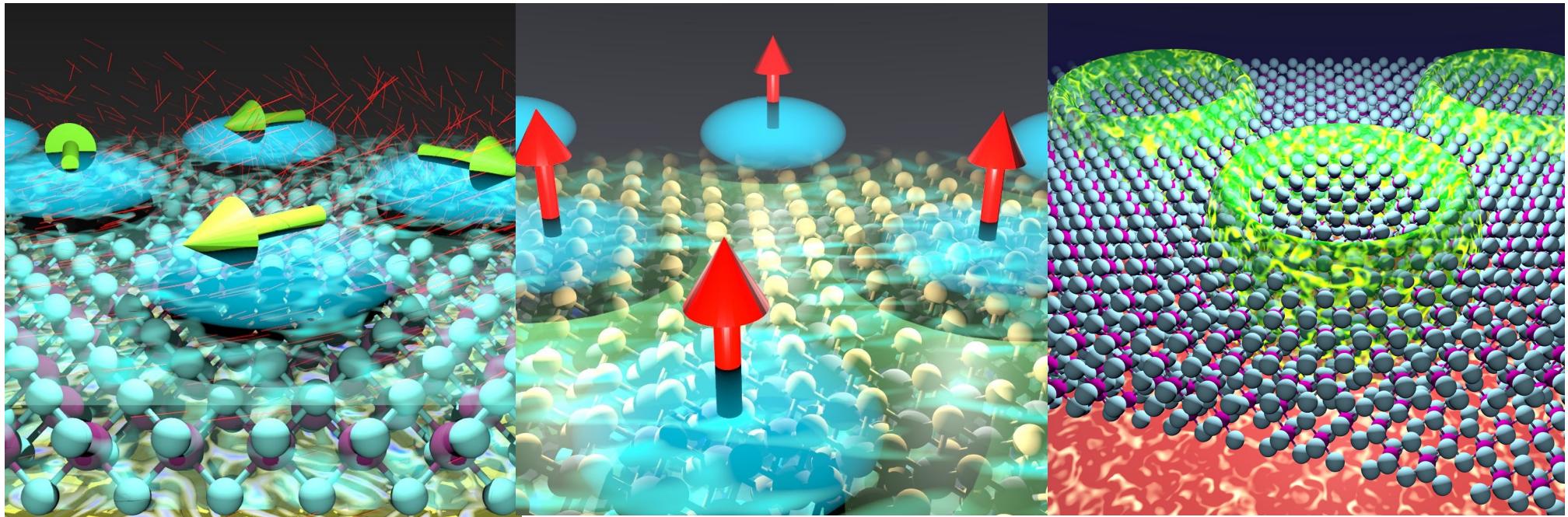


2D magnetism - Theory

Jose Lado

Department of Applied Physics, Aalto University, Finland



Advanced Physics of van der Waals heterostructure, Roscoff, France
30th September 2023

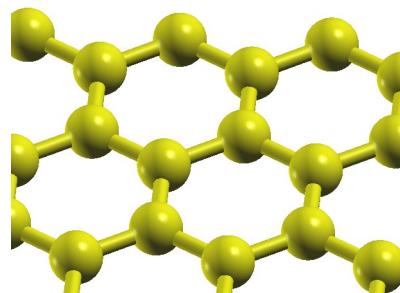
Today's plan

- General introduction to 2d magnets
- Origin of magnetic interactions: exchange and superexchange
- Magnons and the role of magnetic anisotropy
- 2D multiferroics
- 2D spin liquids and spinons
- 2D Kondo magnets

Families of two-dimensional materials

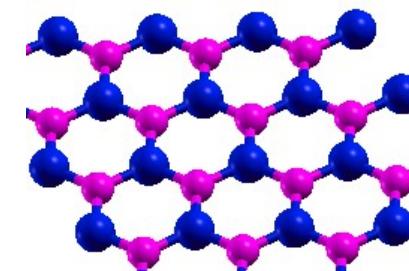
Semimetal

Graphene



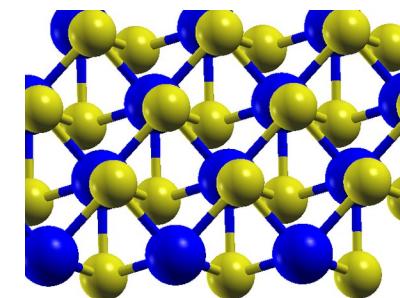
Insulator

BN



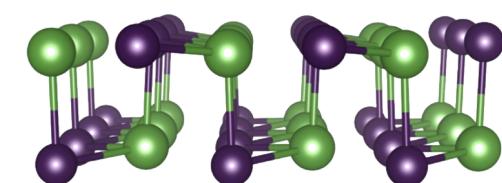
Superconductor

NbSe_2



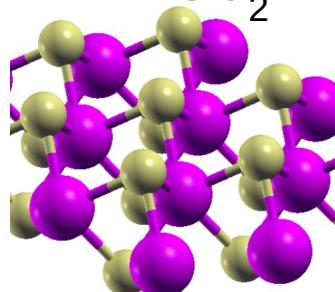
Ferroelectric

SnTe



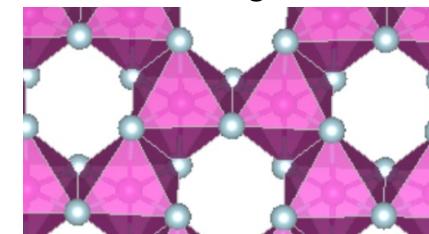
Semiconductor

WSe_2



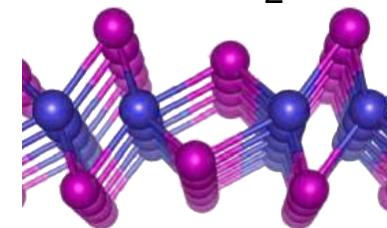
Ferromagnet

CrI_3



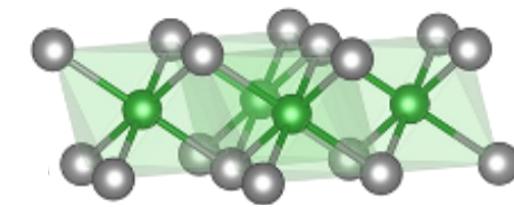
Quantum spin Hall insulator

WTe_2



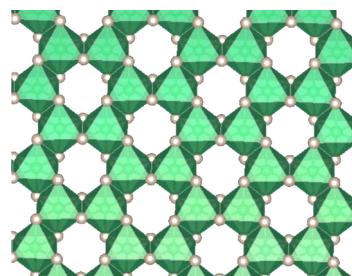
Multiferroic

NiI_2



Van der Waals magnetic materials

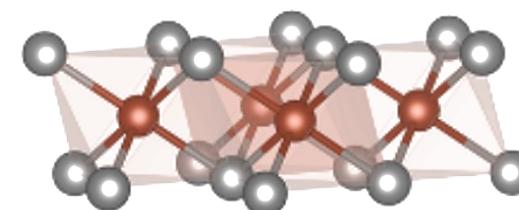
Ferromagnet,
antiferromagnets



CrI_3 , CrCl_3 , CrBr_3

Break time-reversal

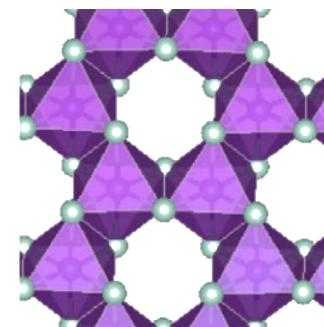
Multiferroics



NiI_2

Break time-reversal
and inversion symmetry

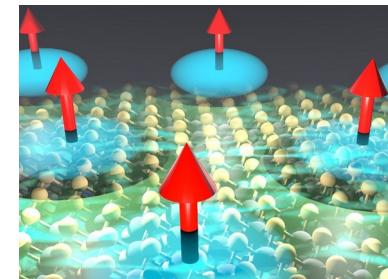
(proximal)
Quantum
spin-liquids



RuCl_3 , 1T-TaS₂

Do not break time-reversal

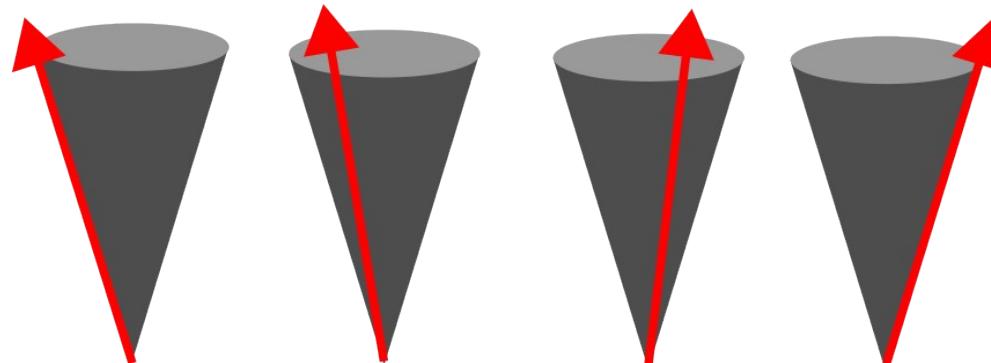
Heavy-fermion
Kondo insulators



1T-TaS₂/1H-TaS₂

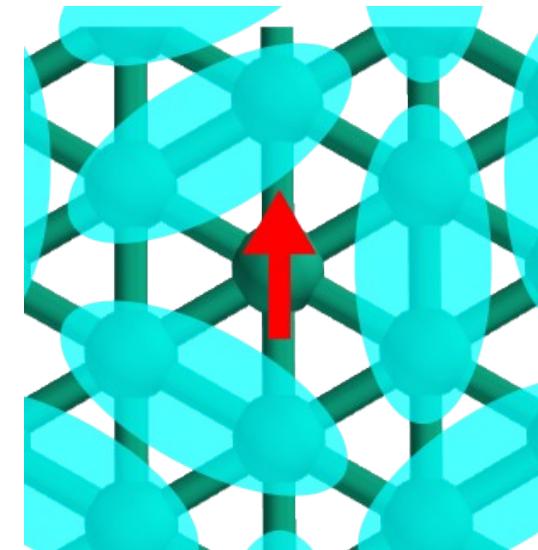
Emergent excitations in van der Waals magnets

Magnons



$S=1$
No charge

Spinons



$S=1/2$
No charge

Open-source software for
magnetic and electronic
properties

Two computational tools

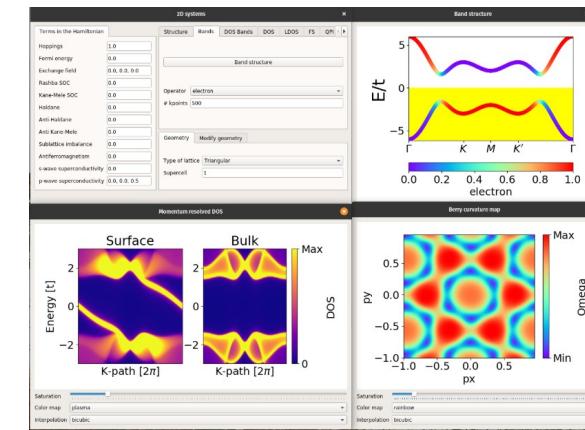
pyqula (Python library)

```
from pyqula import geometry
g = geometry.honeycomb_lattice()
h = g.get_hamiltonian()
h.add_rashba(0.2) # Rashba spin-orbit coupling
h.add_zeeman([0.,0.,0.6]) # Zeeman field
from pyqula import topology
(kx,ky,omega) = h.get_berry_curvature() # compute Berry curvature
c = h.get_chern() # compute the Chern number
```

- Python library
- Ideal for complex models/calculations
- For writing in Python

<https://github.com/joselado/pyqula>

Quantum-lattice (user interface)



- User-friendly interface for tight binding models
- Ideal for simple models and quick checks
- Fully interface-based, no scripting

<https://github.com/joselado/quantum-lattice>

Pyqula: a Python library for interacting electronic models

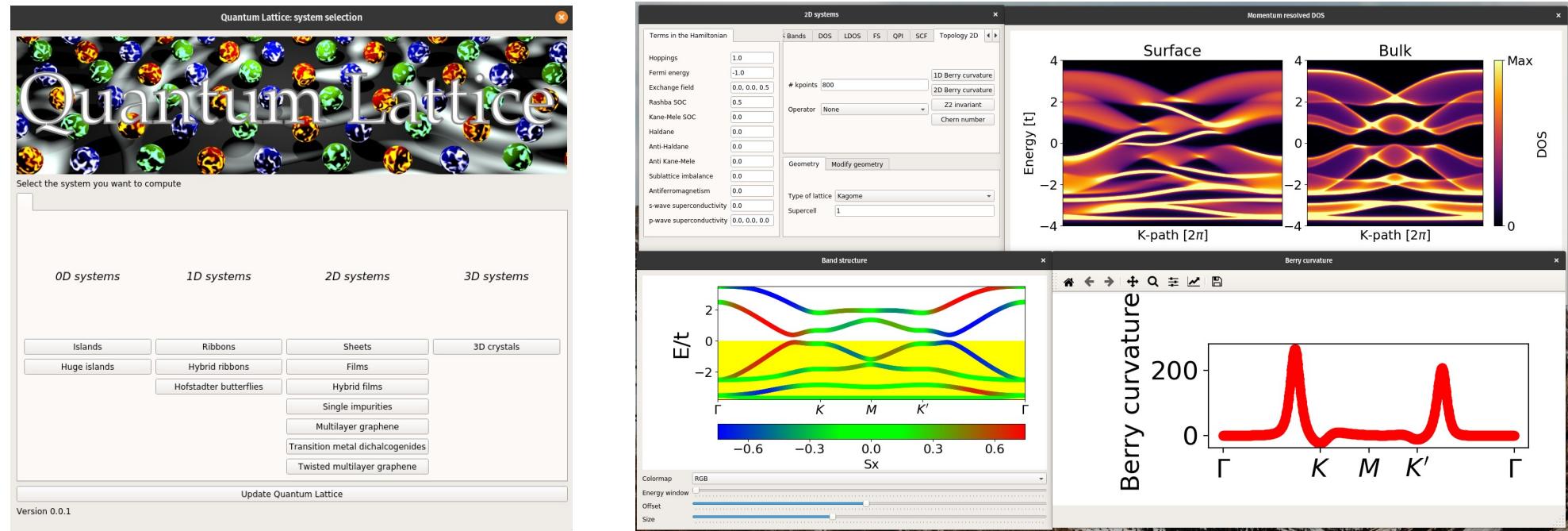
- High level user friendly library for Python to solve tight-binding models
- Focus on:
 - Easy to use
 - Performing complex calculations with minimal effort
 - Full compatibility of different calculation modes

Jupyter-Notebook for magnetic 2d materials

https://github.com/joselado/roscff_summer_school_2023



Quantum Lattice: A user interface to compute electronic properties



Quantum Lattice: open source interactive interface for tight binding modeling

<https://github.com/joselado/quantum-lattice>



A reminder about theory tools:
second quantization,
electronic structure
and mean-field theory

How do we describe quantum matter?

We use quantum mechanics to understand electrons in materials

$$H|\Psi\rangle = -i\partial_t|\Psi\rangle$$

Hamiltonian Wavefunction of the system

Two main kinds of phenomena can emerge

Single particle phenomena

Many-body phenomena

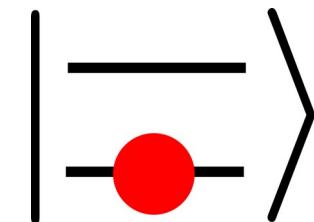
Two different kinds of quantum mechanical formalism

Systems where our number of particles is constant

First quantization, description based on a Hilbert space

Describes metals, semiconductors, insulators

Highly successful and easy formalism

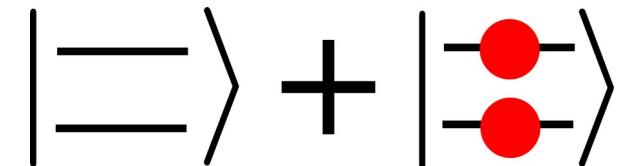


Systems where the number of particles fluctuates

Second quantization, description based on a Fock space

Describes superconductors, superfluids, correlated matter

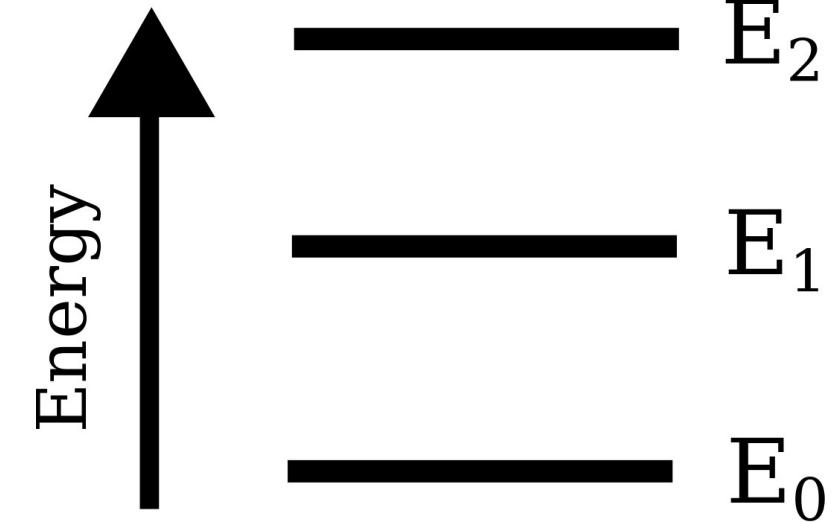
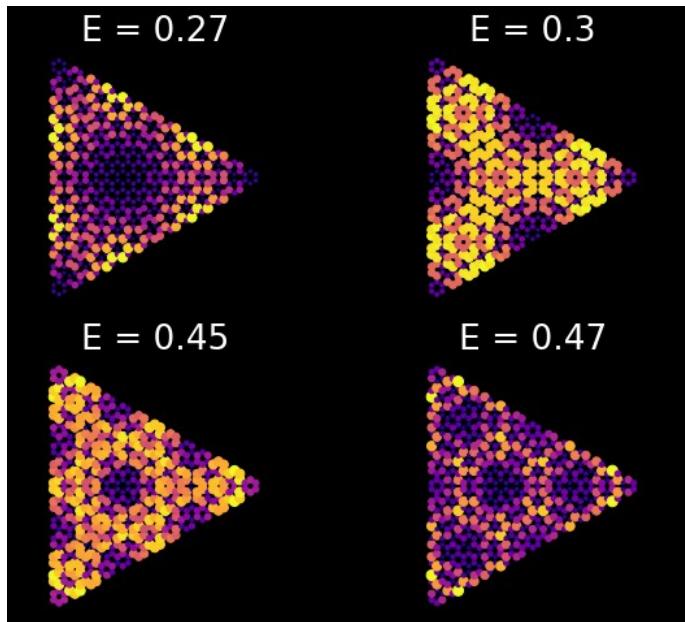
Leads to much exotic phenomena, yet also more challenging



A reminder of a simple single particle state

Particle in a box

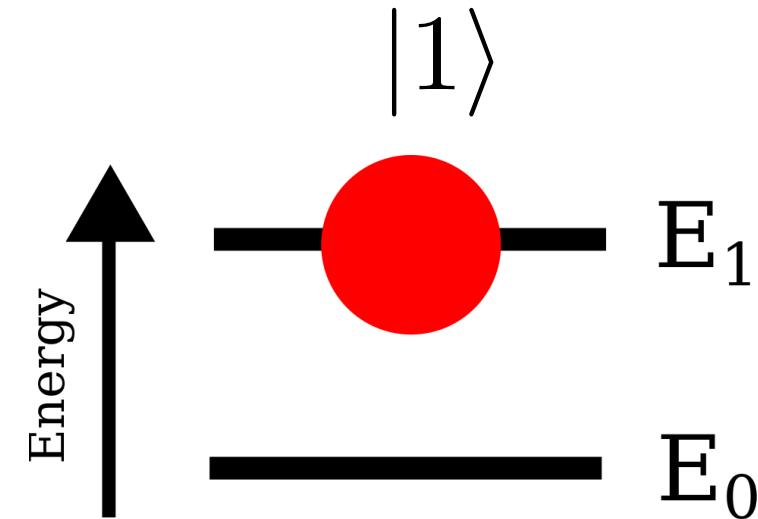
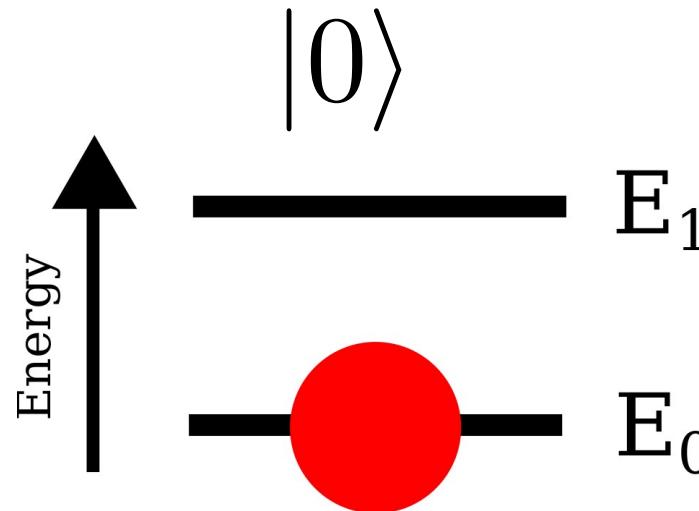
$$H|\Psi\rangle = E_n|\Psi\rangle$$



A reminder of a simple single particle state

Particle in a box

$$H|\Psi\rangle = E_n|\Psi\rangle$$

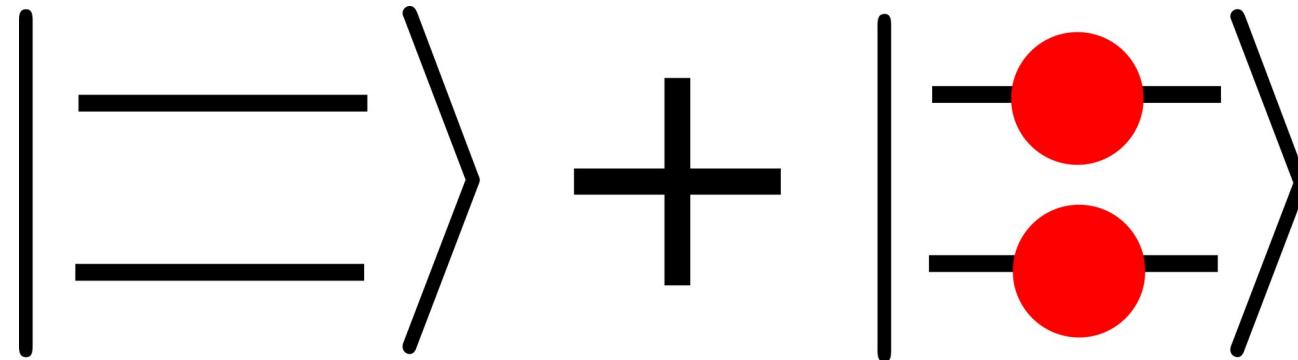


These two states describe having one particle, in one of the possible energy level

From single particle to many body

But what if our state is a combination of states with different numbers of particles?

A state having both 0 particles and 2 particles

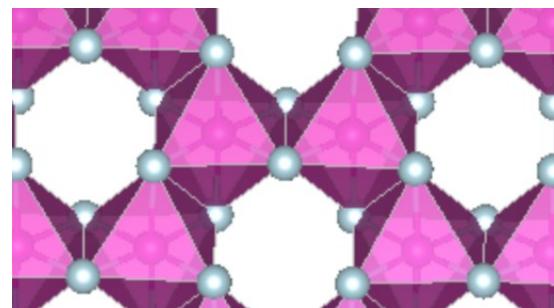


How do we describe states like these?

