On the origin of magnetic anisotropy in two dimensional CrI_3

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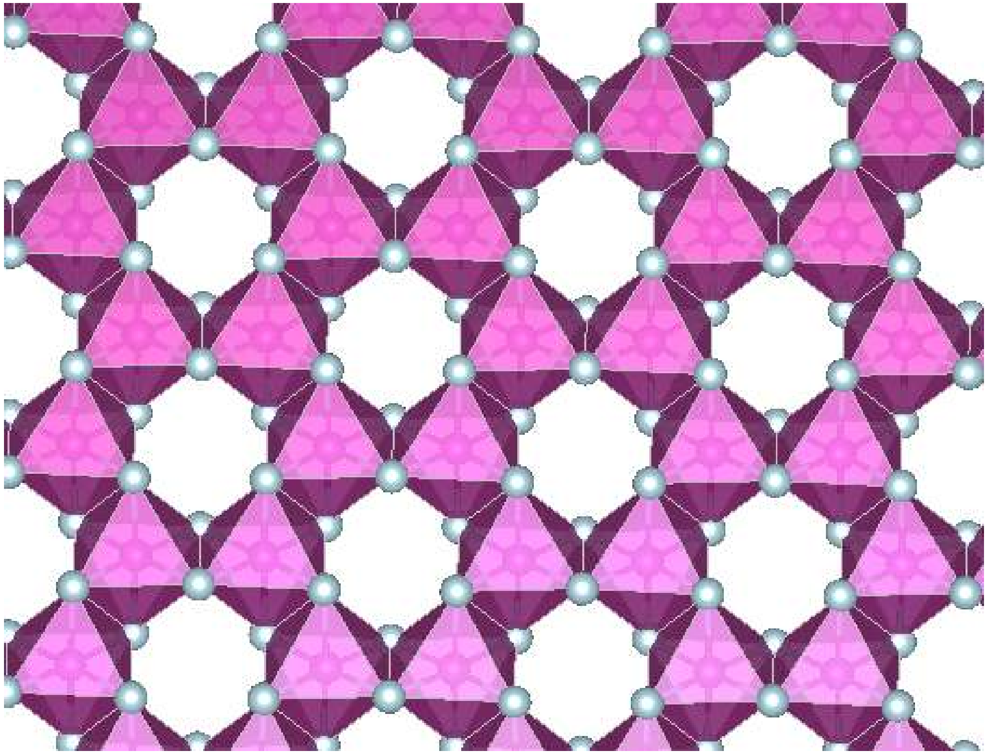


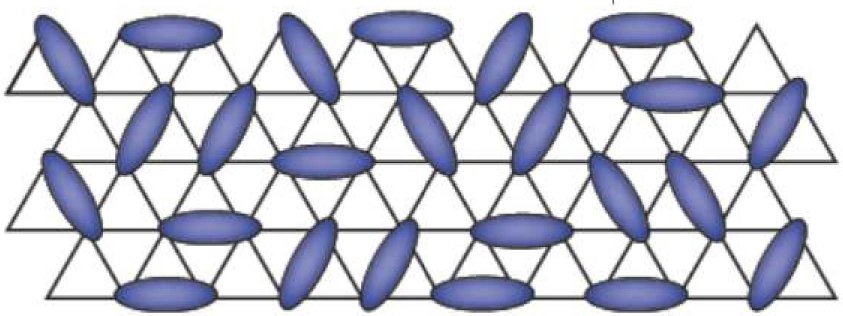
arXiv:1704.03849

Thursday 15th June, CDT Conference, Cambridge 2017

2d magnets join the game

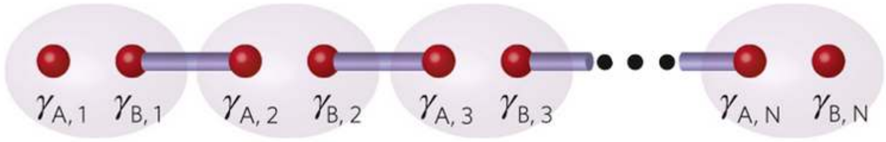
Quantum spin liquid

2d magnet + frustration



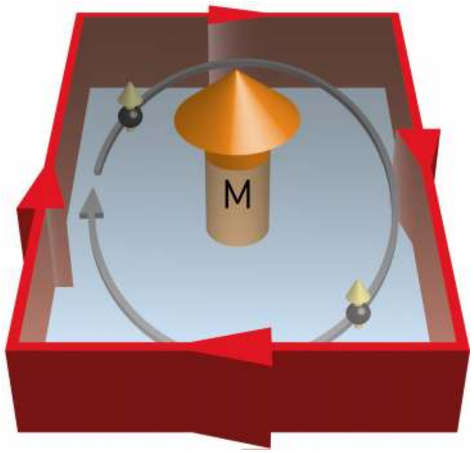
2d topological superconductor

2d magnet + Rashba+ superconductor



Quantum Anomalous Hall insulator

2d magnet + 3d topological insulator



Magnetism in two dimensions

A perfect isotropic system does not order at finite temperature in 2d (Mermin-Wagner theorem)

Magnetic anisotropy is a must for 2d magnetism

Two iso-structural compounds

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?

