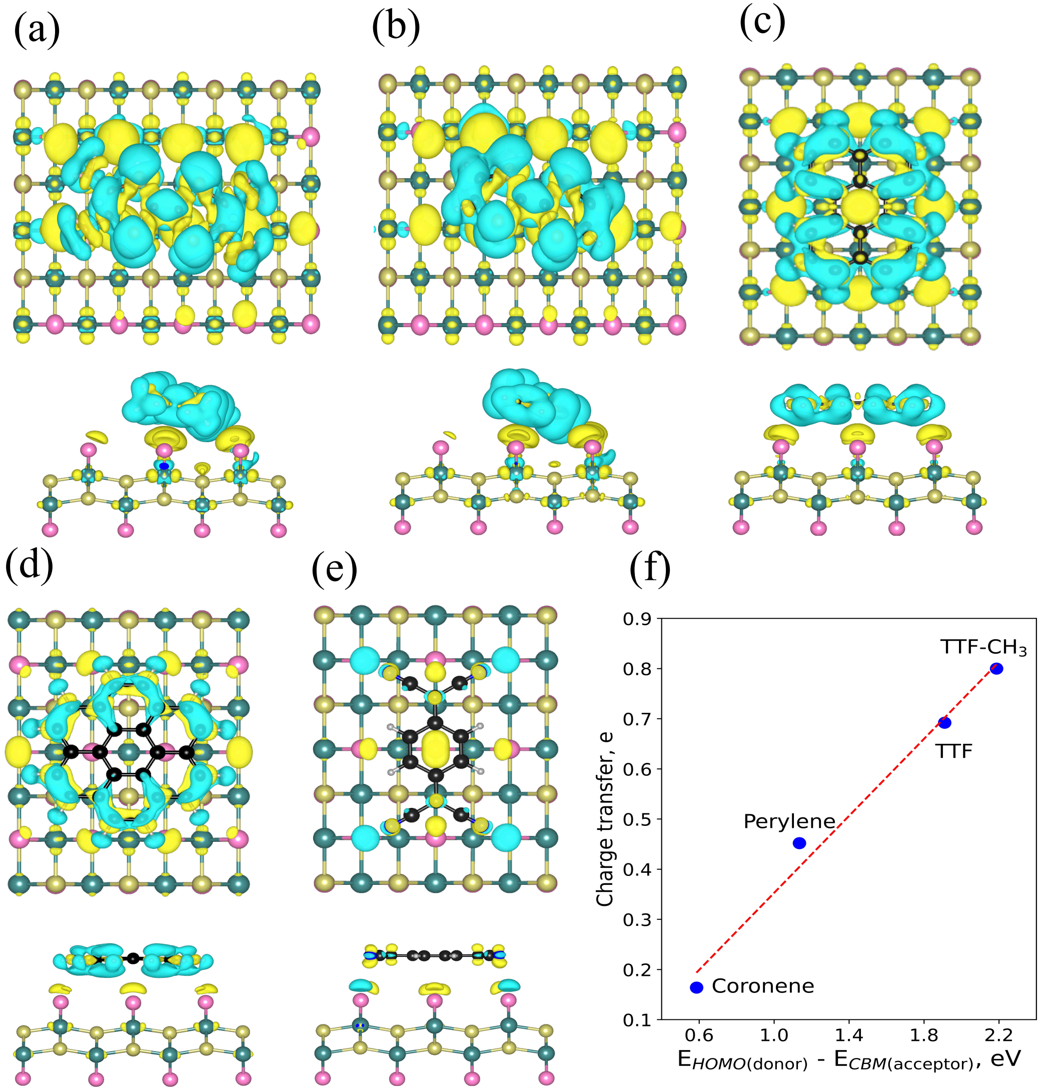
- Full optimization for each molecular orientation





Molecules	Site	E_{ads}, eV	h, Å	e^- transfer
$TTF\text{-}CH_3$	Hollow-top	-2.23	1.32	0.80
TTF	Hollow-top	-1.55	2.14	0.69
Perylene	Br-top	-1.35	2.76	0.45
Coronene	Cr-top	-1.18	2.97	0.16
TCNQ	Br-top	-0.65	3.15	-0.03

Towards molecular controlled magnonics



Ruiz, Rivero-Carracedo, Rybakov, Dey and Baldoví*
Nanoscale Adv. 2024, DOI: 10.1039/D4NA00230J

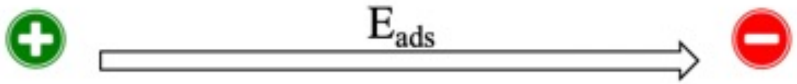
	CrSBr	CrSBr_TTF	CrSBr_Per	CrSBr_Cor	CrSBr_TCNQ
Cr-S-Cr (δ) / (°)	157.591	162.712	161.782	160.271	159.959

Distorted CrSBr

Exchange	CrSBr	$TTF-CH_3$	TTF	Perylene	Coronene	TCNQ
J_1 (meV)	3.11	3.14	3.14	3.14	3.14	3.15
J_2 (meV)	3.87	3.84	3.84	3.85	3.84	3.84
J_3 (meV)	2.76	1.90	1.91	1.96	1.98	2.08

CrSBr + Molecules

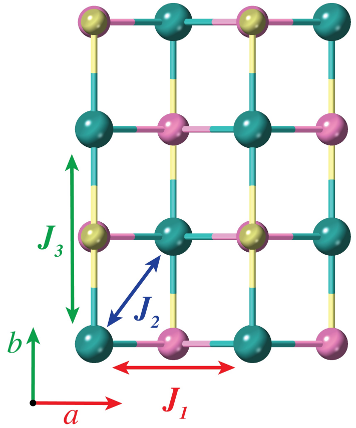
Exchange	CrSBr	$TTF-CH_3$	TTF	Perylene	Coronene	TCNQ
J_1 (meV)	3.11	3.33	3.30	3.23	3.17	3.15
J_2 (meV)	3.87	3.90	3.89	3.87	3.85	3.84
J_3 (meV)	2.76	3.17	2.92	2.43	2.06	2.08



Towards molecular controlled magnonics

Gonzalo Rivero

Alberto M. Ruiz



Ruiz, Rivero, Rybakov, Dey and Baldoví* *Nanoscale Adv.* 2024, DOI: 10.1039/D4NA00230J

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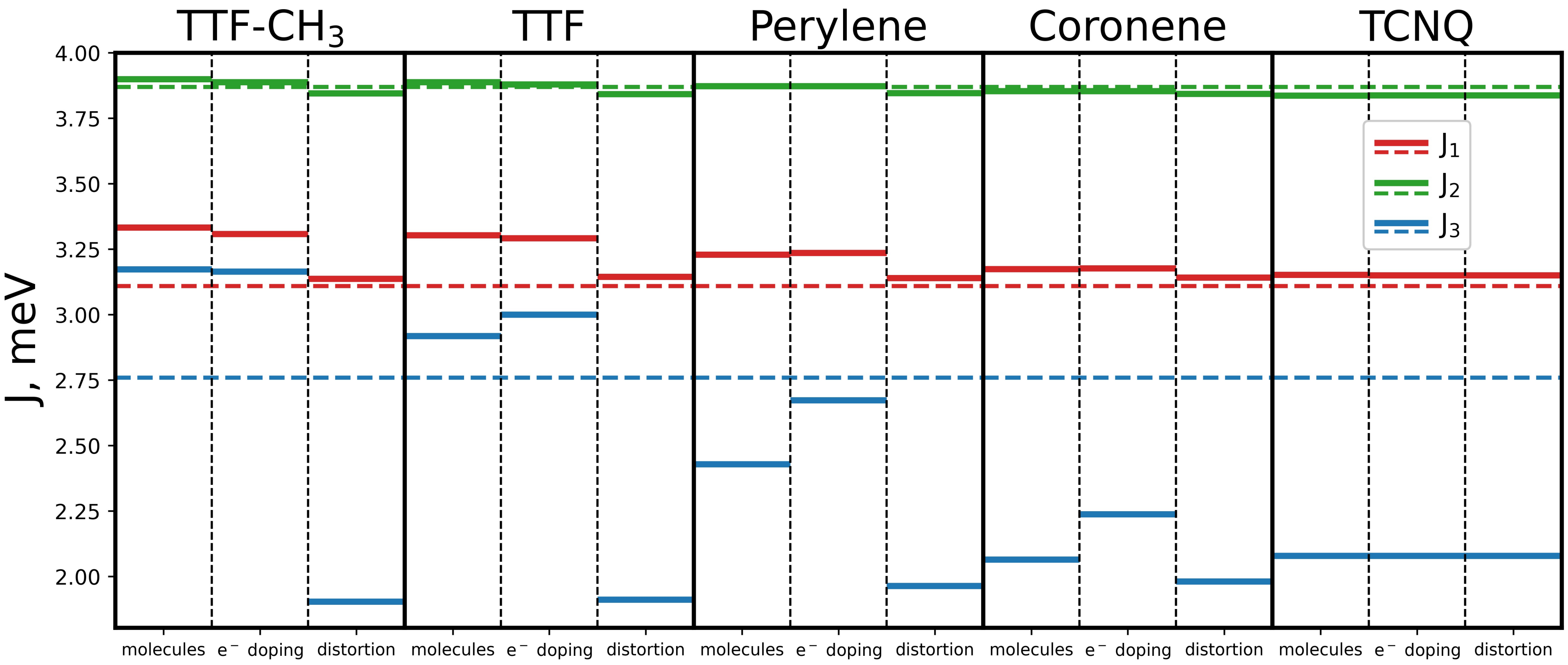
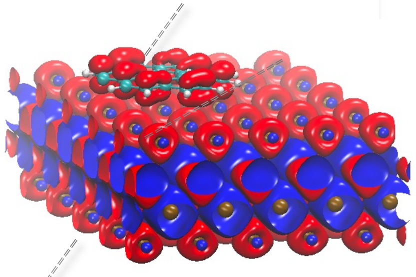


Towards molecular controlled magnonics

Gonzalo Rivero

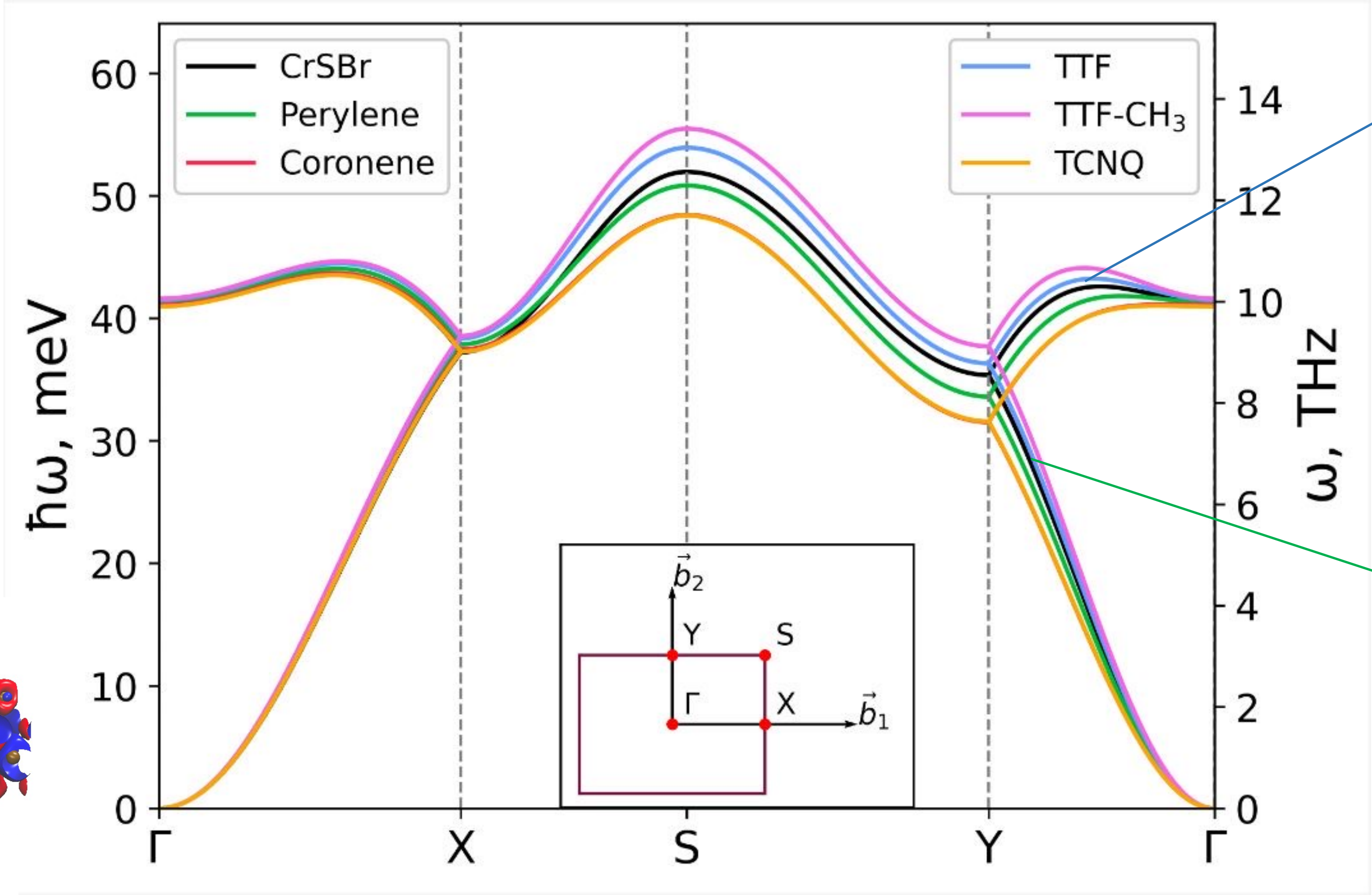
Alberto M. Ruiz

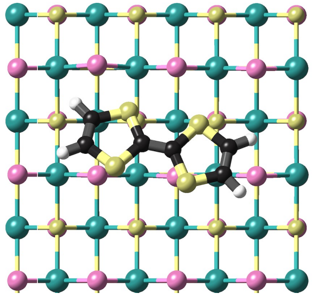
$0.15\ \mu_B$



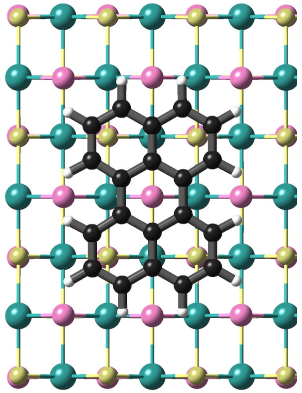
ΔM_{Cr} = -0.21%

Ruiz, Rivero-Carracedo, Rybakov, Dey and Baldoví*
Nanoscale Adv. **2024**, DOI: 10.1039/D4NA00230J