Non-constant number of particles

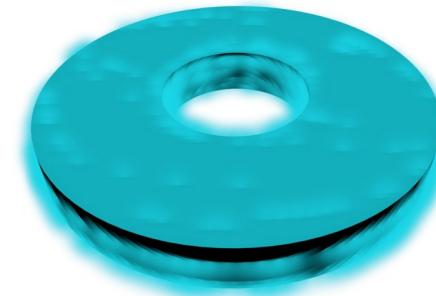
Why is this even relevant to understand materials?

Magnetic ordering of materials depends on processes in which the number of electrons fluctuates



Goodenough-Kanamori rules

The effective description of superconductors does not conserve electron number



Bogoliubov-de Gennes formalism

The idea of second quantization

Define operators that can create or destroy particles

c_i Annihilation operator, destroys a particle in site i

c_i^\dagger Creation operator, creates a particle in site i

The empty vacuum state $|\Omega\rangle$ is defined as $c_i |\Omega\rangle = 0$

The Hamiltonian is written in terms of creation and annihilation operators

$$H = c_0^\dagger c_1 + h.c.$$

The idea of second quantization

Lets see some examples using the two-levels presented before

$$|\Omega\rangle = \left| \begin{array}{c} \text{---} \\ \text{---} \end{array} \right\rangle$$

The “vacuum” state

$$c_0^\dagger |\Omega\rangle = \left| \begin{array}{c} \text{---} \\ \text{---} \\ \text{---} \end{array} \right\rangle$$

One particle in level #0

$$c_1^\dagger |\Omega\rangle = \left| \begin{array}{c} \text{---} \\ \text{---} \\ \text{---} \\ \text{---} \end{array} \right\rangle$$

One particle in level #1

$$c_0^\dagger c_1^\dagger |\Omega\rangle = \left| \begin{array}{c} \text{---} \\ \text{---} \\ \text{---} \\ \text{---} \\ \text{---} \end{array} \right\rangle$$

Two particles in level #0 & #1

Fermionic quantum statistics in second quantization

Fermi-Dirac statistics for electrons

- Wavefunctions are antisymmetric with respect to interchanging labels
- There can only be 0 or 1 fermion per level

$$\{c_i^\dagger, c_j\} = c_i^\dagger c_j + c_j c_i^\dagger = \delta_{ij} \quad \{c_i, c_j\} = 0$$

Anti-symmetric wavefunction

$$c_0^\dagger c_1^\dagger |\Omega\rangle = -c_1^\dagger c_0^\dagger |\Omega\rangle$$

At most one fermion per site

$$c_0^\dagger c_0^\dagger |\Omega\rangle = 0$$

Different kinds of Hamiltonians

Single particle Hamiltonians

$$H = \sum_{ij} t_{ij} c_i^\dagger c_j$$

Insulators, semiconductors, metals

Many-body Hamiltonian

$$H = \sum_{ijkl} V_{ijkl} c_i^\dagger c_j c_k^\dagger c_l$$

Fractional quantum Hall states, superconductors, quantum magnets

With second quantization, both cases can be treated on the same footing

Interactions and mean field

$$H = \sum_{ij} t_{ij} c_i^\dagger c_j + \sum_{ijkl} V_{ijkl} c_i^\dagger c_j c_k^\dagger c_l$$

Free Hamiltonian *Interactions*

What are these interactions coming from?

- Electrostatic (repulsive) interactions
- Mediated by other quasiparticles (phonons, magnons, plasmons,...)

The net effective interaction can be attractive or repulsive

Magnetism is promoted by repulsive interactions

A simple interacting Hamiltonian

Free Hamiltonian

*Interactions
(Hubbard term)*

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

What is the ground state of this Hamiltonian?

$U < 0$ Superconductivity

$U > 0$ Magnetism

The weakly interacting
limit

The mean-field approximation

Mean field: Approximate four fermions by two fermions times expectation values

Four fermions
(not exactly solvable)

$$U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} \approx U \langle c_{i\uparrow}^\dagger c_{i\uparrow} \rangle c_{i\downarrow}^\dagger c_{i\downarrow} + \dots + h.c.$$

Two fermions
(exactly solvable)

$$U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} \approx M \sigma_{ss'}^z c_{i,s}^\dagger c_{i,s'} + h.c.$$

For $U > 0$
i.e. repulsive interactions

Magnetic order

$$M \sim \langle c_{i\uparrow}^\dagger c_{i\uparrow} \rangle - \langle c_{i\downarrow}^\dagger c_{i\downarrow} \rangle$$

The mean-field approximation

The non-collinear mean-field Hamiltonian

$$U c_{n\uparrow}^\dagger c_{n\uparrow} c_{n\downarrow}^\dagger c_{n\downarrow} \approx M_n^\alpha \sigma_{ss'}^\alpha c_{n,s}^\dagger c_{n,s'} + h.c.$$

Non-collinear magnetic order

$$M_n^z \sim \langle c_{n\uparrow}^\dagger c_{n\uparrow} \rangle - \langle c_{n\downarrow}^\dagger c_{n\downarrow} \rangle$$

$$M_n^x \sim \langle c_{n\uparrow}^\dagger c_{n\downarrow} \rangle + \langle c_{n\downarrow}^\dagger c_{n\uparrow} \rangle$$

$$M_n^y \sim i \langle c_{n\uparrow}^\dagger c_{n\downarrow} \rangle - i \langle c_{n\downarrow}^\dagger c_{n\uparrow} \rangle$$

A Hamiltonian for a weakly correlated magnet

Free Hamiltonian

Exchange term

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + M \sum_i \sigma_{s,s'}^z c_{i,s}^\dagger c_{i,s'}$$

Here we assume that interactions are weak (in comparison with the kinetic energy)

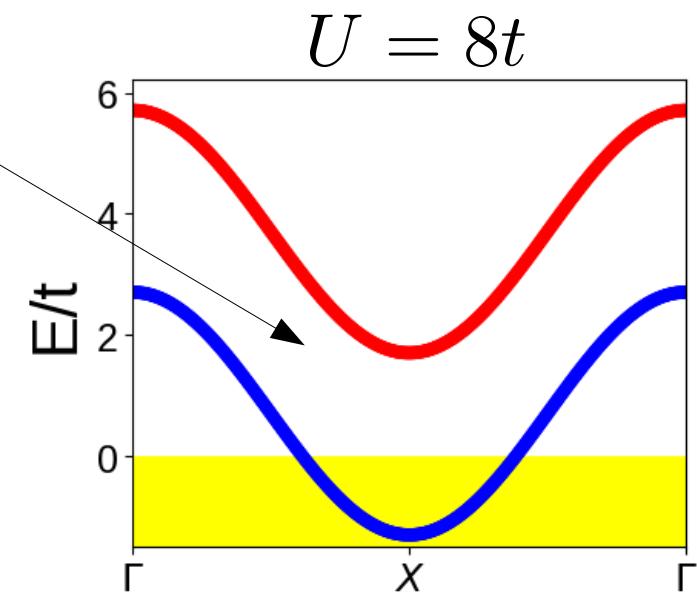
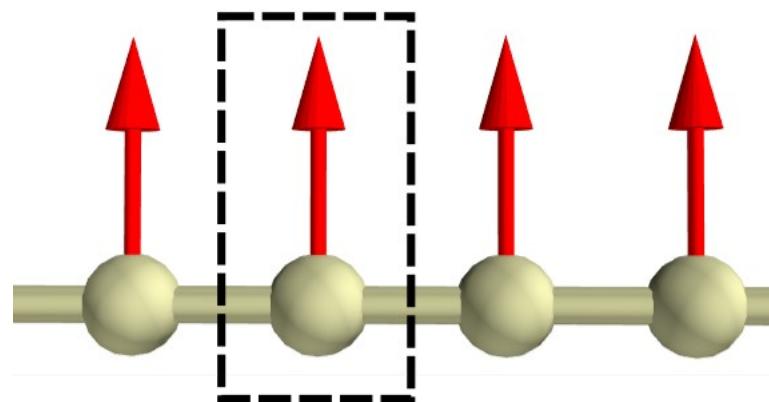
What if interactions are much stronger than the kinetic energy?

Solving the interacting model at the mean-field level in a 1D chain

We will take the interacting model and solve it at the mean field level

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

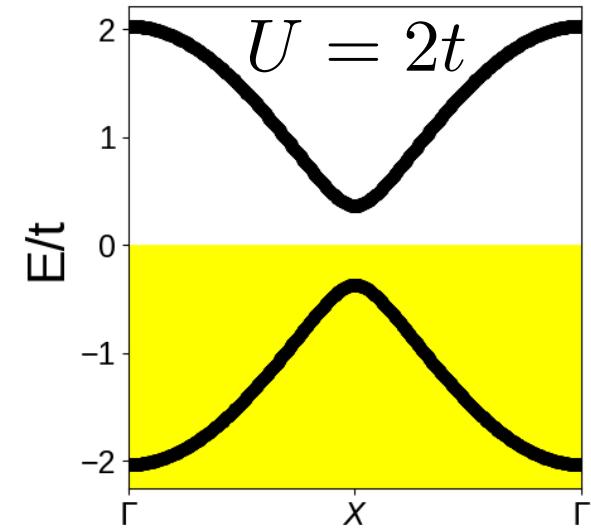
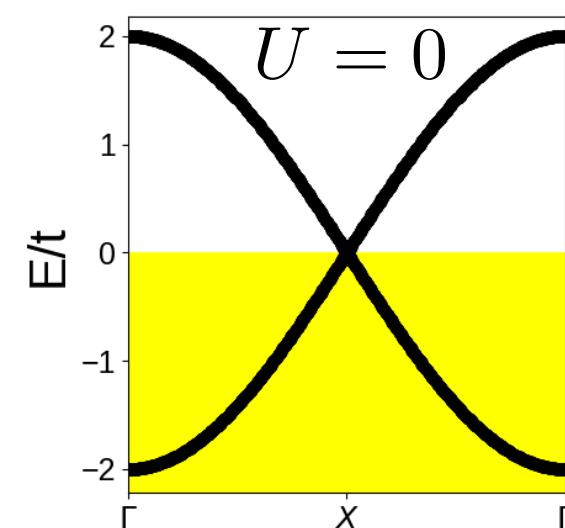
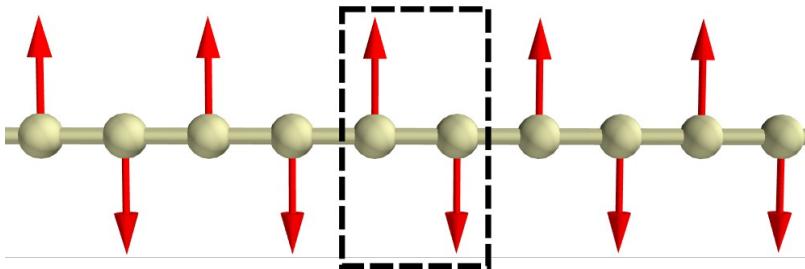
Filling 0.2 (full would be 1)
Interaction-induced splitting



Solving the interacting model at the mean-field level in a 1D chain

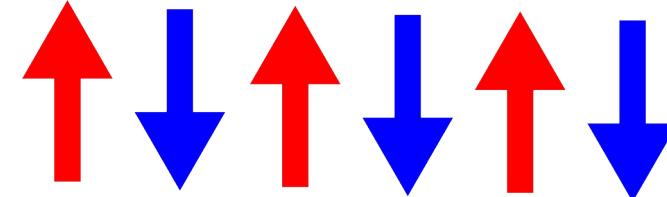
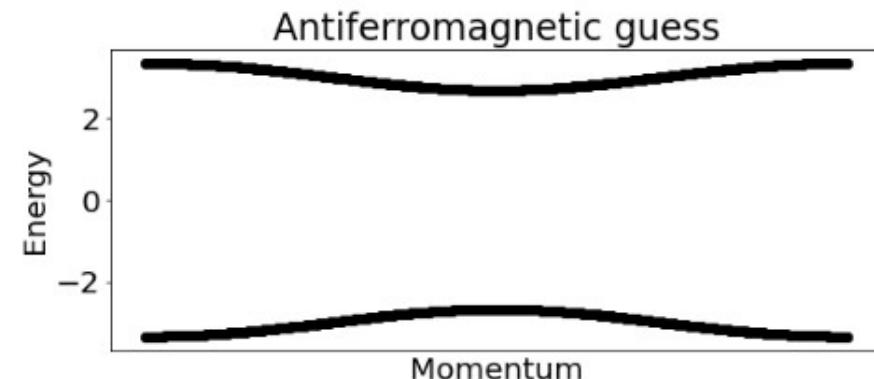
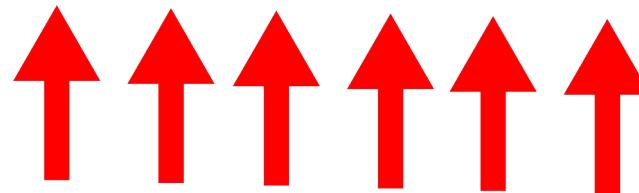
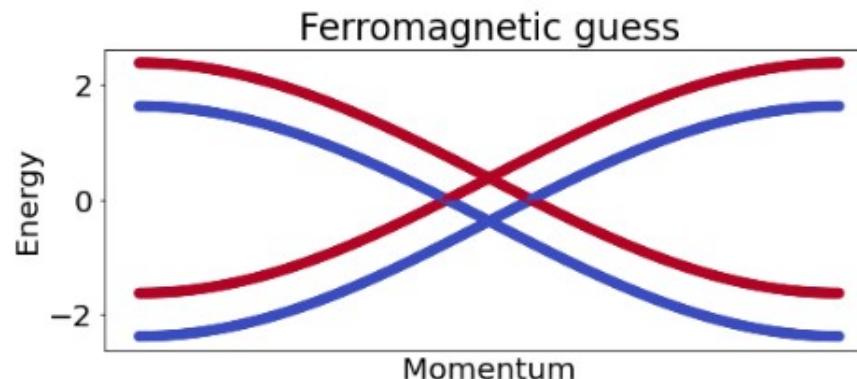
Let us do again a 1D, but now with 2 sites per unit cell and at half filling

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$



Competing solutions for a magnetic state

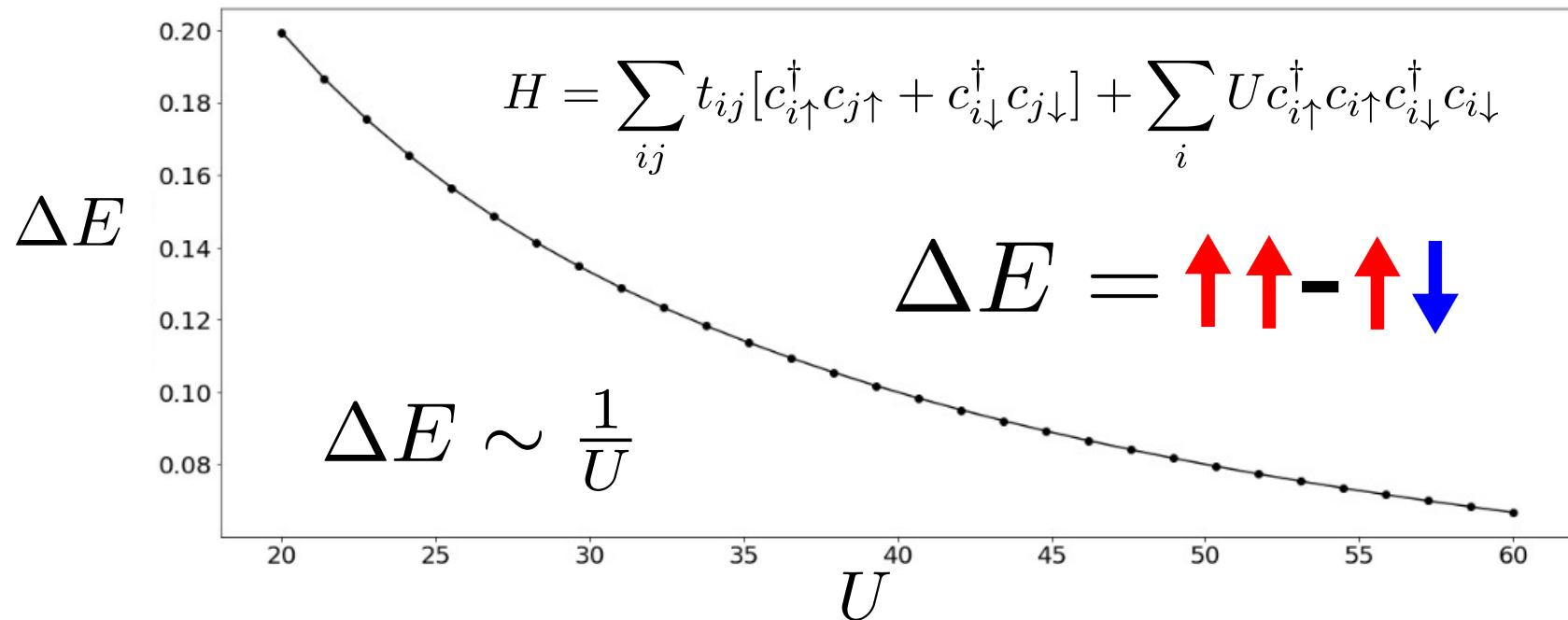
Let us now consider two selfconsistent solutions for the interacting model



Only one of them is the true ground state, but which one it is?

Competing solutions for a magnetic state

Let us now compute the energy difference between the two configurations



For strong interactions, the AF configuration always has lower energy

The critical interaction for magnetic ordering

Lets take the Hamiltonian

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

Do we have magnetism for any value of U ? $\langle S_z \rangle \neq 0$

In general, in the weak coupling limit magnetism appears when

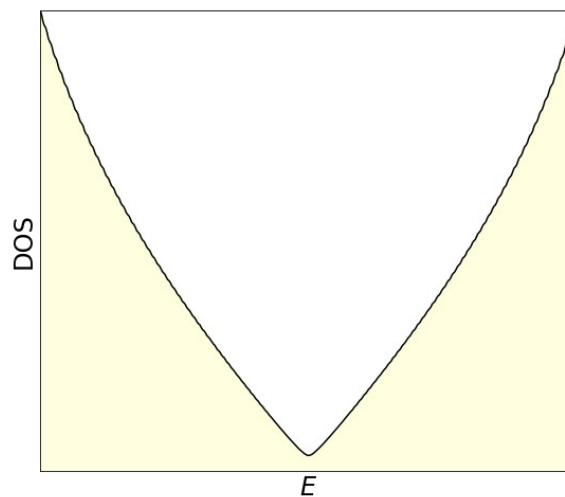
$$UD(\omega) > 1$$

Repulsive interaction

Density of states

The critical interaction for magnetic ordering

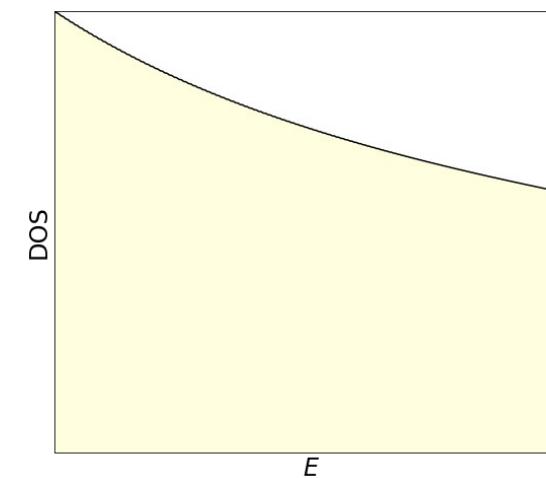
Semimetals



No low coupling instability

$$U_C \gg t$$

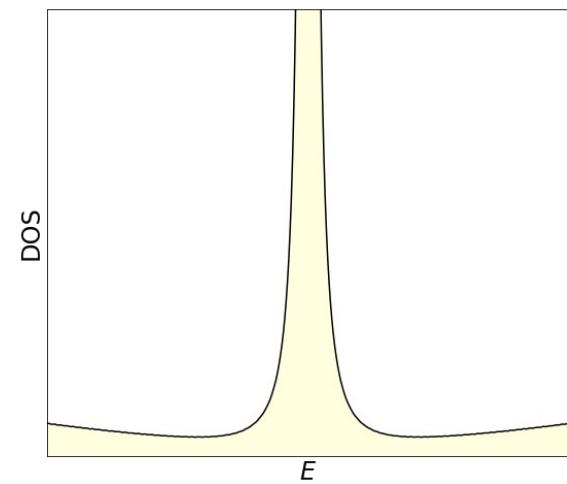
Metals



Controlled by DOS

$$U_C \sim \frac{1}{D(E_F)}$$

Flat bands

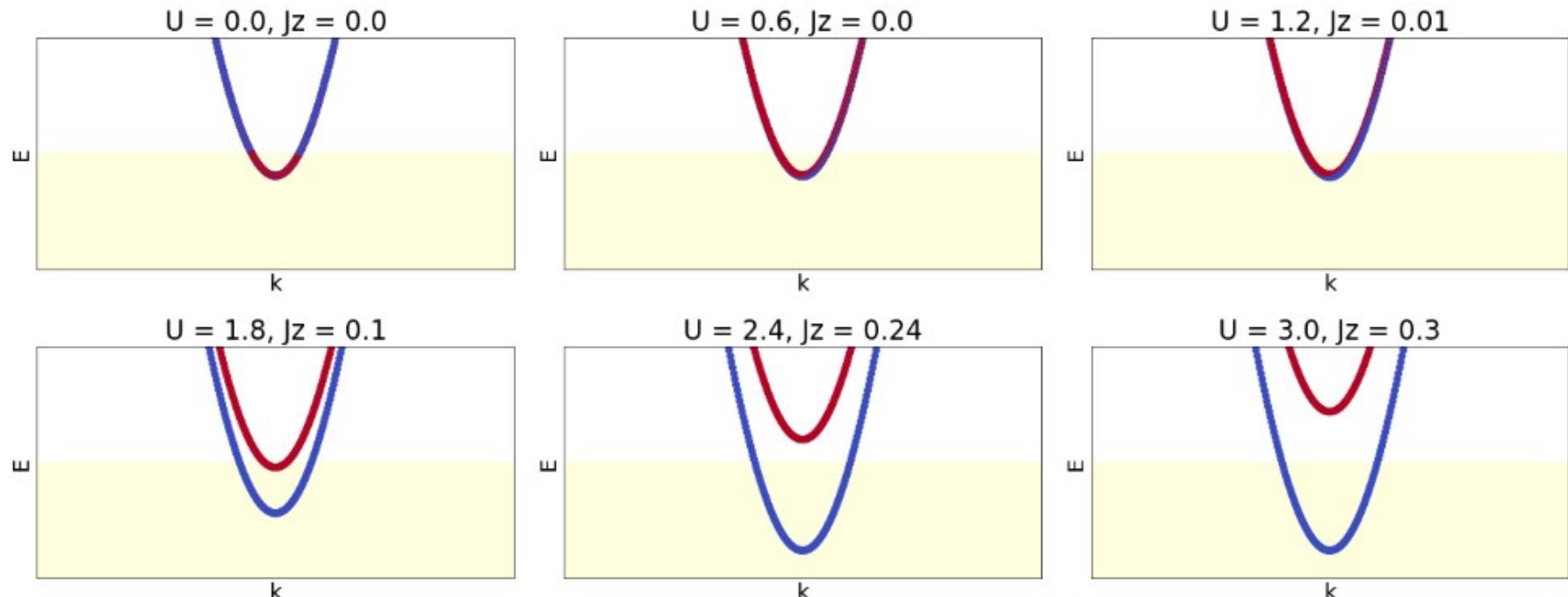


Arbitrarily small interactions

$$U_C \rightarrow 0$$

The critical interaction for magnetic ordering

Magnetic instabilities occur once interactions are strong enough



For interactions below a threshold, no magnetic order occurs

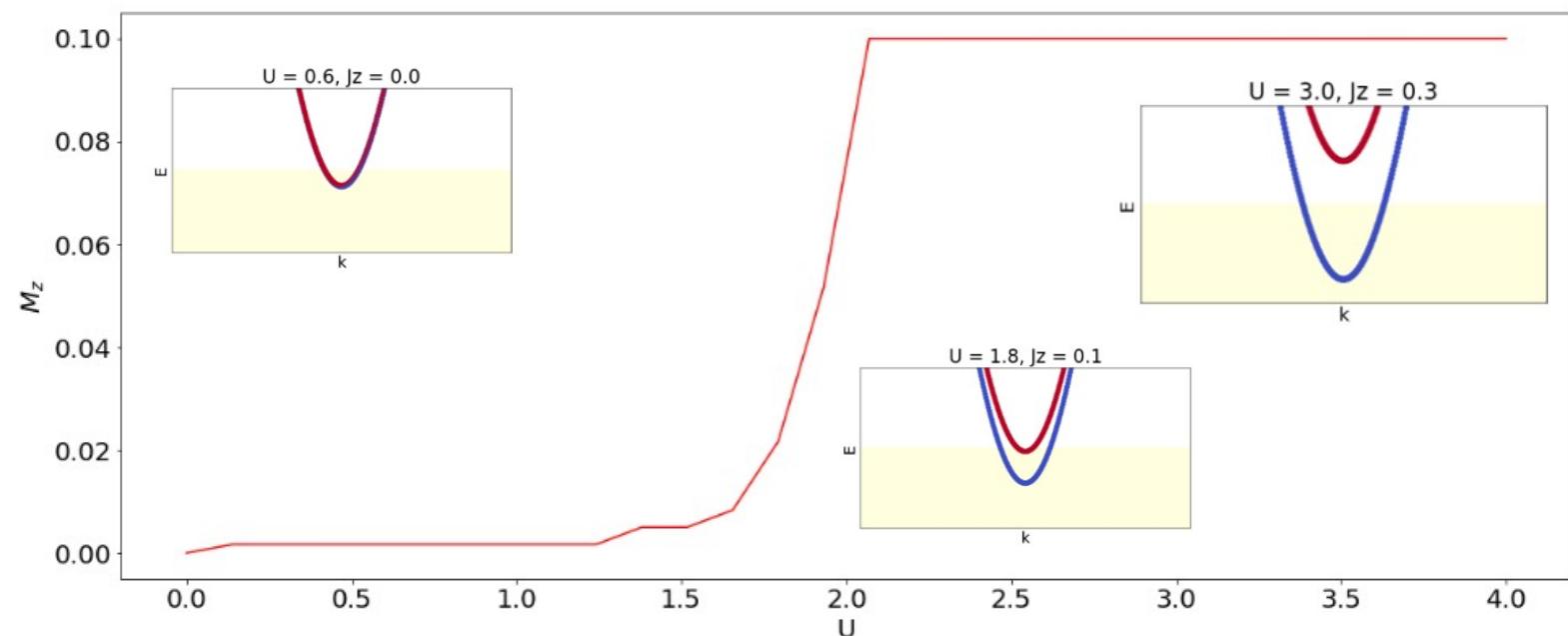
The critical interaction for magnetic ordering

