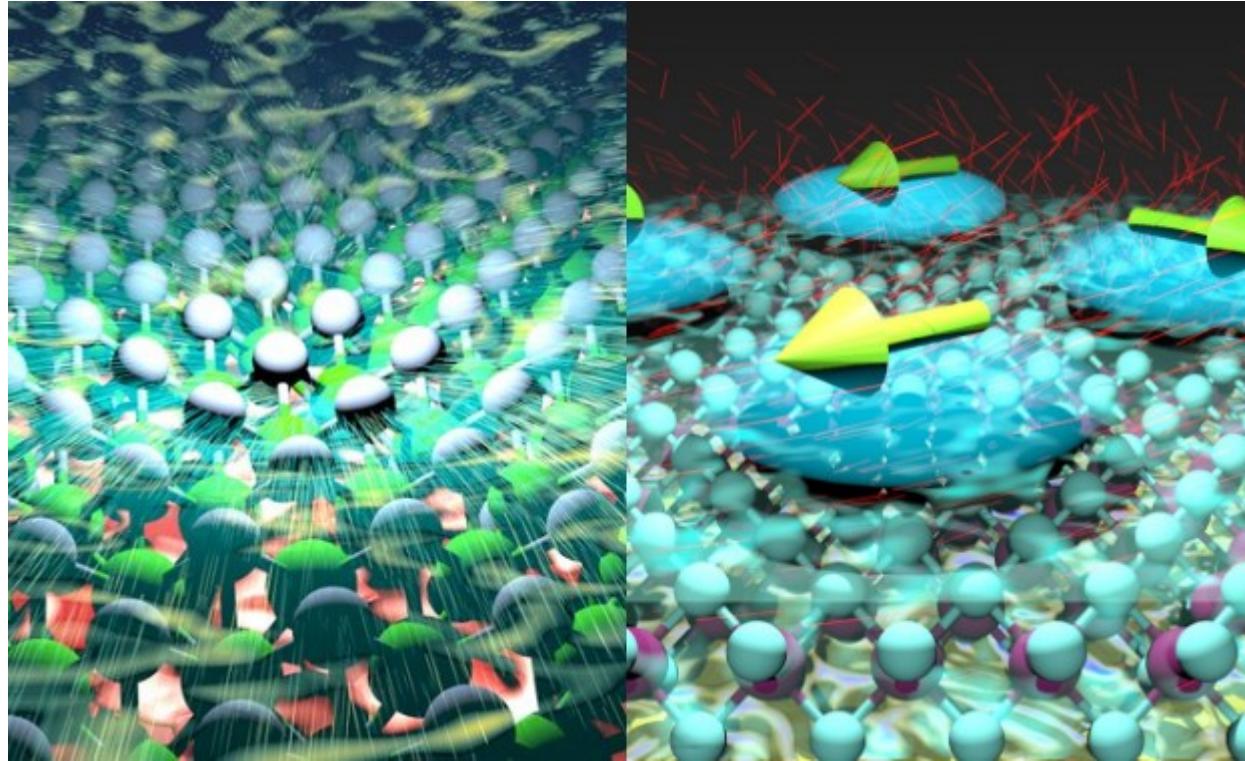


Lecture 2: Superconductivity and magnetism in 2D materials



Pedagogical School “Emergent phenomena in van der Waals heterostructures”

January 7th 2023, Tata Institute of Fundamental Research (TIFR), India

Plan for the lecture

- Conventional and unconventional van der Waals superconductors
- Impurities in 2D superconductors
- Topological superconductivity
- Weakly and strongly localized magnets
- Frustrated magnetism and quantum spin liquids
- Heavy-fermion van der Waals materials

Schedule for the lecture

- 30 min lecture
- 5 min break
- 30 min lecture

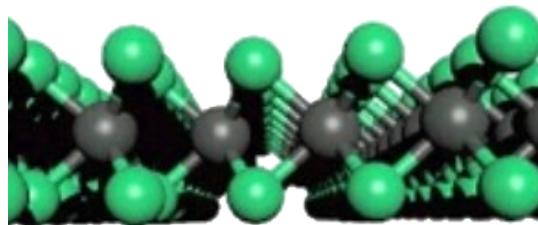
You can download the slides and software from