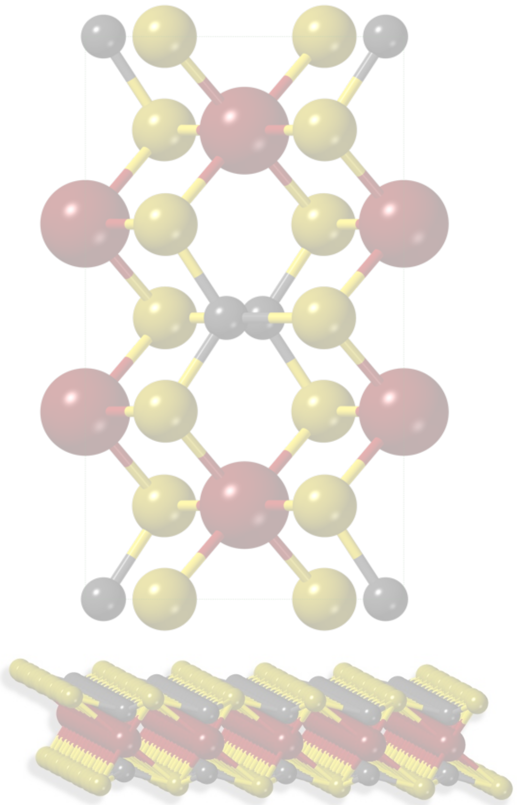
E_{ads} = -1.55 eV



E_{ads} = -1.35 eV

2D van der Waals magnetic materials

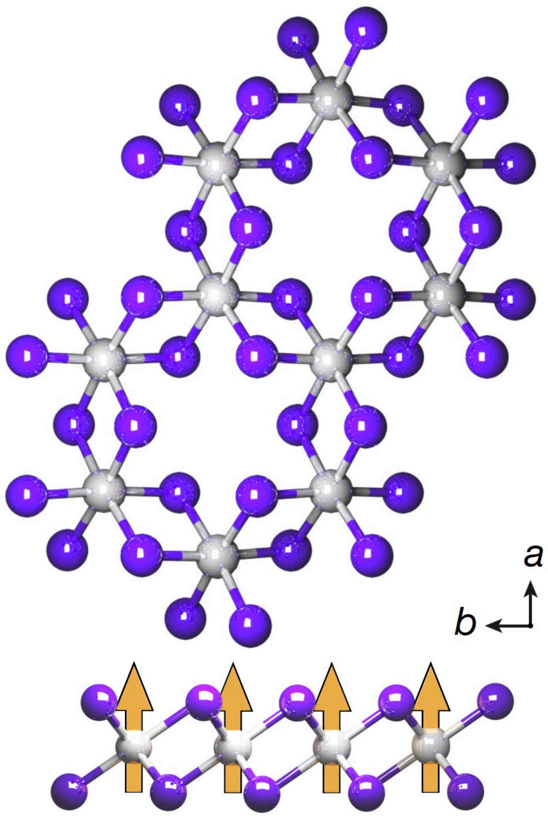
MPS_3



$T_N = 70 - 150$ K

Lee *et al.*, *Nano Lett.* 2016, **16(12)**, 7433–7438

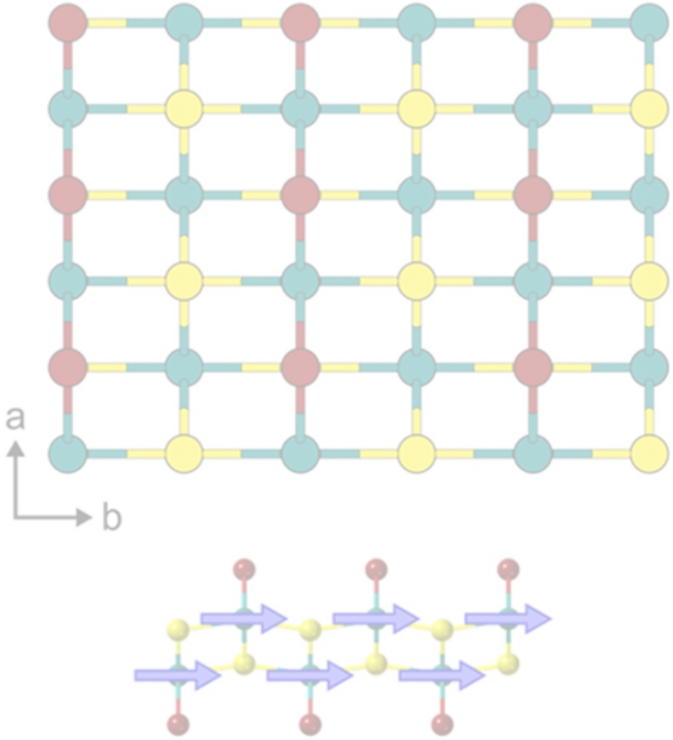
CrI_3



$T_C = 45$ K

Huang *et al.*, *Nature* 2017, **546**, 270–273

CrSBr

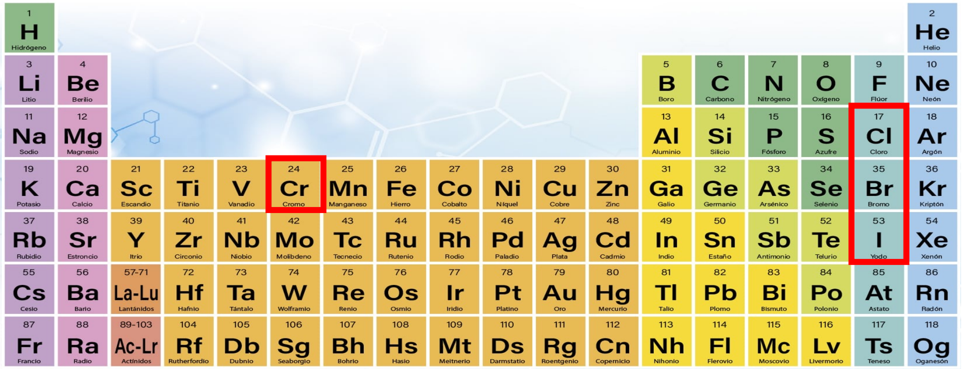


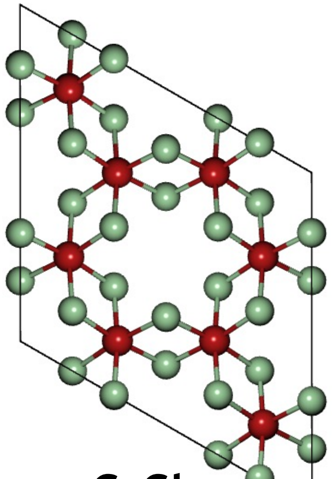
$T_C = 146$ K

Lee *et al.*, *Nano Lett.* 2021, **21(8)**, 3511

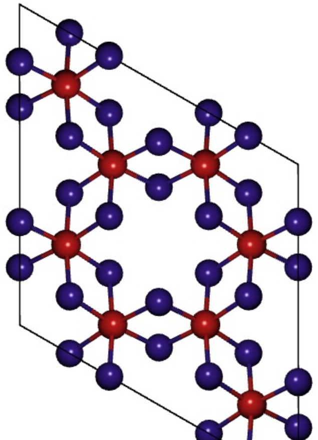
Topological magnons: recent research on CrX_3

Esteras, Baldoví*,
Materials Today Electronics, **2023**, *6*, 100072





$CrCl_3$



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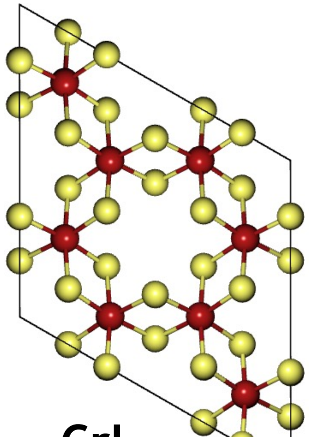
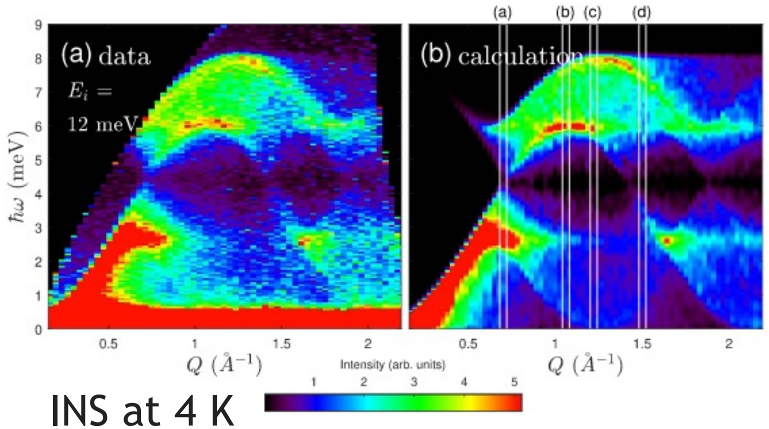


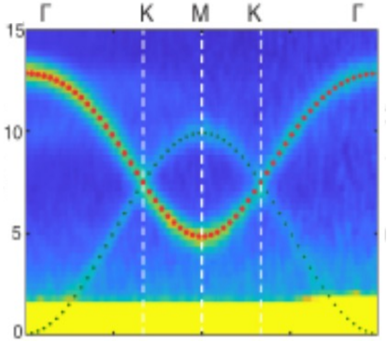
CrI_3

$CrCl_3$

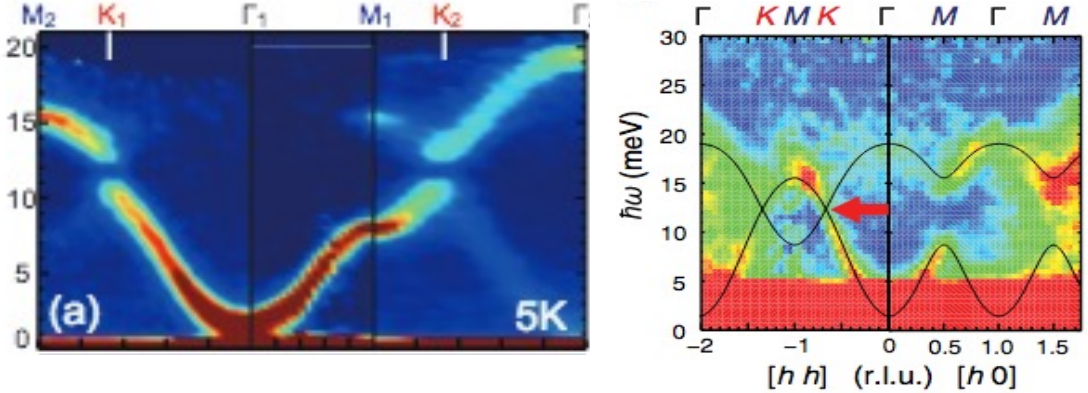


INS at 4 K

$CrBr_3$



CrI_3



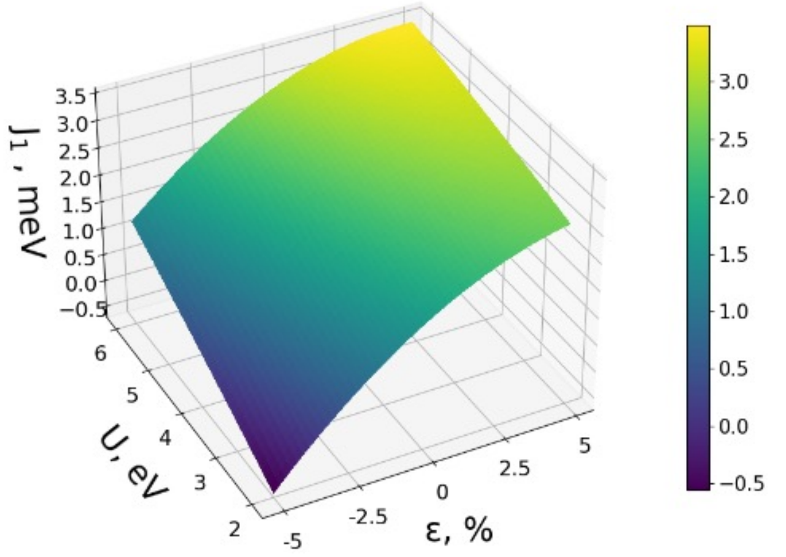
Chen, L. (2021). *2D Materials*, *9*(1), 015006.
Nikitin, et al. (2022). *PRL*, *129*(12), 127201.
Chen, L et al. (2021). *PRX*, *11*(3), 031047.