Ferromagnetism in 2d CrI_3

nature

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

B.Huang et al, Nature 2017

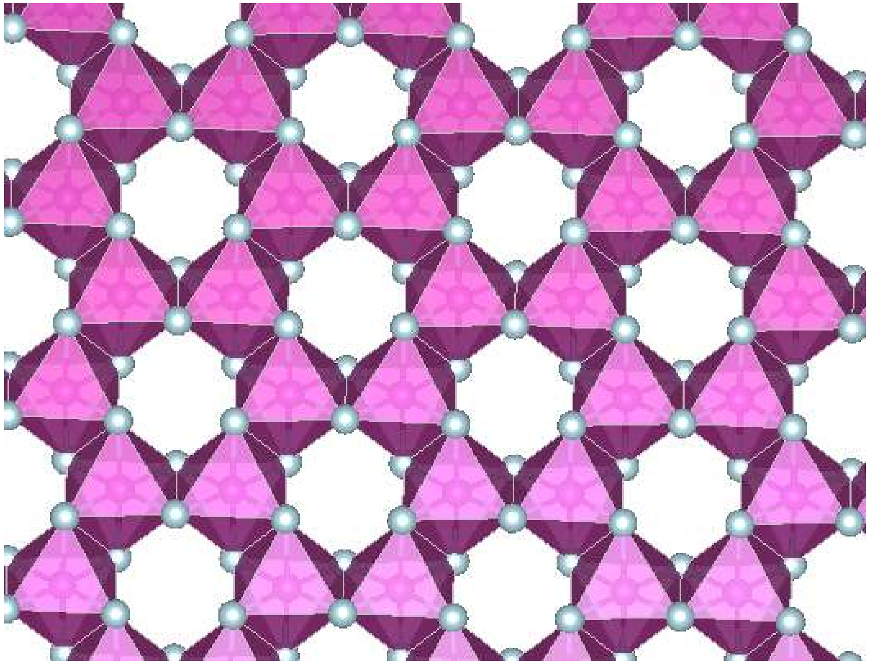
Two simple open questions

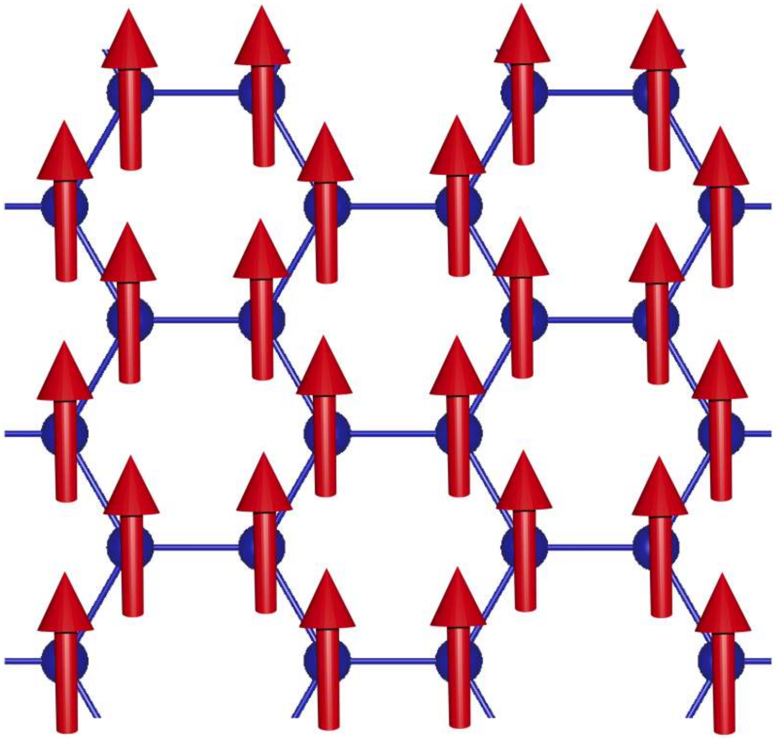
<u>Off-plane easy axis, are they Ising spins?</u>
<u>Critical temperature of 45 K, but what controls this?</u>

Is CrI_3 a two dimensional Ising model?