https://github.com/joselado/emergent_phenomena_in_van_der_Waals_school_tifr_2023



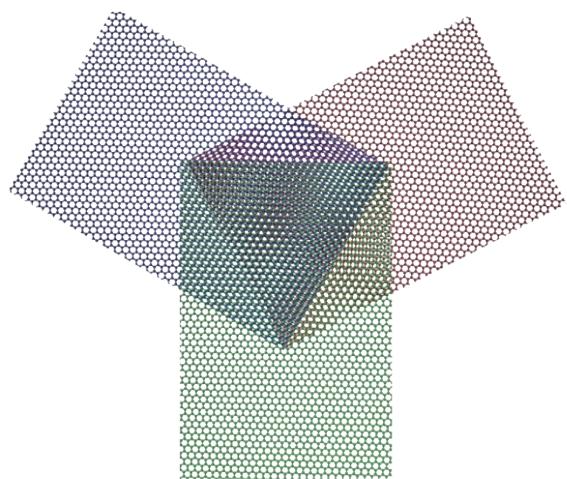
Superconducting van der Waals materials

NbSe_2



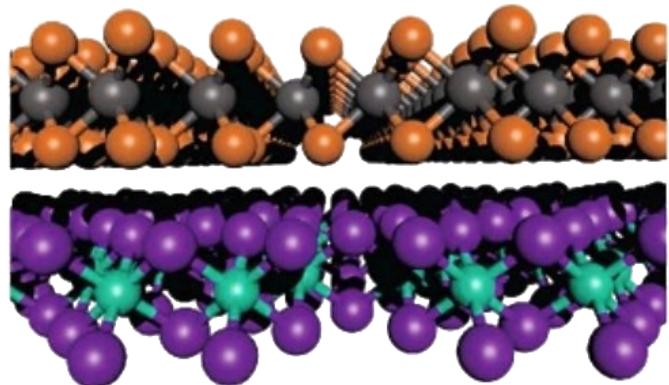
Gapped trivial
superconductors

Twisted graphene
multilayers



Nodal superconductivity
Spin-triplet superconductivity

$\text{CrBr}_3/\text{NbSe}_2$

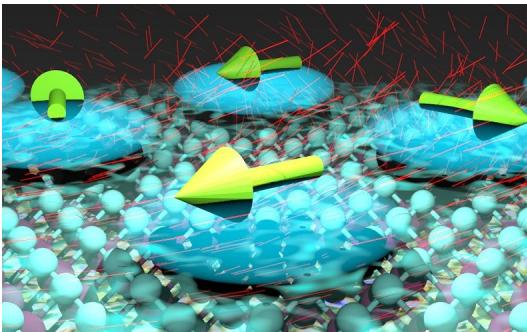


Gapped topological
superconductors

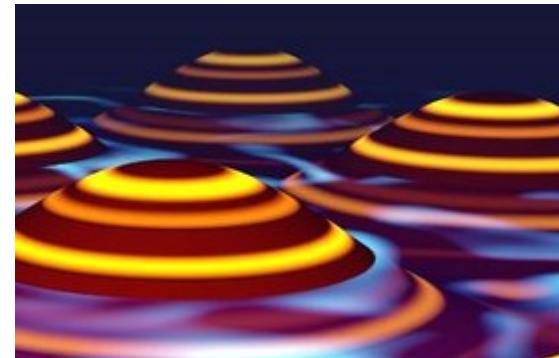
The role of electronic interactions

Electronic interactions are responsible for symmetry breaking

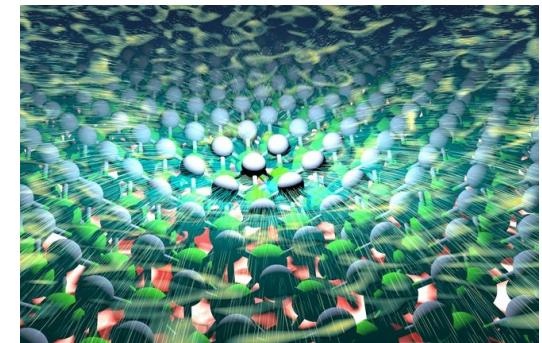
**Broken
time-reversal symmetry**
Classical magnets



**Broken
crystal symmetry**
Charge density wave



**Broken
gauge symmetry**
Superconductors



$$\mathbf{M} \rightarrow -\mathbf{M}$$

$$\mathbf{r} \rightarrow \mathbf{r} + \mathbf{R}$$

$$\langle c_{\uparrow} c_{\downarrow} \rangle \rightarrow e^{i\phi} \langle c_{\uparrow} c_{\downarrow} \rangle$$

Macroscopic quantum phenomena

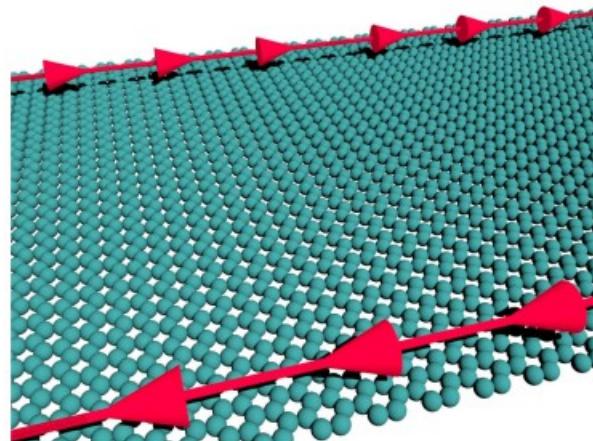
Superconductivity



$$\Phi = \frac{h}{2e}$$

Many-body state
Quantization of flux

Quantum Hall effect



$$\sigma_{xy} = \nu \frac{e^2}{h}$$

Single-particle state
Quantization of conductance

The theoretical description of superconductivity

A simple interacting Hamiltonian

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

From now on lets consider we have a spin degree of freedom \uparrow, \downarrow

What is the ground state of this Hamiltonian?

$U < 0$ Superconductivity

$U > 0$ Magnetism

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

Superconductivity requires effective attractive interactions

Origin of attractive interactions

Interactions between electrons can be effectively attractive when mediated by other quasiparticles

Conventional superconductors

Phonons

Unconventional superconductors

Antiferromagnetic magnons

Ferromagnetic magnons

Plasmons

Valence fluctuations

Charge fluctuations

...