Depending on the strength of interactions, we can have three different regimes

No magnetism

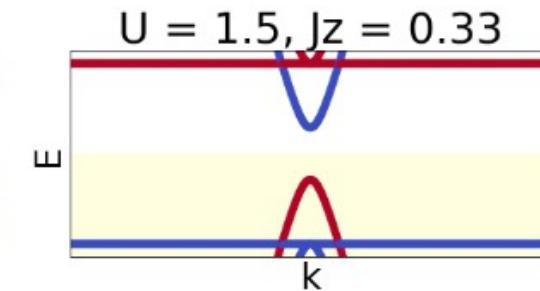
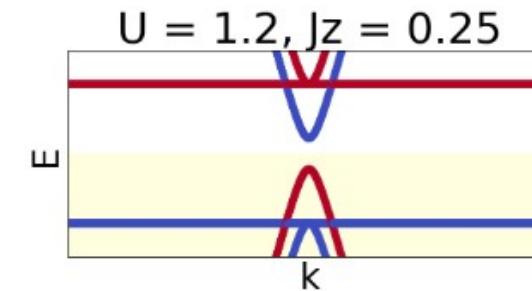
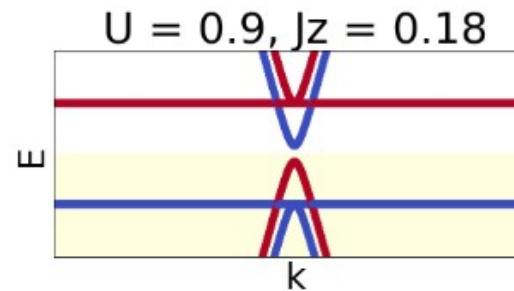
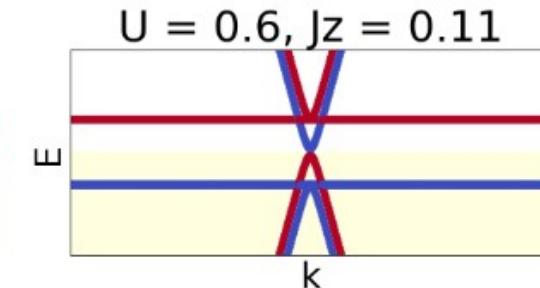
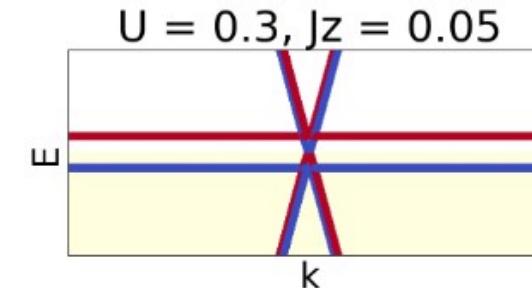
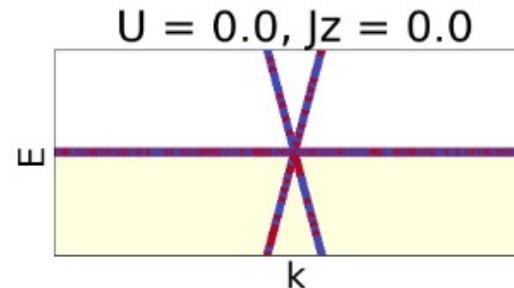
Non-saturated magnetization

Saturated magnetization (half metal)



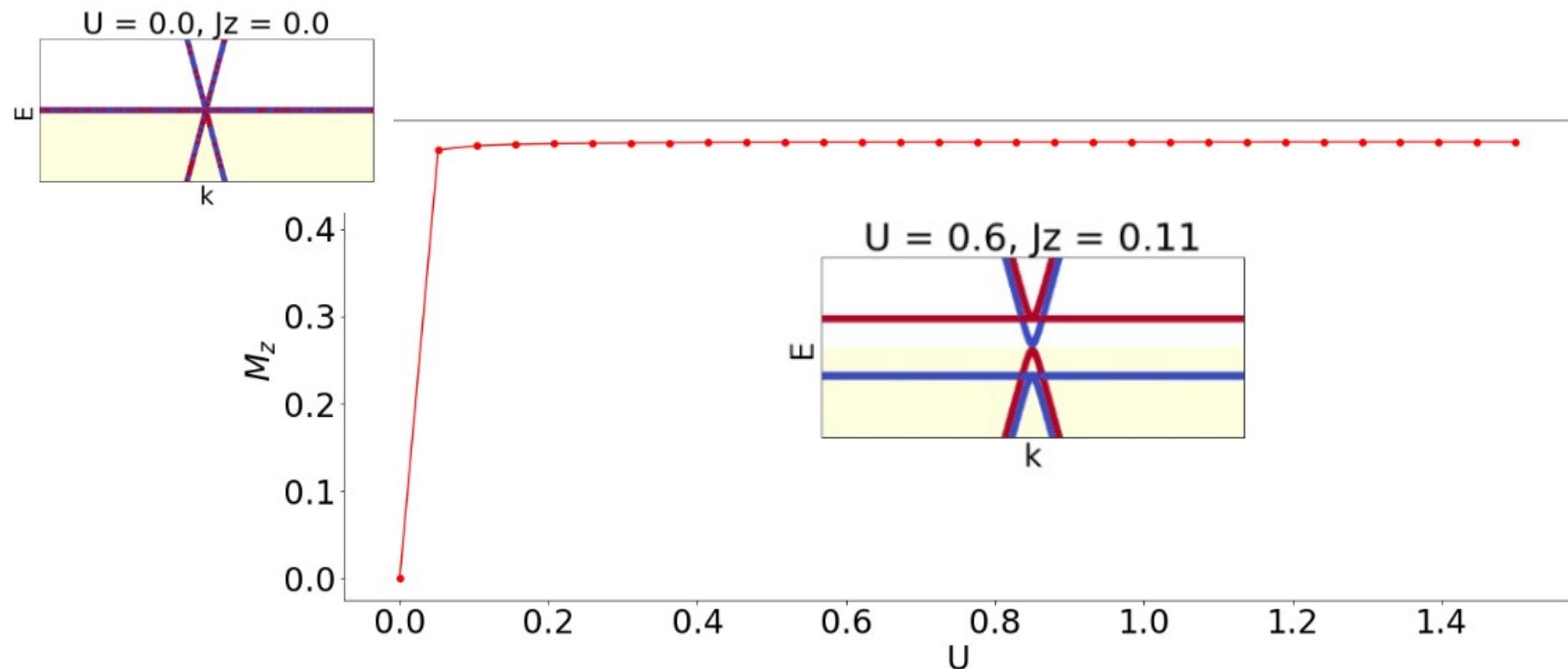
Magnetic instabilities in a flat band system

Magnetic instabilities occur for arbitrarily small interactions



Magnetic instabilities in a flat band system

In the flat band regime, any non-zero interaction gives rise to a magnetic instability

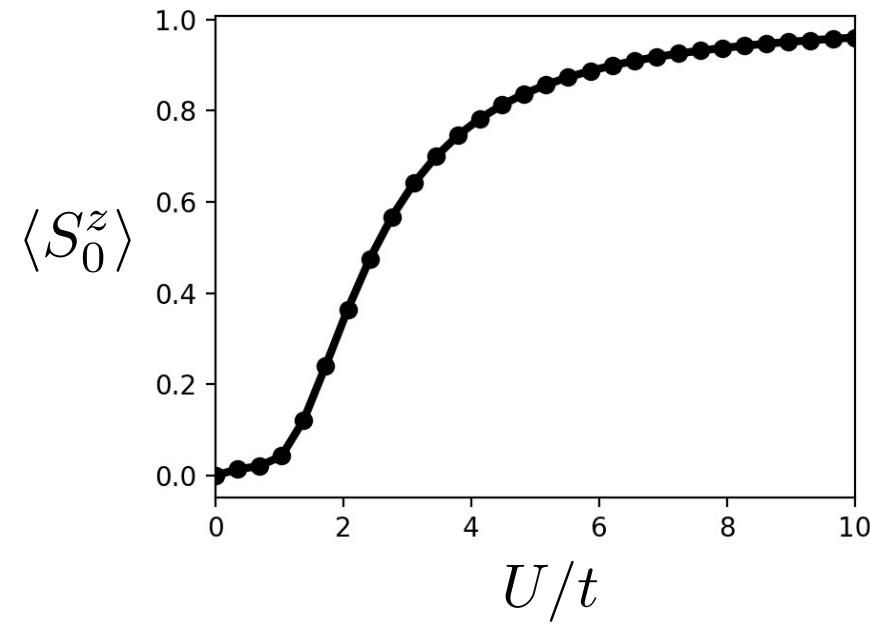
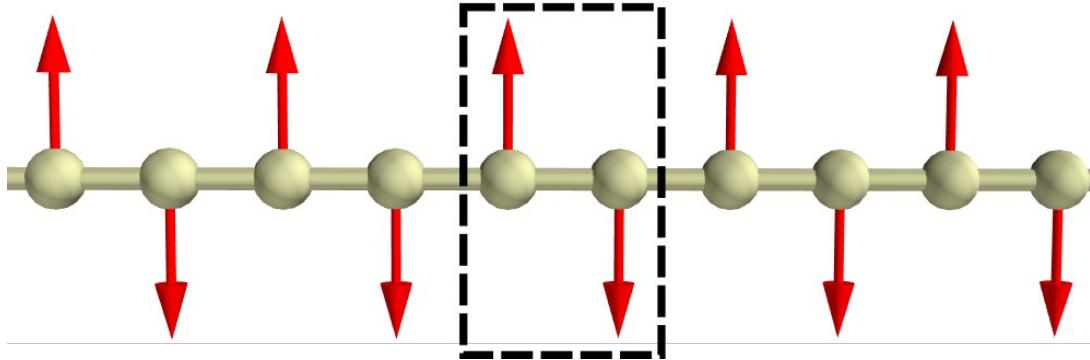


The strongly localized limit
and the Heisenberg model

From a weak magnet to the strongly localized limit

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

For large interaction strength, the system develops a local quantized magnetic moment



The strongly localized limit

Let us start with a Hubbard model dimer

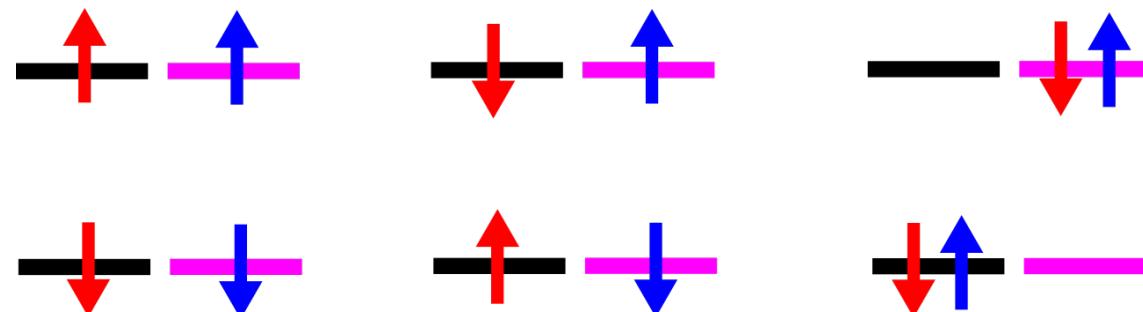
$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

Now in the limit

$$U \gg t$$



The full Hilbert space at half filling is

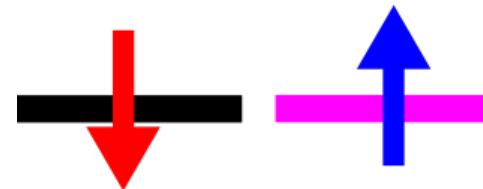


The strongly localized limit

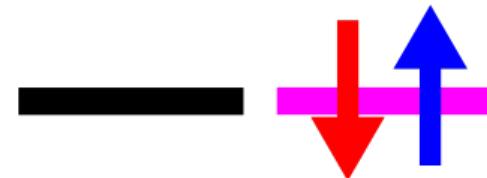
Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

The energies in the strongly localized limit are $U \gg t$



~ 0

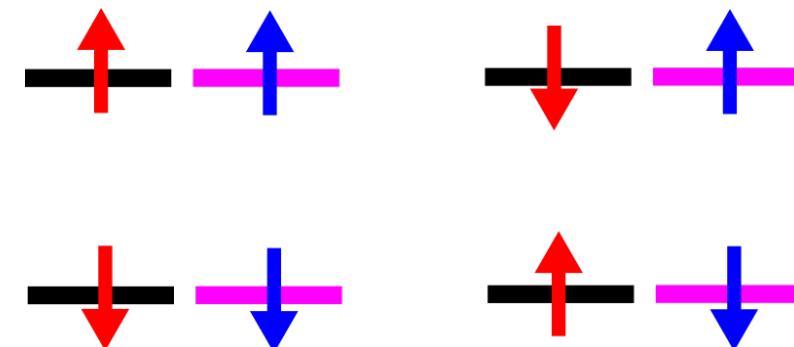


$\sim U$

The strongly localized limit

Let us start with a Hubbard model dimer

$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$



The low energy manifold is

Just one electron in each site for $U \gg t$

Local S=1/2 at each site

The strongly localized limit

Effective Heisenberg model in the localized limit

$$\mathcal{H} = J \vec{S}_0 \cdot \vec{S}_1$$

We can compute \mathbf{J} using second order perturbation theory

$$H = H_0 + V$$

$$H_0 = \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

“pristine” Hamiltonian
(Hubbard)

$$V = t [c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \text{h.c.}$$

“perturbation” Hamiltonian
(hopping)

The strongly localized limit

Effective Heisenberg model in the localized limit

$$\mathcal{H} = J \vec{S}_0 \cdot \vec{S}_1$$

We can compute J using second order perturbation theory

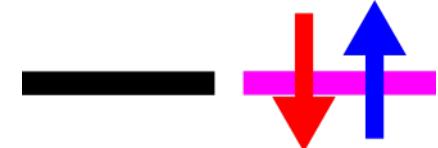
$$H = H_0 + V$$

$$J \sim \frac{t^2}{U}$$

Ground state



Virtual state



The Heisenberg model

For a generic Hamiltonian in a generic lattice

$$H = \sum_{ij} t_{ij} [c_{i\uparrow}^\dagger c_{j\uparrow} + c_{i\downarrow}^\dagger c_{j\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow}$$

In the strongly correlated (half-filled) limit we obtain a Heisenberg model

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \qquad J_{ij} \sim \frac{|t_{ij}|^2}{U}$$

The Heisenberg model

Non-Hubbard (multiorbital) models also yield effective Heisenberg models

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

In those generic cases, the exchange couplings can be positive or negative

$$J_{ij} > 0$$

Antiferromagnetic coupling

$$J_{ij} < 0$$

Ferromagnetic coupling

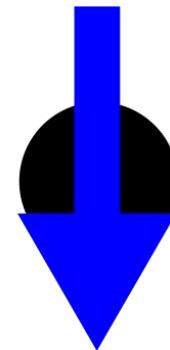
Spin-orbit coupling introduces anisotropic couplings

$$\mathcal{H} = \sum_{ij} J_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta$$

The Heisenberg model

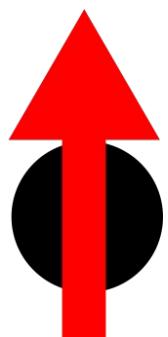
$$J_{ij} > 0$$

Antiferromagnetic coupling



$$J_{ij} < 0$$

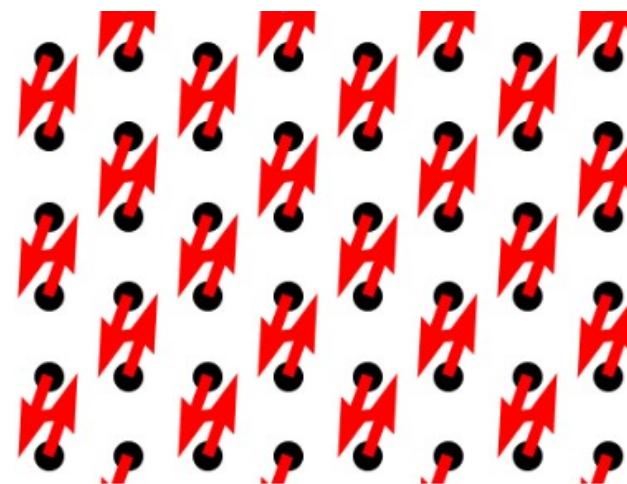
Ferromagnetic coupling



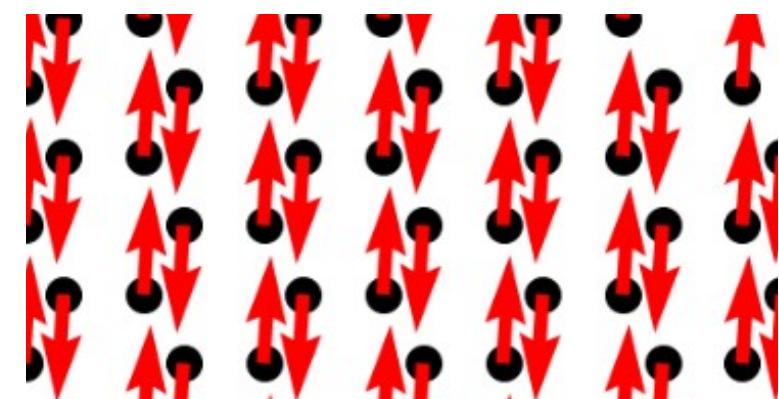
Classical ground states

Antiferromagnetism driven by superexchange

In the square lattice



In the honeycomb lattice

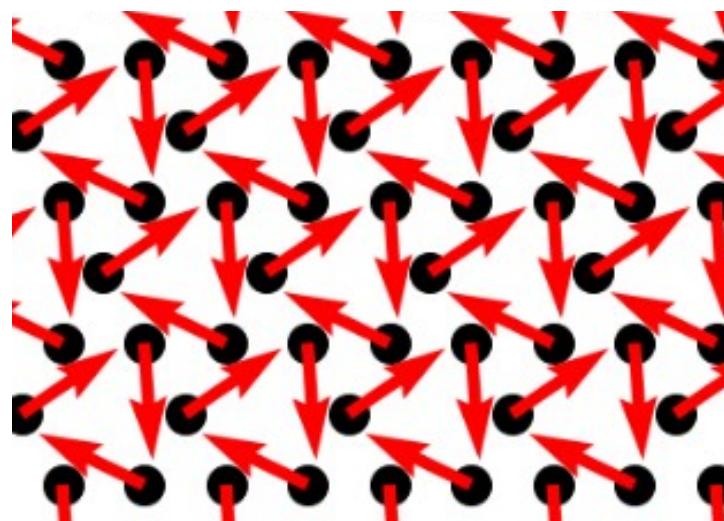


$$H = \sum_{ij,s} t_{ij} c_{i,s}^\dagger c_{j,s} + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

In bipartite lattices, the magnetization is collinear

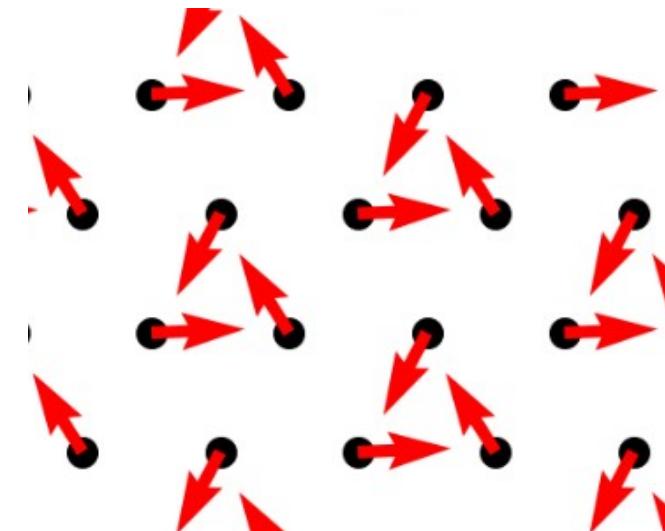
Antiferromagnetism driven by superexchange

In the Kagome lattice



$$H = \sum_{ij,s} t_{ij} c_{i,s}^\dagger c_{j,s} + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

In the triangular lattice

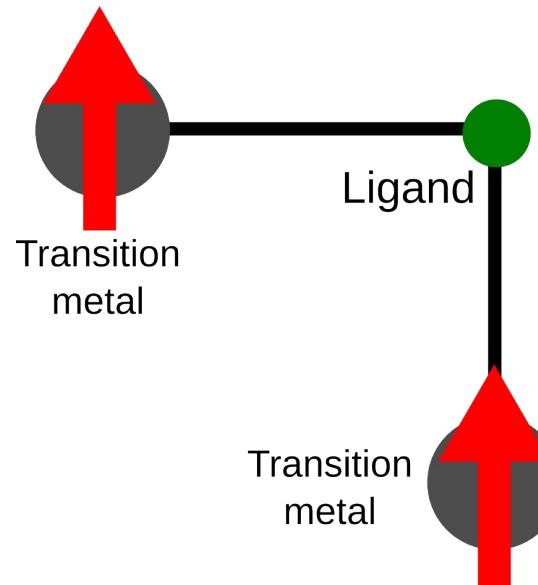


Geometric frustration promotes non-collinear order at the mean-field level

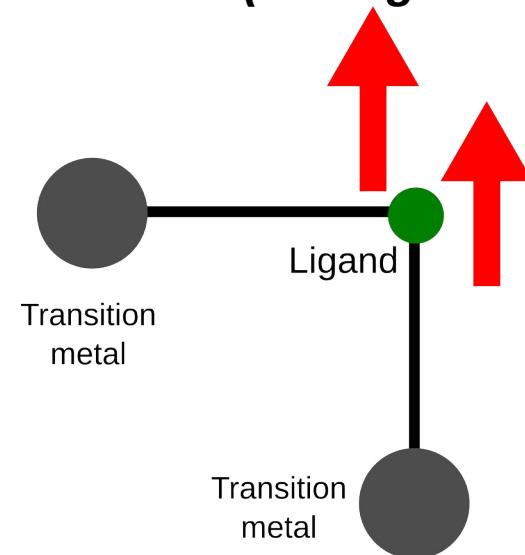
The origin of ferromagnetic coupling

Exchange interactions can be ferromagnetic if mediated by an intermediate site

Low energy manifold



Virtual state (among others)