If not, what is the right model for CrI_3?

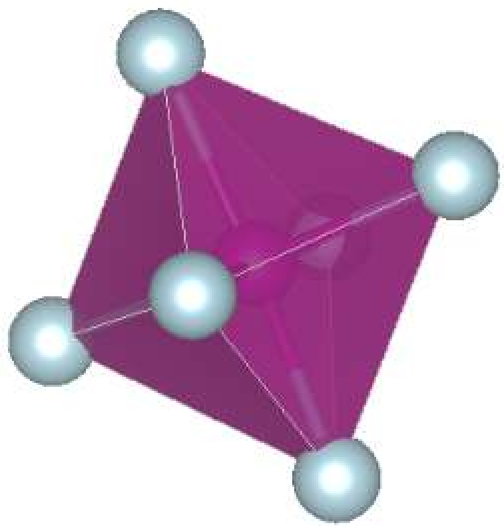
CrI_3, ferromagnetic honeycomb lattice



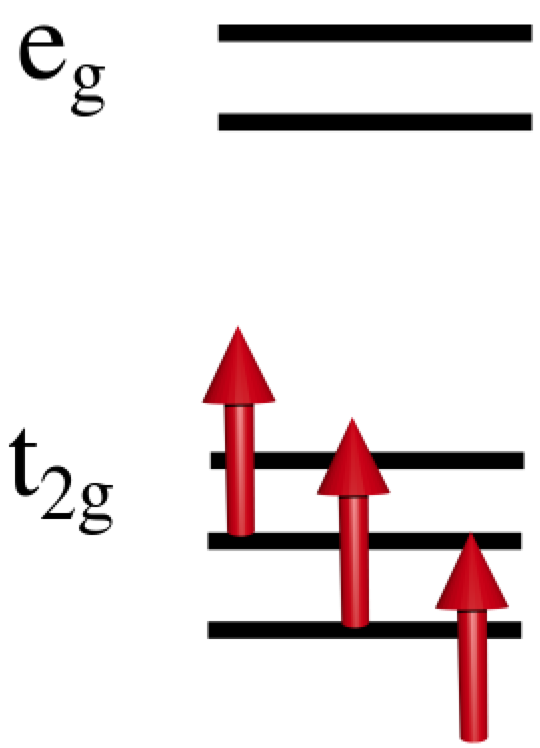


Each chromium atom contributes with S=3/2

Electronic filling and anisotropy