The mean-field approximation, superconductivity

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

**Four fermions
(not exactly solvable)**

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

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

For $U < 0$
i.e. attractive interactions

$\Delta \sim \langle c_{i\uparrow}^\dagger c_{i\downarrow}^\dagger \rangle$ is the superconducting order

Superconductivity and gauge symmetry breaking

Gauge symmetry

$$c_n \rightarrow e^{i\phi} c_n$$

$$c_n^\dagger \rightarrow e^{-i\phi} c_n^\dagger$$

How does the superconducting order transform under a gauge transformation?

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

$$\Delta \rightarrow e^{-2i\phi} \Delta$$

A superconductor breaks gauge symmetry

The electronic structure of
a superconductor

The Nambu representation

How do we solve a Hamiltonian of the form $H = \sum_{\mathbf{k},s} \epsilon_{\mathbf{k}} c_{\mathbf{k},s}^\dagger c_{\mathbf{k},s} + \sum_{\mathbf{k}} \Delta c_{\mathbf{k},\uparrow}^\dagger c_{-\mathbf{k},\downarrow}^\dagger + h.c.$

Define a Nambu spinor

$$\Psi_{\mathbf{k}} = \begin{pmatrix} c_{\mathbf{k}\uparrow} \\ c_{\mathbf{k}\downarrow} \\ c_{-\mathbf{k}\downarrow}^\dagger \\ -c_{-\mathbf{k}\uparrow}^\dagger \end{pmatrix}$$

← Electron sector

← Hole sector

The Hamiltonian in the Nambu basis is quadratic and can be diagonalized

$$H = \Psi_{\mathbf{k}}^\dagger \mathcal{H} \Psi_{\mathbf{k}}$$

A single orbital superconductor

The original Hamiltonian

$$H = \sum_{\mathbf{k},s} \epsilon_{\mathbf{k}} c_{\mathbf{k},s}^\dagger c_{\mathbf{k},s} + \sum_{\mathbf{k}} \Delta c_{\mathbf{k},\uparrow}^\dagger c_{-\mathbf{k},\downarrow}^\dagger + h.c.$$

Can be rewritten as

$$H = \frac{1}{2} \Psi_{\mathbf{k}}^\dagger \mathcal{H} \Psi_{\mathbf{k}}$$

$$\Psi_{\mathbf{k}} = \begin{pmatrix} c_{\mathbf{k}\uparrow} \\ c_{\mathbf{k}\downarrow} \\ c_{-\mathbf{k}\downarrow}^\dagger \\ -c_{-\mathbf{k}\uparrow}^\dagger \end{pmatrix}$$

Electron sector

Hole sector

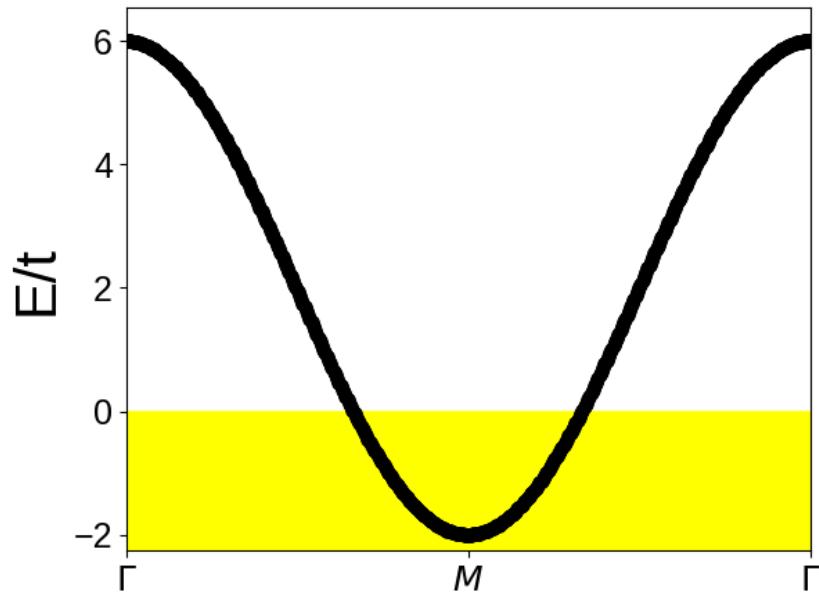
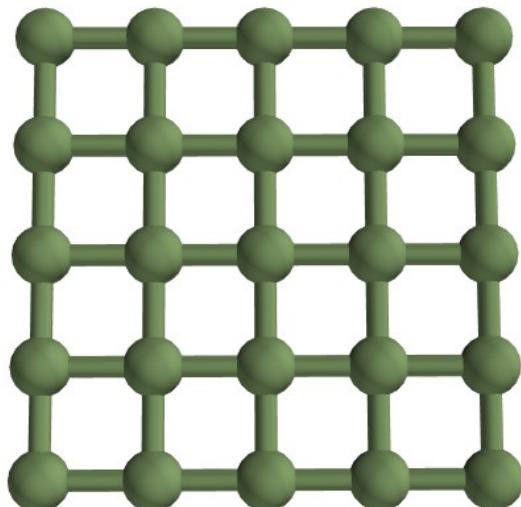
with

$$\mathcal{H} = \begin{pmatrix} \epsilon_{\mathbf{k}} & 0 & \Delta & 0 \\ 0 & \epsilon_{\mathbf{k}} & 0 & \Delta \\ \Delta & 0 & -\epsilon_{\mathbf{k}} & 0 \\ 0 & \Delta & 0 & -\epsilon_{\mathbf{k}} \end{pmatrix}$$

