# On the origin of magnetic anisotropy in two dimensional $CrI_3$

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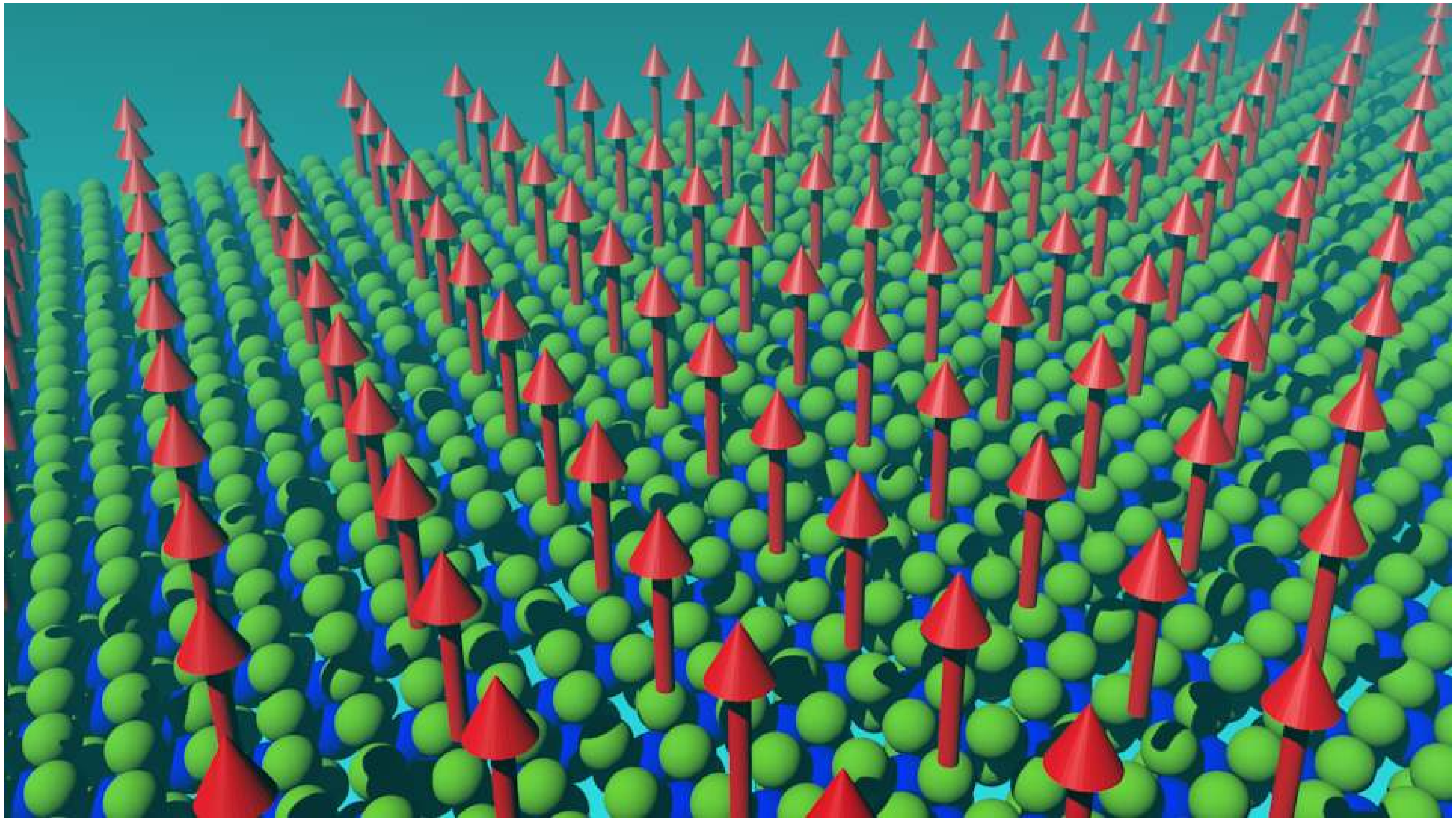
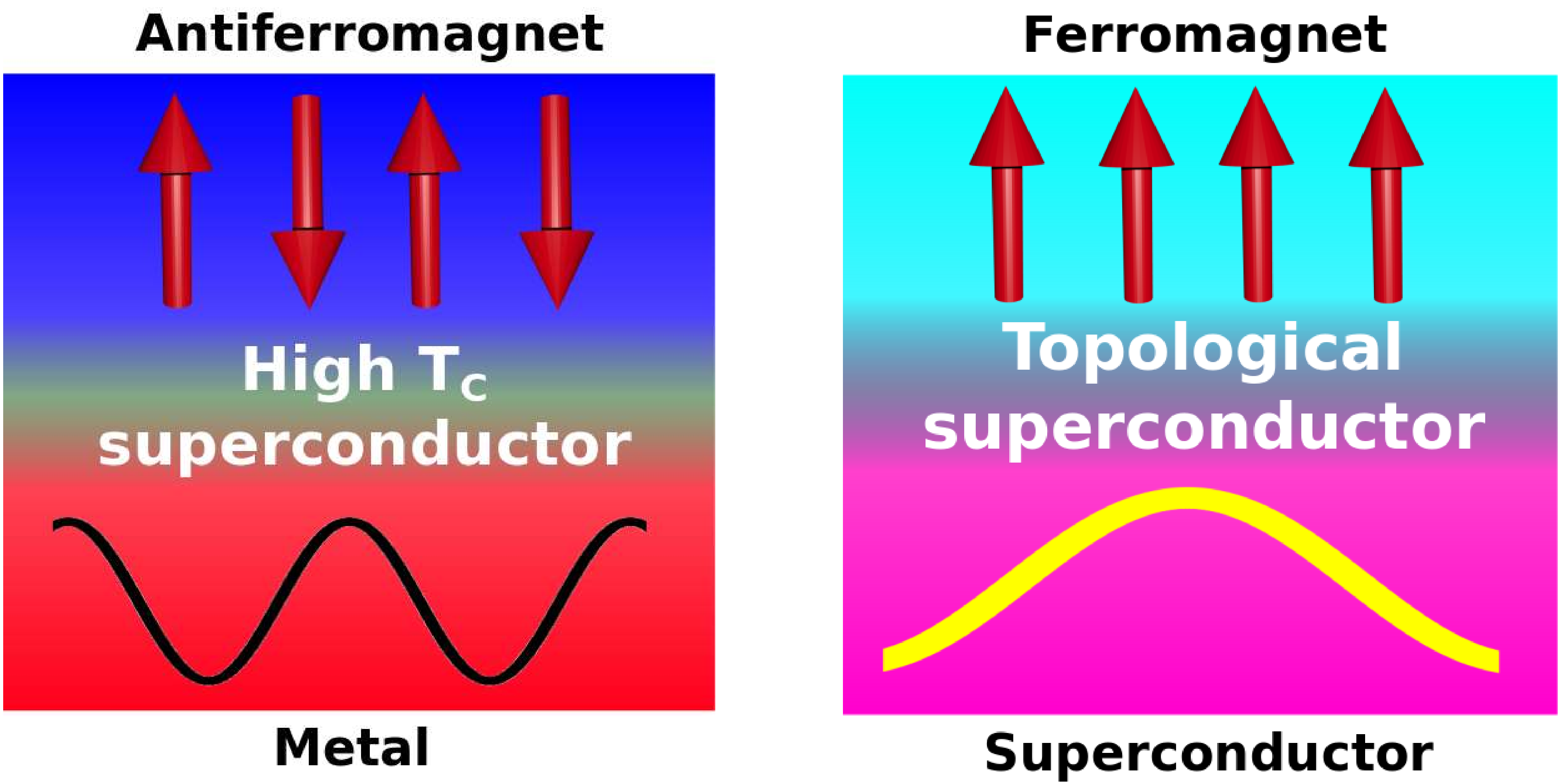