The sign of the coupling depends on the filling of the d-shell and the angle

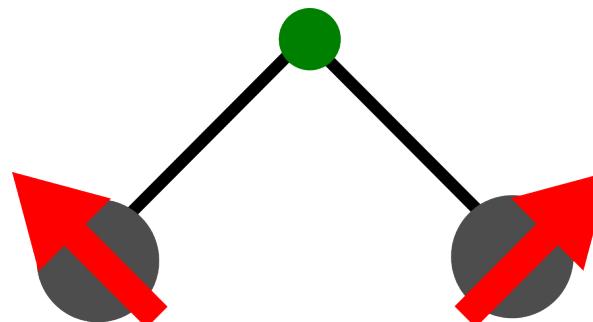
Goodenough-Kanamori rules

Non-isotropic exchange coupling

In the presence of spin-orbit coupling, new terms can appear in the Hamiltonian

Antisymmetric exchange

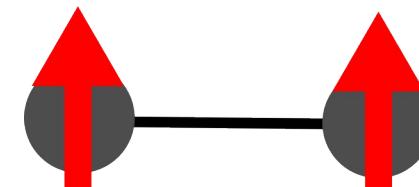
$$(\mathbf{r}_{ik} \times \mathbf{r}_{kj}) \cdot \vec{S}_i \times \vec{S}_j$$



Promotes
non-collinear order

Anisotropic exchange

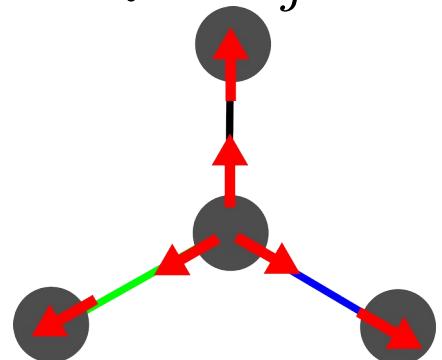
$$S_i^z S_j^z$$



Promotes
easy axis/plane

Kitaev interaction

$$S_i^{\alpha(i)} S_j^{\alpha(j)}$$



Promotes
frustration

Break

2 min break

(optional) to discuss during the break

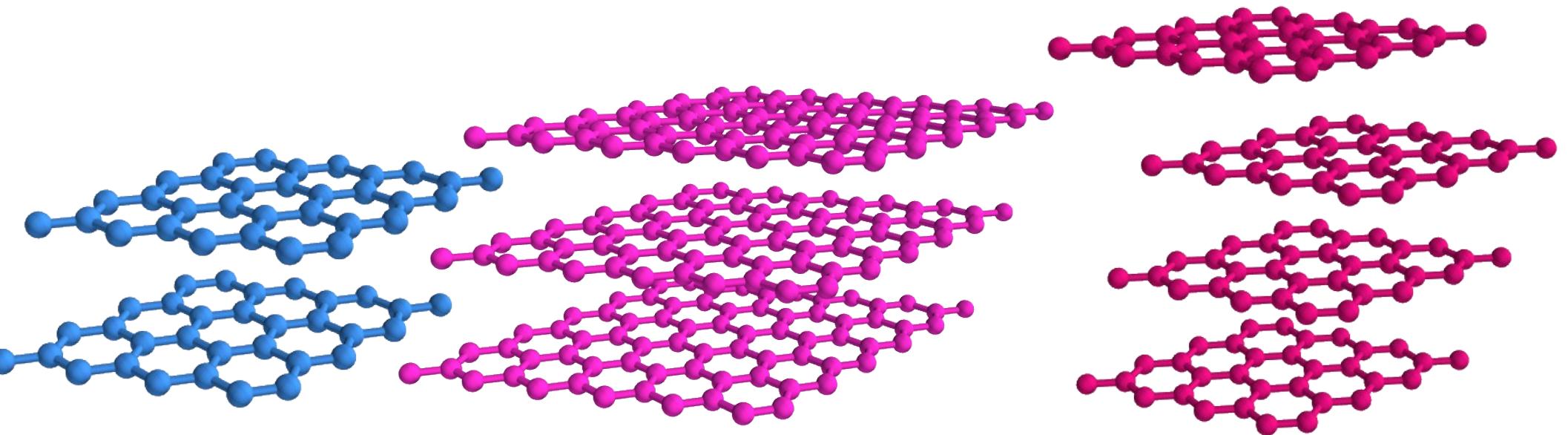
Which type of magnetic order fulfills

$$\langle c_{n\uparrow}^\dagger c_{n\downarrow} \rangle \neq 0$$

$$Im \left[\langle c_{n\uparrow}^\dagger c_{n\downarrow} \rangle \right] = 0 \quad Re \left[\langle c_{n\uparrow}^\dagger c_{n\downarrow} \rangle \right] = 0$$

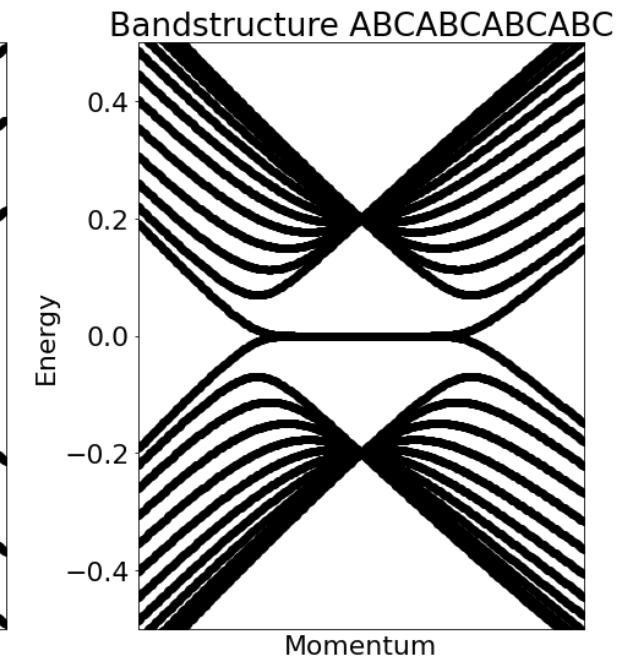
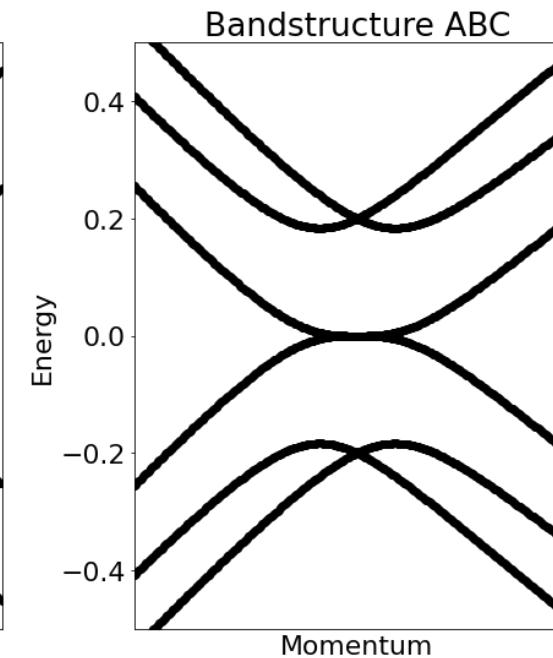
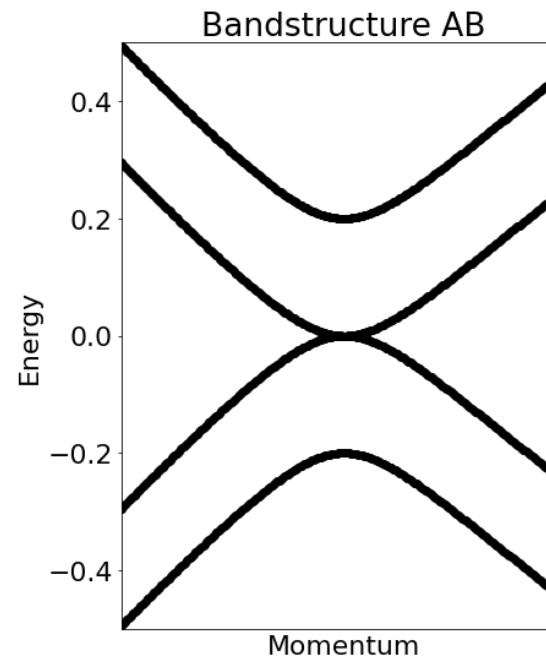
Symmetry breaking in graphene multilayers

Magnetic symmetry breaking in graphene multilayers



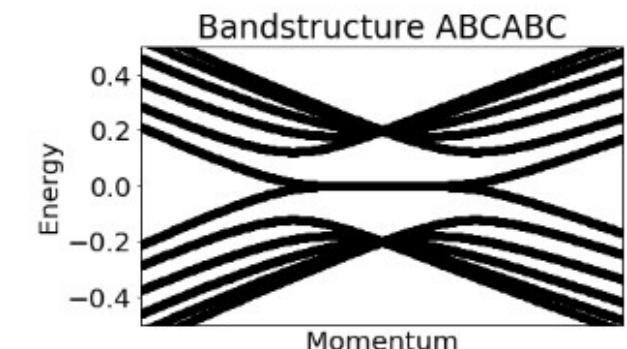
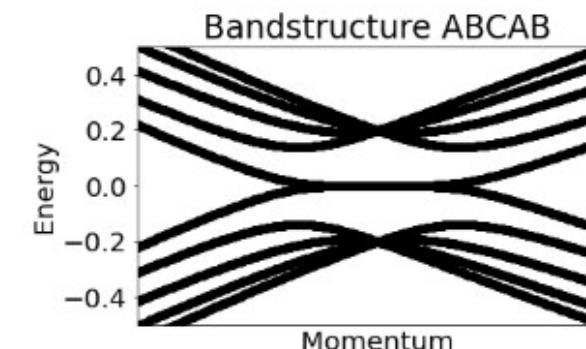
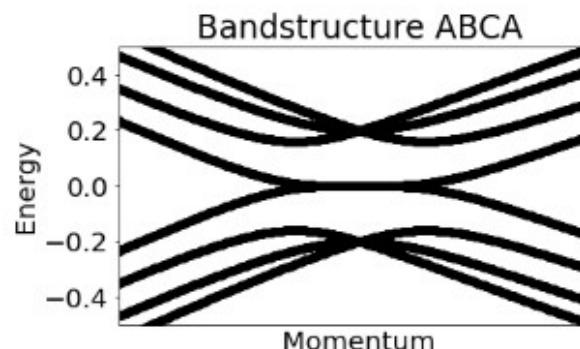
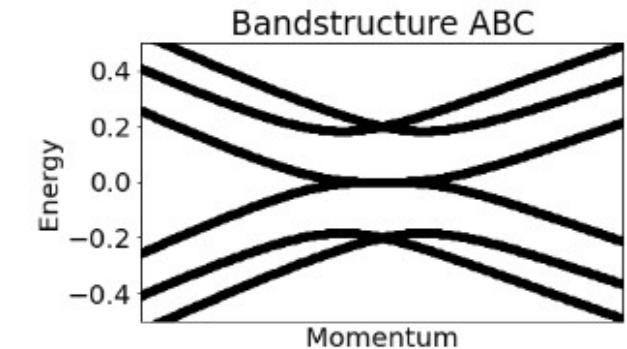
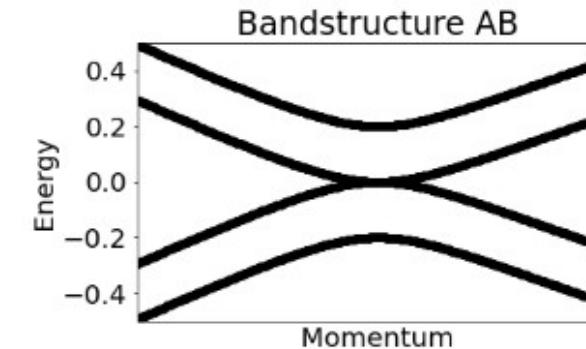
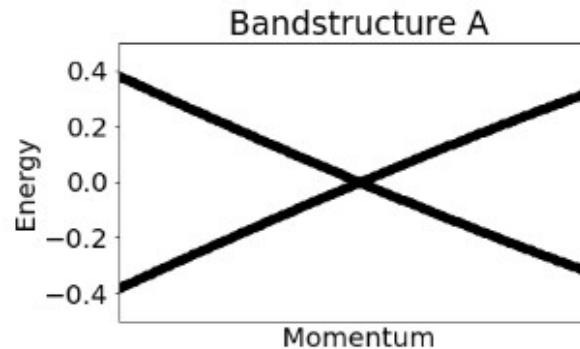
Magnetism in graphene multilayers

Graphene trilayers can have magnetic instabilities driven by repulsive interactions



The more layers a stacking has, the flatter the dispersion

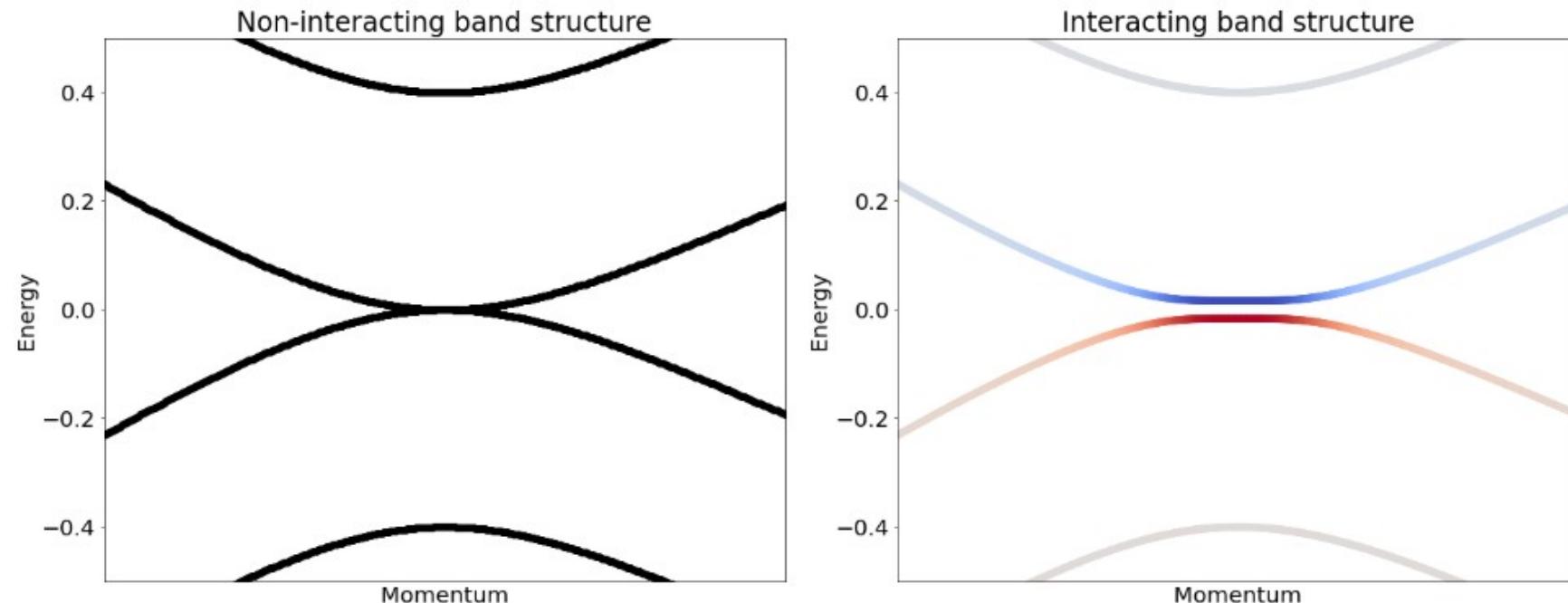
Magnetism in graphene multilayers



The more layers a stacking has, the flatter the dispersion

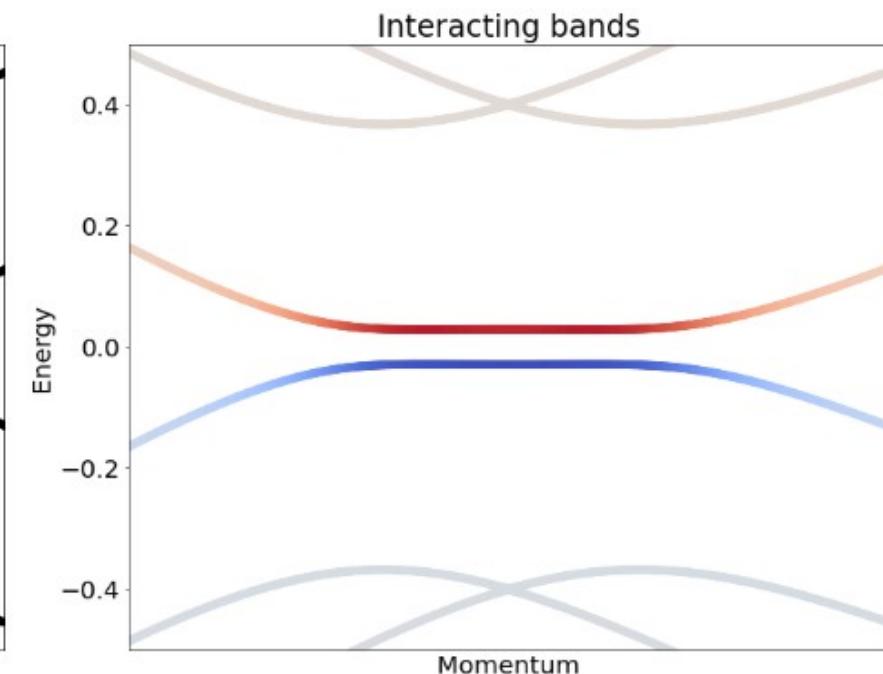
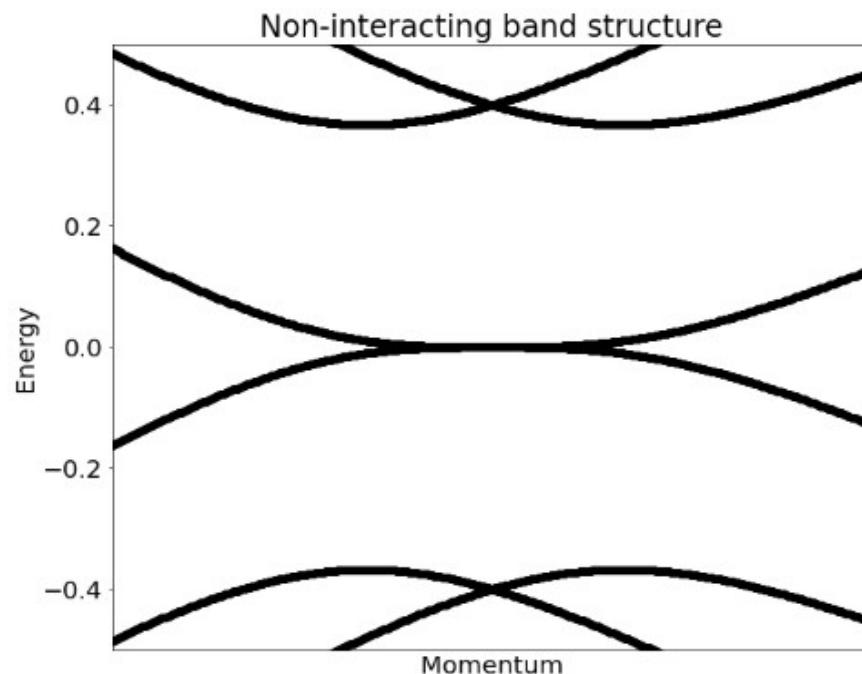
Magnetism in graphene bilayers

Graphene bilayers can have magnetic instabilities driven by repulsive interactions



Magnetism in graphene trilayers

Graphene trilayers can have magnetic instabilities driven by repulsive interactions

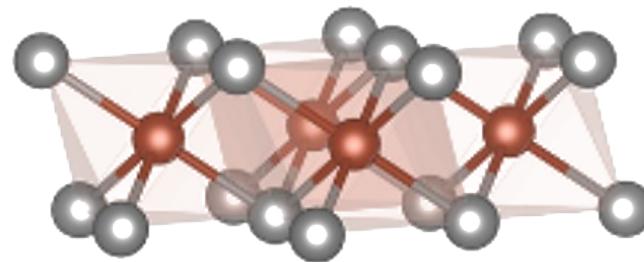


ABC graphene trilayer

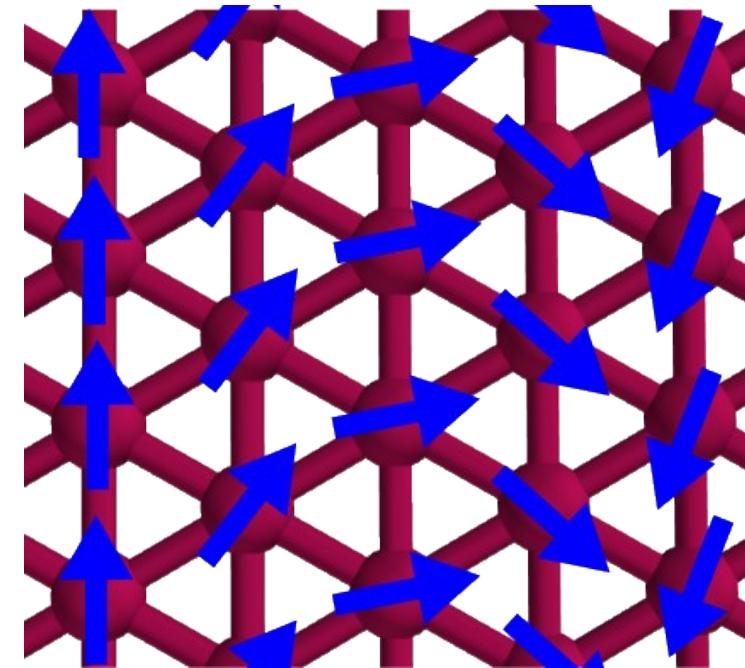
Multiferroic van der Waals materials

Electric polarization in a magnetic material

Multiferroics host, simultaneously, magnetism and electric polarization



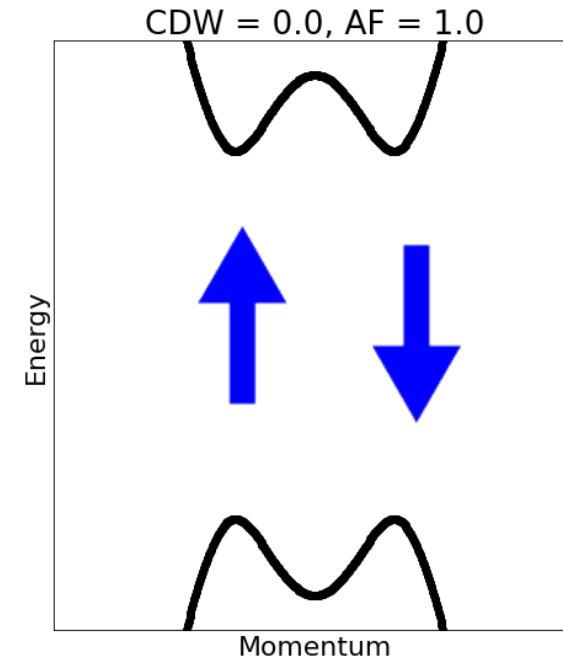
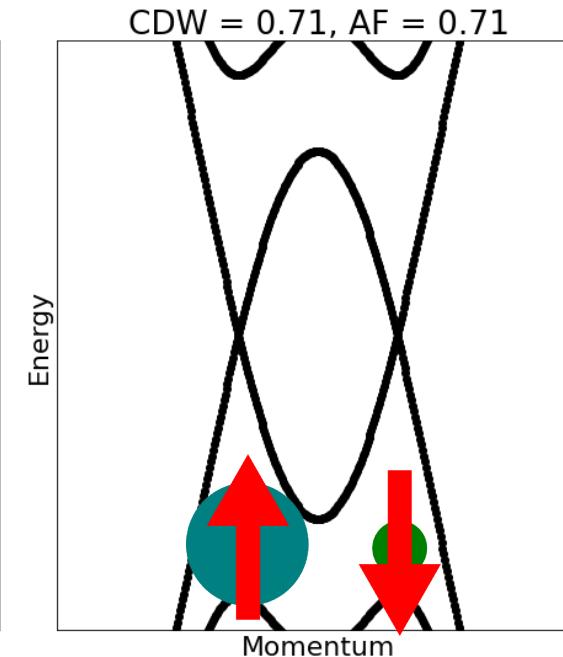
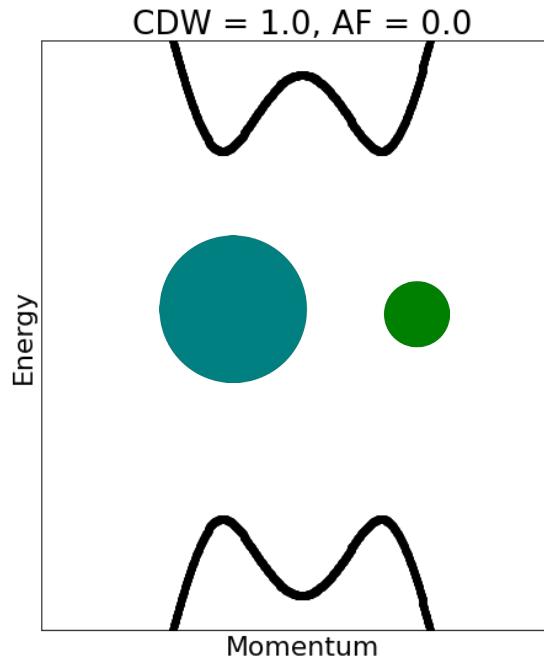
NiI_2