A single orbital superconductor

Single orbital in the square lattice

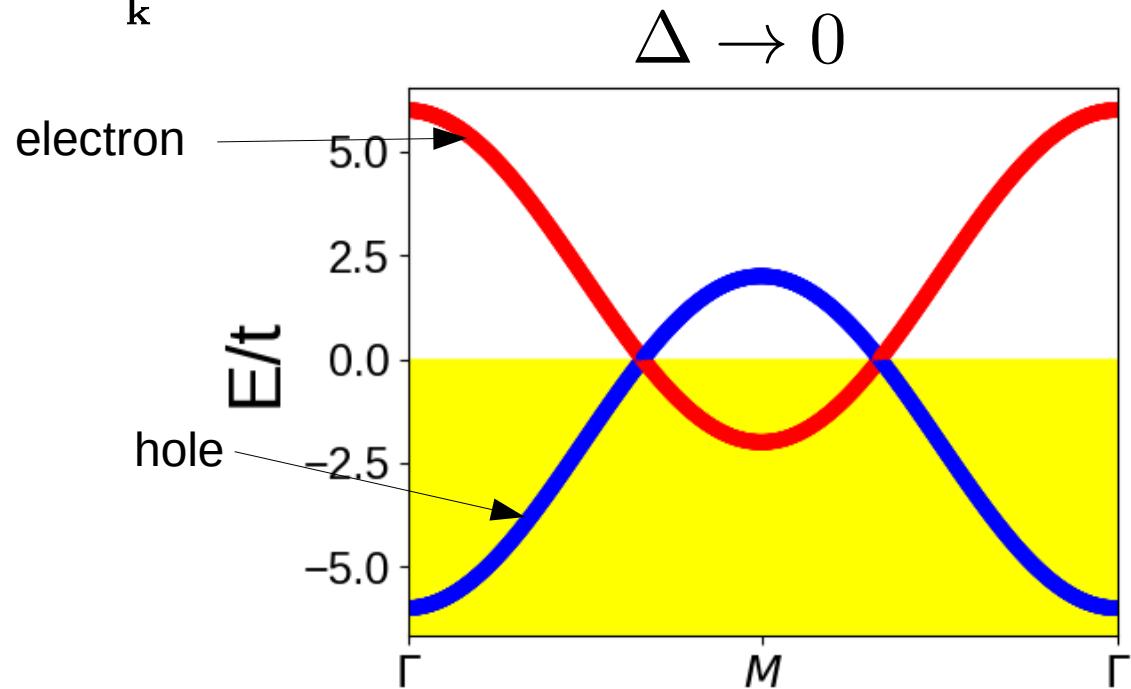
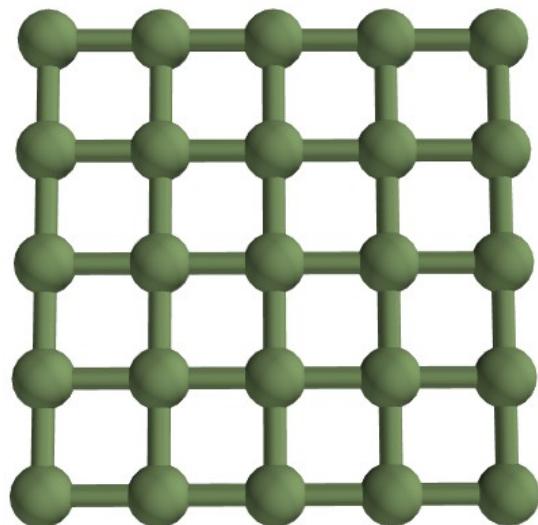
$$H = \sum_{\mathbf{k}, s} \epsilon_{\mathbf{k}} c_{\mathbf{k}, s}^{\dagger} c_{\mathbf{k}, s}$$



A single orbital superconductor

Single orbital in the square lattice

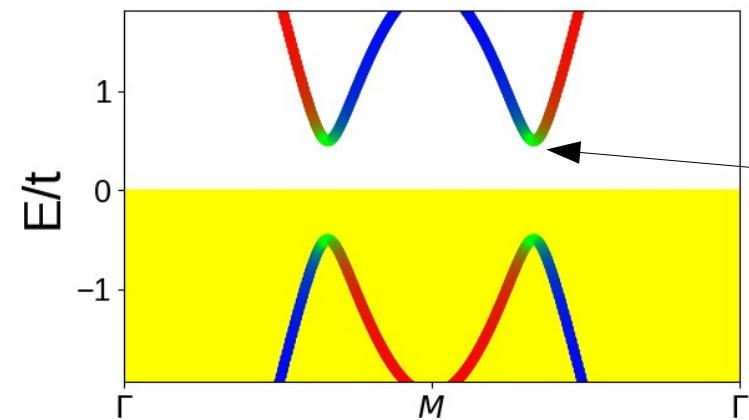
$$H = \sum_{\mathbf{k}, s} \epsilon_{\mathbf{k}} c_{\mathbf{k}, s}^\dagger c_{\mathbf{k}, s} + \sum_{\mathbf{k}} \Delta c_{\mathbf{k}, \uparrow}^\dagger c_{-\mathbf{k}, \downarrow}^\dagger + h.c.$$