**Quantum Materials and Technologies, Santiago de Compostela (Spain)**

*Tuesday 18th July 2017*

# Emergence in electronic systems

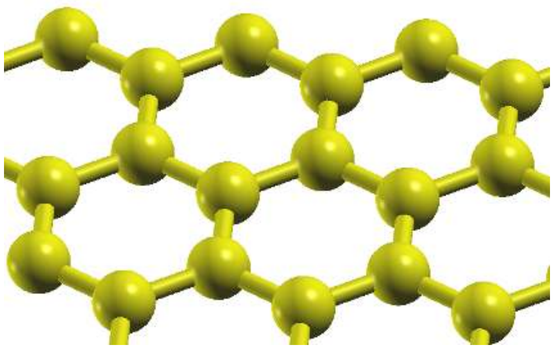



**Emergent electronic phases show up when competing orders coexist**

# Electronic orders in the 2d world

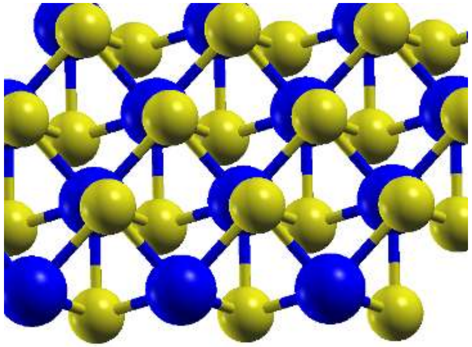
**Semimetal**

Graphene



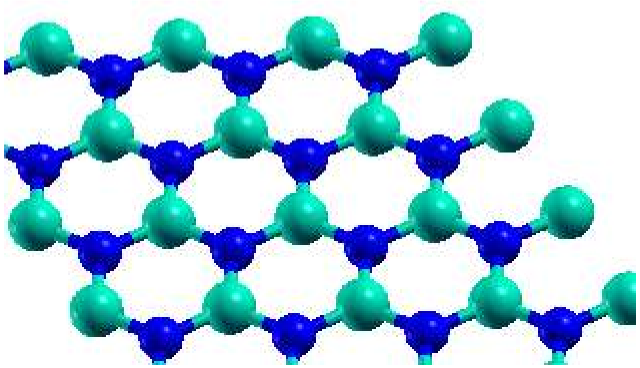
**Superconductor**

$NbSe_2$



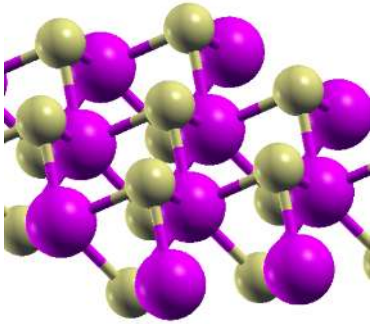
**Insulator**

BN

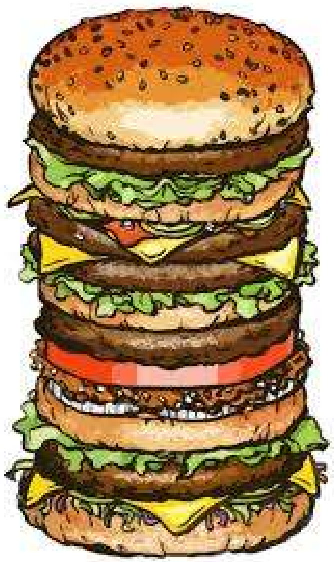


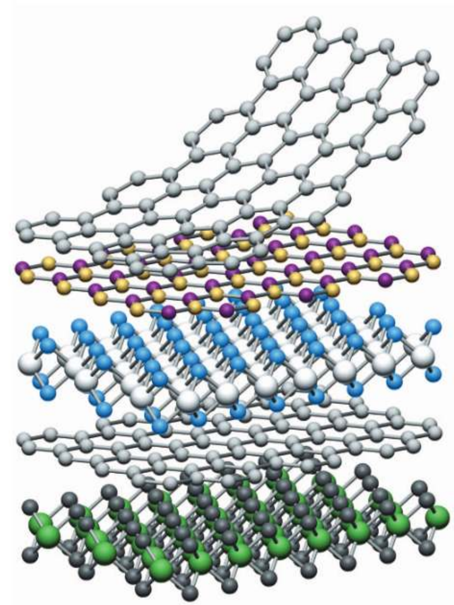
**Semiconductor**

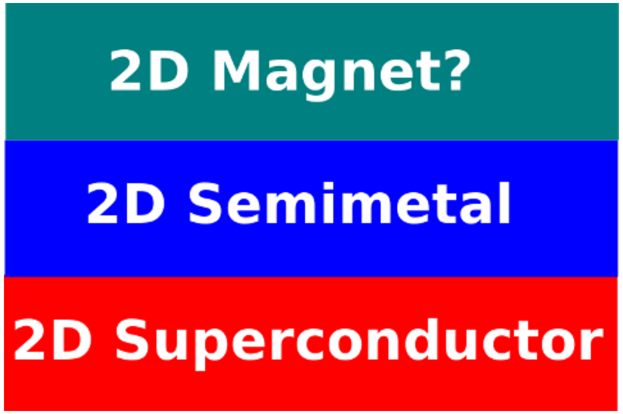
$MoS_2$



# Coexisting orders in 2d





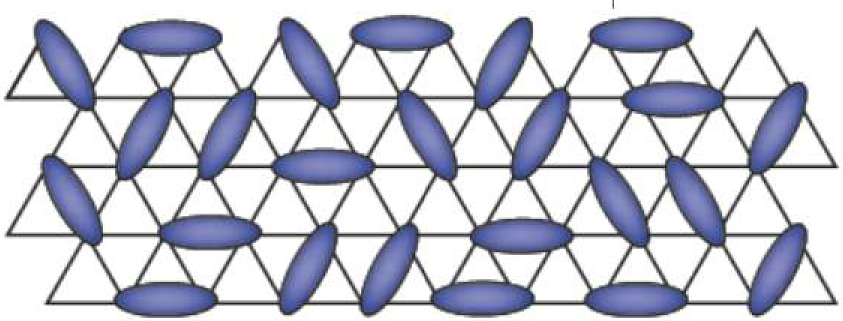



**2D materials offer an extraordinary playground to explore interplay between antagonist electronic orders**

# Possibilities with 2d magnets

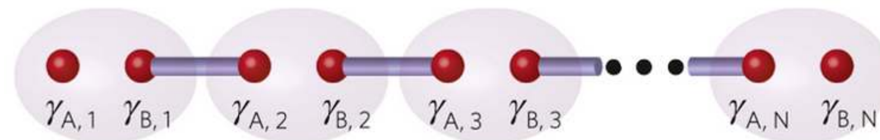
## Quantum spin liquid

**2d magnet + frustration**



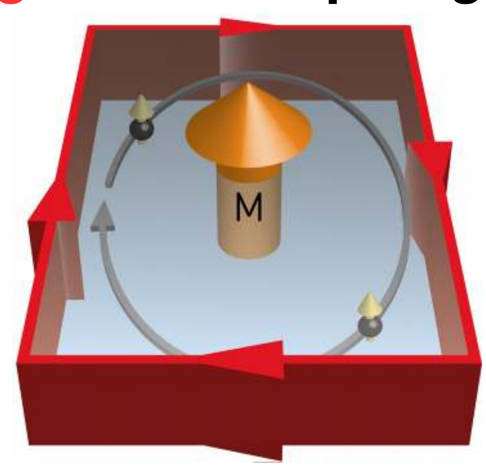
## 2d topological superconductor

**2d magnet + Rashba+ superconductor**




## Quantum Anomalous Hall insulator

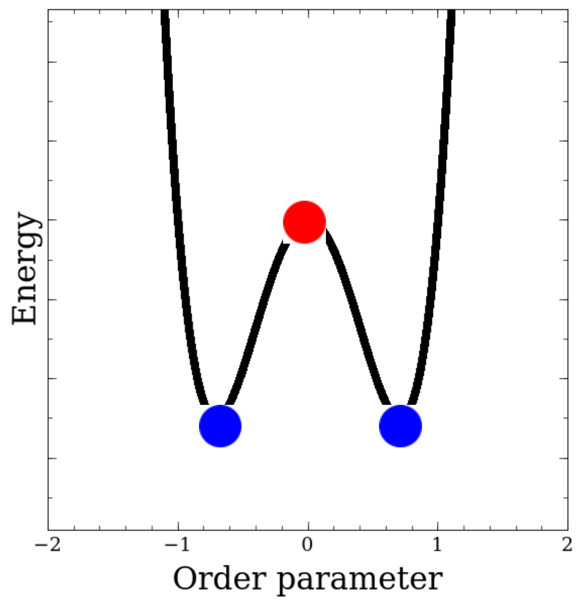
**2d magnet + 3d topological insulator**




# Spontaneous symmetry breaking

**Energy landscape of a system with spontaneous symmetry breaking**

$$E(M) = \alpha(M^2 - \beta)^2$$




*Order parameter:*
→ **Magnetization (M)**
→ Some field expectation value
→ Some correlation function