Why are multiferroics rare

If we want both electric and magnetic polarization, interactions must drive both simultaneously

However, magnetic and charge order often compete to open gaps

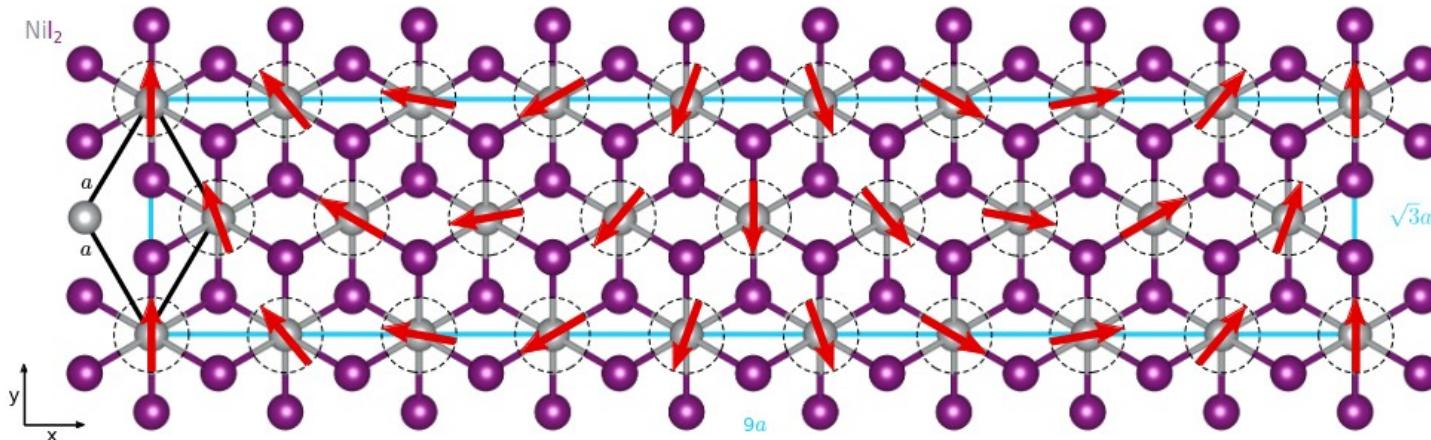


The origin of ferroelectricity in Nil_2

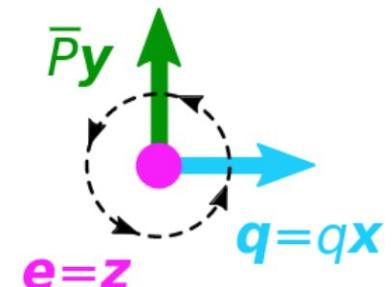
Magnetically driven ferroelectric order

$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e},$$

$$\mathbf{M}(\mathbf{r}) = e^{i(\mathbf{e} \cdot \mathbf{S})(\mathbf{q} \cdot (\mathbf{r} - \mathbf{r}_0))} \mathbf{M}(\mathbf{r}_0)$$

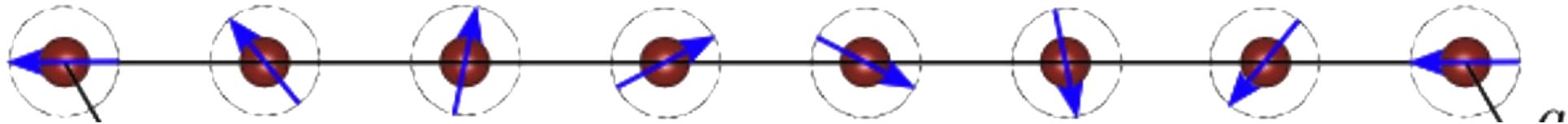


$$\bar{\mathbf{P}} \propto \lambda_{SOC}(\mathbf{e} \times \mathbf{q})$$



The emergence of non-collinear order is the driving of ferroelectricity

The origin of ferroelectricity in Ni_2



$$\mathbf{P} = \xi \mathbf{q} \times \mathbf{e}$$

Electric polarization

SOC driven

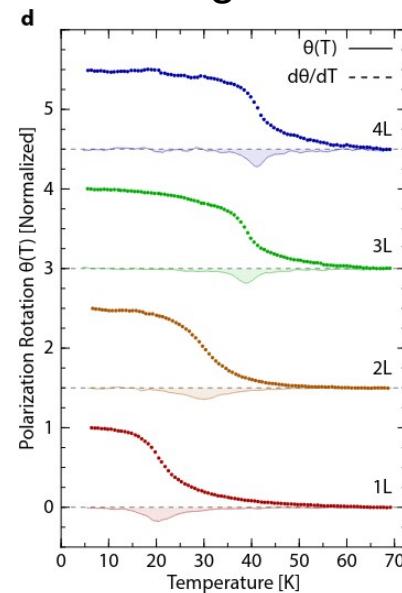
Spin rotation axis

Wavevector of the spin spiral

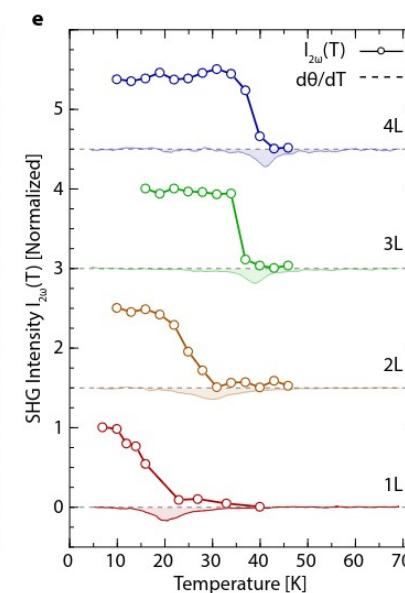
Multiferroicity in NiI_2 (experiment)

Magnetic and ferroelectric order
detected with optics

Magnetic

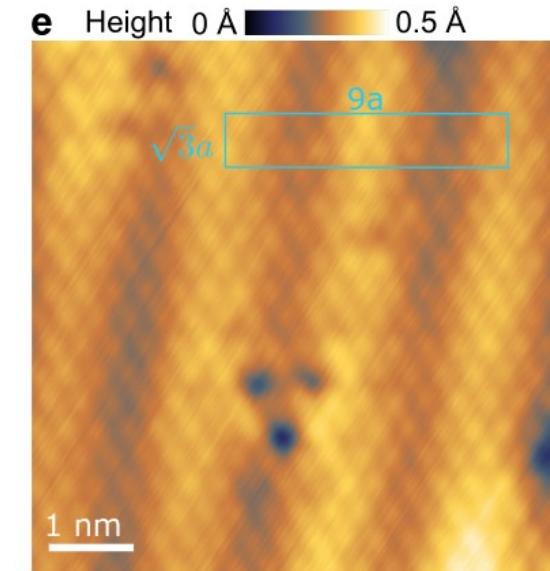


Ferroelectric

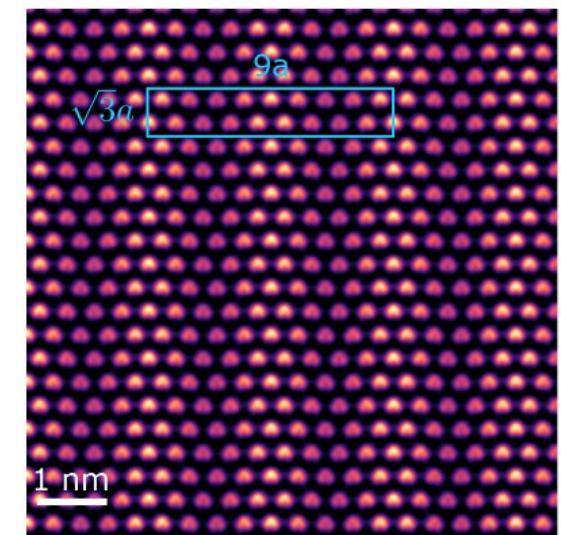


Atomic-scale visualization of multiferroic
order with STM

Experiment



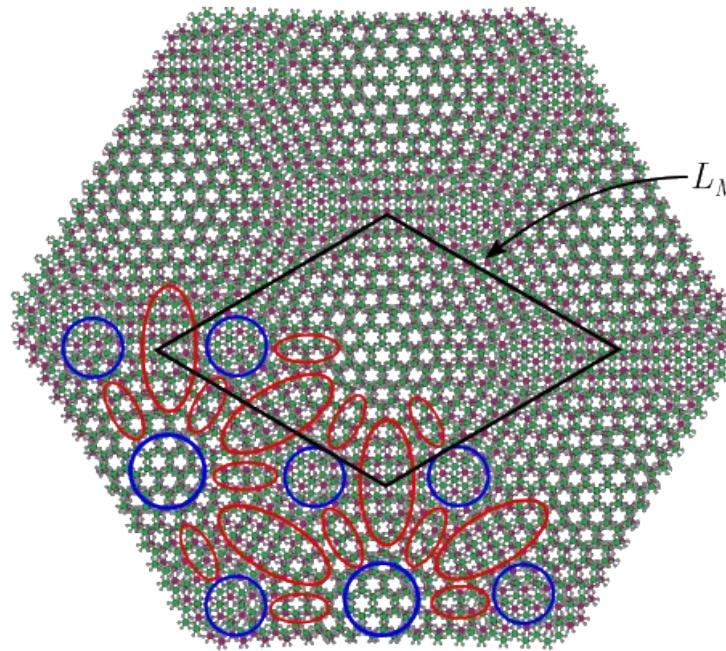
Theory



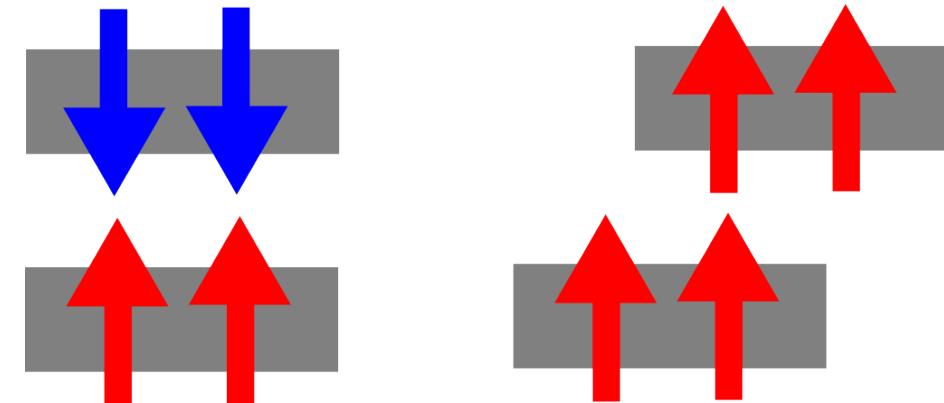
Nature 602, 601-605(2022)

arXiv:2309.11217

Twisted 2D trihalides



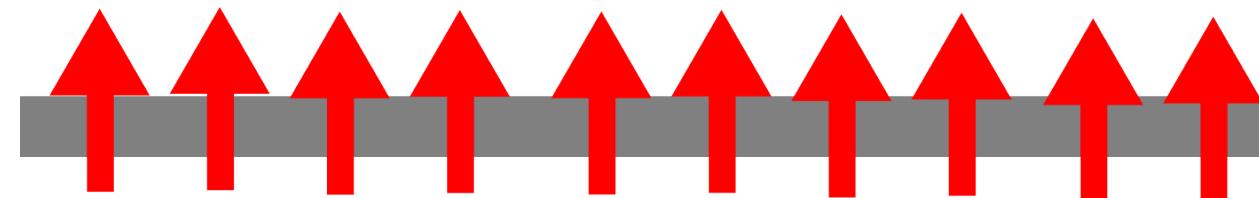
Bilayer Stacking {
M Monoclinic
R Rhombohedral



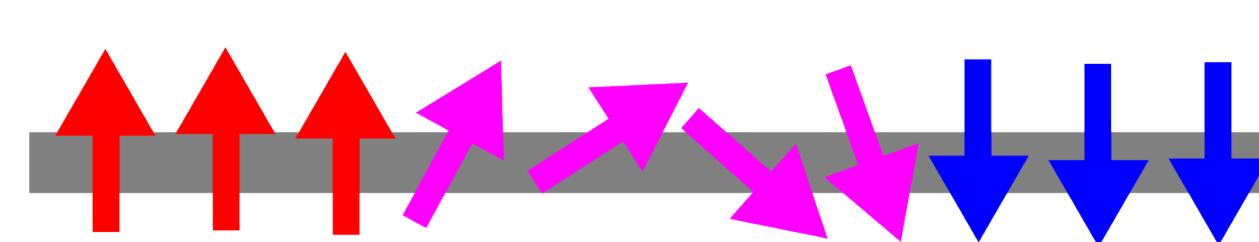
The local stacking determines
the coupling between layers

Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism at domain walls



Stacking #1



Stacking #2

Mixed FE/AF domains

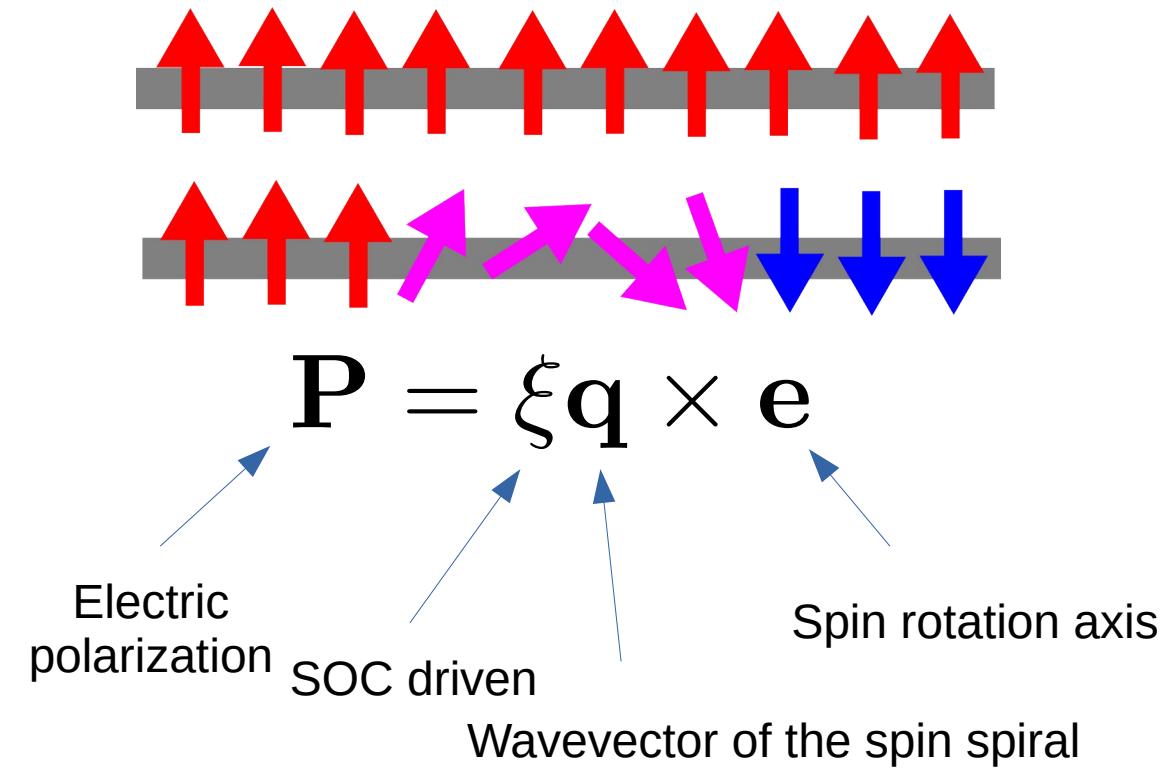
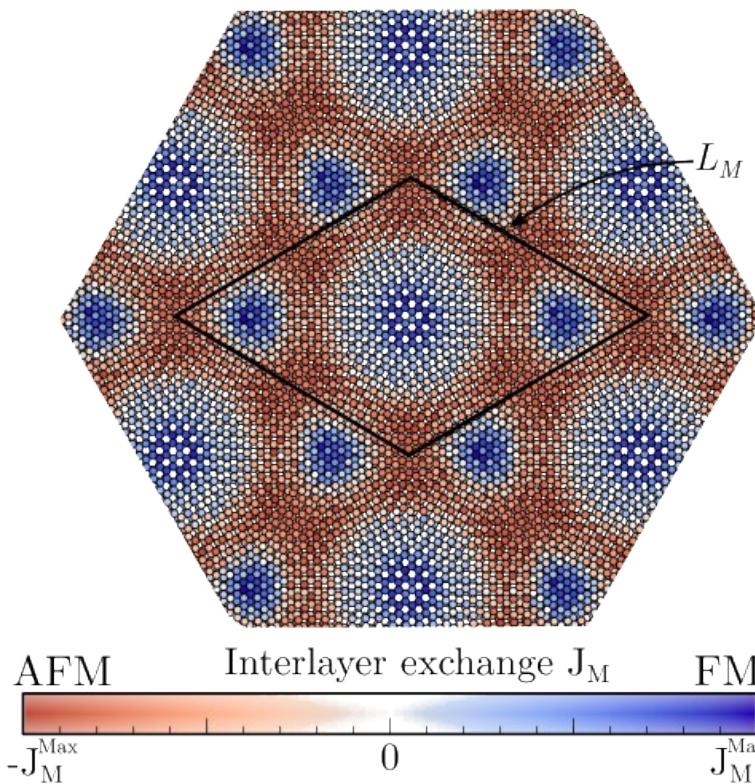
Nature Nanotechnology 17, 143–147 (2022)

Non-collinear magnetism

arXiv:2204.01636

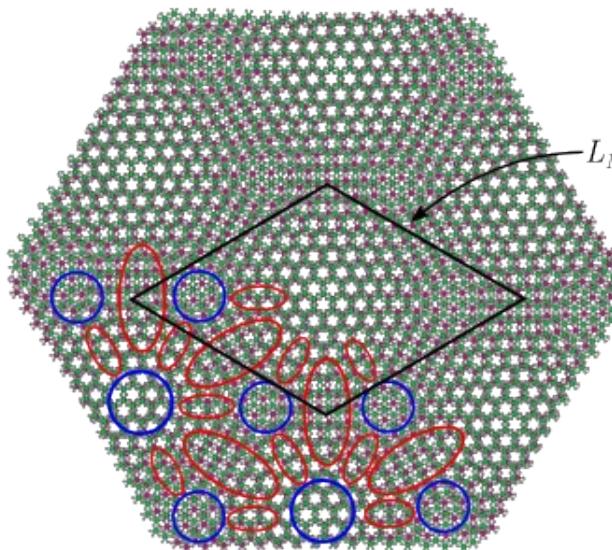
Twisted 2D magnetic materials

Non-collinear magnetism and multiferroic order appear due to the moire

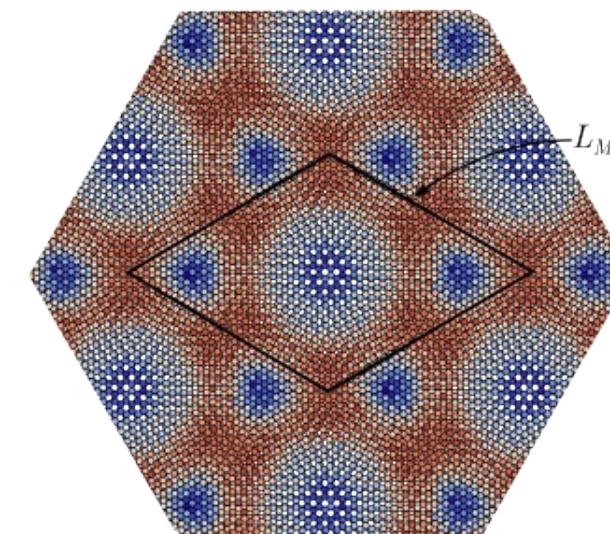


Stacking, magnetic order and electric polarization

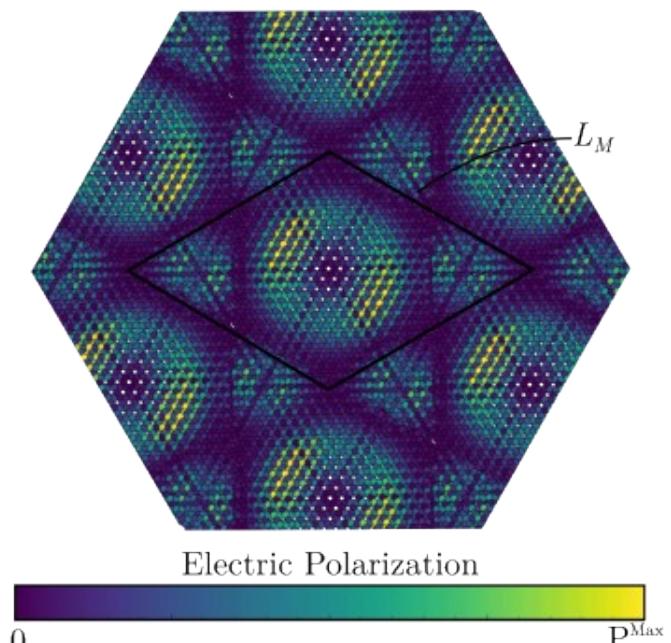
Structure



Magnetic order



Ferroelectric order

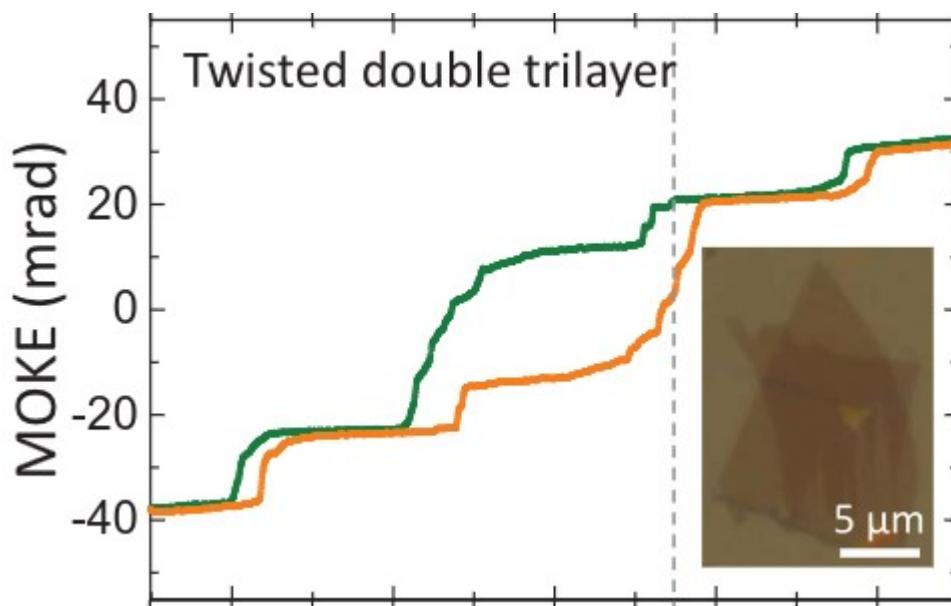


Bilayer Stacking
(M) Monoclinic
(R) Rhombohedral

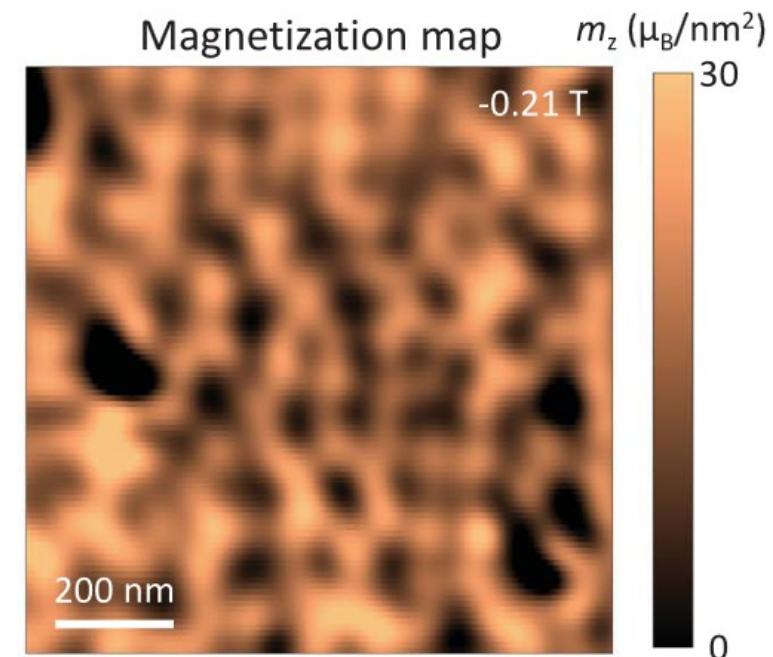
Stacking, magnetic structure and ferroelectric order are correlated