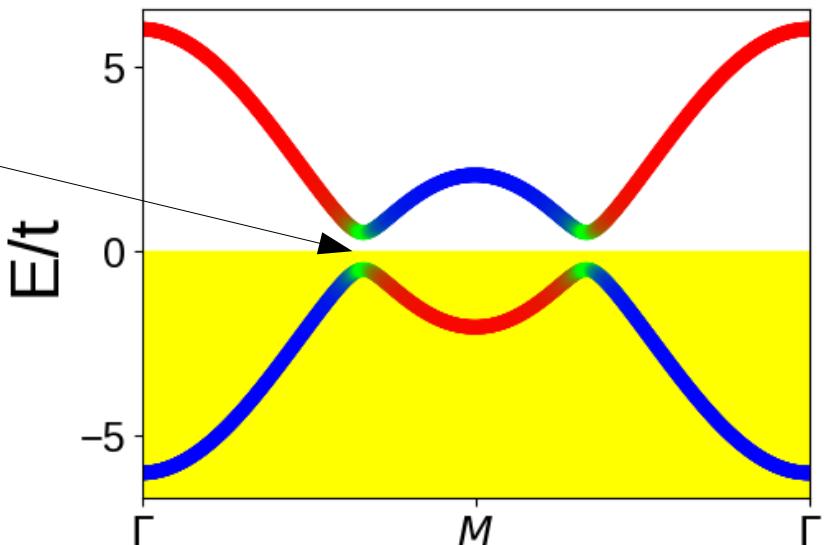
A single orbital superconductor

Single orbital in the square lattice

$$H = \sum_{\mathbf{k}, s} \epsilon_{\mathbf{k}} c_{\mathbf{k}, s}^\dagger c_{\mathbf{k}, s} + \sum_{\mathbf{k}} \Delta c_{\mathbf{k}, \uparrow}^\dagger c_{-\mathbf{k}, \downarrow}^\dagger + h.c.$$

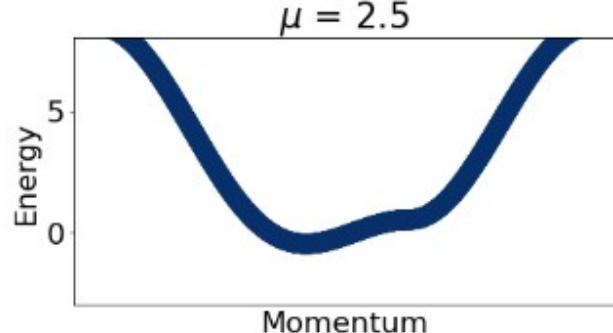
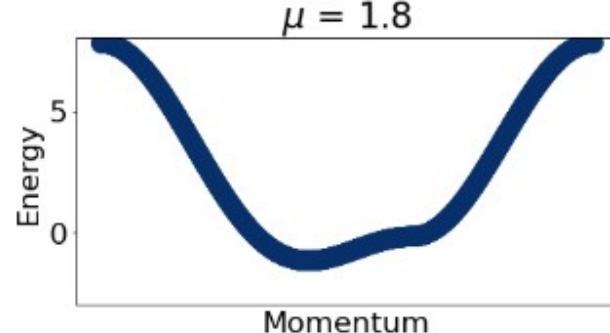
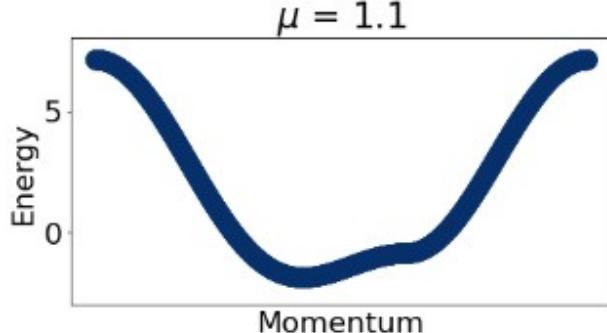
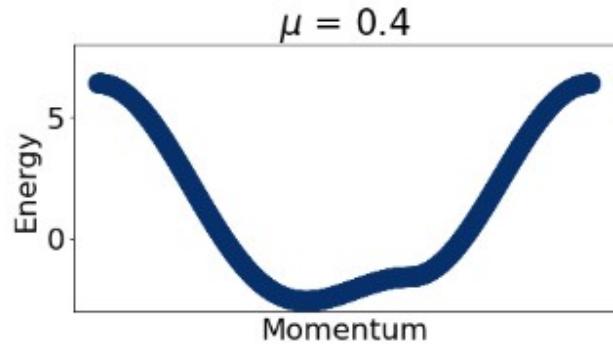
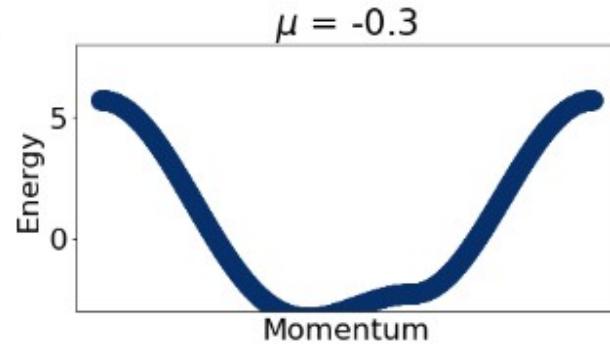
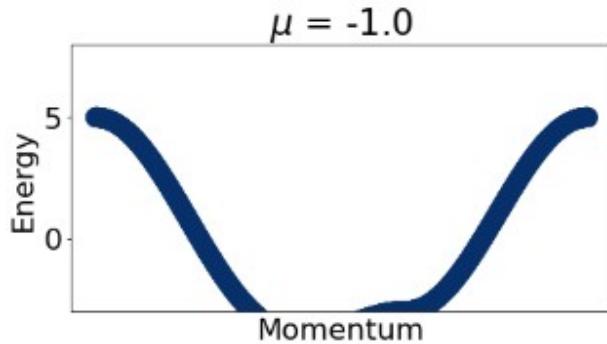