**Energy is minimized for a finite order parameter M**


**But!!!!!**

**Spontaneous symmetry breaking can be destroyed by**

→ **Quantum fluctuations**

<u>Example: Haldane chain</u>

→ **Thermal fluctuations**

<u>Example: any paramagnet</u>

# Magnetism and dimensionality

**The Ising model** $\sigma_i = \pm 1$

$$H = -J \sum_{\langle i,j \rangle} \sigma_i \sigma_j + h \sum_i \sigma_i$$

*Exchange* *Magnetic field*

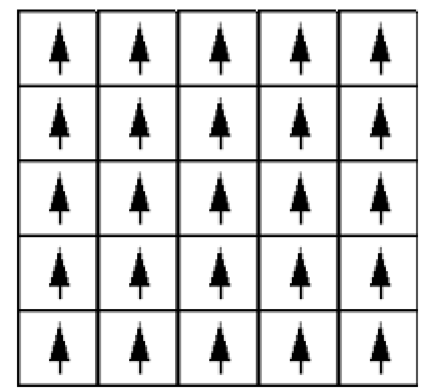
***Spontaneous symmetry breaking if***

$$\lim_{h \to 0} \langle \sigma \rangle (T, h) \neq 0$$

**Ising model in 1d**

**No magnetization at non zero T**

**Ising model in 2d**



**Magnetization at non zero T**

**Dimensionality determines if there is spontaneous symmetry breaking at finite temperature**

# Continuous symmetry breaking in 2d

**Mermin-Wagner theorem**

**A perfect isotropic system <u>does not order</u> at finite temperature in 2d**

In particular, for this Hamiltonian in a 2d lattice:

$$H = -J \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j$$

The expectation value of the magnetization is zero at any finite temperature

$$\langle \vec{S}_i \rangle = 0 \quad \text{for} \quad T > 0$$

**How to avoid the Mermin-Wagner theorem?**
→ *Add anisotropic interactions in the Hamiltonian*

**Magnetic anisotropy is a must for 2d magnetism**

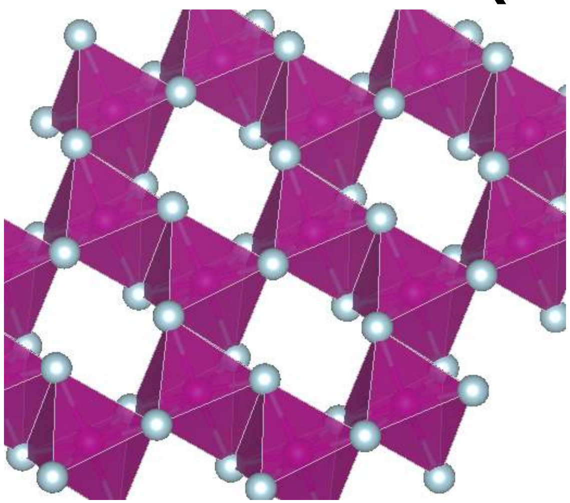
# How to find 2d magnets?


- Take some layered 3d magnet, and **pick a single layer**
- Look for materials with **strong magnetic anisotropy**