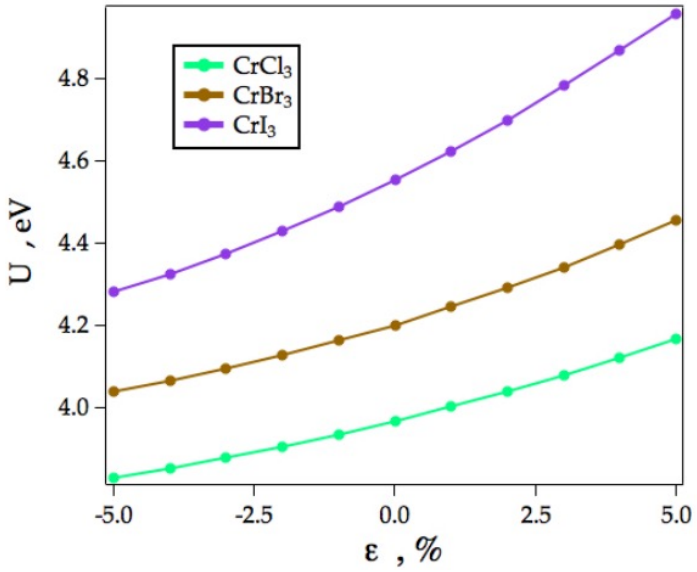
Magnons in strained CrX_3: strain and scf U

Esteras, Baldoví*, *Materials Today Electronics*, **2023**, *6*, 100072

- **First principles**: Zero free parameter methodology

- **Biaxial strain**: scf Hubbard U for each structure (Linear response DFPT)





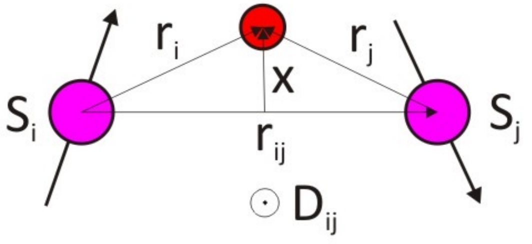
Magnons in strained CrX_3: strain and scf U

Esteras, Baldoví*, *Materials Today Electronics*, **2023**, *6*, 100072

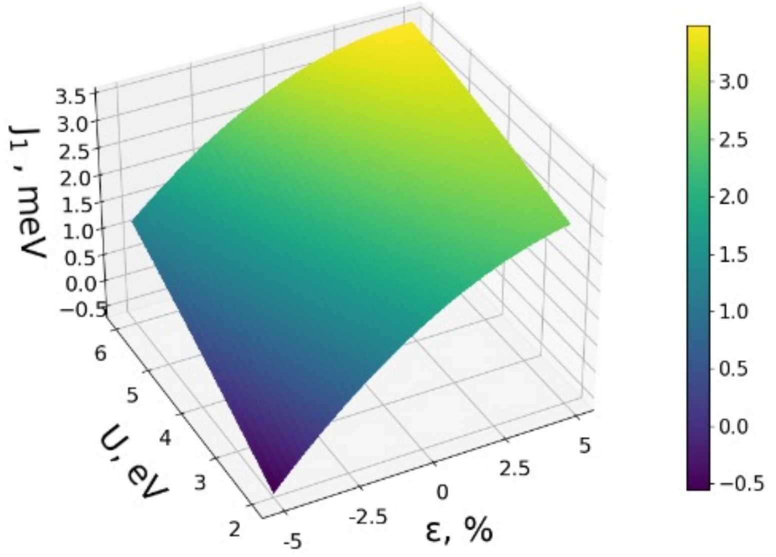
- **First principles**: Zero free parameter methodology

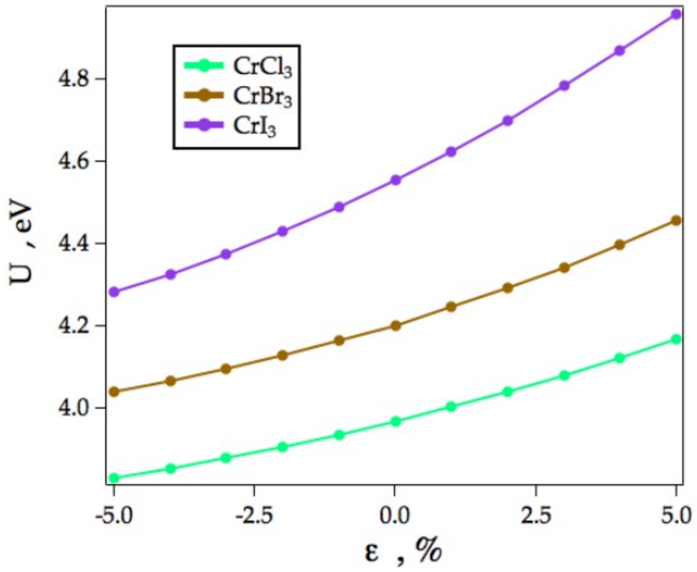
- **Biaxial strain**: scf Hubbard U for each structure (Linear response DFPT)

- In absence of DMI (antisymmetric exchange), **there is no gap**

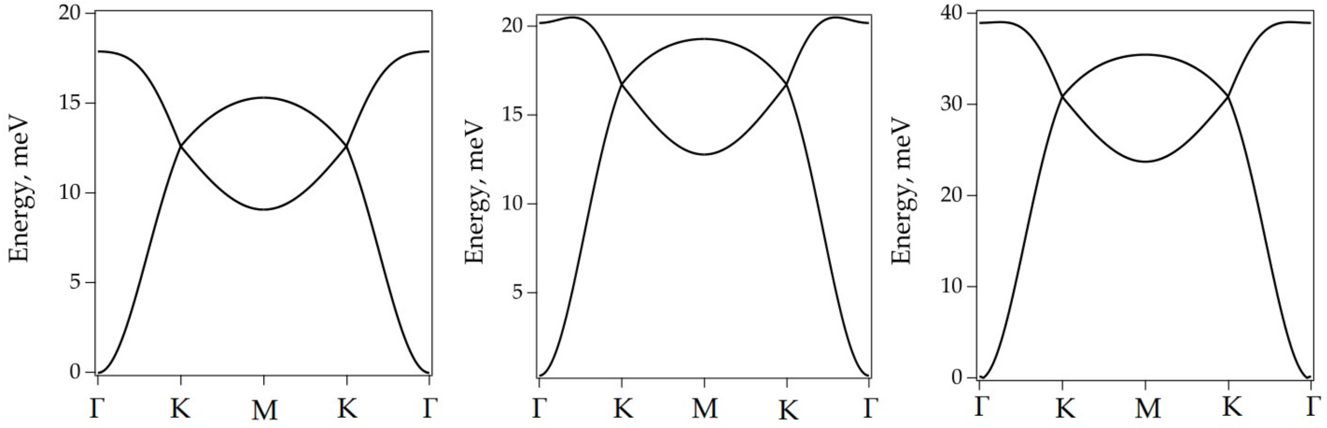


$$H_{spin} = -\sum_i A_i(\vec{S}_i \cdot \vec{e}_i)^2 - \sum_{i \neq j} [\vec{S}_i J_{ij} \vec{S}_j + \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)]$$

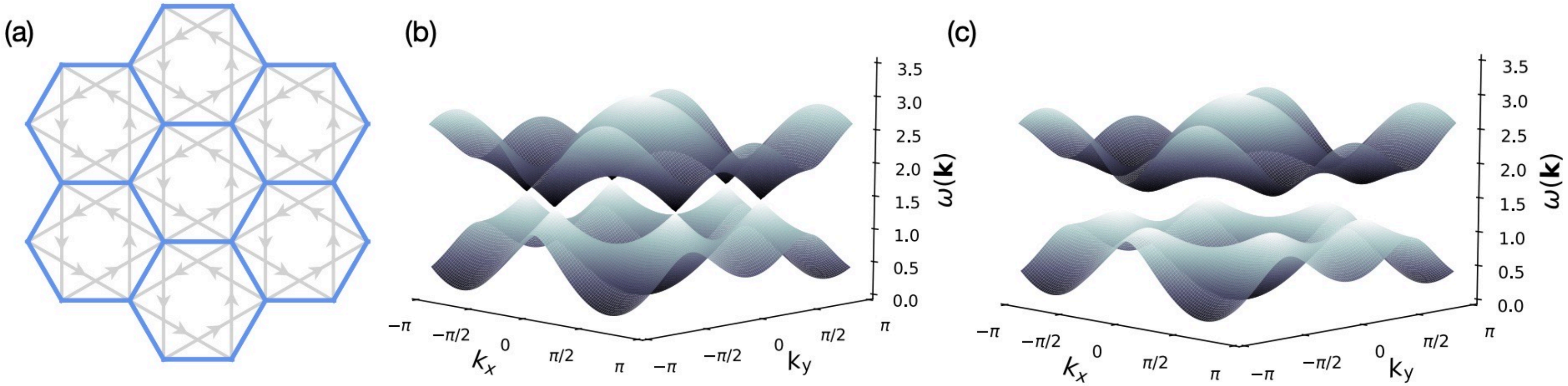




$CrCl_3$ $CrBr_3$ CrI_3

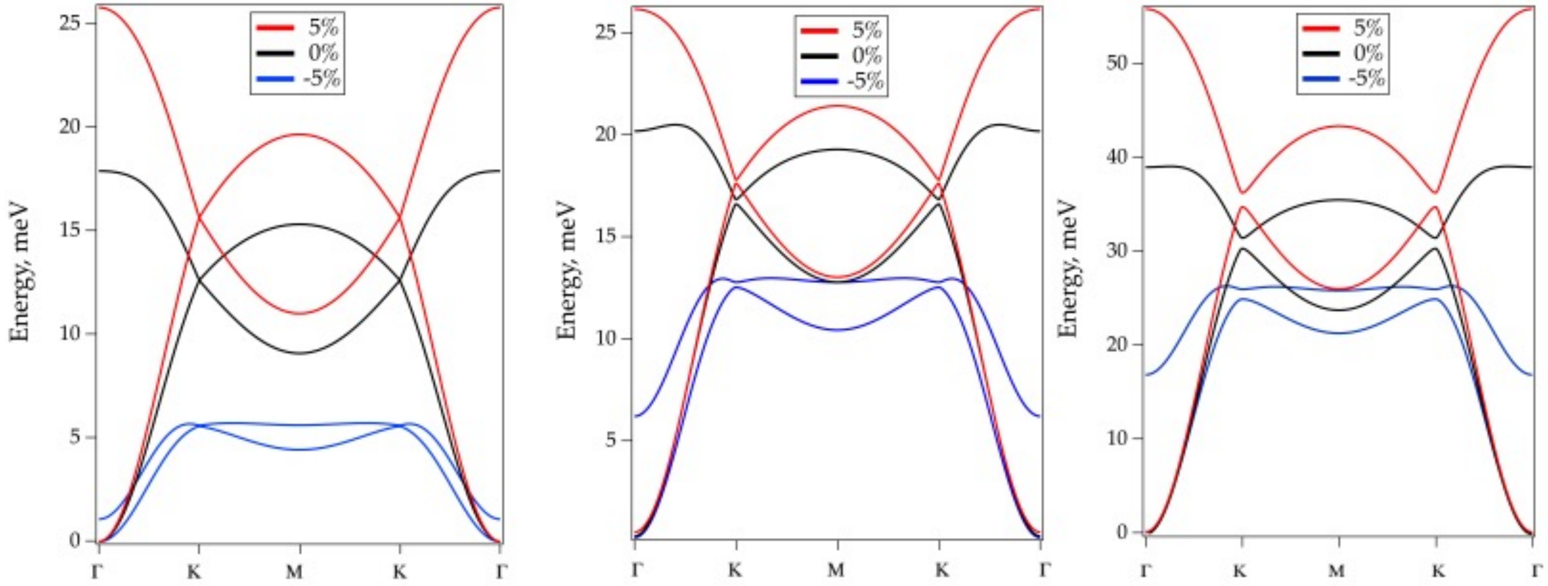


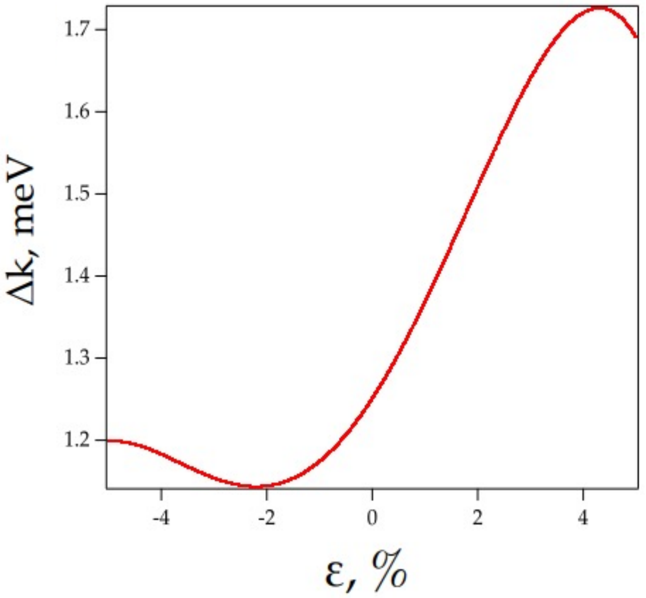
Tuning the magnon gap with strain



- **DMI gaps out the Dirac points** in the magnon dispersion of CrI_3 monolayer
- Strain engineering of DMI: **enhancement of magnon gap** (by 40 %)