SC gap



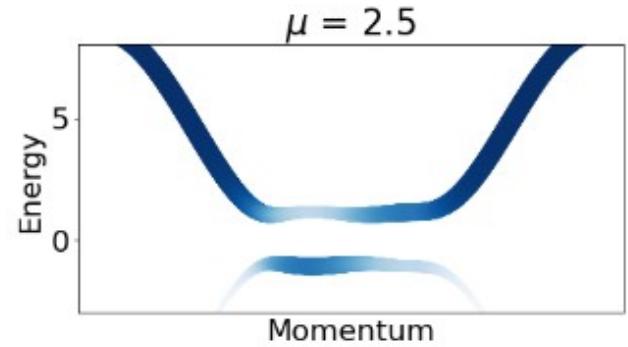
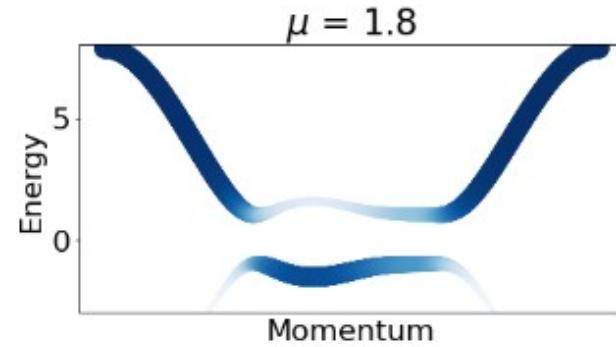
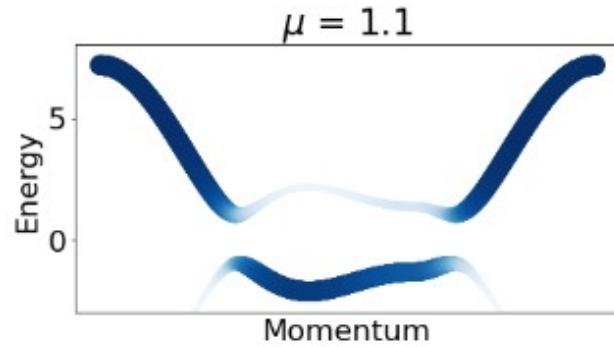
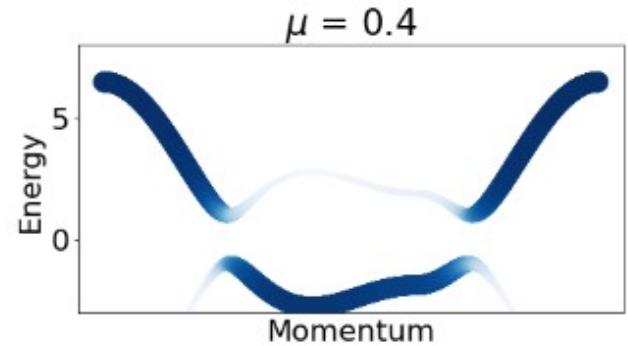
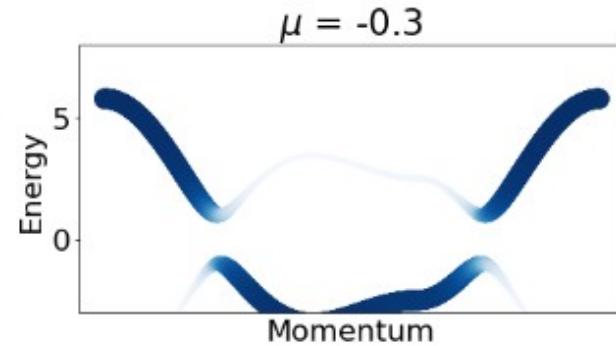
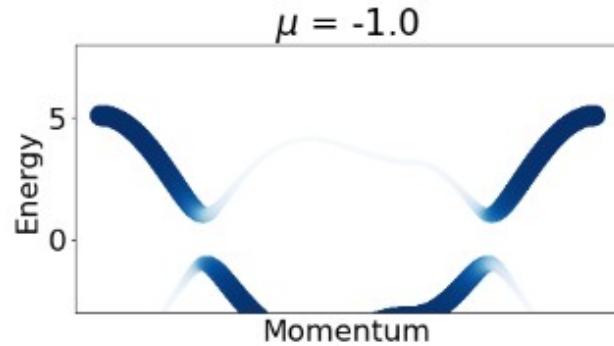
Impact of superconductivity in the electronic structure