# Two (bulk) halide compounds

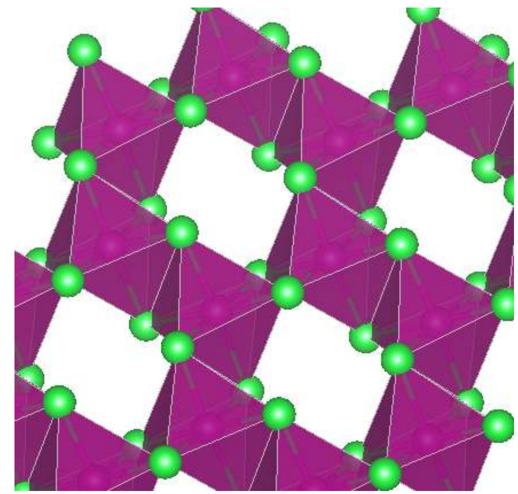
## <u>Two iso-structural compounds</u>

**Chromium iodine ($CrI_3$)**



**Off-plane** ferromagnet
Anisotropy field of 3 T

**Chromium chlorine ($CrCl_3$)**



**In-plane** ferromagnet
Anisotropy field of 0.1 T

*Michael A. McGuire Crystals 7.5 (2017): 121.*

***Too similar, yet too different***

**What is the ultimate origin of the magnetic anisotropy in $CrI_3$?**

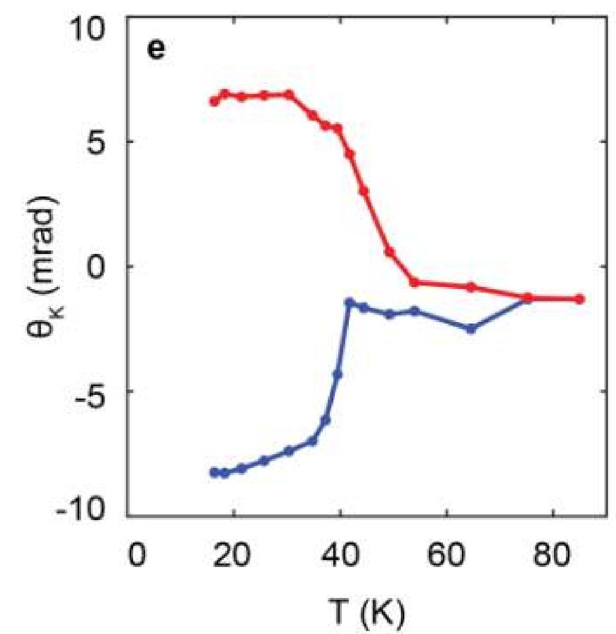
# Purely 2d ferromagnet: $CrI_3$

nature

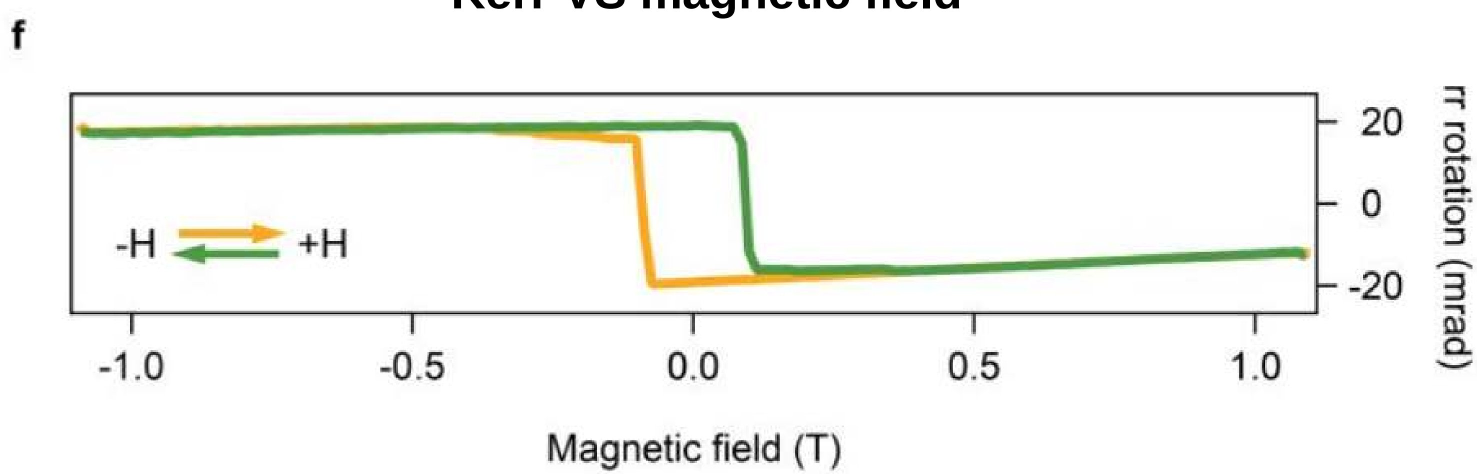
Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit

*Huang et al (2017)*

**Kerr VS temperature**




**Kerr VS magnetic field**



# Ferromagnetism in 2d $CrI_3$

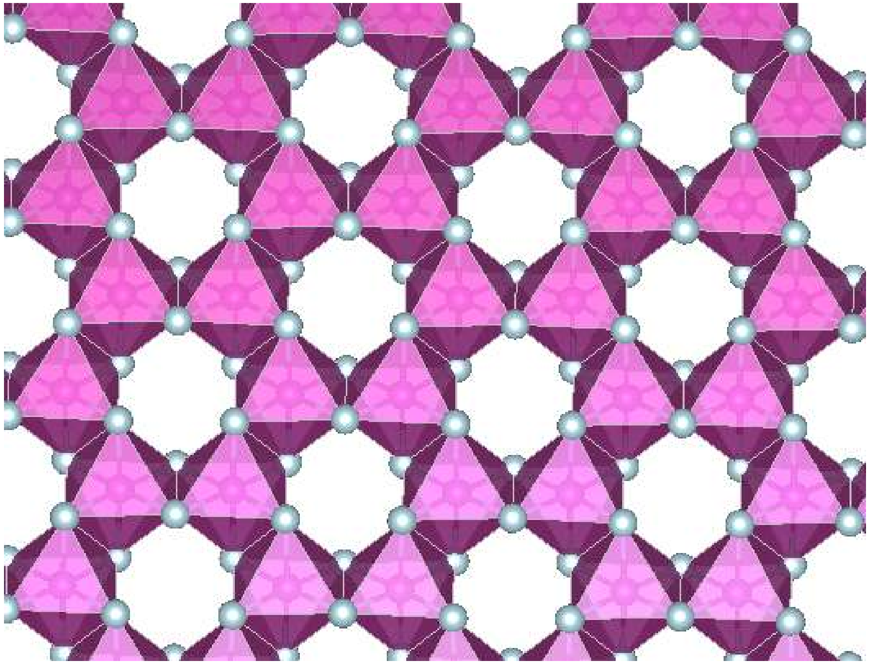
## Two simple open questions

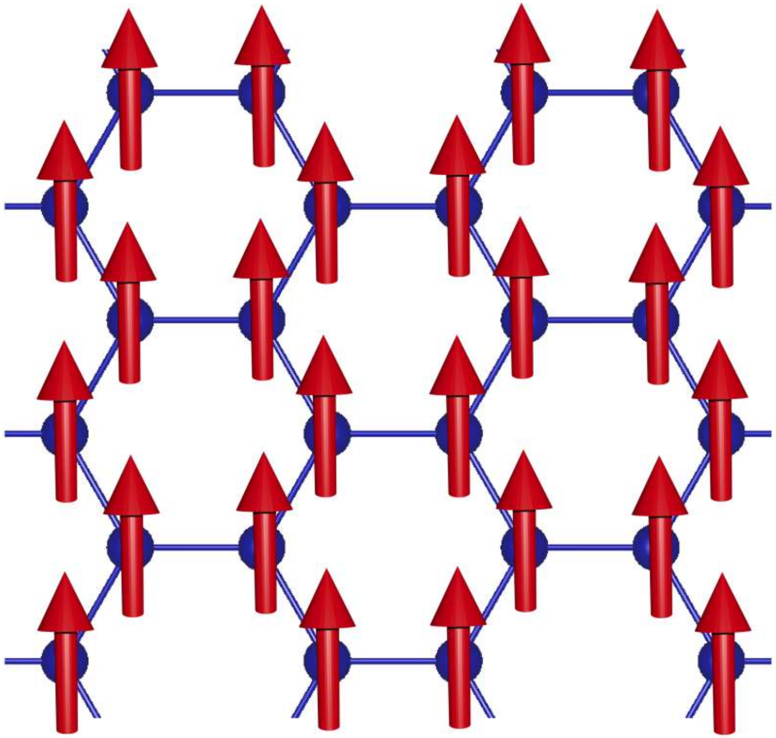
**Off-plane easy axis, are they Ising spins?**
**Critical temperature of 45 K, but what controls this?**

Is $CrI_3$ a two dimensional Ising model?

If not, what is the right model for $CrI_3$?

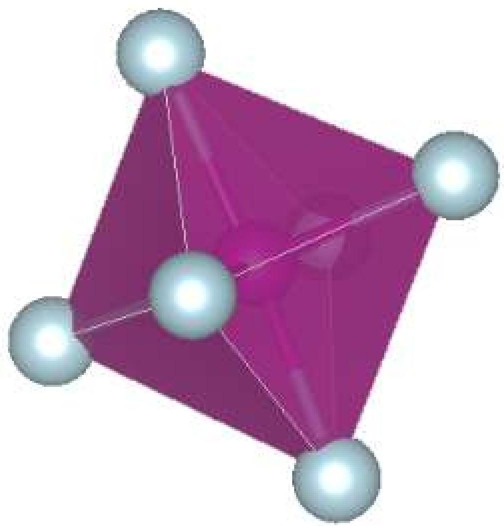
# $CrI_3$, ferromagnetic honeycomb lattice