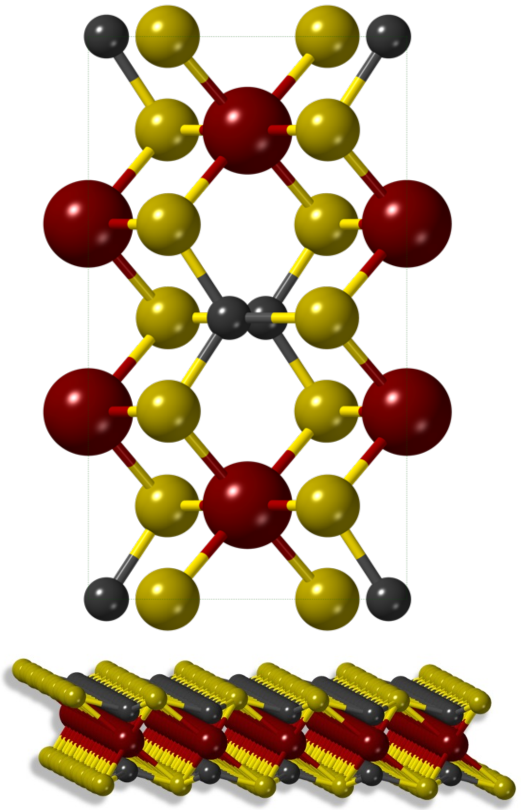
$CrCl_3$ $CrBr_3$ CrI_3





2D van der Waals magnetic materials

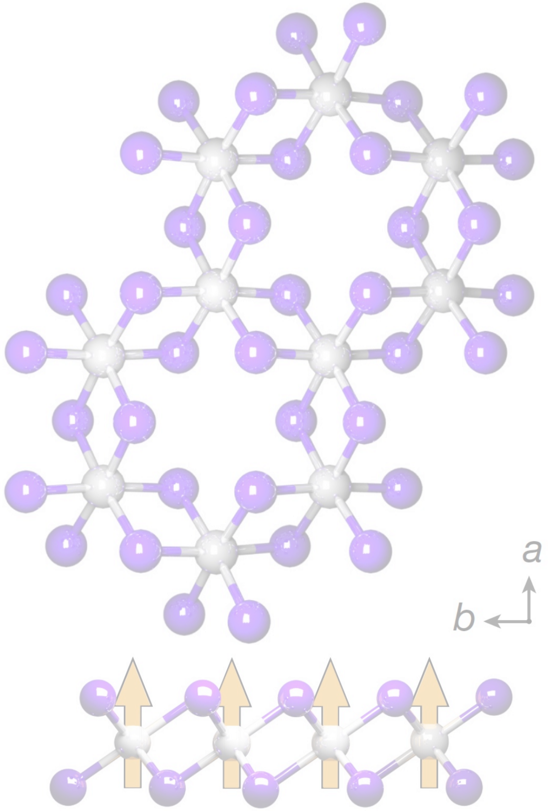
MPS_3



T_N = 70 - 150 K

Lee *et al.*, *Nano Lett.* 2016, **16(12)**, 7433–7438

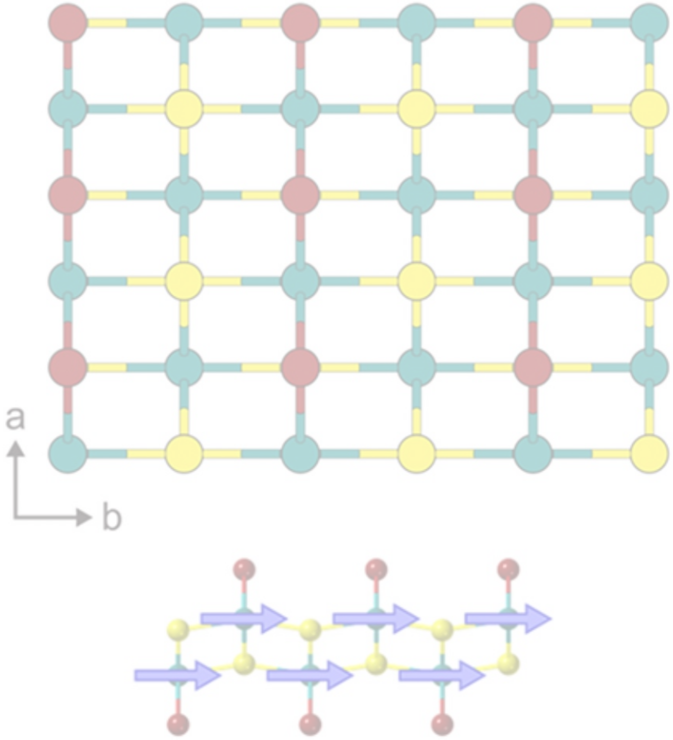
CrI_3



T_C = 45 K

Huang *et al.*, *Nature* 2017, **546**, 270–273

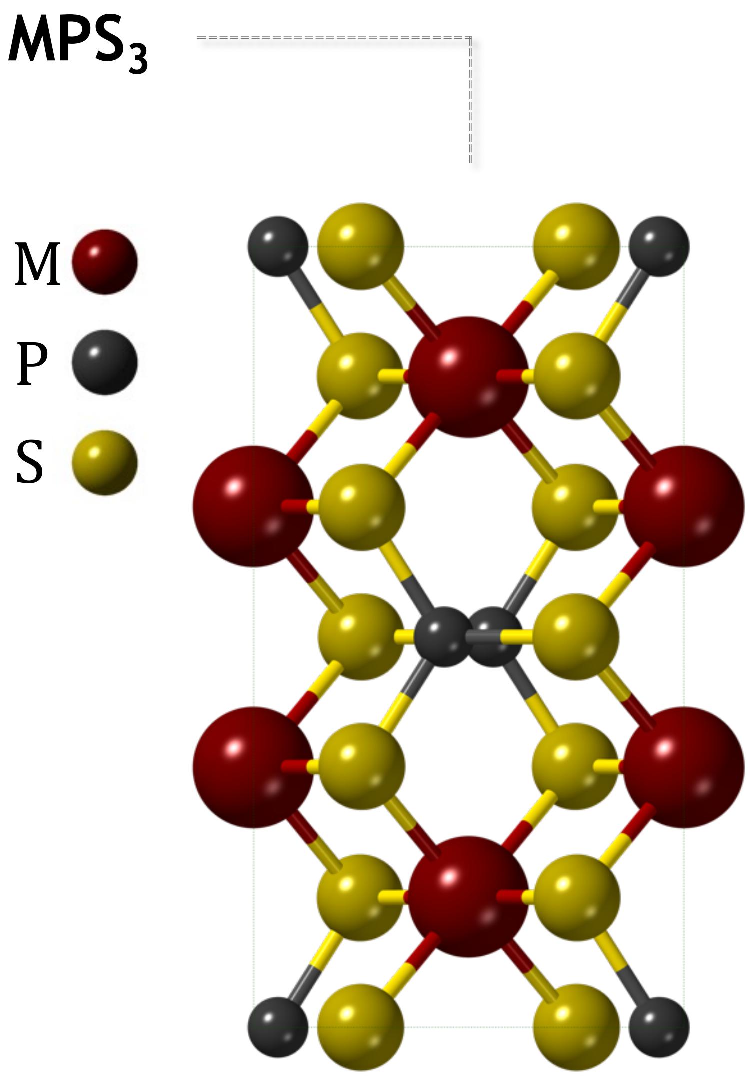
CrSBr

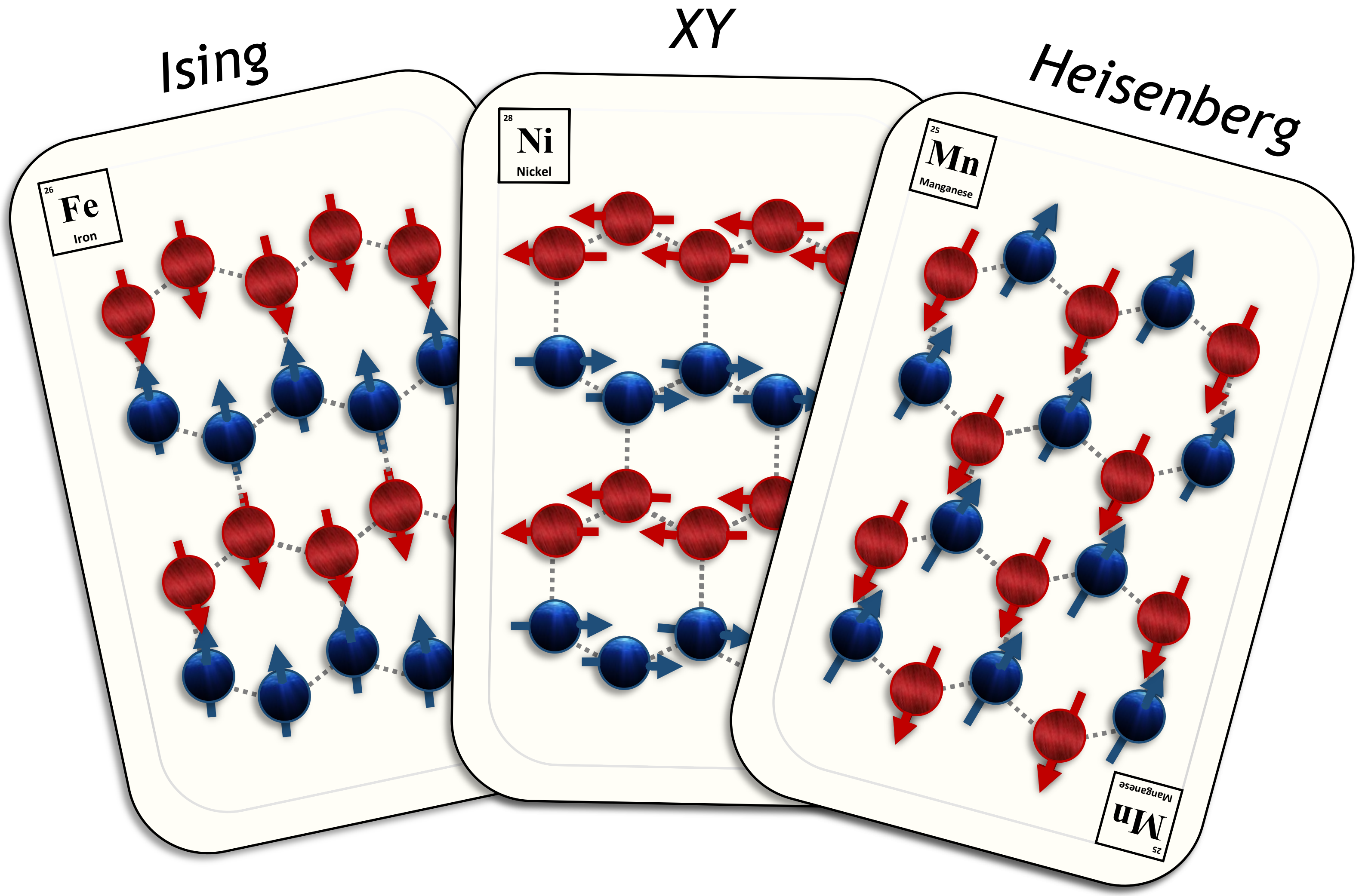


T_C = 146 K

Lee *et al.*, *Nano Lett.* 2021, **21(8)**, 3511

2D antiferromagnetic materials





PRX **2019**, 9, 011026; *PRB* **2020**, 101, 064416; *PRB* **2021**, 103, 064431; *Nano Lett.* **2021**, 21, 5045; *PRL* **2021**, 127, 097401; *PRB* **2021**, 104, L100404

ARPES simulation of $FePS_3$

Valence band electronic structure of the van der Waals antiferromagnet $FePS_3$

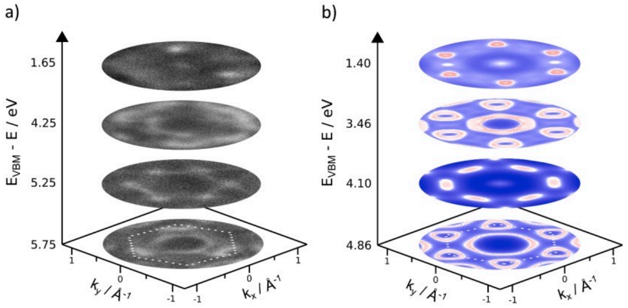
Jonah Elias Nitschke [a,1], Dorye L. Esteras [b,1], Michael Gutnikov [a], Karl Schiller [a], Samuel Mañas-Valero [b], Eugenio Coronado [b], Matija Stupar [a], Giovanni Zamborlini [a], Stefano Ponzoni [a], José J. Baldoví [b,*], Mirko Cinchetti [a,*]

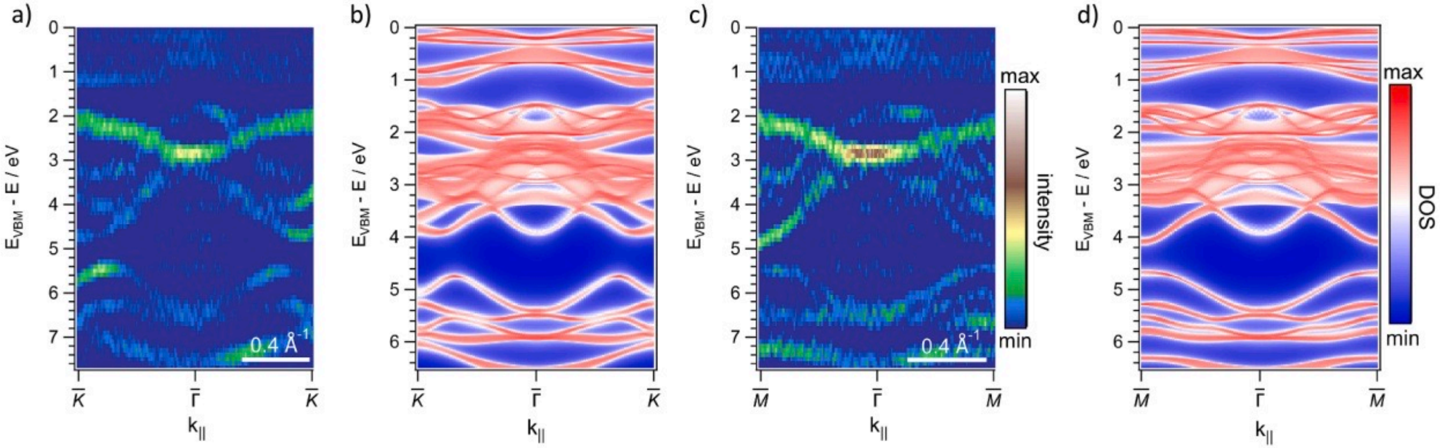
Materials Today Electronics, **6**, 100061 (2023)