Let us take the electronic structure of a triangular lattice



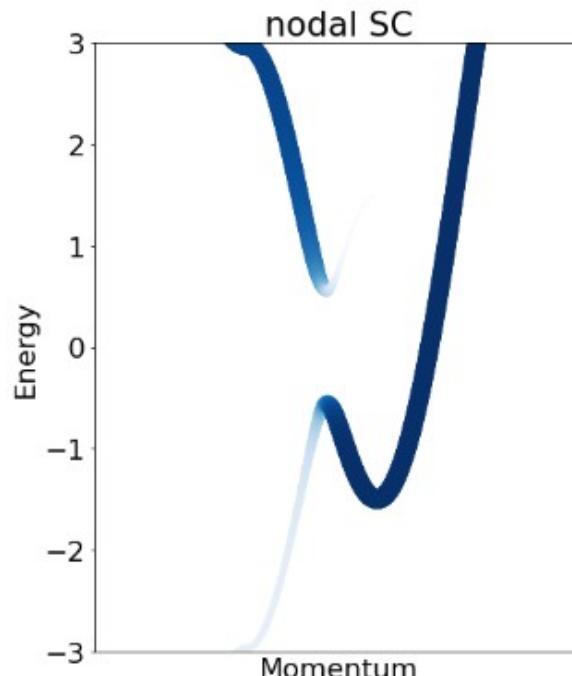
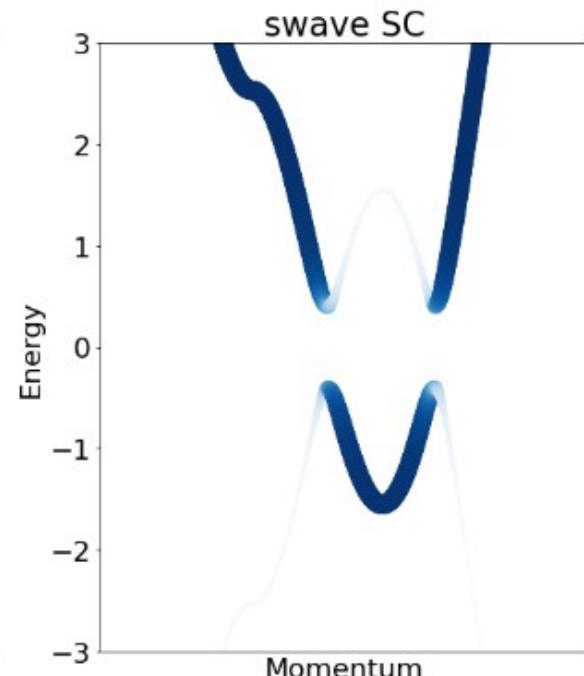
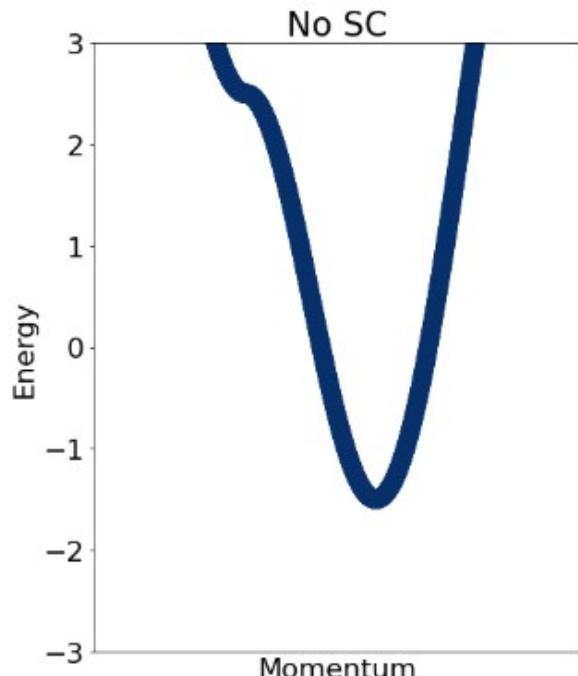
Impact of superconductivity in the electronic structure

A uniform superconducting terms opens up a gap in the electronic structure



Gapped and gapless superconductivity

The electronic structure is modified differently depending on the type of superconductivity



Square lattice

Unconventional superconductivity

Superconducting momentum symmetries

A generic type of a superconductor is characterized by the order parameter

Real space

$$\Delta_{\uparrow\downarrow}(\mathbf{r}, \mathbf{r}') \sim \langle c_{\mathbf{r}\uparrow} c_{\mathbf{r}'\downarrow} \rangle$$

Reciprocal space

$$\Delta_{\uparrow\downarrow}(\mathbf{k}) \sim \langle c_{\mathbf{k}\uparrow} c_{-\mathbf{k}\downarrow} \rangle$$

The superconducting state can be characterized by the symmetry of $\Delta_{\uparrow\downarrow}(\mathbf{k})$