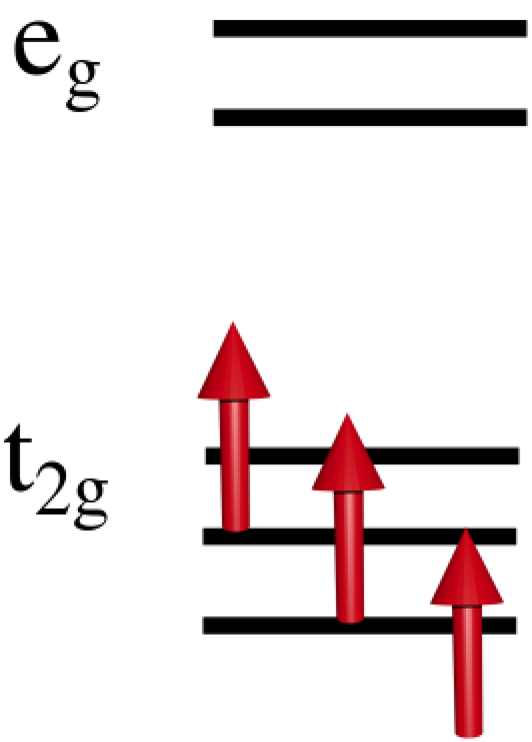


**Each chromium atom contributes with S=3/2**

# Electronic filling and anisotropy



Octahedral iodine environment

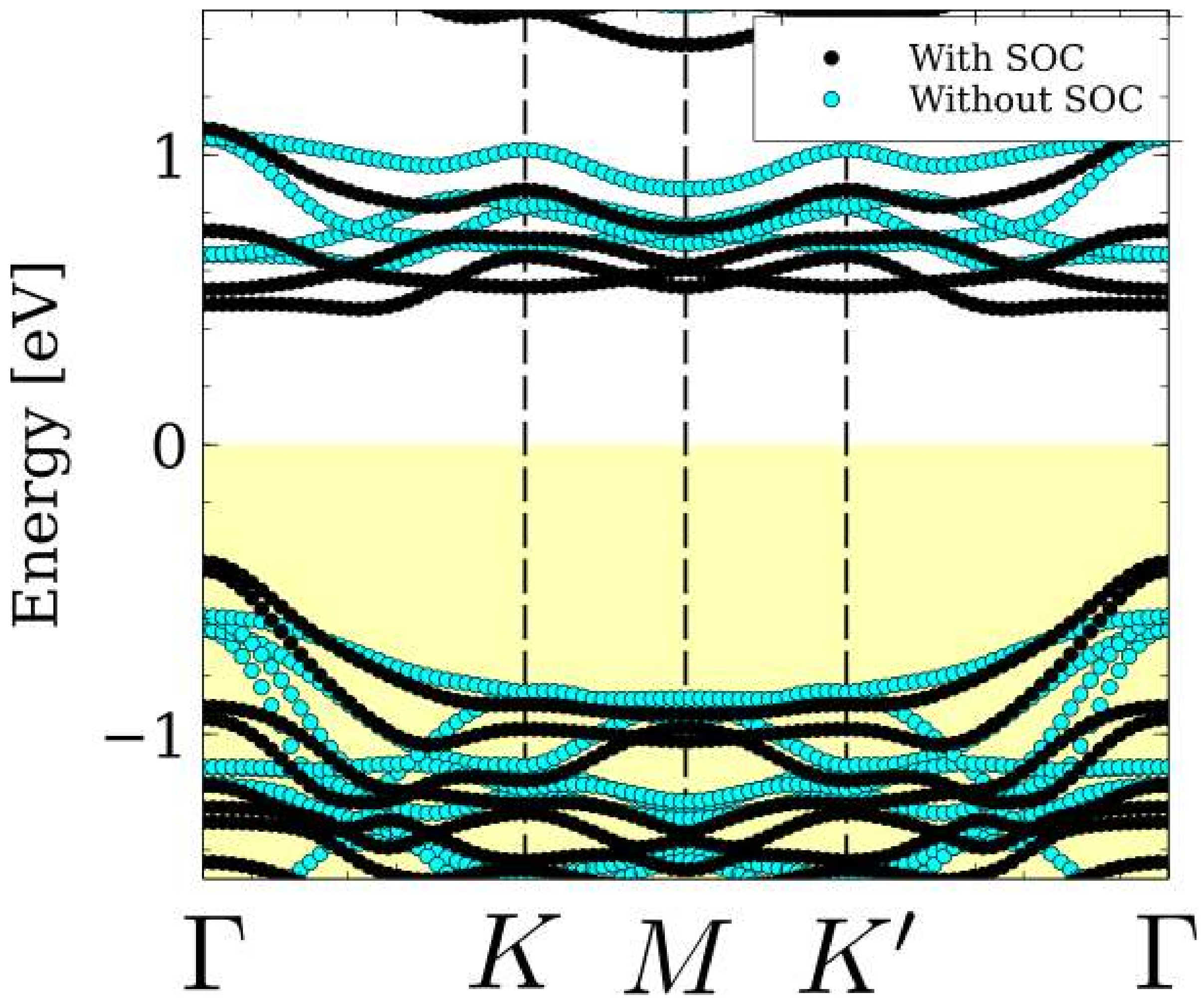



Quenched angular momenta

**With quenched angular momenta, single ion anisotropy is extremely small**

**How is there a sizable magnetic anisotropy in $CrI_3$?**

# Effect of spin orbit coupling in $CrI_3$

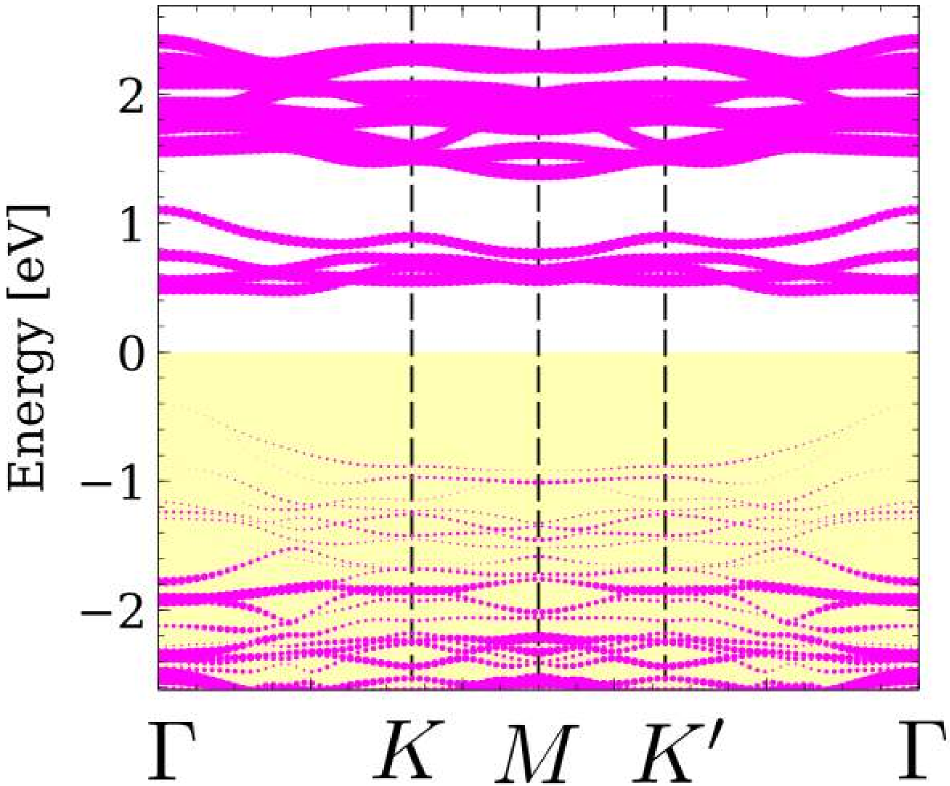



Elk LAPW code

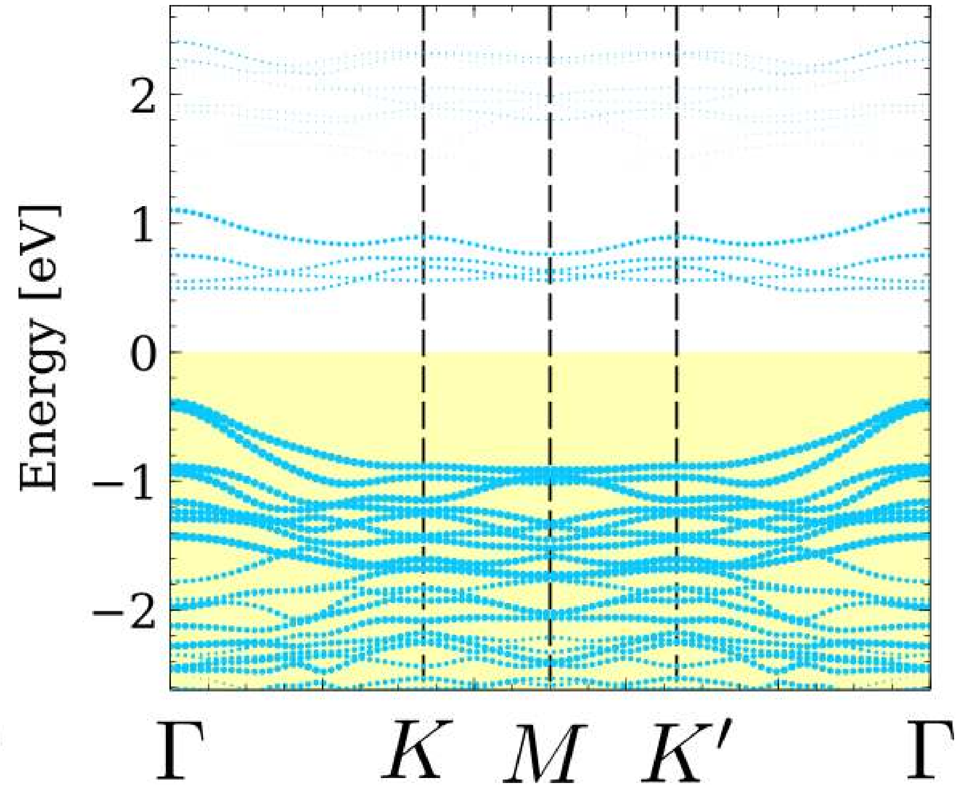