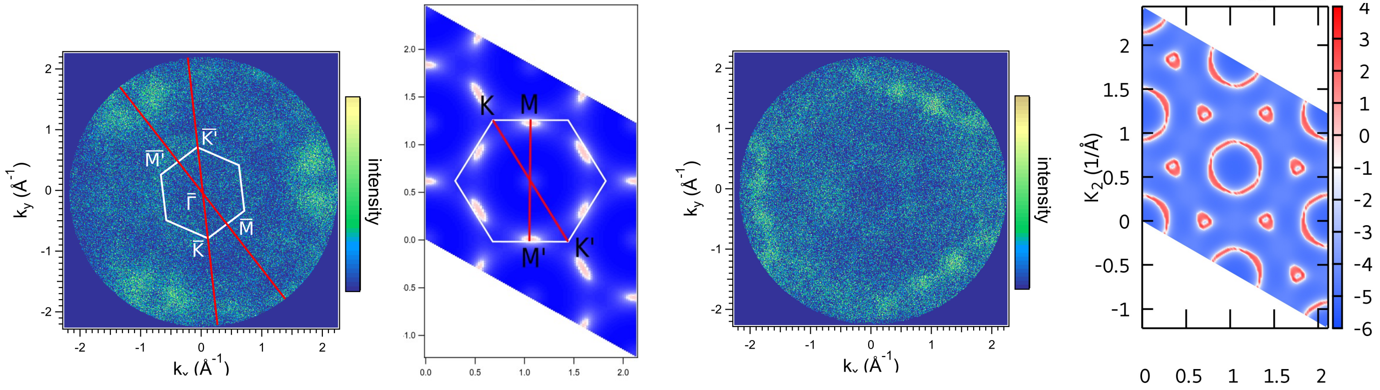
Jonah Nitschke

Mirko Cinchetti
TU Dortmund

- Exp. vs theo. momentum maps for various energies
- Phenomenological Hubbard U = 1.9 eV that matches energy gap between 1st and 2nd group of bands
- Rich band structure with multiple overlapping bands (agreement with DFT+U)
- Bands closer to the VBM dominated by Fe 3d orbitals







Spontaneous magnetostriction in MPS_3

nature communications

Article

https://doi.org/10.1038/s41467-023-44180-4

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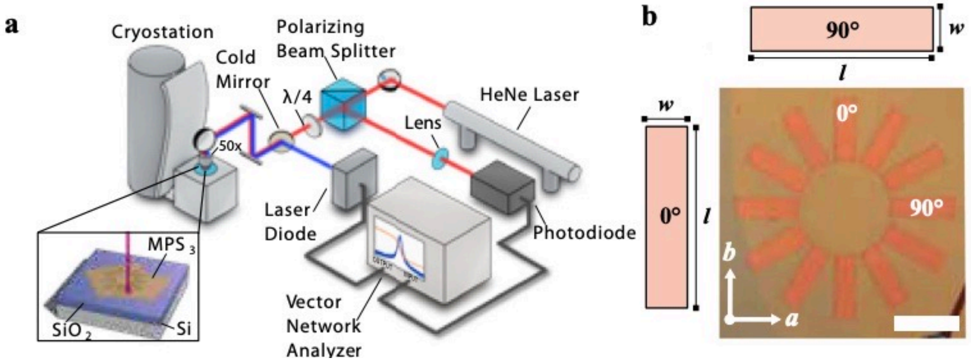
Magnetic order in 2D antiferromagnets revealed by spontaneous anisotropic magnetostriction



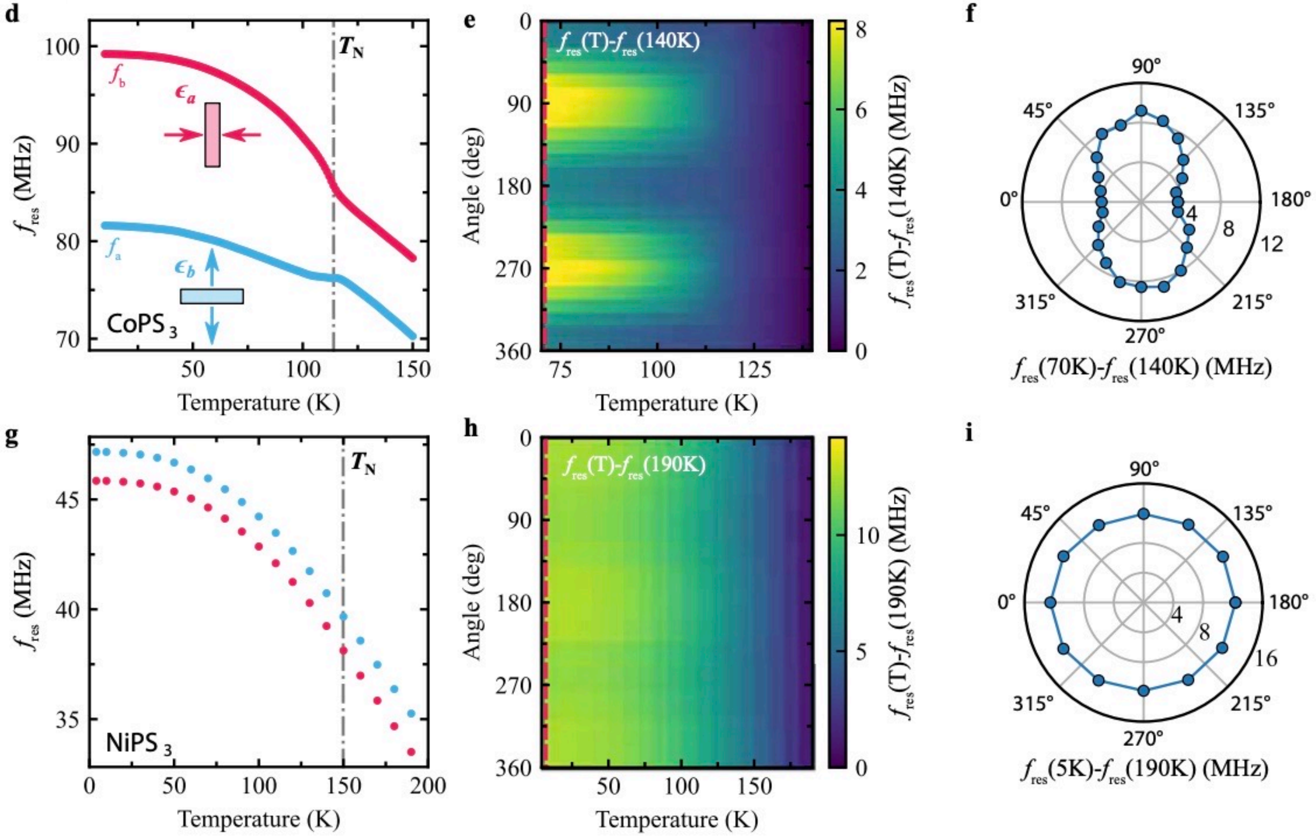
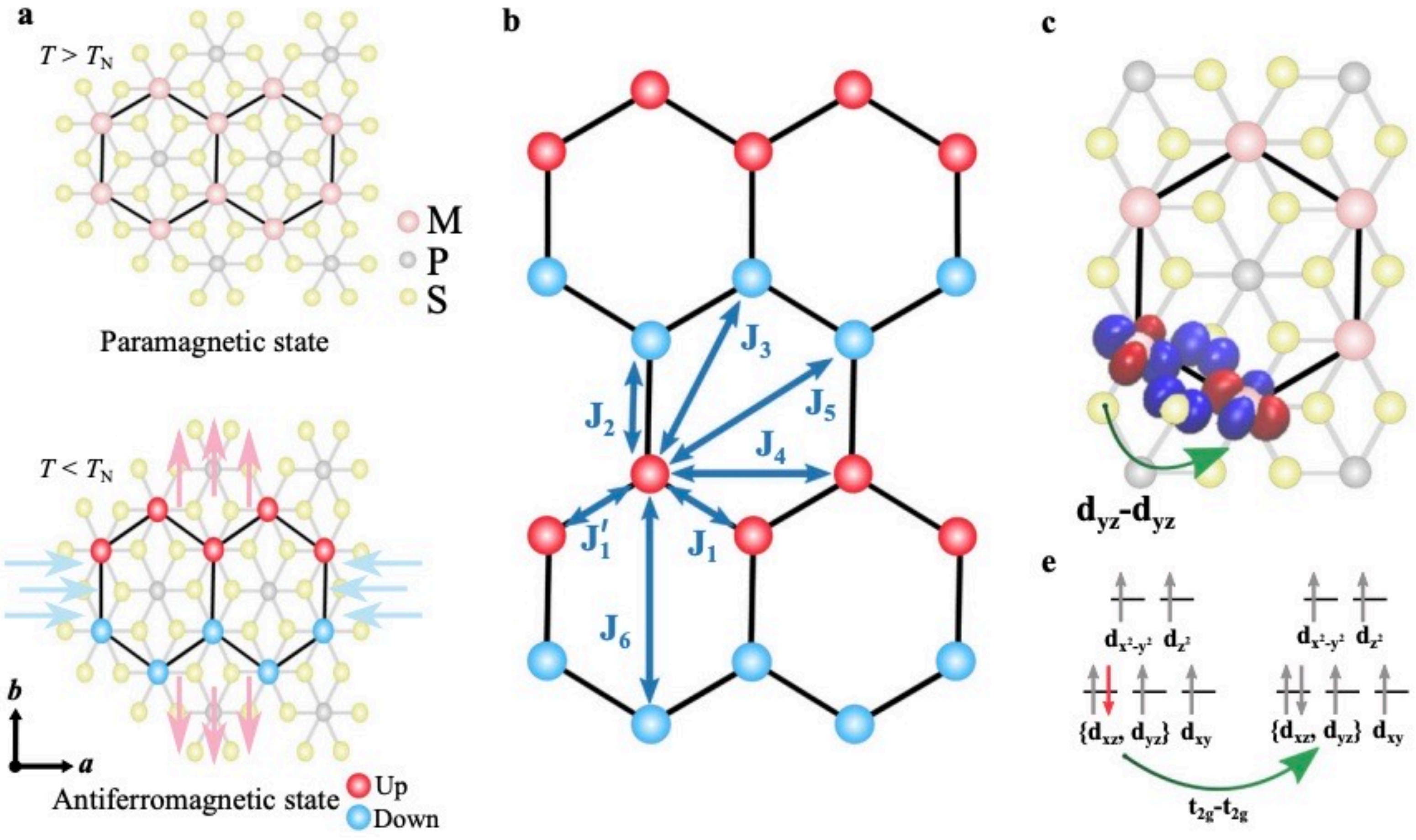


Table 1 | $CoPS_3$, $FePS_3$ and $NiPS_3$ lattice parameters of the crystallographic non-magnetic (NM) and fully optimized zig-zag antiferromagnetic (AF-zigzag) configurations, as calculated by DFT (see Supplementary Note 1)

	$CoPS_3$		$FePS_3$		$NiPS_3$	
Lattice parameter (Å)	*a*	*b*	*a*	*b*	*a*	*b*
NM	5.895	10.19	5.947	10.301	5.812	10.07
AF-zigzag	5.745	10.231	5.868	10.338	5.817	10.061
Change (%)	−2.545	+0.402	−1.328	+0.359	+0.086	−0.089

Magnetic structure & electron configuration

Houmes, Baglioni, Siskins, Lee, Esteras, Ruiz, Mañas-Valero, Boix-Constant, Baldoví, Coronado, Blanter, Steeneken and van der Zant, *Nature Commun.*, **14**, 8503 (2023)



Atomic-layer substitution in MPS_3

Janus 2D materials

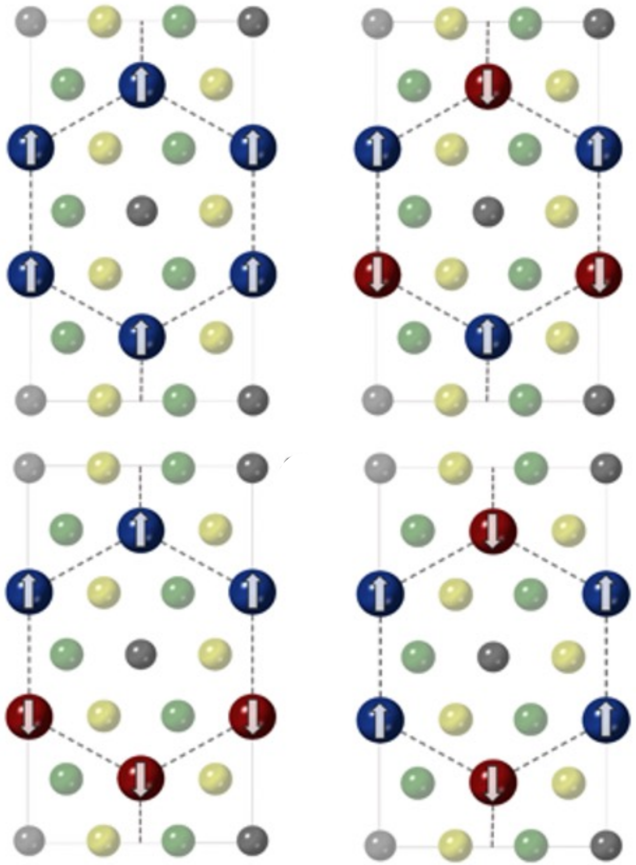
Two faces of the material are asymmetric

What are the effects of mirror broken symmetry?