Octahedral iodine environment

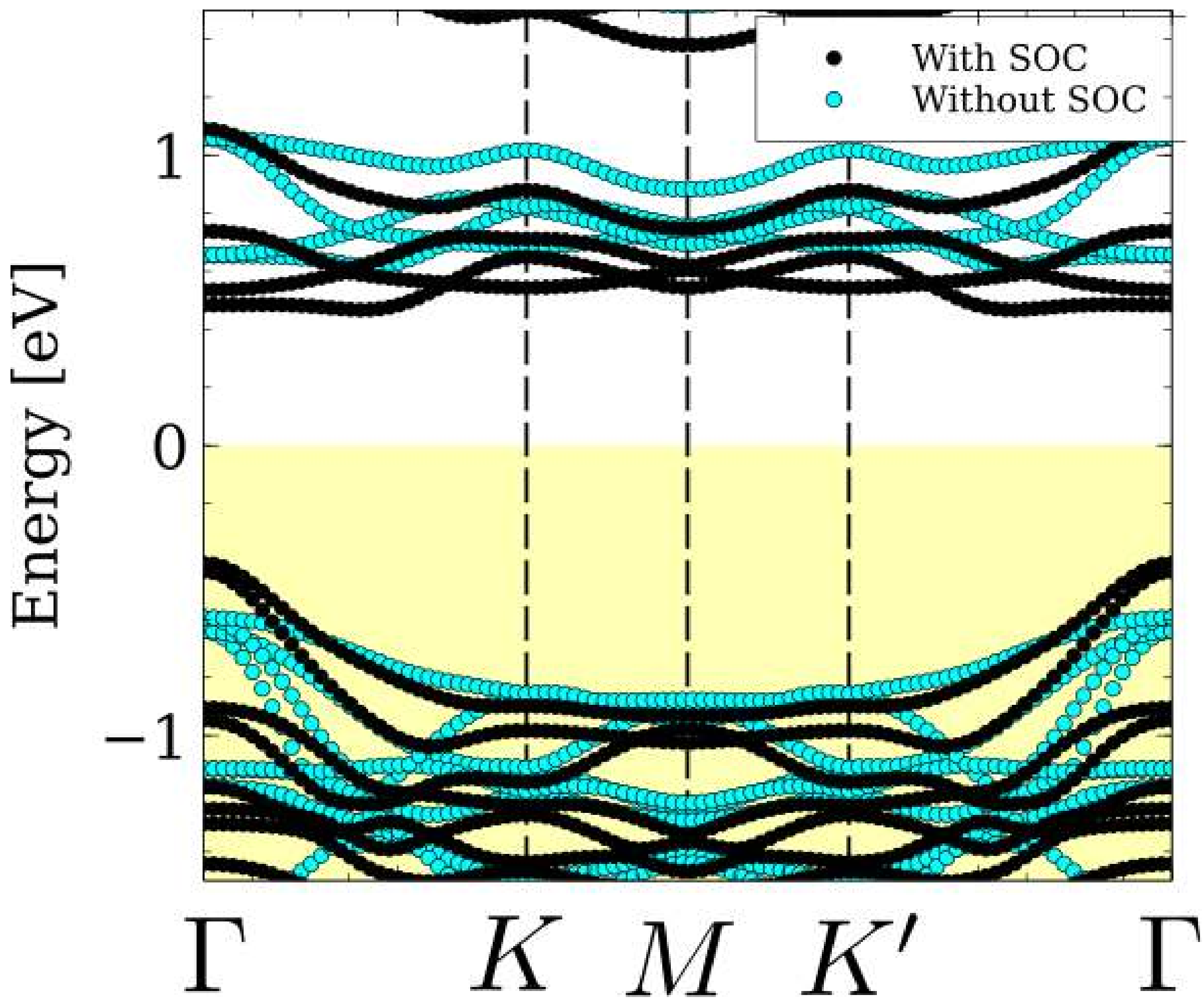


Quenched angular momenta

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

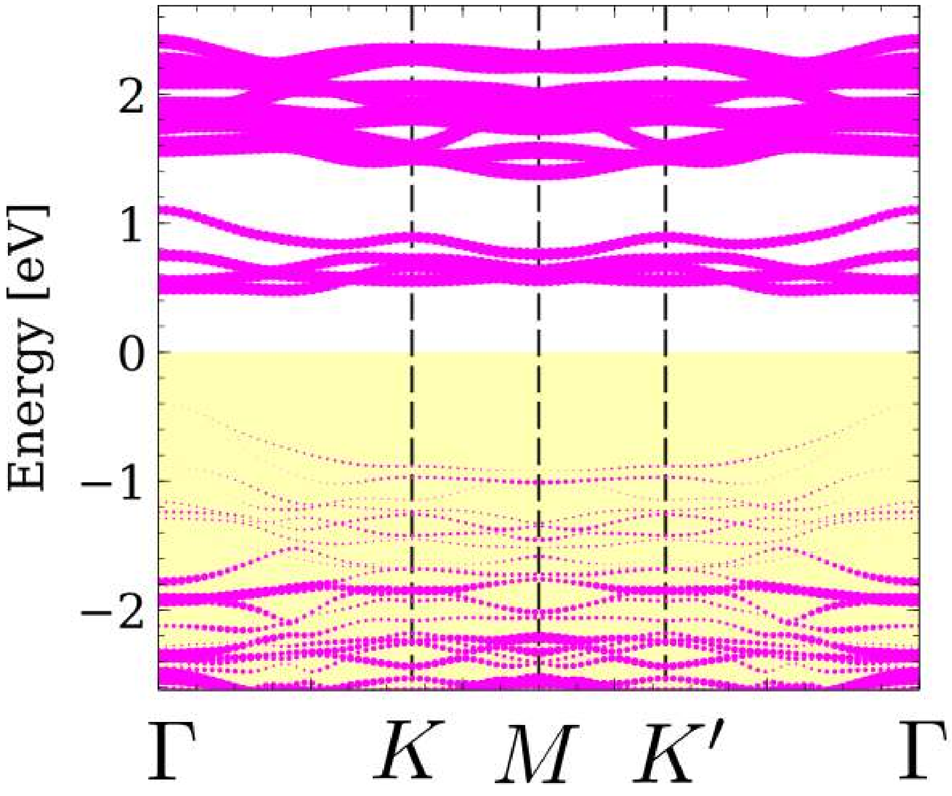


Elk LAPW code

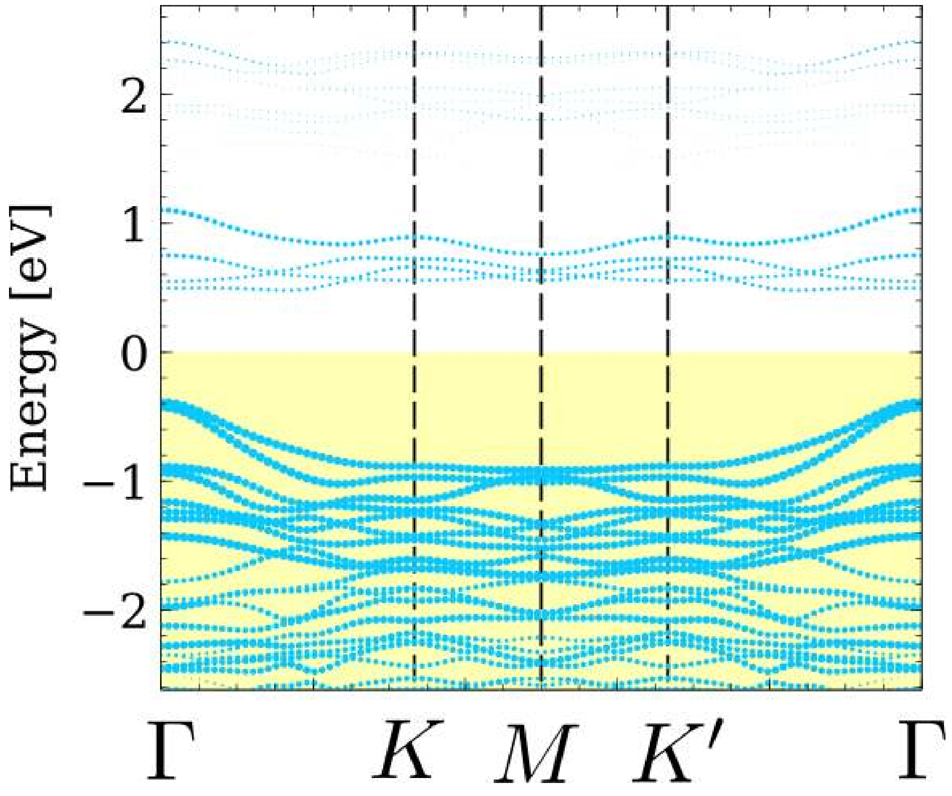