**Effect of SOC in the band structure is extremely large**

# Weights over the atoms in $CrI_3$

**Weight on chromium**




**Weight on iodine**



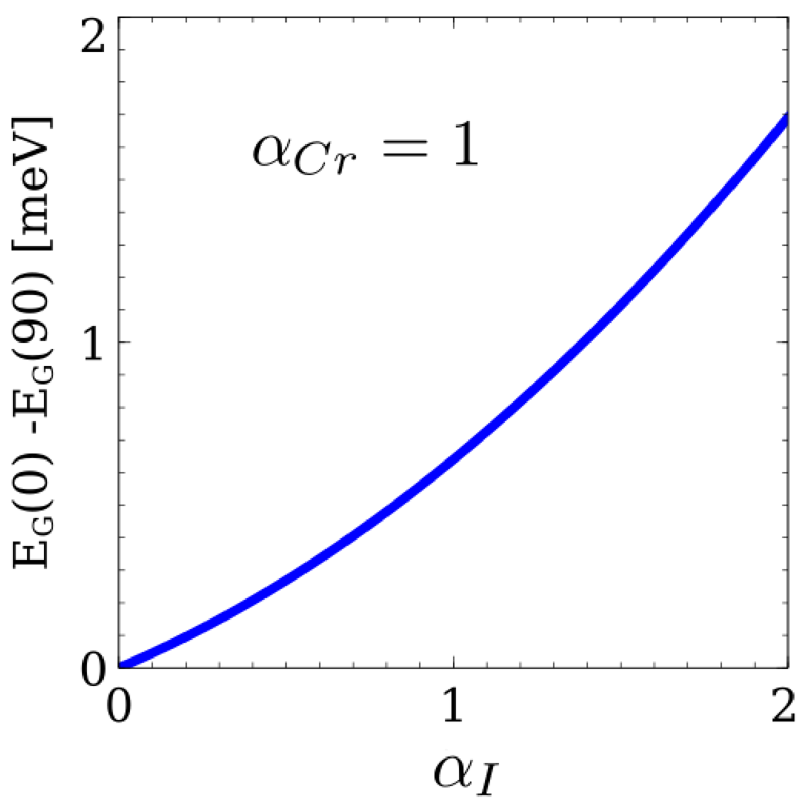

There is a sizable contribution of iodine above and below the gap

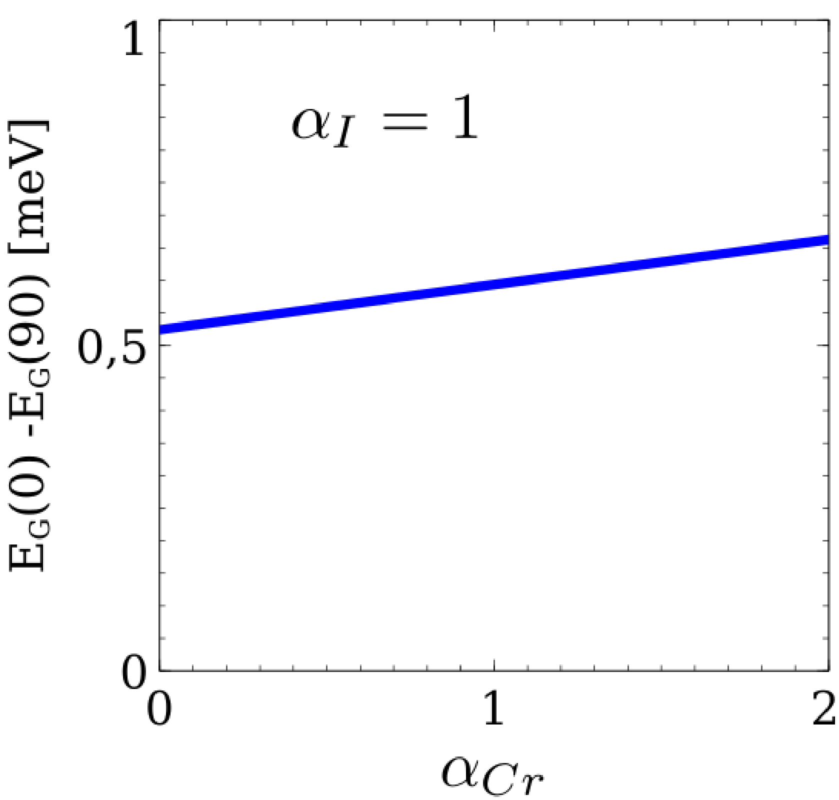
**Is iodine a key to determine the magnetic properties?**

# Magnetic anisotropy in $CrI_3$

**We can separately see the effect of the two atoms to magnetic anisotropy**

$$\mathcal{H}_{\mathrm{DFT}}(\alpha_I, \alpha_{Cr}) = \mathcal{H}_0 + \alpha_I \mathcal{H}_I^{SOC} + \alpha_{Cr} \mathcal{H}_{Cr}^{SOC}$$