Ruiz, Esteras, Rybakov and Baldoví* *Dalton Trans*. 2022, **51**, 16816

Atomic-layer substitution in MPS_3

- **Crystal**, **electronic** and **magnetic structure** of selenized Janus monolayers



Ruiz, Esteras, Rybakov and Baldoví* *Dalton Trans.* 2022, **51**, 16816

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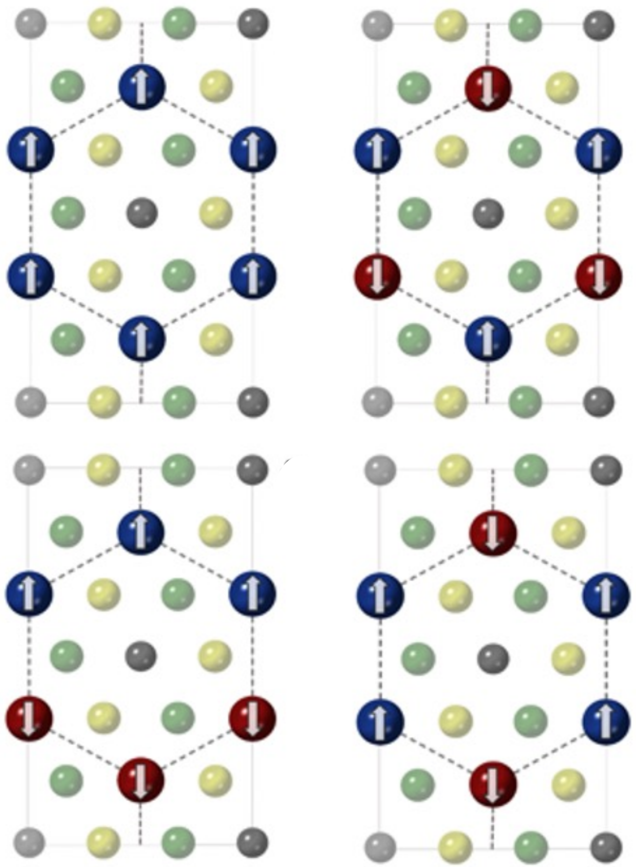
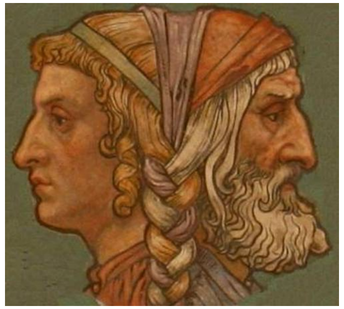


	$MnPS_3$	$Mn_2P_2S_3Se_3$	$NiPS_3$	$Ni_2P_2S_3Se_3$
FM	16.79	16.96	28.56	42.57
AFM-Néel	0.00	0.00	7.26	12.09
AFM-Zigzag	7.90	6.99	0.00	0.00
AFM-Stripy	4.24	7.25	41.64	52.38
MAE	-67.55	-355.52	-97.48	414.29



Change of magnetic easy axis from **in-plane** ($NiPS_3$) to **out-of-plane** ($Ni_2P_2S_3Se_3$)

Drastic enhancement of **magnetic anisotropy** (induced broken inversion symmetry)

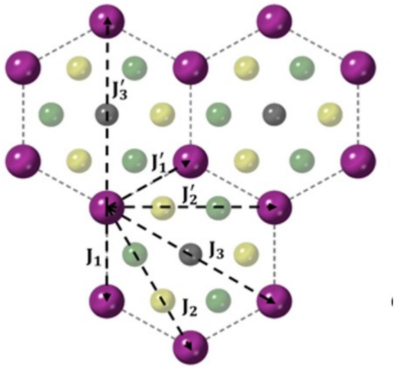
Ruiz, Esteras, Rybakov and Baldoví* *Dalton Trans.* 2022, **51**, 16816

Effects on magnetic properties

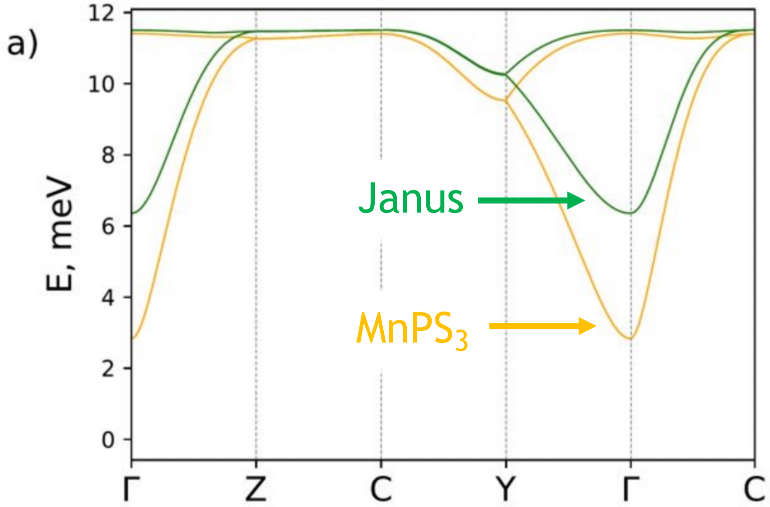
- **Wannier Hamiltonian** using *d* orbitals of M and *s*, *p* orbitals of P, S and Se
- Exchange interactions determined by Green's function method; Single-anion anisotropy extracted from MAE

$$E = -\sum_i K_i(\vec{S}_i \cdot \vec{e}_i)^2 - \sum_{i \neq j} [\vec{S}_i \mathbf{J}_{ij} \vec{S}_j + \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)]$$

- Induced broken symmetry creates **large DMI**



	$MnPS_3$	$Mn_2P_2S_3Se_3$	$NiPS_3$	$Ni_2P_2S_3Se_3$
$\lvert D_1/J_1 \rvert$	0.0007	0.0263	0.0018	0.0367
$\lvert D_1'/J_1' \rvert$	0.0041	0.0260	0.0005	0.3677
$\lvert D_2/J_2 \rvert$	0.0445	0.1499	0.0470	0.2999
$\lvert D_2'/J_2' \rvert$	0.0445	0.1450	0.1065	0.5643
$\lvert D_3/J_3 \rvert$	0.0023	0.0321	0.0003	0.0495
$\lvert D_3'/J_3' \rvert$	0.0009	0.0348	0.0002	0.0254



Ruiz, Esteras, Rybakov and Baldoví* *Dalton Trans.* 2022, **51**, 16816

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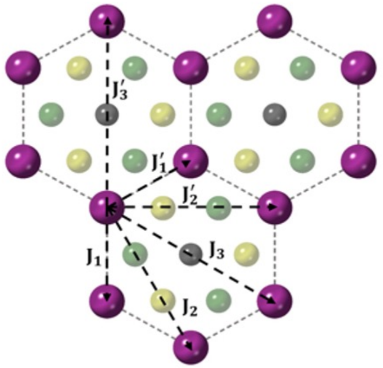
- Induced broken symmetry creates **large DMI**

Janus



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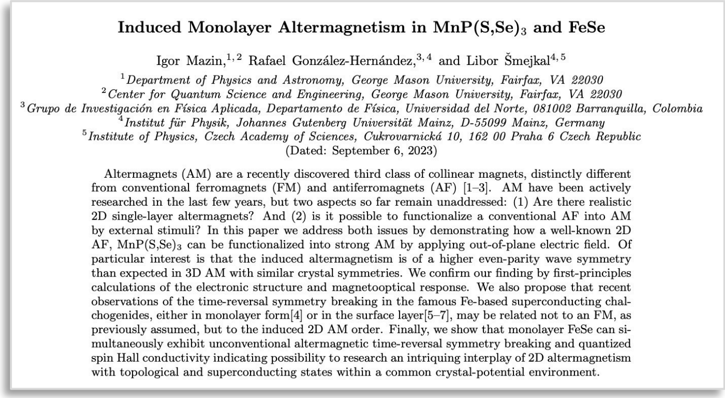
$\lvert D_1'/J_1' \rvert$	0.0041	0.0260	0.0005	0.3677
$\lvert D_2/J_2 \rvert$	0.0445	0.1499	0.0470	0.2999
$\lvert D_2'/J_2' \rvert$	0.0445	0.1450	0.1065	0.5643
$\lvert D_3/J_3 \rvert$	0.0023	0.0321	0.0003	0.0495
$\lvert D_3'/J_3' \rvert$	0.0009	0.0348	0.0002	0.0254



Induced Monolayer Altermagnetism in $MnP(S,Se)_3$ and FeSe

Igor Mazin,[1,2] Rafael González-Hernández,[3,4] and Libor Šmejkal[4,5]

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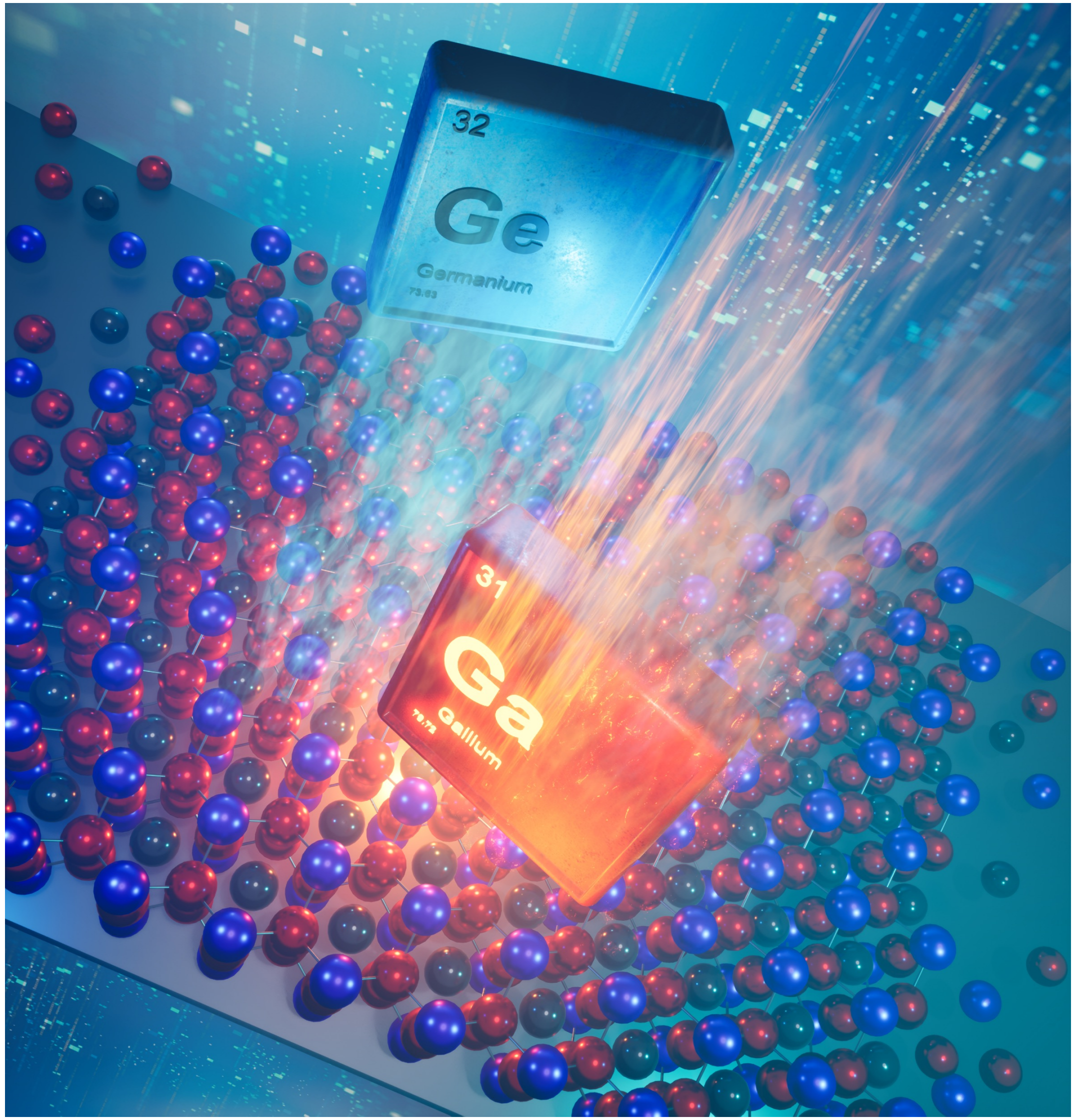
[2] *Center for Quantum Science and Engineering, George Mason University, Fairfax, VA 22030*
[3] *Grupo de Investigación en Física Aplicada, Departamento de Física, Universidad del Norte, 081002 Barranquilla, Colombia*
[4] *Institut für Physik, Johannes Gutenberg Universität Mainz, D-55099 Mainz, Germany*
[5] *Institute of Physics, Czech Academy of Sciences, Cukrovarnická 10, 162 00 Praha 6 Czech Republic*
(Dated: September 6, 2023)

Altermagnets (AM) are a recently discovered third class of collinear magnets, distinctly different from conventional ferromagnets (FM) and antiferromagnets (AF) [1–3]. AM have been actively researched in the last few years, but two aspects so far remain unaddressed: (1) Are there realistic 2D single-layer altermagnets? And (2) is it possible to functionalize a conventional AF into AM by external stimuli? In this paper we address both issues by demonstrating how a well-known 2D AF, $MnP(S,Se)_3$ can be functionalized into strong AM by applying out-of-plane electric field. Of particular interest is that the induced altermagnetism is of a higher even-parity wave symmetry than expected in 3D AM with similar crystal symmetries. We confirm our finding by first-principles calculations of the electronic structure and magnetooptical response. We also propose that recent observations of the time-reversal symmetry breaking in the famous Fe-based superconducting chalcogenides, either in monolayer form[4] or in the surface layer[5–7], may be related not to an FM, as previously assumed, but to the induced 2D AM order. Finally, we show that monolayer FeSe can simultaneously exhibit unconventional altermagnetic time-reversal symmetry breaking and quantized spin Hall conductivity indicating possibility to research an intriquing interplay of 2D altermagnetism with topological and superconducting states within a common crystal-potential environment.

Ruiz, Esteras, Rybakov and Baldoví* *Dalton Trans.* 2022, **51**, 16816

Mazin et al. arXiv:2309.02355

Above-Room-Temperature T_C in Fe_3GaTe_2



Fe_3GaTe_2

Isostructural with Fe_3GeTe_2

What is the origin of above-room-temperature magnetism in Fe_3GaTe_2?