Effect of SOC in the band structure is extremely large

Weights over the atoms in CrI_3

Weight on chromium



Weight on iodine



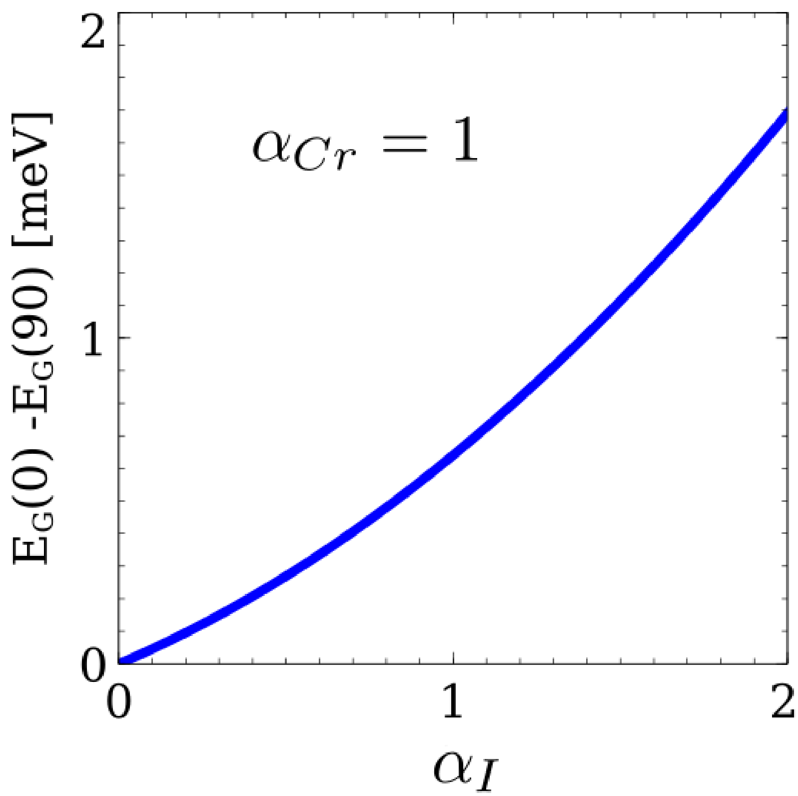
There is a sizable contribution of iodine above and below the gap