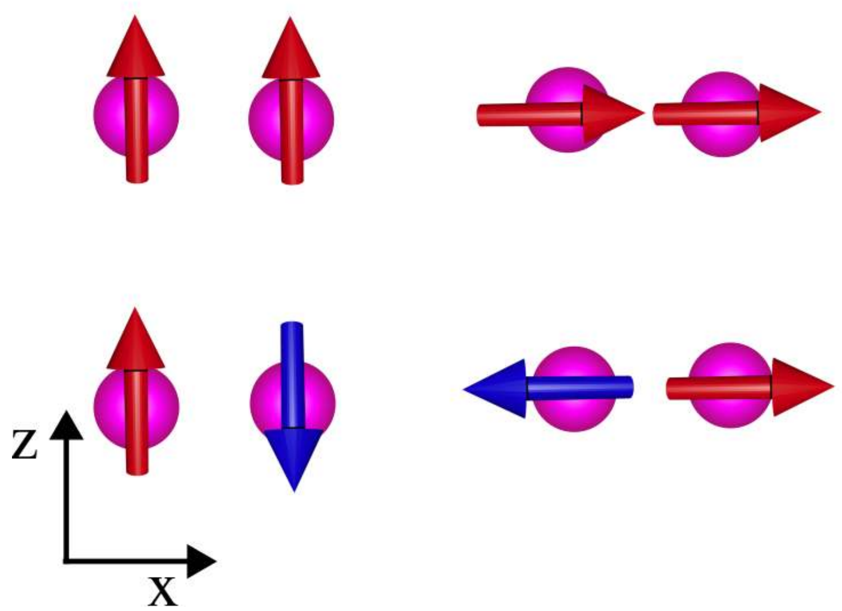

**The major contribution to magnetic anisotropy comes from iodine**

# Disentangling the spin Hamiltonian

## Minimal Spin Hamiltonian

$$\mathcal{H} = -\left( \sum_i D (S_i^z)^2 + \frac{J}{2} \sum_{i,i'} \vec{S}_i \cdot \vec{S}_{i'} + \frac{\lambda}{2} \sum_{i,i'} S_i^z S_{i'}^z \right).$$

**Single ion anisotropy** | **Isotropic Heisenberg** | **Anisotropic interaction**




$$\mathcal{E}_{FM,z} = -2S^2 D - 3S^2(J+\lambda)$$

$$\mathcal{E}_{AF,z} = -2S^2 D + 3S^2(J+\lambda)$$

$$\mathcal{E}_{FM,x} = -3S^2 J$$

$$\mathcal{E}_{AF,x} = +3S^2 J$$

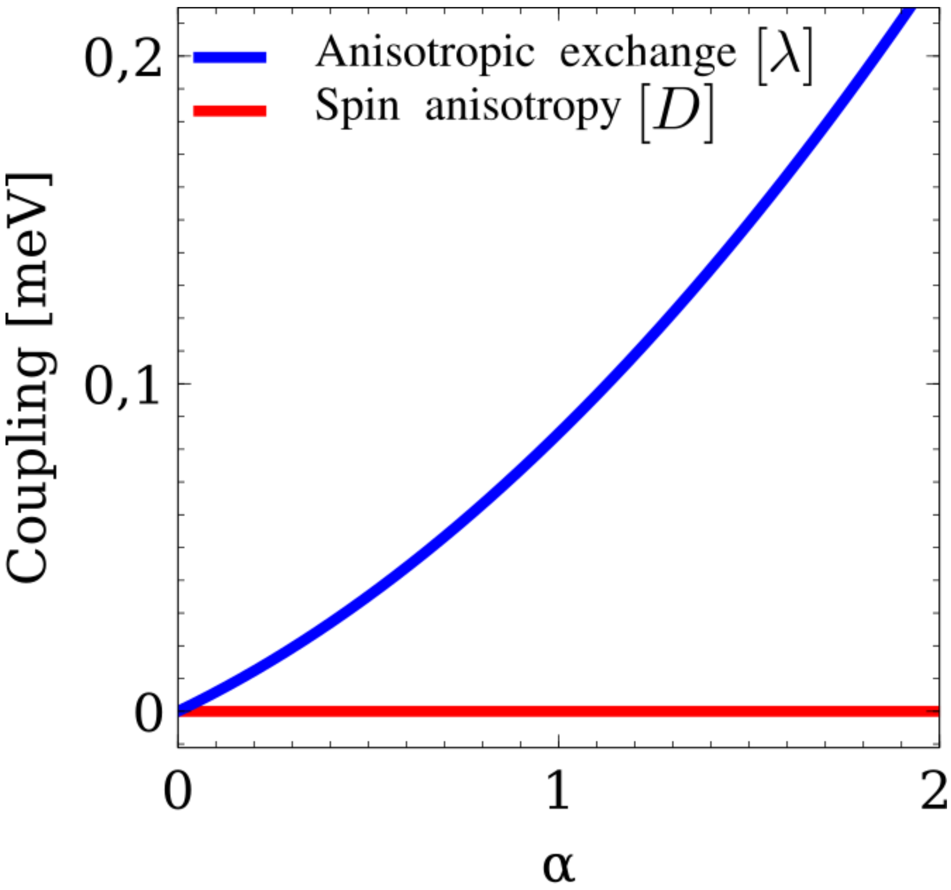
# The true spin model for $CrI_3$

### Hamiltonian

$$\mathcal{H} = -\left( \sum_i D (S_i^z)^2 + \frac{J}{2} \sum_{i,i'} \vec{S}_i \cdot \vec{S}_{i'} + \frac{\lambda}{2} \sum_{i,i'} S_i^z S_{i'}^z \right),$$

### Energetics

$$\mathcal{E}_{FM,z} = -2S^2 D - 3S^2 (J + \lambda)$$
$$\mathcal{E}_{AF,z} = -2S^2 D + 3S^2 (J + \lambda)$$
$$\mathcal{E}_{FM,x} = -3S^2 J$$
$$\mathcal{E}_{AF,x} = +3S^2 J$$




Single ion anisotropy is negligible → **not a 2d Ising system**

Sizable anisotropic exchange → **$CrI_3$ is a 2d realization of the XXZ model**

# Magnetic fluctuations: spin waves

**Spin Hamiltonian**

$$\mathcal{H} = -\left( \sum_i D(S_i^z)^2 + \frac{J}{2} \sum_{i,i'} \vec{S}_i \cdot \vec{S}_{i'} + \frac{\lambda}{2} \sum_{i,i'} S_i^z S_{i'}^z \right)$$