Visualization of magnetic domains in twisted CrBr₃ (experiment)

Magneto-optical Kerr anisotropy



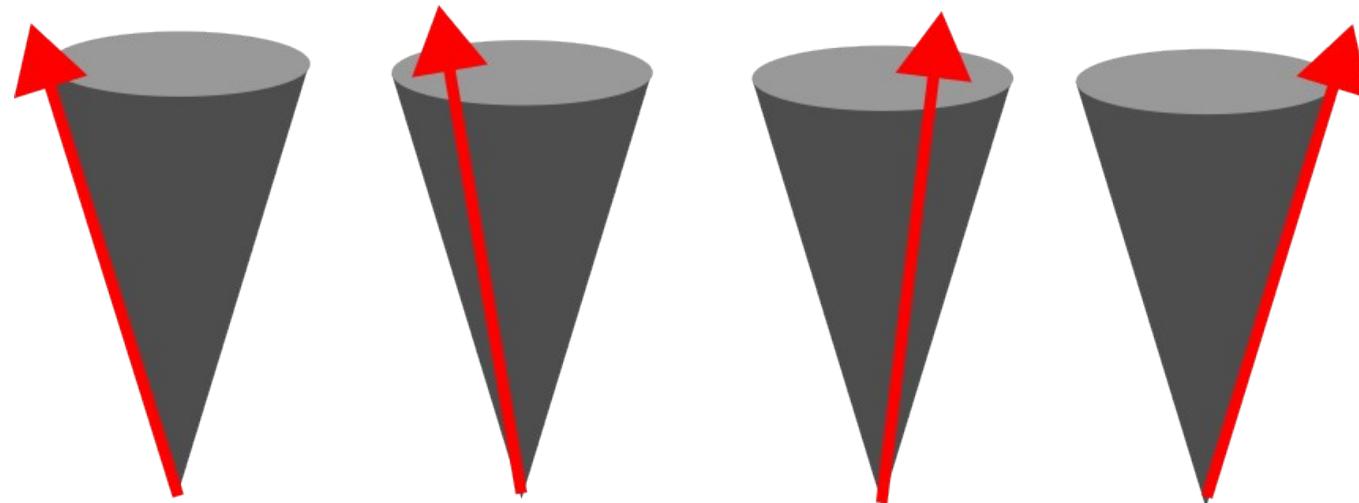
Single spin magnetometry



Excitations in 2D magnets

Excitations in a ferromagnet

Qualitatively, magnons are the fluctuations of the order parameter



Excitations in the Heisenberg model

The Heisenberg model is a full-fledged many-body problem

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

Algebraic commutation relations

$$[S_j^\alpha, S_j^\beta] = i\epsilon_{\alpha\beta\gamma} S_j^\gamma$$

$$S = 1/2, 1, 3/2, 2, \dots$$

How do we compute its many-body excitations?

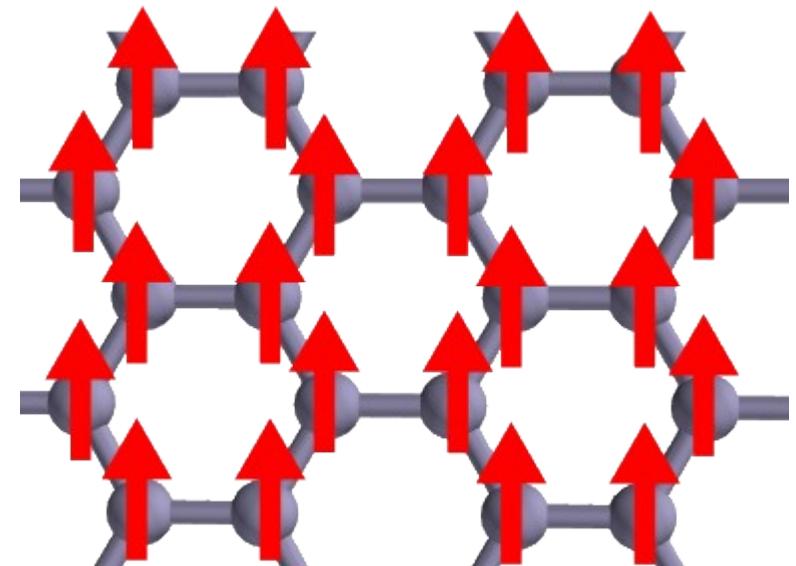
The ferromagnetic Heisenberg model

In the case of a ferromagnetic Heisenberg model, we know the ground state

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$J_{ij} < 0$$

$$|GS\rangle = | \uparrow\uparrow\uparrow\uparrow\uparrow \dots \rangle$$



But how do we compute the excitations?

The Holstein–Primakoff transformation

Replace the spin Hamiltonian by a bosonic Hamiltonian

$$S_+ = \sqrt{2s} \sqrt{1 - \frac{a^\dagger a}{2s}} a, \quad S_- = \sqrt{2s} a^\dagger \sqrt{1 - \frac{a^\dagger a}{2s}}, \quad S_z = (s - a^\dagger a)$$

Make the replacement and decouple with mean-field assuming $\langle a_i^\dagger a_i \rangle \ll s$

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \longrightarrow \mathcal{H} = \sum_{ij} \gamma_{ij} a_i^\dagger a_j$$

Spins

Magnon

Magnons in a nutshell

Increase the spin

$$S_i^+ \sim a_i$$

Destroy a magnon

Decrease the spin

$$S_i^- \sim a_i^\dagger$$

Create a magnon

Net magnetization

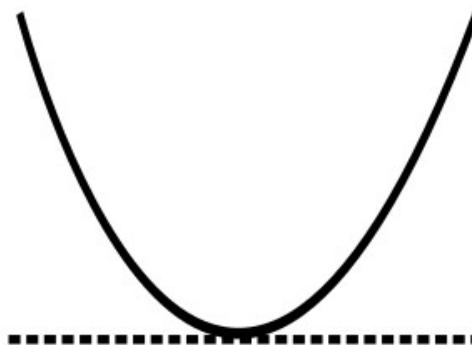
$$\langle S_i^z \rangle = S - \langle a_i^\dagger a_i \rangle$$

Maximal minus the magnons

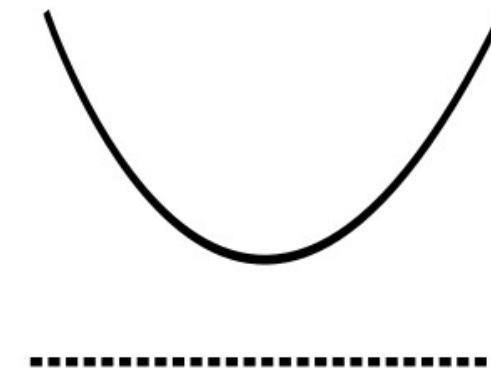
Magnons are $S=1$ excitations that exist over the symmetry broken state

Magnon dispersions

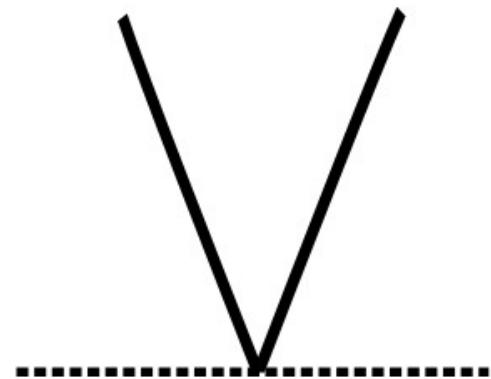
Gapless magnons



Gapped magnons



Dirac magnons



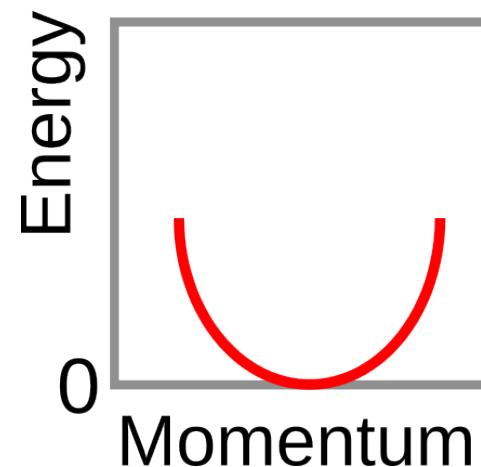
$$\mathcal{H} = \sum_{ij} \gamma_{ij} a_i^\dagger a_j$$

$$\mathcal{H} = \sum_{\mathbf{k}} \epsilon(\mathbf{k}) a_{\mathbf{k}}^\dagger a_{\mathbf{k}}$$

Magnons in the presence and absence of anisotropy

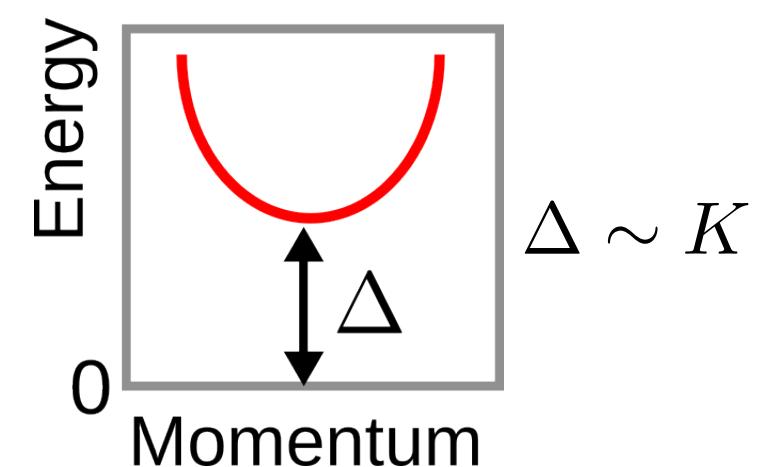
Without anisotropy

$$\mathcal{H} = - \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$



With anisotropy

$$\mathcal{H} = -J \sum_{\langle ij \rangle} \vec{S}_i \cdot \vec{S}_j - K \sum_{\langle ij \rangle} S_i^z S_j^z$$



Anisotropy in the spin model generates a magnon gap

The role of magnons in 2D magnets

$$S_z = s - a^\dagger a$$

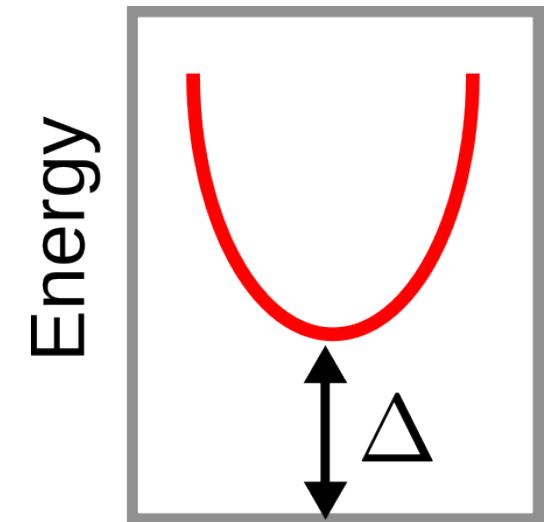
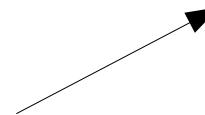
Correction from magnon population

$$\delta M_z = \langle a^\dagger a \rangle$$

Magnons renormalize the total magnetization

$$\delta M_z \sim T \int_0^{k_c} \frac{k dk}{\Delta + k^2}$$

Temperature

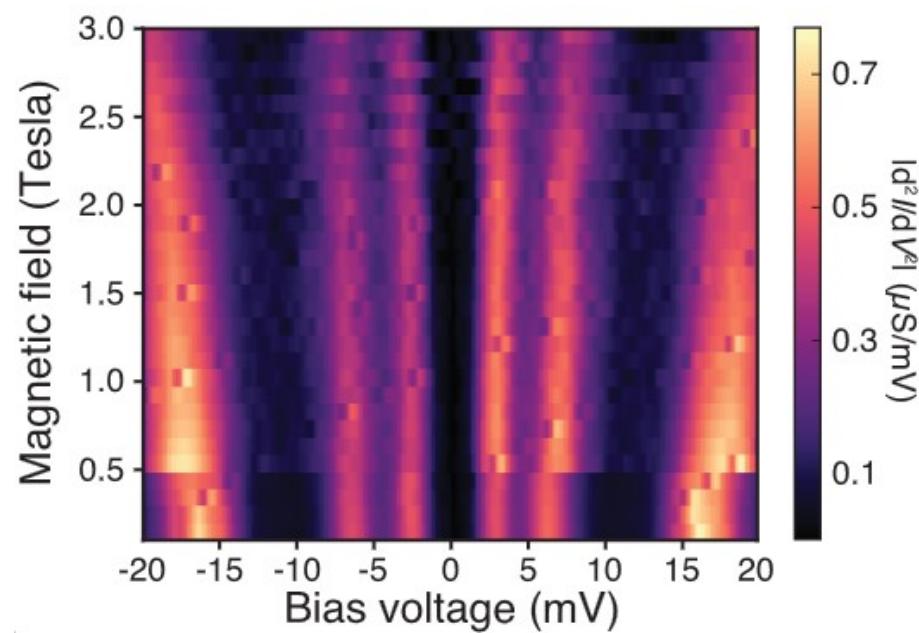


In the absence of a magnon gap, the correction to the magnetization is infinite

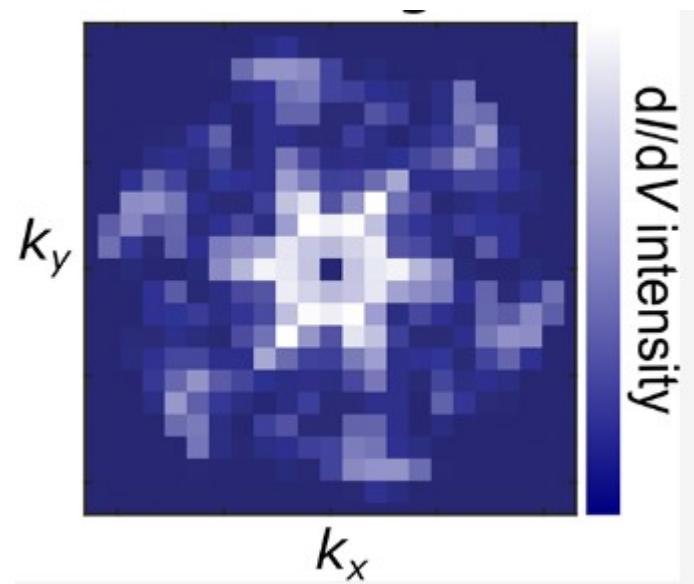
$$\delta M_z \sim T \int_0^{k_c} \frac{dk}{k} \rightarrow \infty$$

Magnons from inelastic spectroscopy (experiment)

Inelastic vertical transport

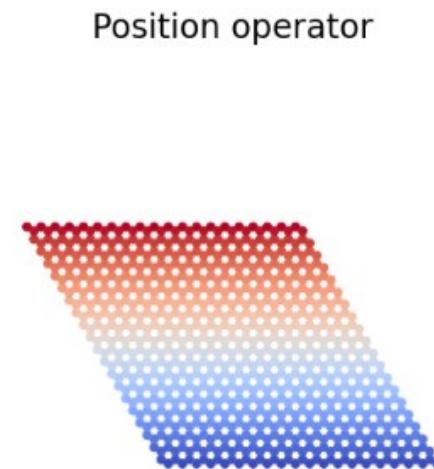
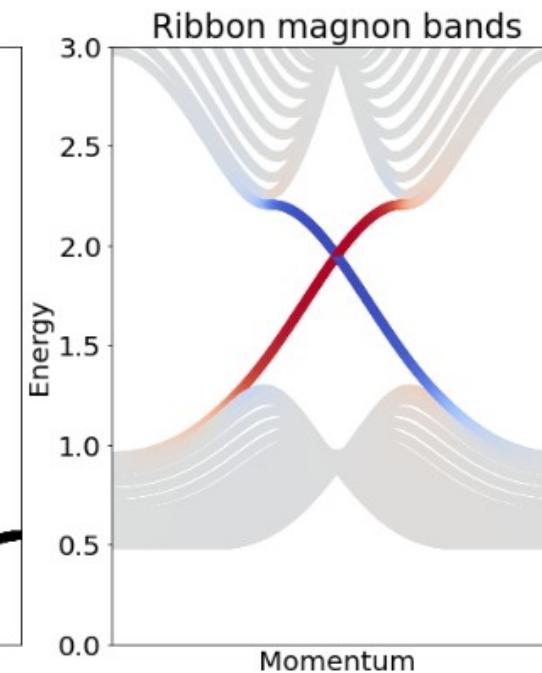
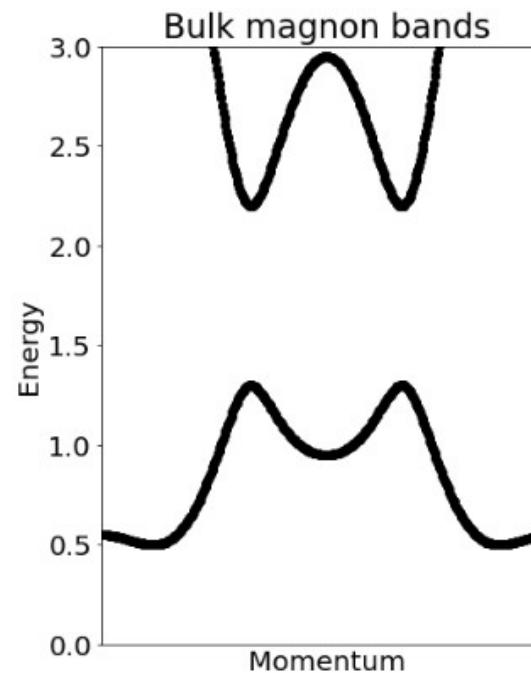


Inelastic quasiparticle interference



Topological magnons

A magnon dispersion can have topological gaps at high energies, leading to topological modes



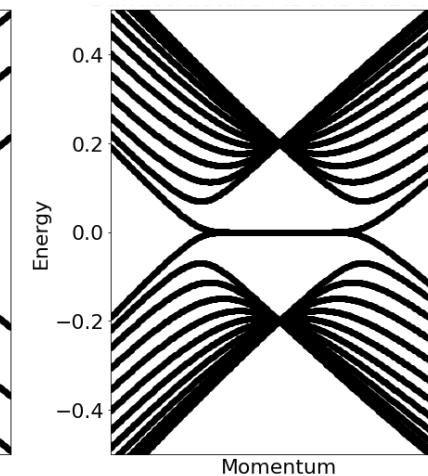
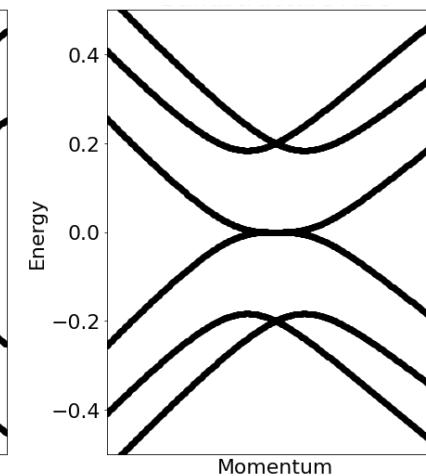
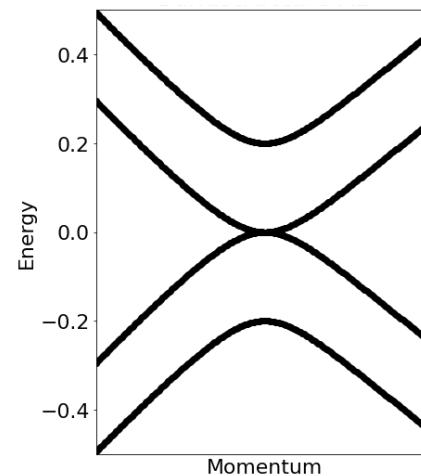
$$\mathcal{H} = \sum_{ij} \gamma_{ij} a_i^\dagger a_j$$

Break

2 min break

(optional) to discuss during the break

Which one of these electronic structures has the strongest magnetic instability?



Van der Waals quantum spin liquids

The Ising dimer

What is the ground state of this Hamiltonian

$$\mathcal{H} = S_0^z S_1^z$$

The Hamiltonian has two ground states (related by time-reversal symmetry)

$$|GS_1\rangle = |\uparrow\downarrow\rangle$$

$$|GS_2\rangle = |\downarrow\uparrow\rangle$$

Each ground state breaks time-reversal symmetry

A symmetry broken antiferromagnet is a macroscopic version of this

The quantum Heisenberg dimer

What is the ground state of this quantum Hamiltonian?

$$\mathcal{H} = \vec{S}_0 \cdot \vec{S}_1$$

The ground state is unique, and does not break time-reversal

$$|GS\rangle = \frac{1}{\sqrt{2}}[|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle] \quad \langle \vec{S}_i \rangle = 0$$

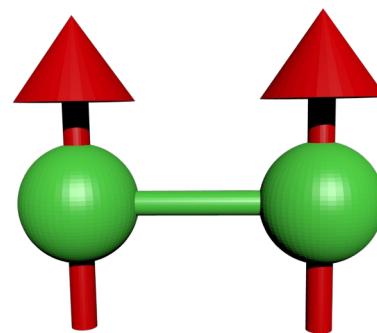
The state is maximally entangled

Can we have a macroscopic version of this ground state?