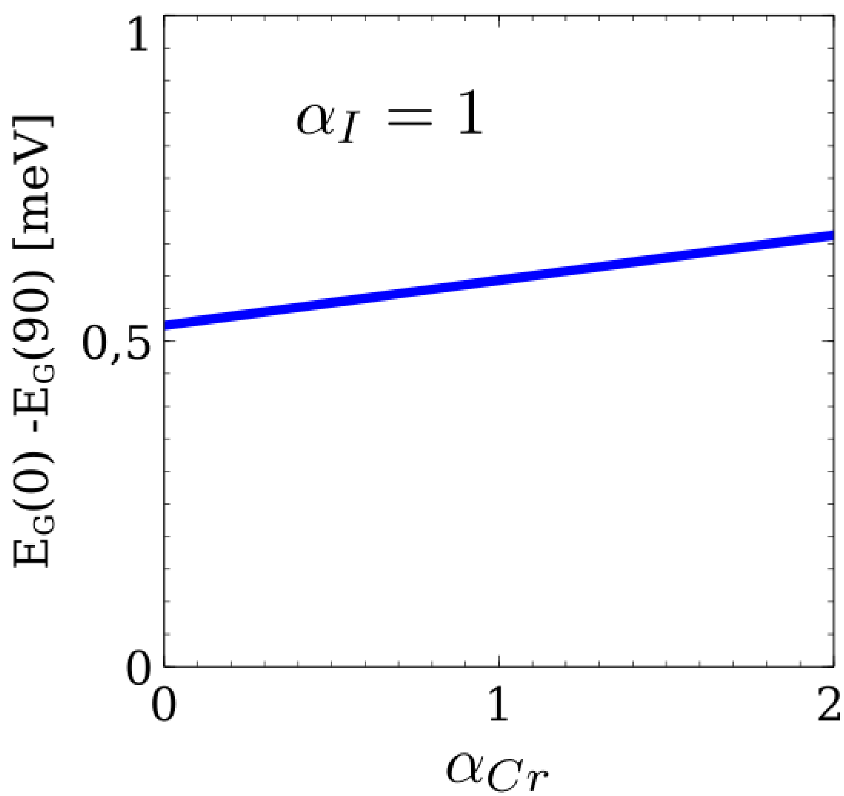
Is iodine a key to determine the magnetic properties?

Magnetic anisotropy in CrI_3

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

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





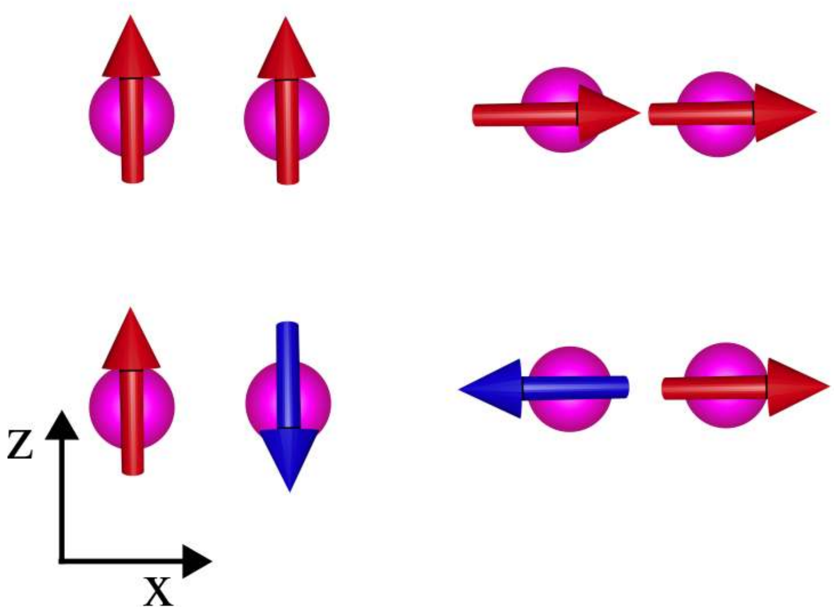
The major contribution to magnetic anisotropy comes from iodine