**Holstein-Primakoff**

$$S_i^+ = \sqrt{2S} \sqrt{1 - \frac{b_i^\dagger b_i}{2S}}\, b_i$$

$$S_i^- = \sqrt{2S}\, b_i^\dagger \sqrt{1 - \frac{b_i^\dagger b_i}{2S}}$$

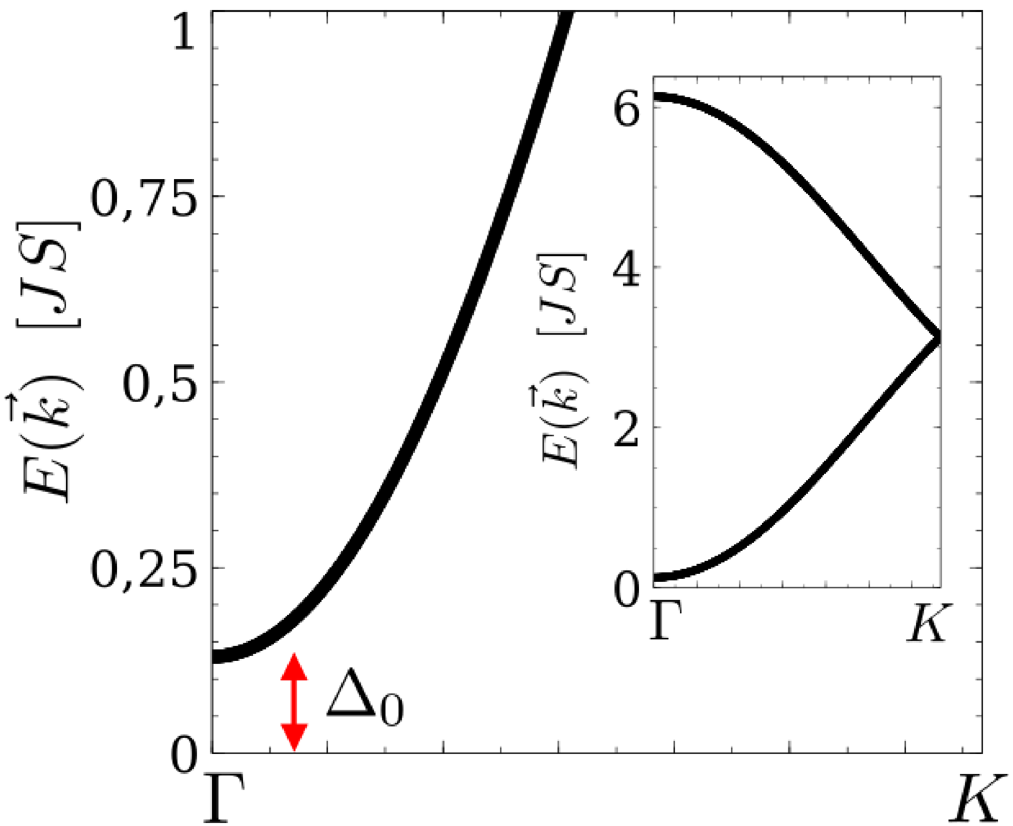
$$S_i^z = S - b_i^\dagger b_i$$

**Spin wave Hamiltonian (bosonic Hamiltonian)**

$$\mathcal{H}_{\text{spin waves}} = \sum_i \left(2DS + 3S(J+\lambda)\right) b_i^\dagger b_i - JS \sum_{\langle ij \rangle} b_i^\dagger b_j$$

# Spin wave spectra

## Spin wave spectra




$$\Delta_0 = 2DS + 3S\lambda$$

## Spin Wave Bloch Hamiltonian

$$\mathcal{H}_{SW}(\vec{k}) = \begin{pmatrix} \epsilon_0 & -JSf(\vec{k}) \\ -JSf^*(\vec{k}) & \epsilon_0 \end{pmatrix}$$

## Correction to magnetization without magnetic anisotropy

$$\delta M = \frac{1}{4\pi}\int_0^{k_c} \frac{kdk}{\beta\rho k^2} \rightarrow \infty$$

# Spin wave Hamiltonian at finite temperature

**First order expansion**

$$S_i^+ \approx \sqrt{2S}\left(1 - \frac{b_i^\dagger b_i}{4S}\right) b_i$$

$$S_i^- \approx \sqrt{2S} b_i^\dagger \left(1 - \frac{b_i^\dagger b_i}{4S}\right)$$

$$S_i^z = S - b_i^\dagger b_i$$

**Mean field approximation**

$$b_i^\dagger b_i b_j^\dagger b_j \approx \langle b_i^\dagger b_i \rangle b_j^\dagger b_j + b_i^\dagger b_i \langle b_j^\dagger b_j \rangle + \mathcal{C}$$

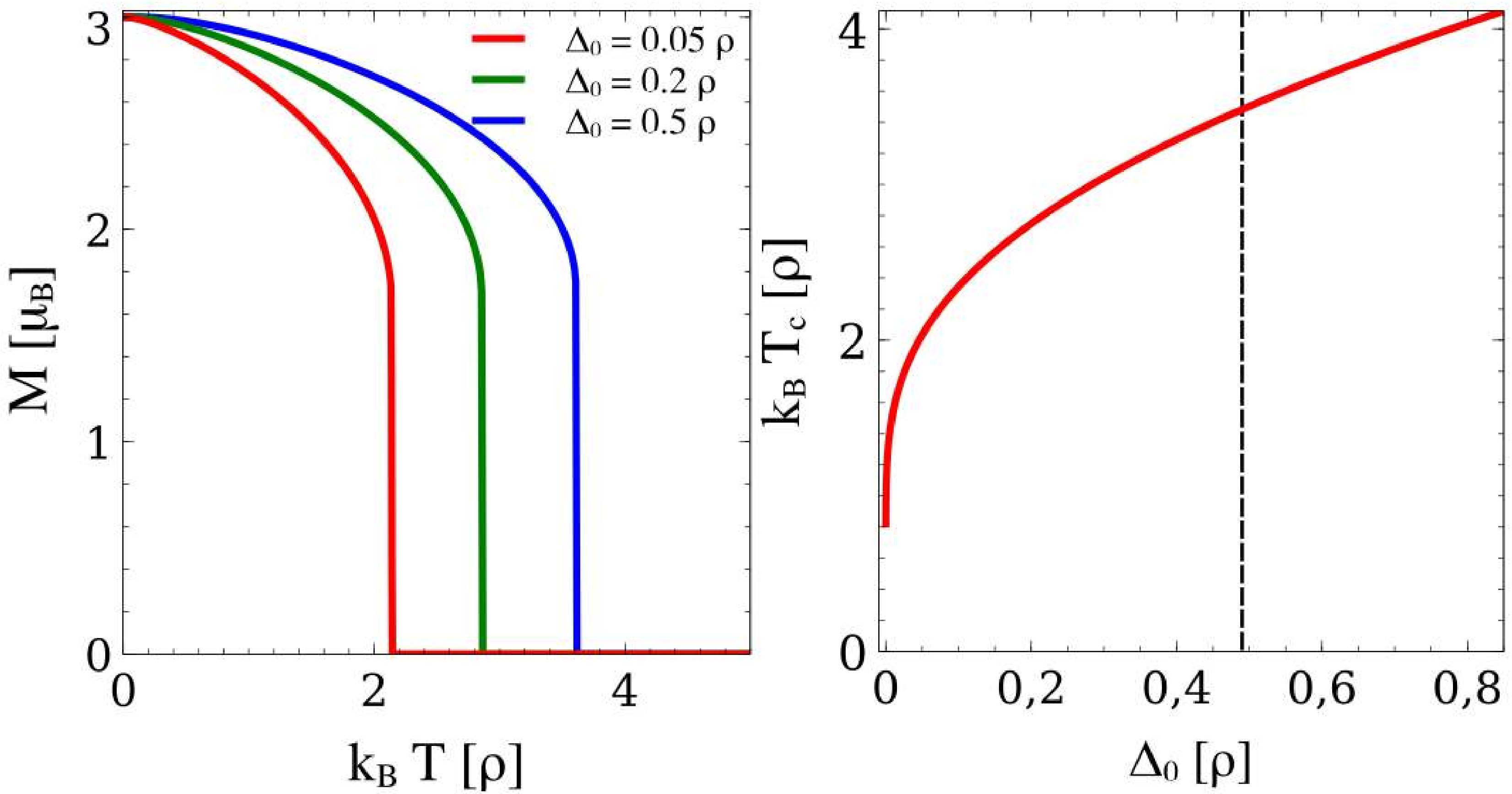
$$JS \to J(S - \langle b^\dagger b \rangle) = JM(T)$$

$$\lambda S \to \lambda(S - \langle b^\dagger b \rangle) = \lambda M(T)$$

**Selfconsistent magnetization at finite temperature**

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

# The critical temperature




**The value of the spin wave gap determines the critical temperature**

# Take home

- $CrI_3$ is a 2d realization of the XXZ model
- Iodine is the ultimate source of magnetic anisotropy in this material

***2D Materials. 4 035002 (2017)***

  - ***2d magnetic materials open the venue to engineer competing orders in 2d stacks***

**Thank you!!!**

Spinograph

Spintronics in Graphene