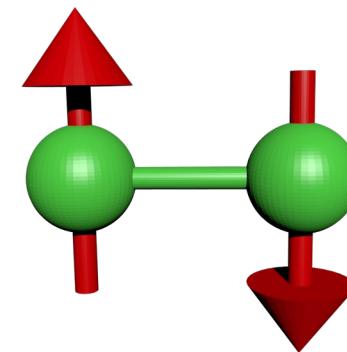
$$\langle \vec{S}_i \rangle = 0$$

Towards quantum-spin liquids

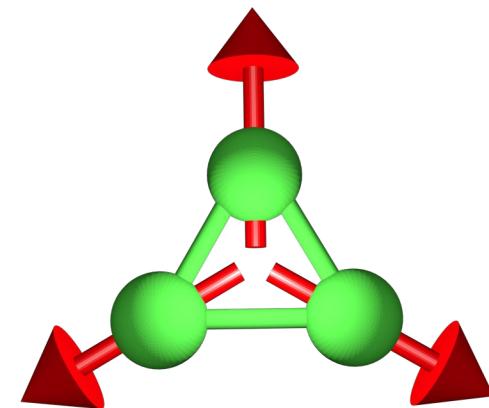
Ferromagnetism



Antiferromagnetism



Frustrated magnetism

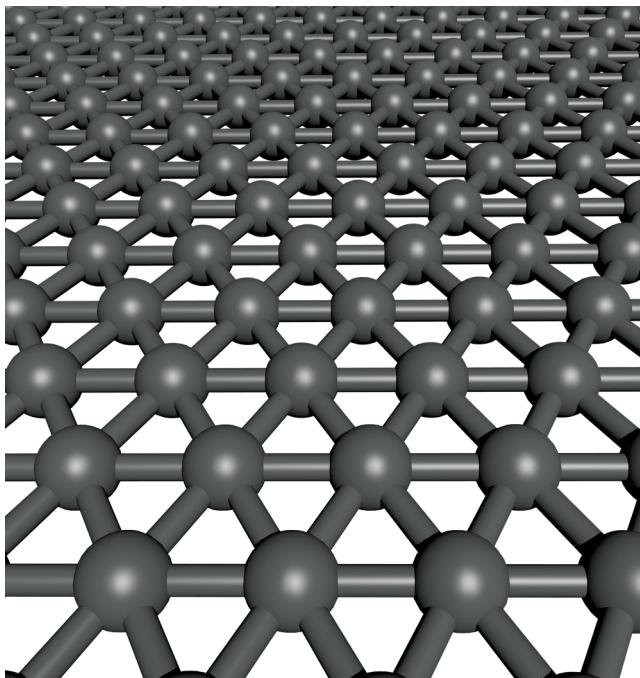


To get a quantum-spin liquid, we should look for frustrated magnetism

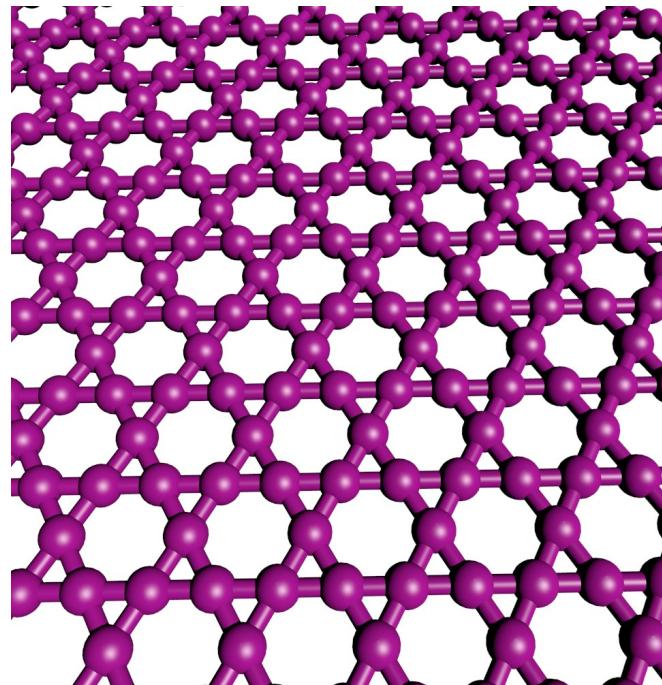
$$\langle \vec{S}_i \rangle = 0$$

Frustrated lattices

Triangular



Kagome



Quasiparticles in a quantum spin-liquid

Let us assume that a certain Hamiltonian realizes a QSL

$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

Quantum spin liquids require $\langle \vec{S}_i \rangle = 0$

The approximation used for magnons breaks down

$$\langle S_i^z \rangle = S - \langle a_i^\dagger a_i \rangle$$

$$\langle a_i^\dagger a_i \rangle \ll S$$

We need a new approximation for the quantum excitations

The parton transformation

Transform spin operators to auxiliary fermions (Abrikosov fermions)

$$S_i^\alpha = \frac{1}{2} \sigma_{s,s'}^\alpha f_{i,s}^\dagger f_{i,s'}$$

The fermions f (spinons) have $S=1/2$ but no charge

This transformation artificially enlarges the Hilbert space, thus we have to put the constraint

$$\sum_s f_{i,s}^\dagger f_{i,s} = 1$$

This transformation allow to turn a spin Hamiltonian into a fermionic Hamiltonian

The spinon Hamiltonian

We can insert the auxiliary fermions $S_i^\alpha \sim \sigma_{s,s'}^\alpha f_{i,s}^\dagger f_{i,s'}$

And perform a mean-field in the auxiliary fermions (spinons)

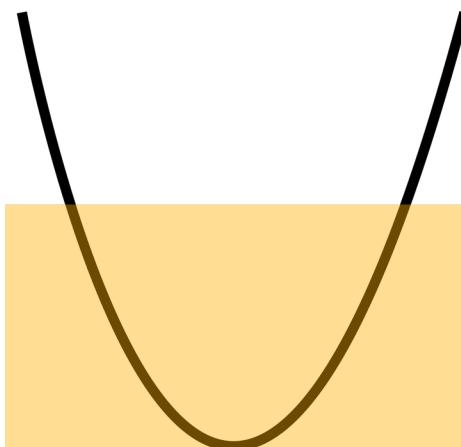
$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \longrightarrow \mathcal{H} = \sum_{ij,s} \chi_{ij} f_{i,s}^\dagger f_{j,s}$$

Enforcing time-reversal symmetry $\langle \vec{S}_i \rangle = 0$

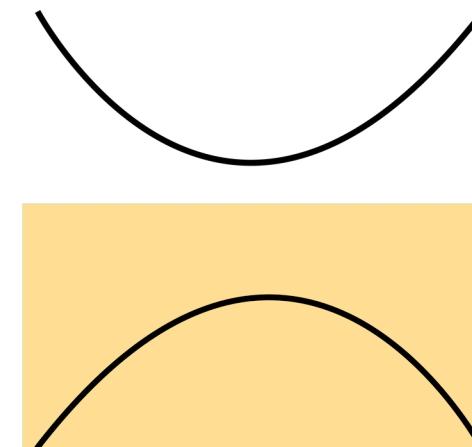
The excitations of the QSL are described by a single particle spinon Hamiltonian

Spinon dispersions

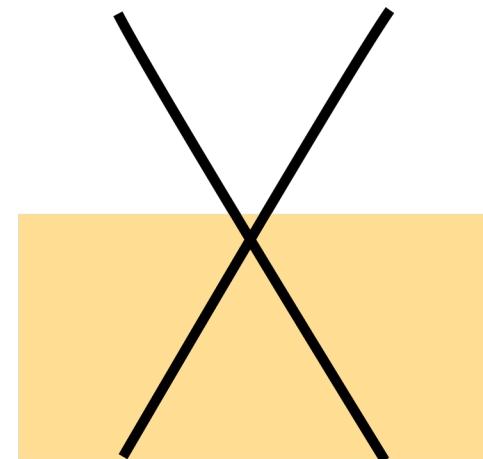
Gapless spinons



Gapped spinons



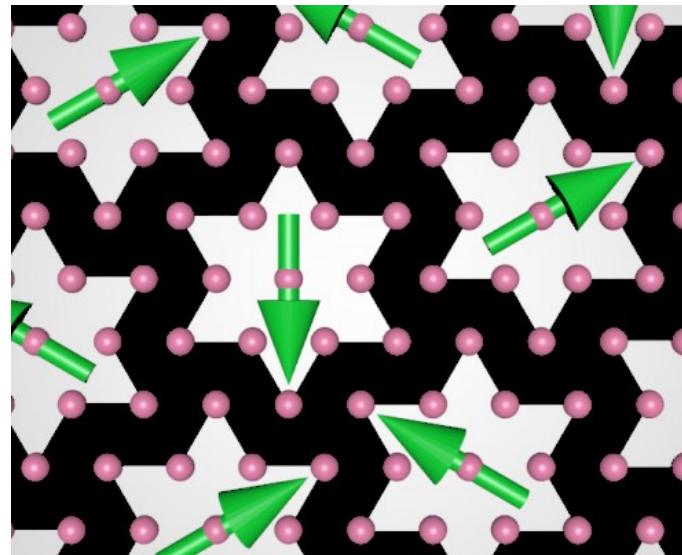
Dirac spinons



$$\mathcal{H} = \sum_{ij,s} \chi_{ij} f_{i,s}^\dagger f_{j,s}$$

Frustrated magnetism in 1T-TaS₂

Charge-density wave reconstruction, leading to a localized orbital in a $\sqrt{13} \times \sqrt{13}$ unit cell

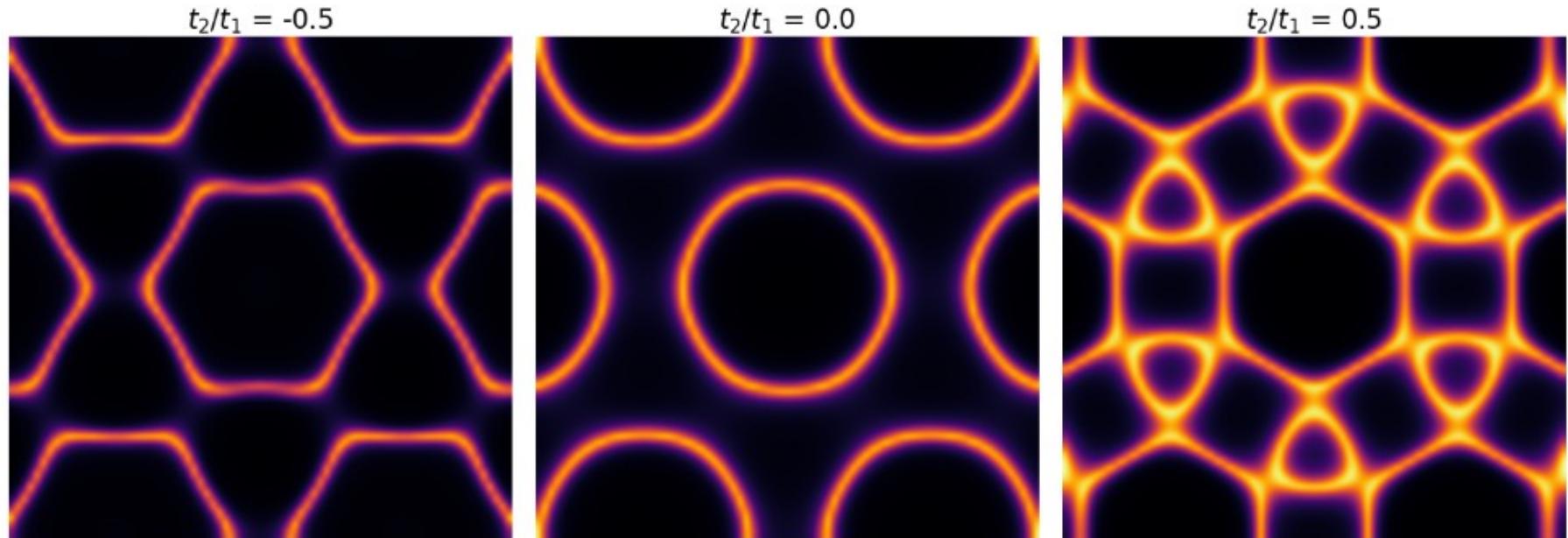


$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

Strong interactions give rise to local moment formation

Effectively described by an S=1/2 Heisenberg model in a triangular lattice

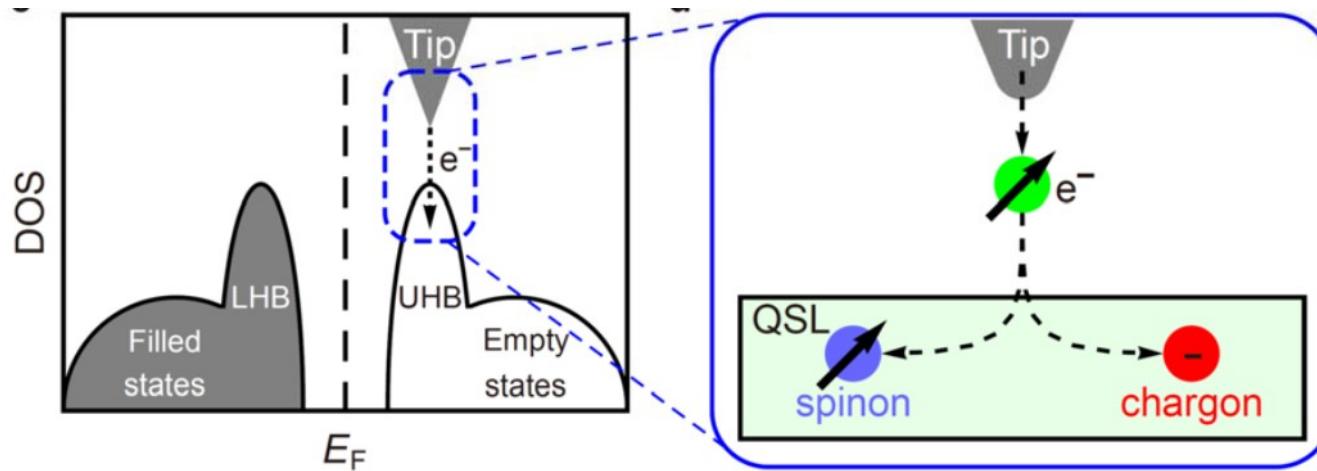
Spinon Fermi surfaces of gapless QSL



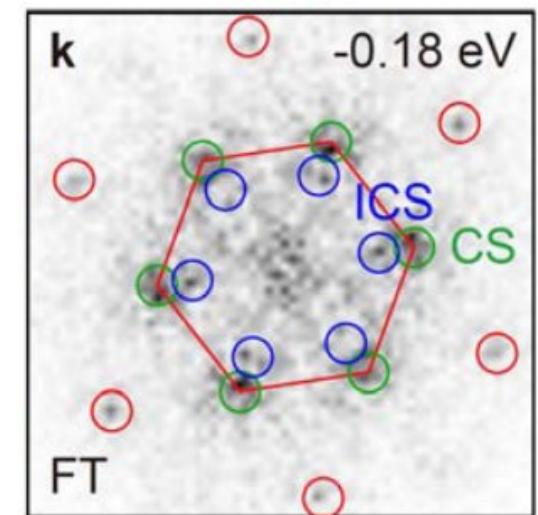
In the class of gapless QSL, different Fermi surfaces can appear depending on details of the Hamiltonian

Potential spinons in TaS₂ (experiment)

Scanning tunneling using spin-charge separation



Signatures of spinon in quasiparticle interference



Heavy-fermions in van der Waals materials

The Kondo problem



Conduction electrons

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right)$$

Kondo coupling

$$H_K = \sum_{\alpha\beta} \left(c_{0\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{0\alpha} \right) \cdot \vec{S}$$

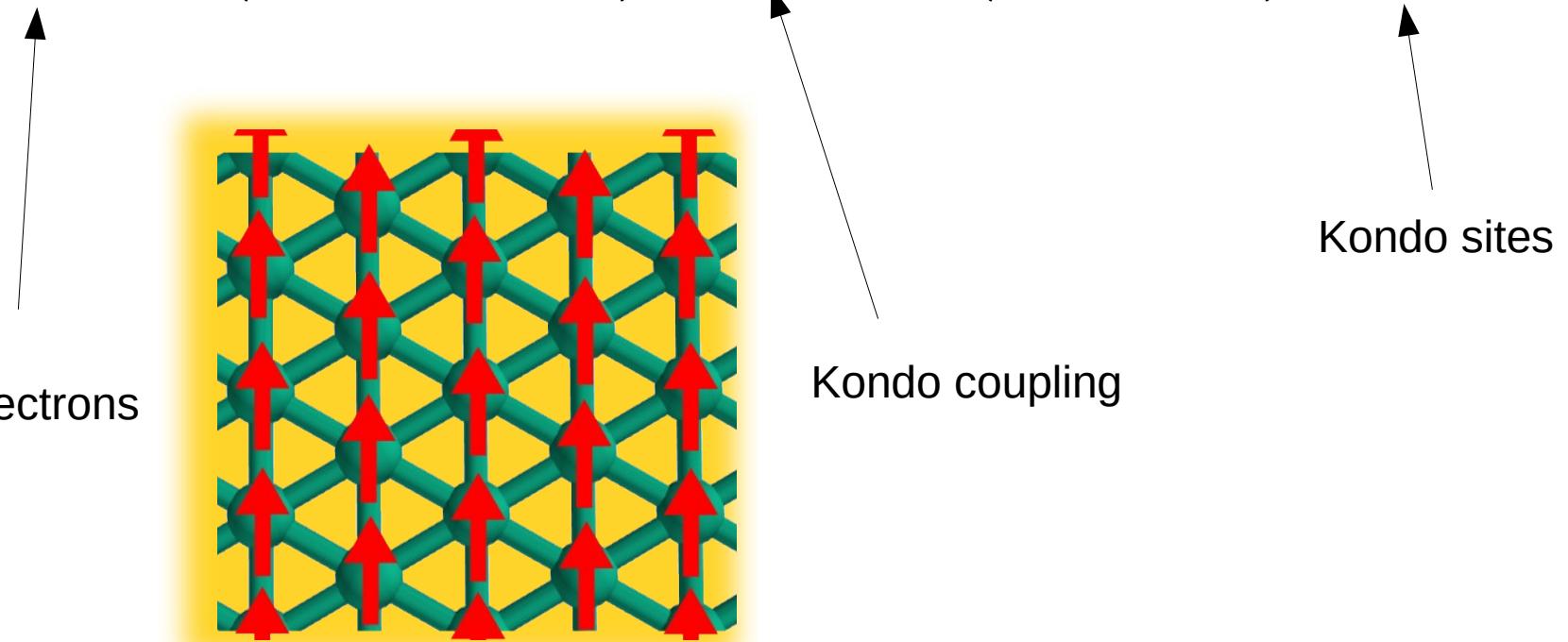
We now take a quantum spin $S=1/2$

$$|GS\rangle \sim \frac{1}{\sqrt{2}} [| \uparrow\downarrow \rangle - | \downarrow\uparrow \rangle]$$

The Kondo lattice problem

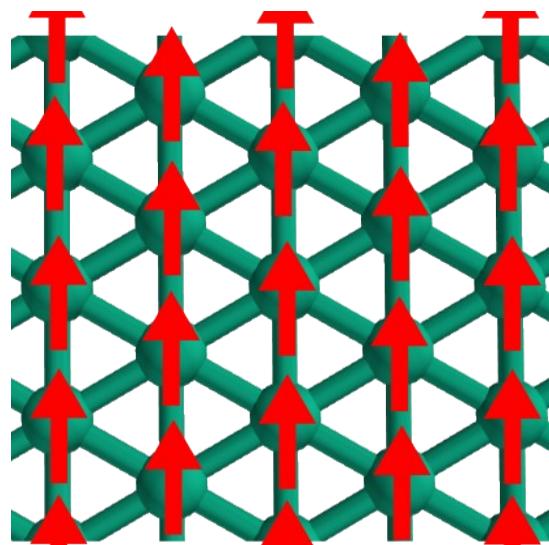
The Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$



Building an artificial heavy fermion state

Lattice of Kondo impurities



Dispersive electron gas

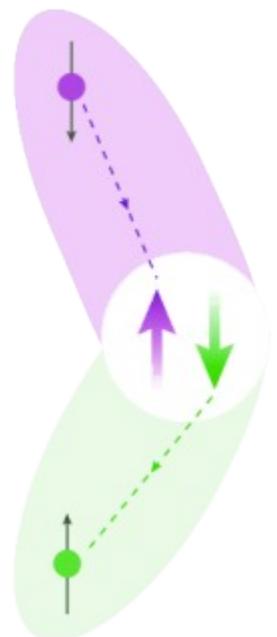
$$J_K \longleftrightarrow$$



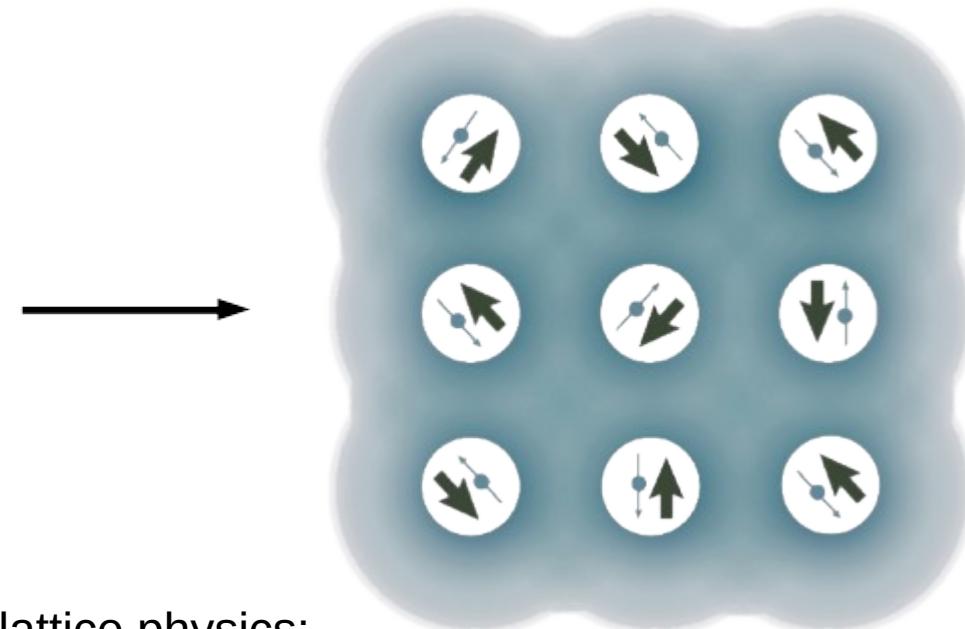
Both ingredients coupled through Kondo coupling

Building an artificial heavy fermion state

Conduction electrons form
Kondo singlets with the impurities



Kondo-lattice model



Associated with Kondo lattice physics:

- Colossal mass enhancement of electrons
- Quantum criticality
- Unconventional (topological) superconductivity

Solving the Kondo lattice problem

$$H = -t \sum_{(i,j)\sigma} \left(c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.} \right) + J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger \vec{\sigma}_{\beta\alpha} c_{j\alpha} \right) \cdot \vec{S}_j$$

Replace the spin sites by auxiliary fermions

$$S_{\alpha\beta}(j) = f_{j\alpha}^\dagger f_{j\beta} - \frac{n_f(j)}{N} \delta_{\alpha\beta}$$