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**



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

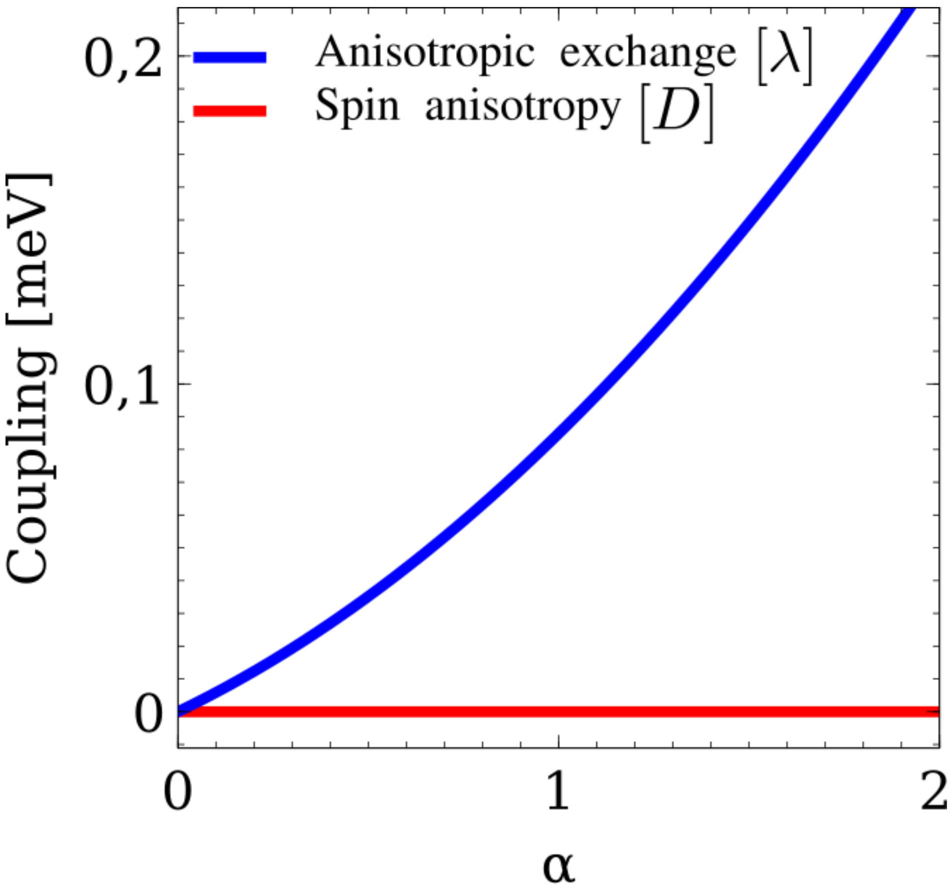
The true spin model for CrI_3

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

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



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

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 Hamiltonian

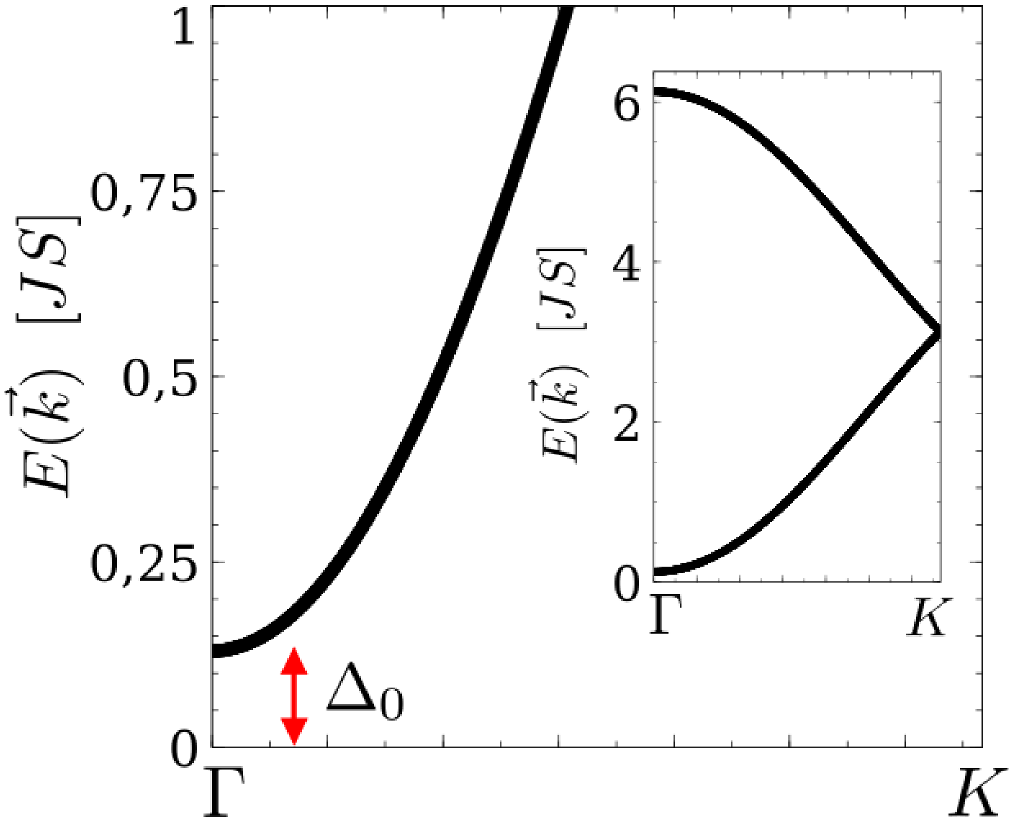
$$\mathcal{H} = -\left(\sum_i D(S_i^z)^2 + \frac{J}{2}\sum_{i,i'}\vec{S}_i\cdot\vec{S}_{i'} + \frac{\lambda}{2}\sum_{i,i'} S_i^z S_{i'}^z\right),$$