Ruiz, Esteras, López-Alcalá and Baldoví*
Nano Lett. 2024, DOI: 0.1021/acs.nanolett.4c01019

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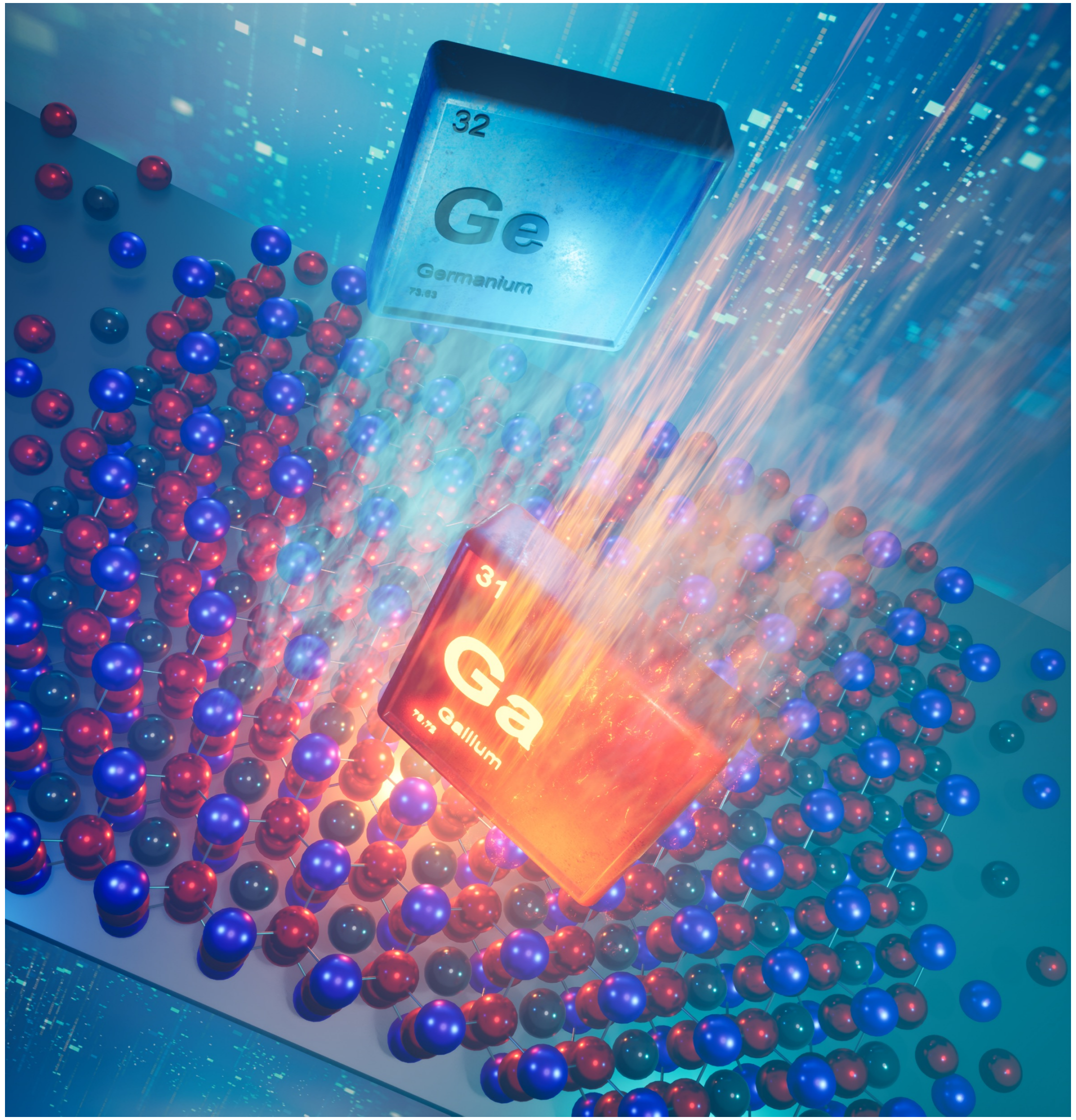

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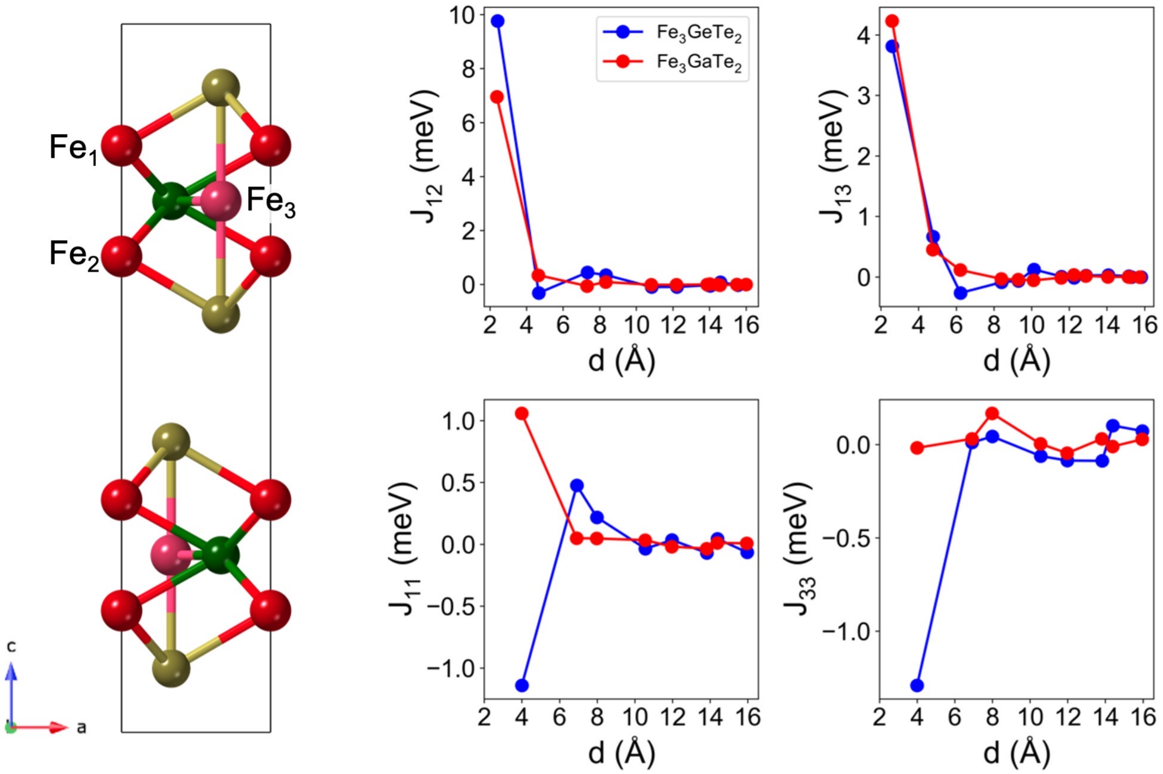
Fe_3GaTe_2 (T_c ~380 K); Fe_3GeTe_2 (T_c ~230 K)



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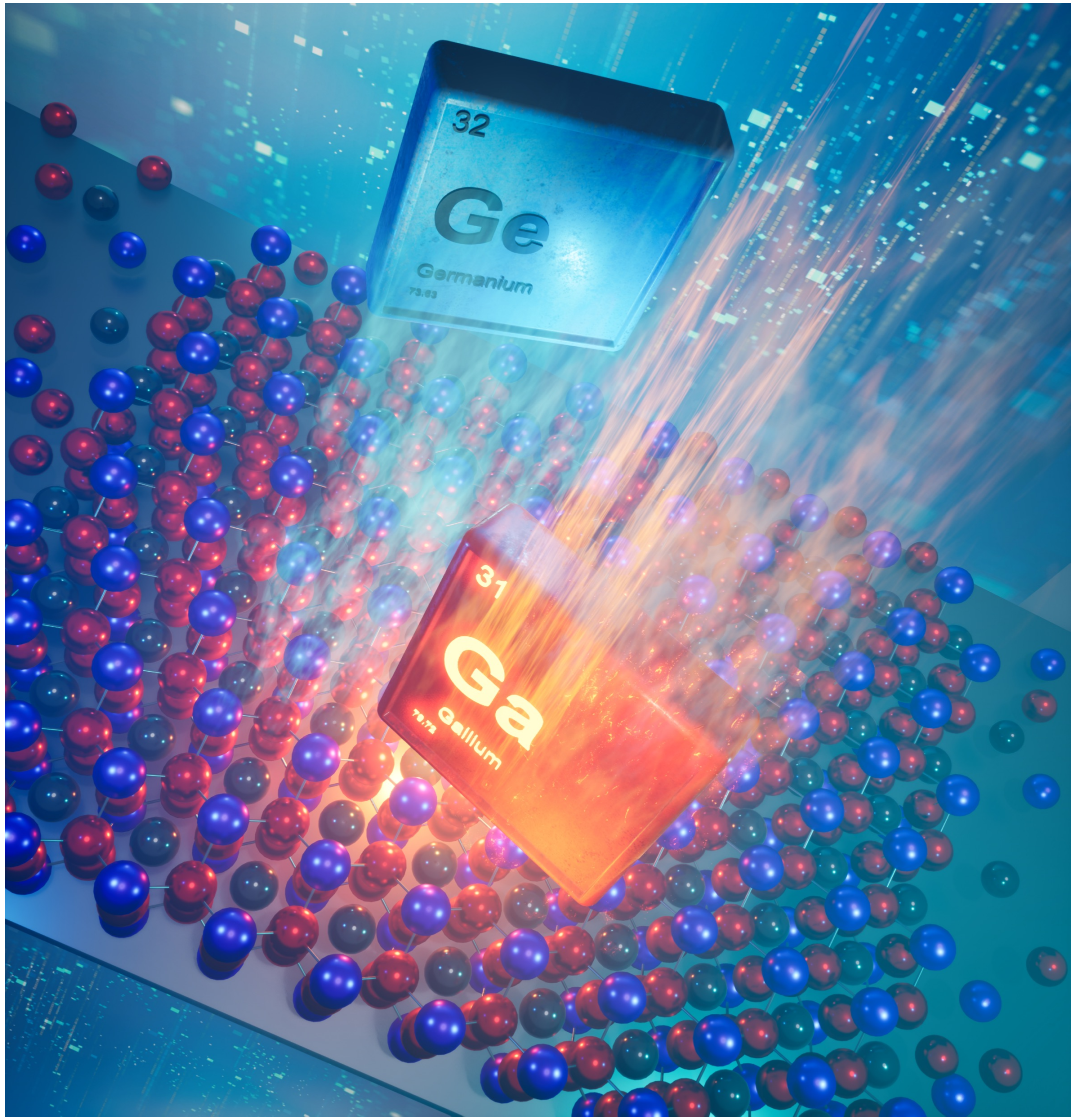

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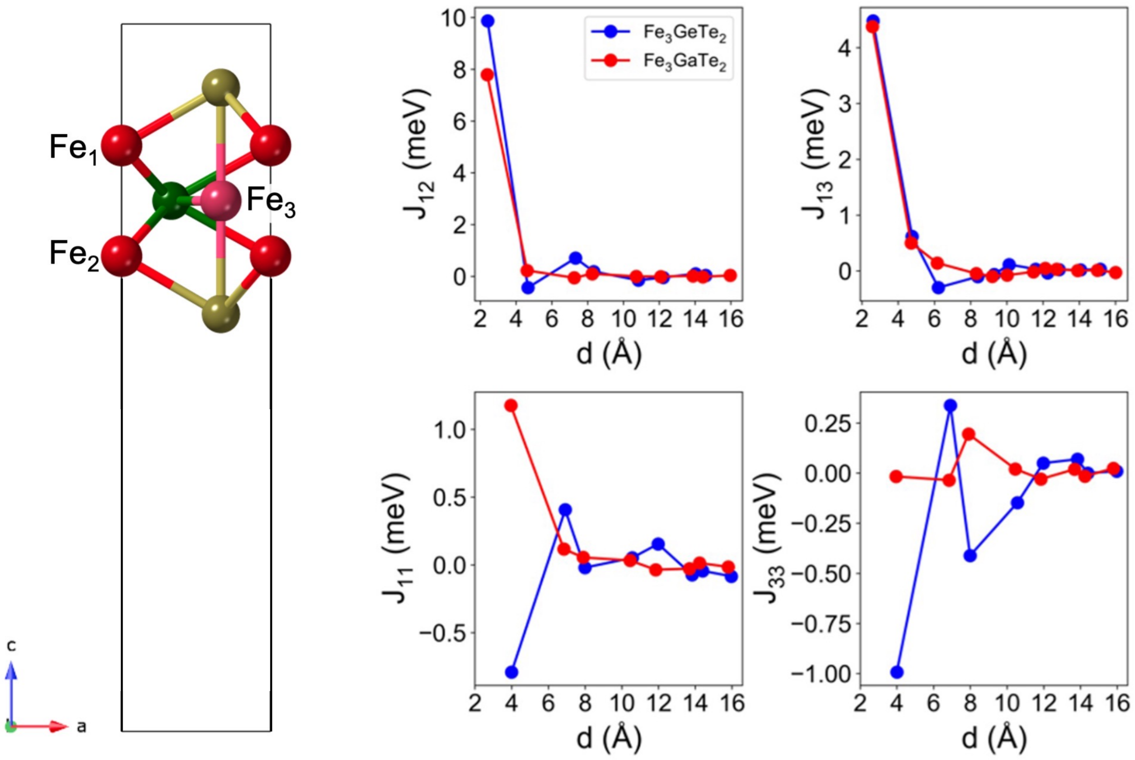