This makes the effective Hamiltonian an “interacting” fermionic Hamiltonian

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} \left(c_{j\beta}^\dagger f_{j\beta} \right) \left(f_{j\alpha}^\dagger c_{j\alpha} \right)$$

Solving the Kondo lattice problem

Now we decouple the fermions with a mean-field approximation

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - J \sum_{j,\alpha\beta} (c_{j\beta}^\dagger f_{j\beta}) (f_{j\alpha}^\dagger c_{j\alpha})$$

Obtaining a quadratic Hamiltonian

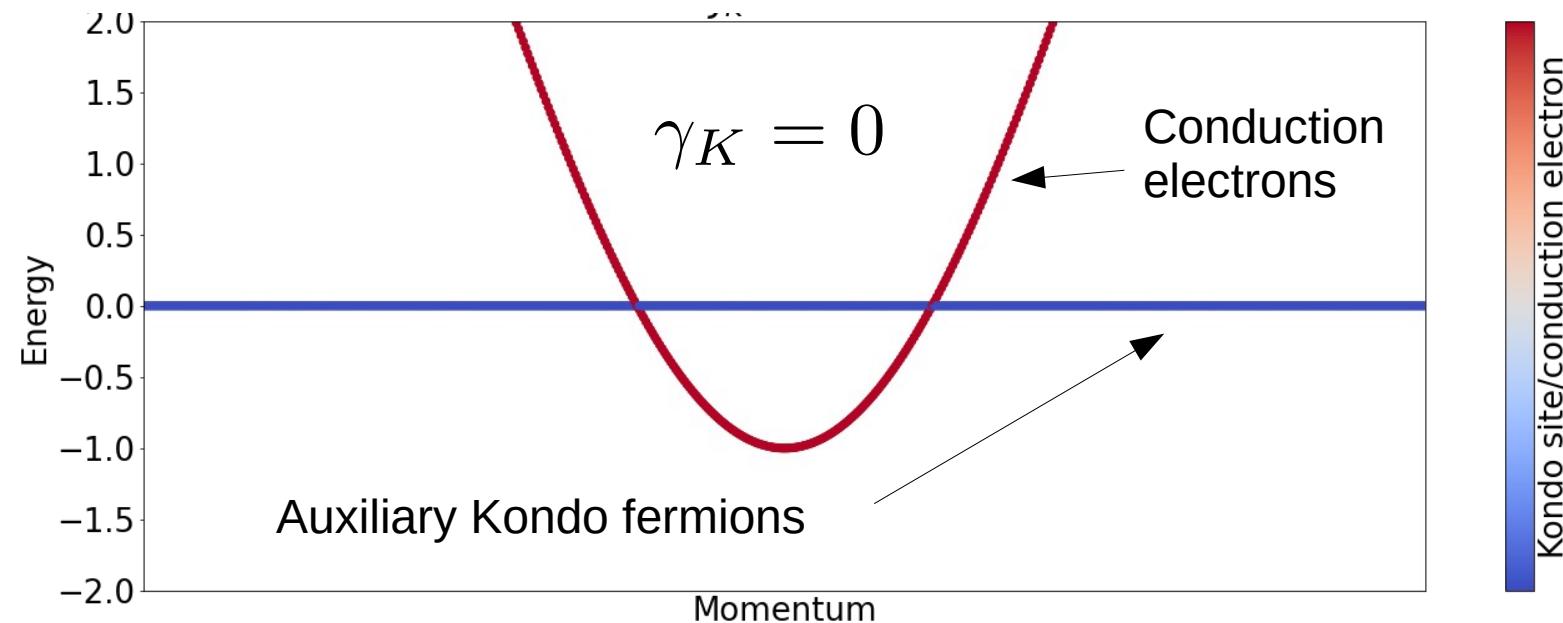
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

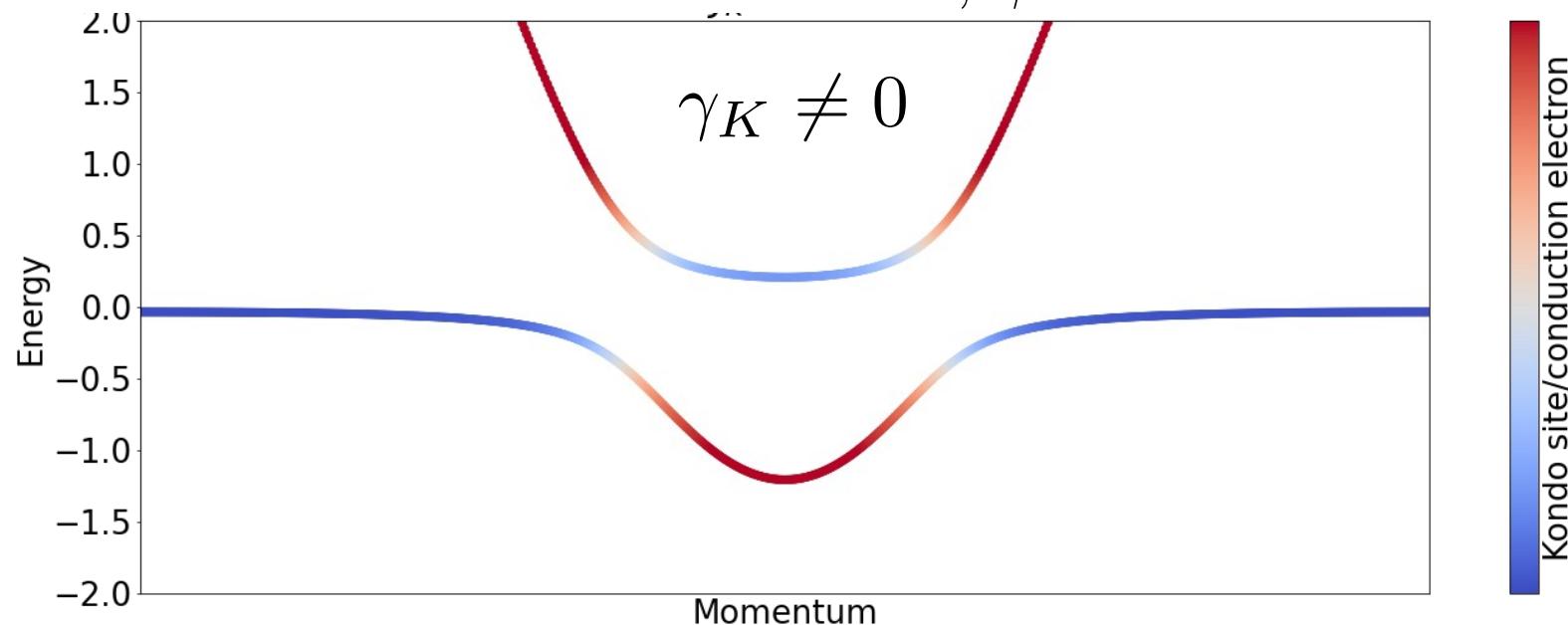
Electronic structure of the Kondo lattice problem

$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} + h.c.$$



Electronic structure of the Kondo lattice problem

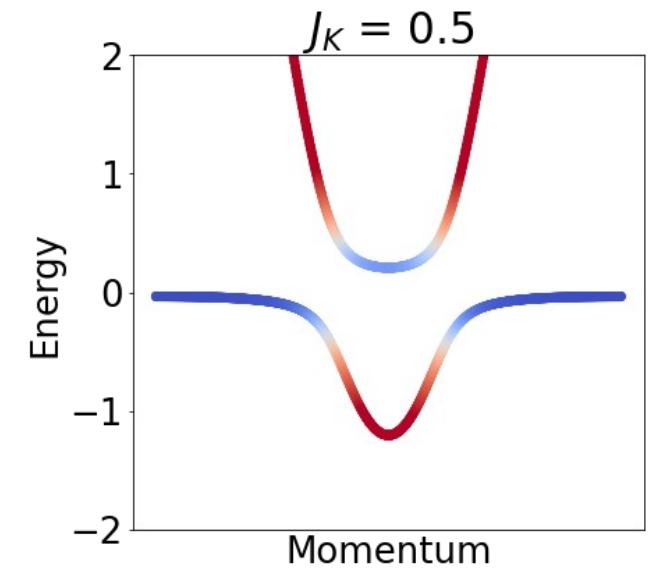
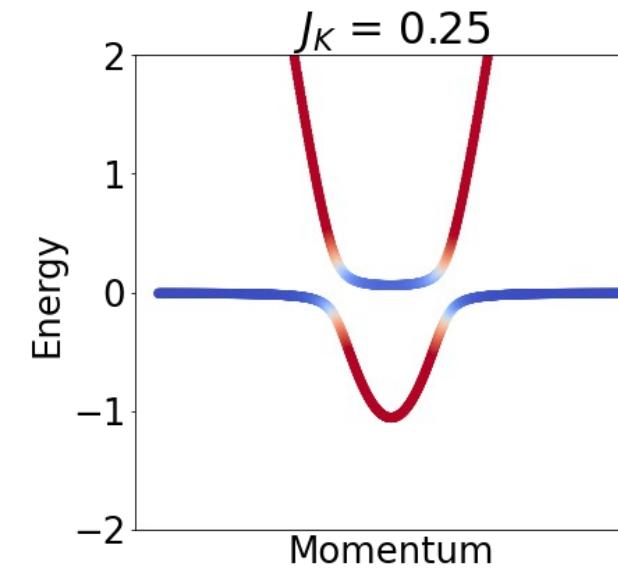
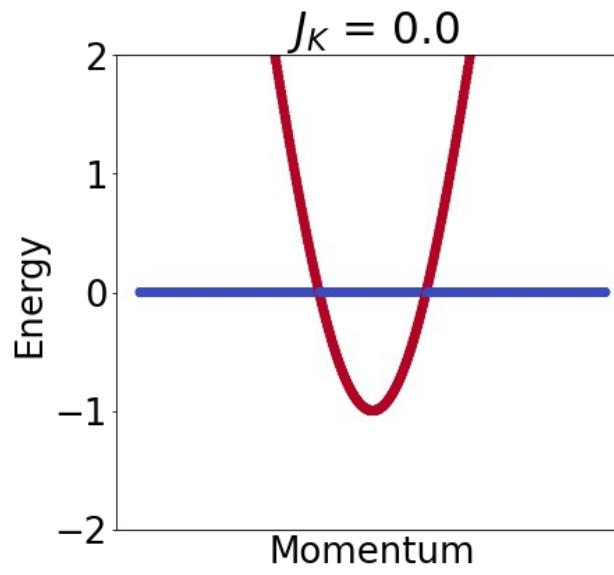
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} + h.c.$$



The Kondo coupling opens up a gap in the electronic structure

Dependence on the Kondo coupling

The heavy-fermion gap becomes bigger as the Kondo coupling increases



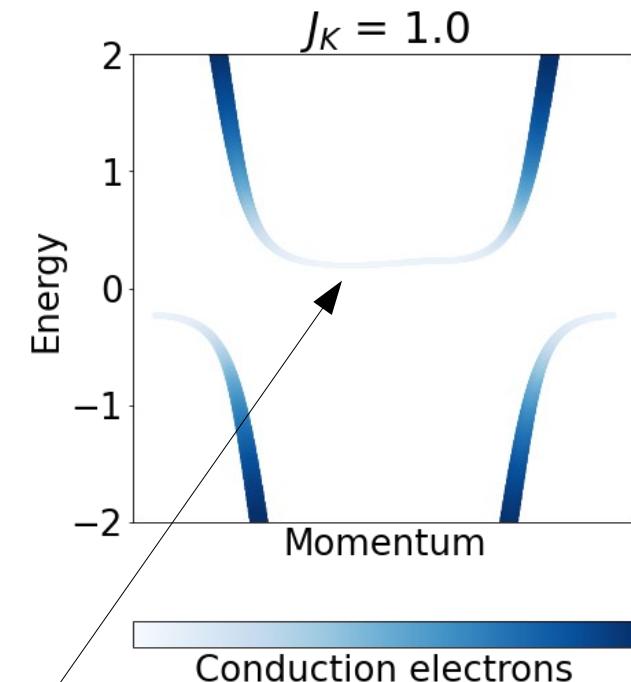
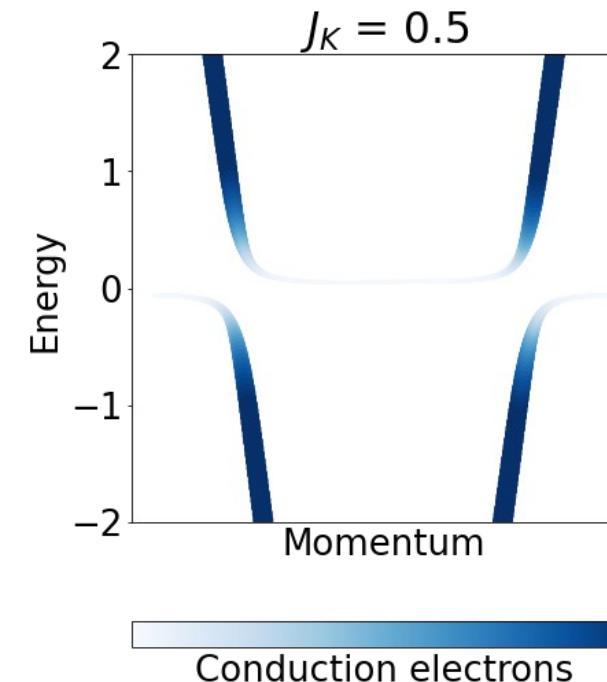
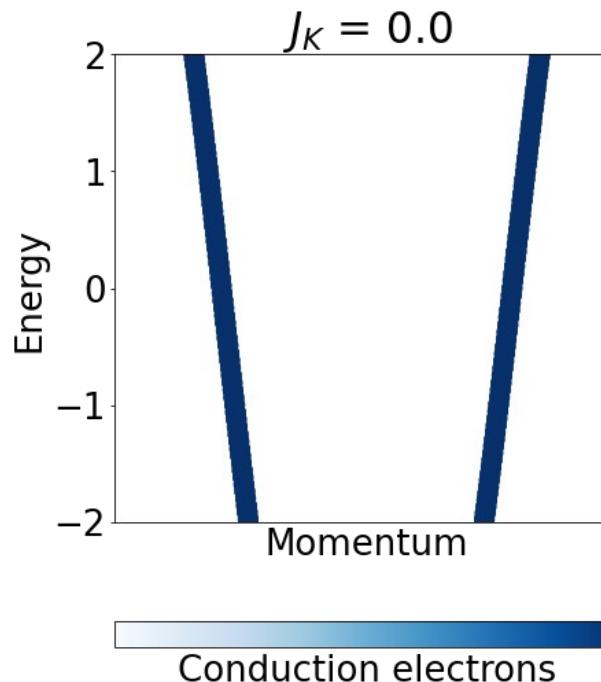
Kondo site/conduction electron

Kondo site/conduction electron

Kondo site/conduction electron

Spectral function of conduction electrons

The conduction electrons develop a heavy mass due to the Kondo coupling

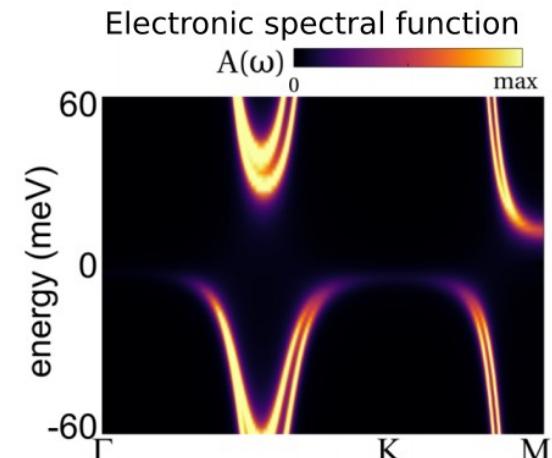
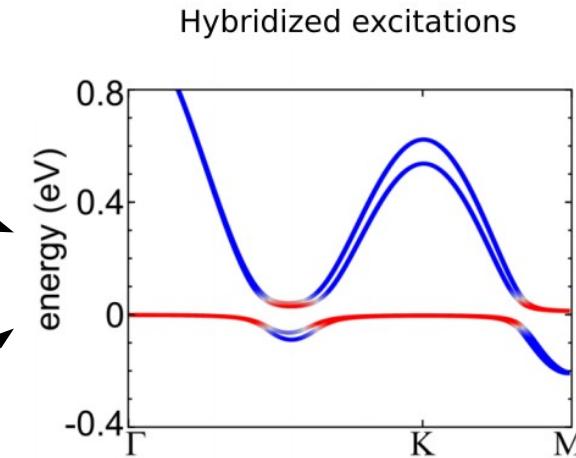
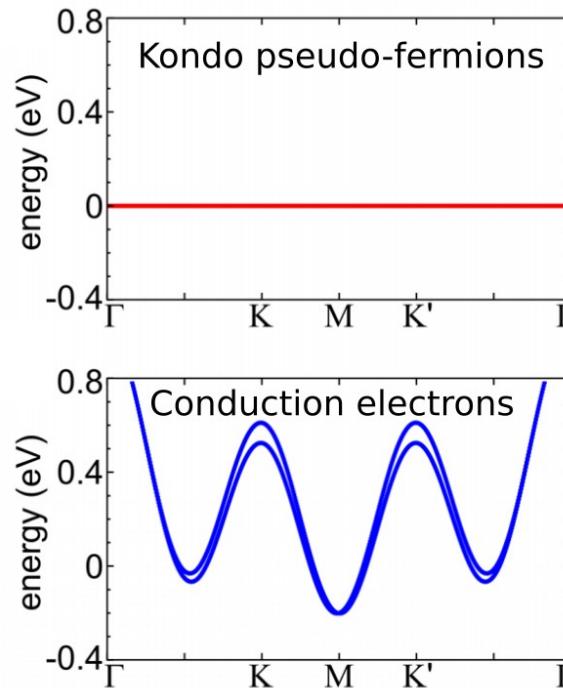


Nearly flat dispersion

Heavy-fermion Kondo insulators

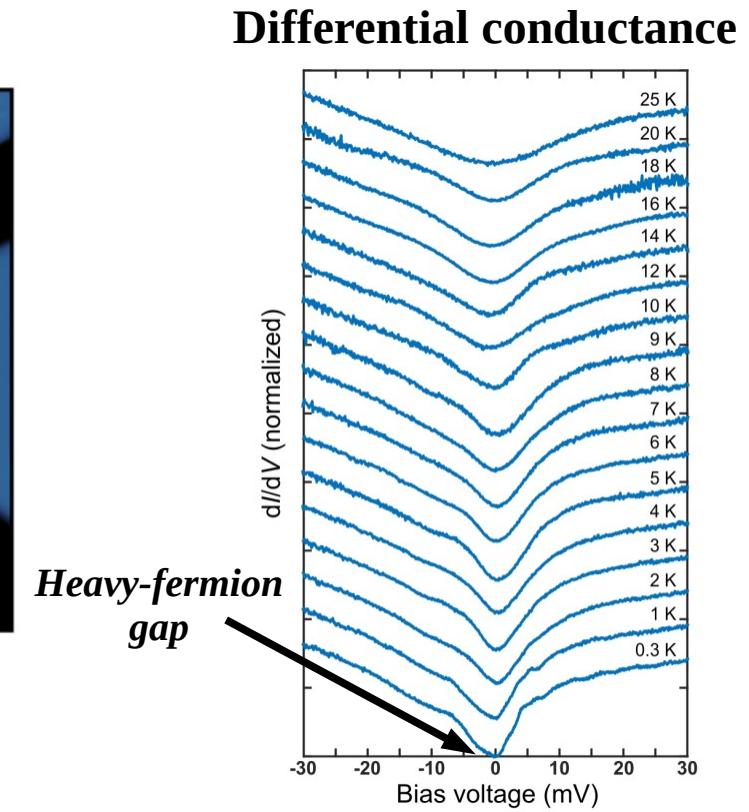
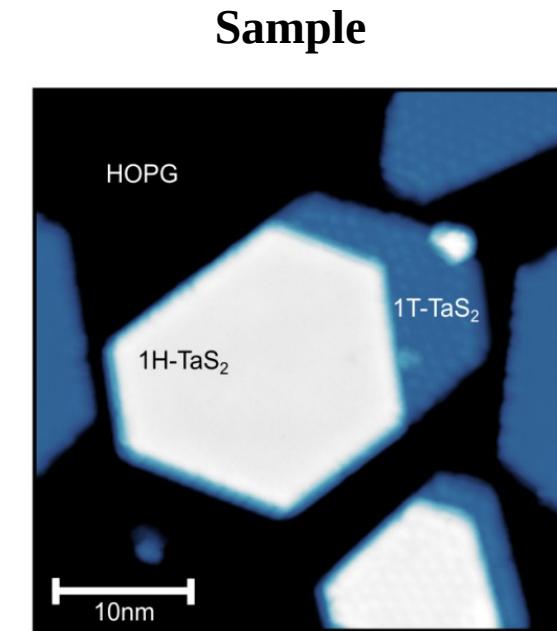
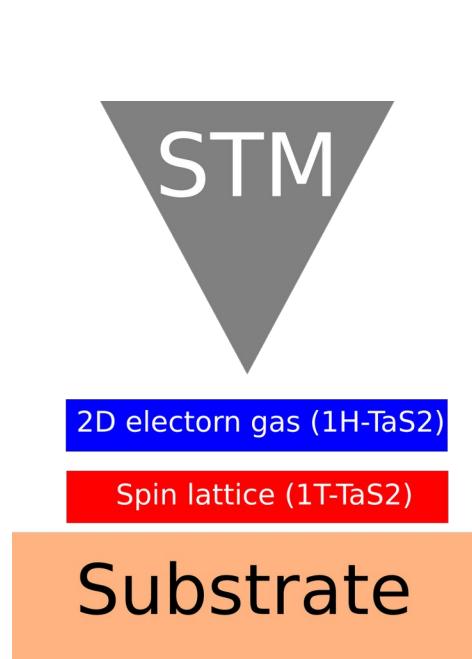
Kondo physics introduces resonant pseudo-fermions at the chemical potential

Leading to the opening of a heavy-fermion gap



Heterostructures of 1H-TaS₂/1T-TaS₂

Heavy-fermion in 1T-1H TaS₂ (experiment)



A heavy-fermion gap appears

Nature 599, 582–586 (2021)

(Optional) Interactive exercises

Download the Jupyter-notebook from

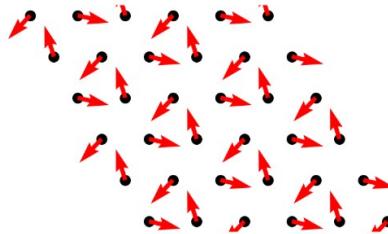
https://github.com/joselado/roscff_summer_school_2023



You will see examples with the code

```
from pygula import geometry
g = geometry.triangular_lattice() # generate a chain
g = g.get_supercell((3,3)) # make a supercell
h = g.get_hamiltonian() # generate the Hamiltonian

# generate the SCF Hamiltonian
U = 10 # strong Hubbard interaction
h = h.get_scf_hamiltonian(U=U,mf="XY",mix=0.9) # solve the interacting problem with a mean-field guess
hs = h.get_supercell(2) # generate a supercell
mx = hs.extract("mx") ; my = hs.extract("my") ; x = hs.geometry.r[:,0] ; y = hs.geometry.r[:,1] # get magnetization
plt.scatter(x,y,c="black",s=400) ; plt.quiver(x,y,mx,my,color="red") # plot magnetization
plt.axis("equal") ; plt.axis("off")
```



You have to modify them, and answer questions

Exercise

- Plot the band structure for the SCF solution for the 3x3 supercell, and estimate its gap
- Plot the band structure for the SCF solution for the 1x1 supercell, and estimate its gap
- Can you infer which one is the lowest energy solution, and why?

Take home

- Van der Waals magnets show highly rich phenomena
 - Collinear magnets (CrI_3)
 - Multiferroics (NiI_2)
 - Quantum spin liquid candidates (TaSe_2 , RuCl_3)
 - Heavy fermion Kondo insulators (1H/1T TaS_2)
- Realizing exotic quantum excitations
 - Magnons
 - Spinons

Thank you!