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

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

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

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

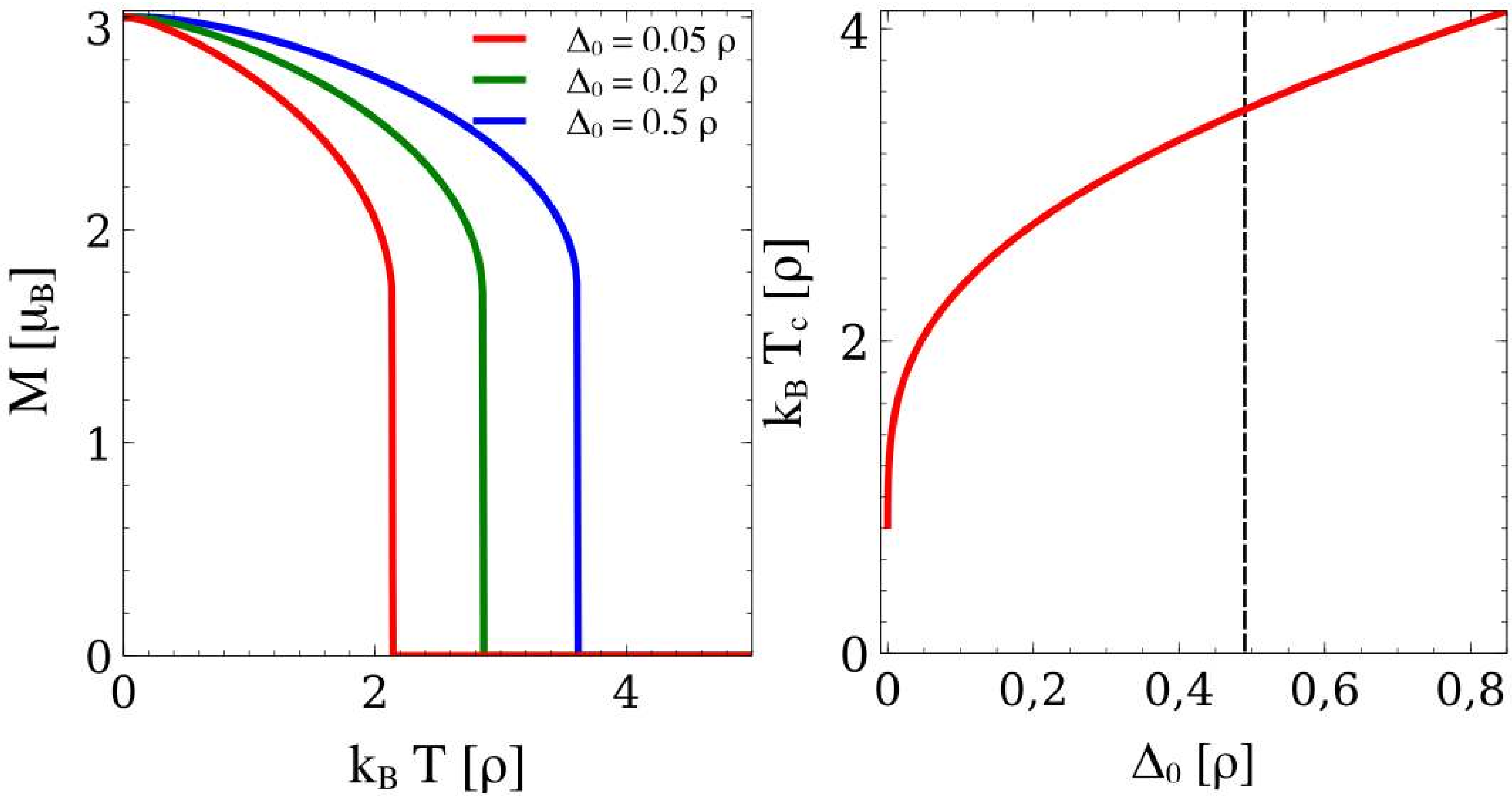
$$JS \rightarrow J(S - \langle b^\dagger b \rangle) = JM(T)$$

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

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

Thank you!!!

Spinograph
Spintronics in Graphene