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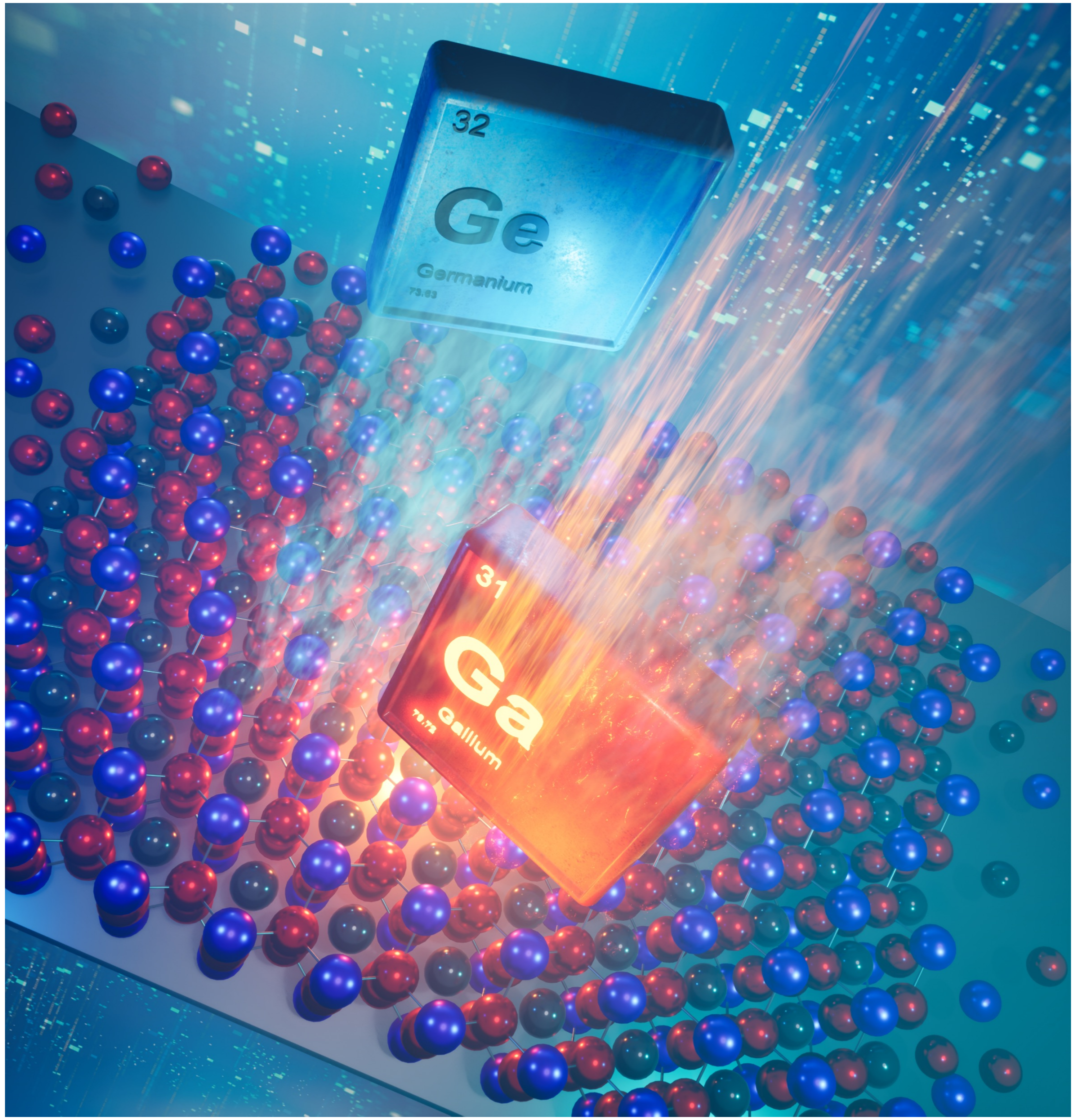

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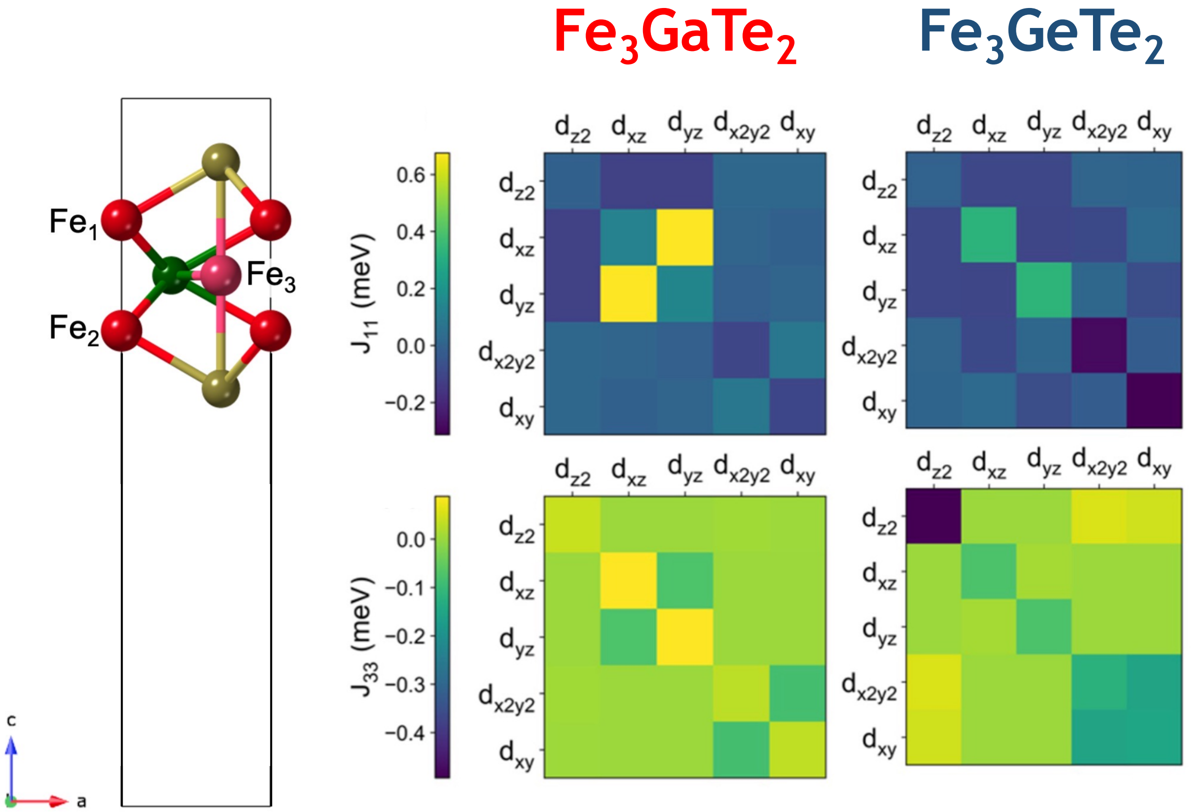
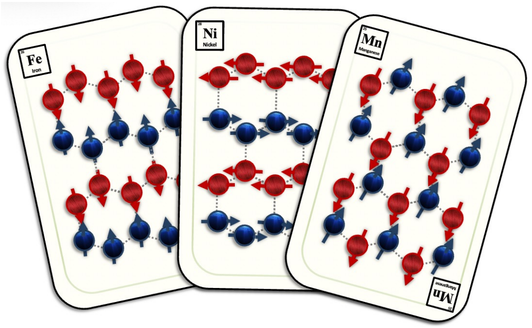
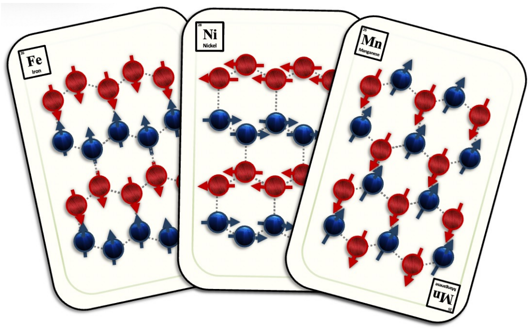


Take home messages



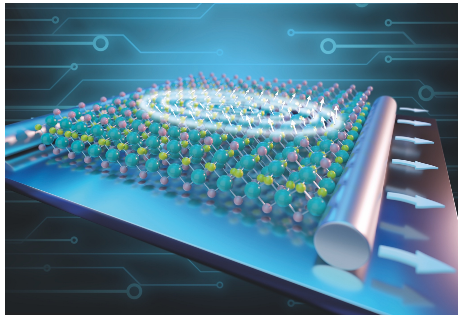
1. **2D magnetic materials**: A fascinating playground for exploring and controlling magnetic properties.

Take home messages



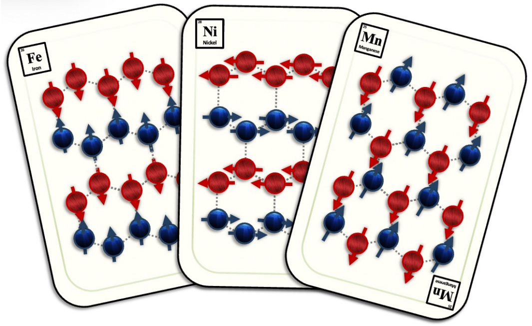
1. **2D magnetic materials**: A fascinating playground for exploring and controlling magnetic properties.

2. ***Ab initio*** **calculations** provide fundamental understanding and practical guidelines for the rational design and optimization of novel materials.



Take home messages

1. **2D magnetic materials**: A fascinating playground for exploring and controlling magnetic properties.



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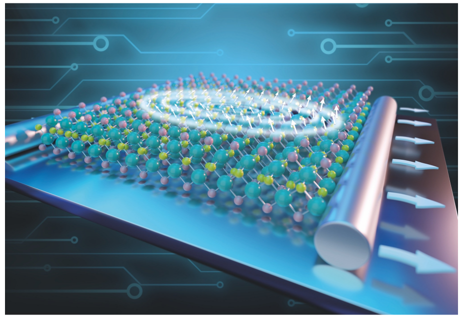
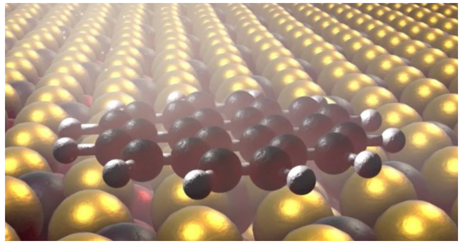
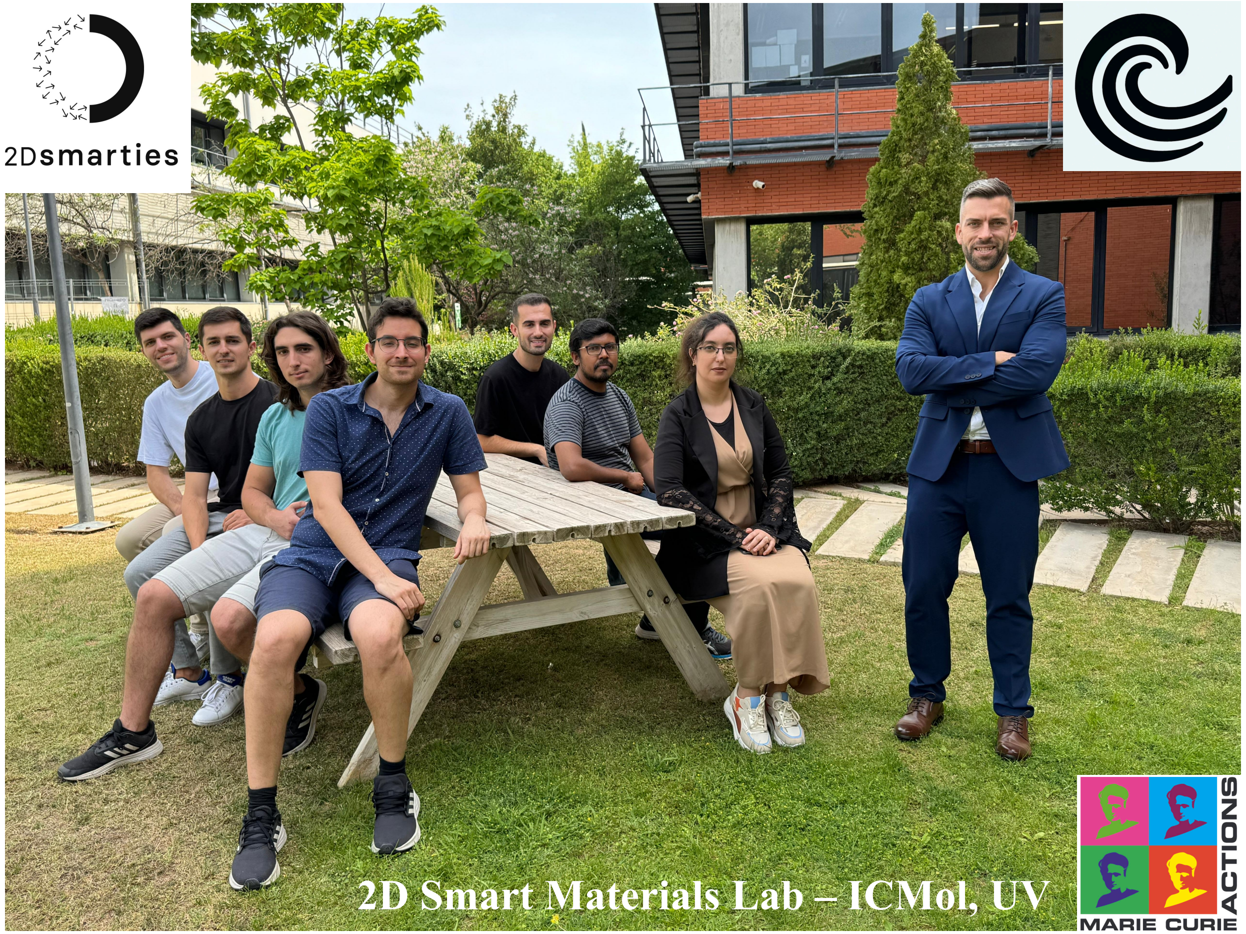


3. **The versatility of chemistry** to tune the magnetic properties and offer new opportunities for designing ultrathin films with tailored properties for magnonic applications.



Acknowledgements



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Thank you!

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