Singlet and triplet superconductors

The superconducting order inherits a symmetry property

$$\Delta_{\vec{k},s_1s_2} = -\Delta_{-\vec{k},s_2s_1} = \begin{cases} \Delta_{-\vec{k},s_1s_2} = -\Delta_{\vec{k},s_2s_1} & \text{even} \\ -\Delta_{-\vec{k},s_1s_2} = \Delta_{\vec{k},s_2s_1} & \text{odd} \end{cases}$$

Spin-singlet (even)

$$\Delta_{\uparrow\downarrow}(\mathbf{k}) = \Delta_{\uparrow\downarrow}(-\mathbf{k})$$

Spin-triplet (odd)

$$\Delta_{\uparrow\uparrow}(\mathbf{k}) = -\Delta_{\uparrow\uparrow}(-\mathbf{k})$$

The symmetry of the superconducting order characterizes the superconductor

Generating a spin-triplet superconductor

Let us take a Hamiltonian breaking time-reversal with attractive interactions

$$H = t \sum_{\langle ij \rangle} c_{i,s}^\dagger c_{j,s} + J_z \sum_i \sigma_z^{s,s'} c_{i,s}^\dagger c_{j,s'} + V_1 \sum_{\langle ij \rangle} \sum_s (c_{i,s}^\dagger c_{i,s}^\dagger) \sum_{s'} (c_{j,s'}^\dagger c_{j,s'}^\dagger)$$

kinetic *exchange* *attractive interactions*

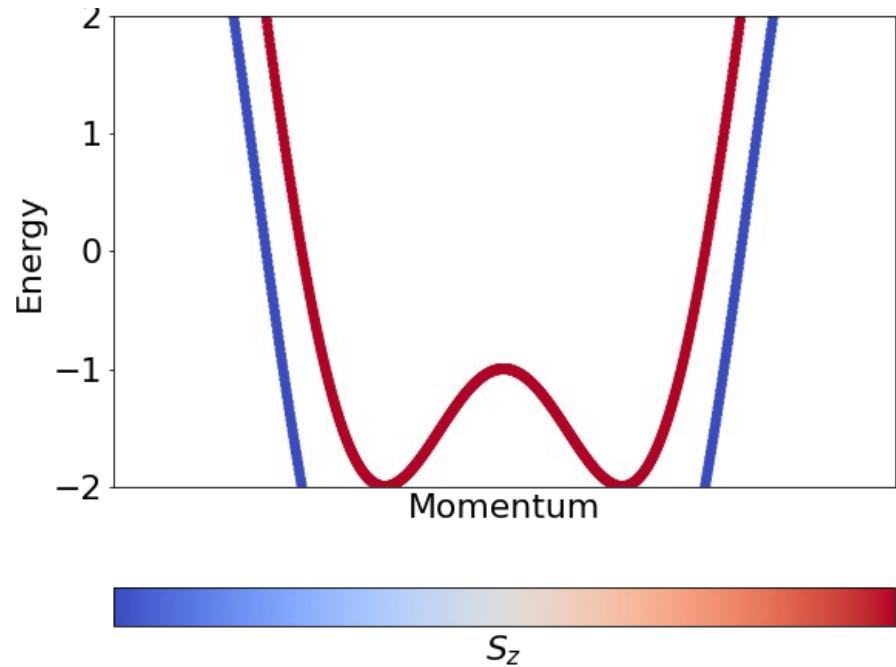
At the mean-field level, this may generate

$$H \sim \sum_{\langle ij \rangle} \Delta_{\uparrow\uparrow} c_{i,\downarrow}^\dagger c_{j,\downarrow}^\dagger + h.c. + \dots$$

Generating a spin-triplet superconductor

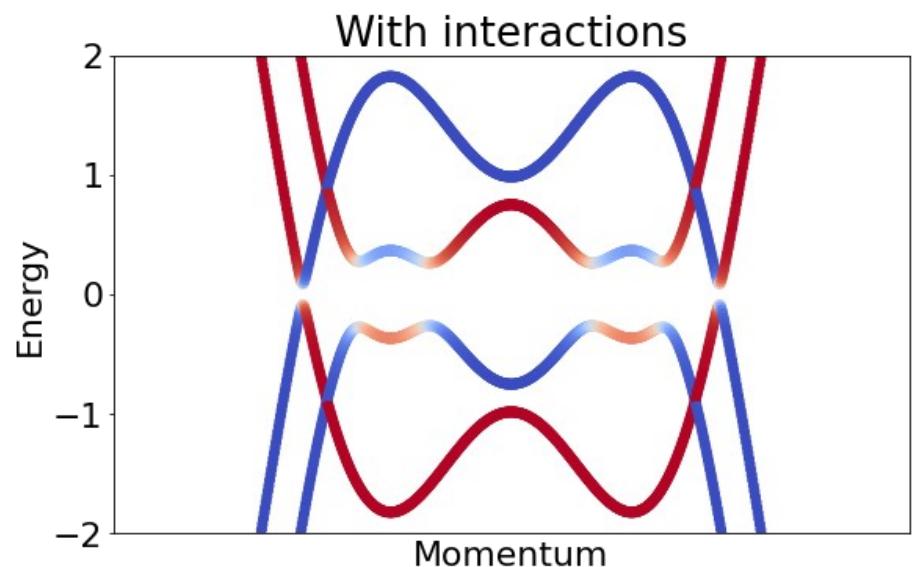
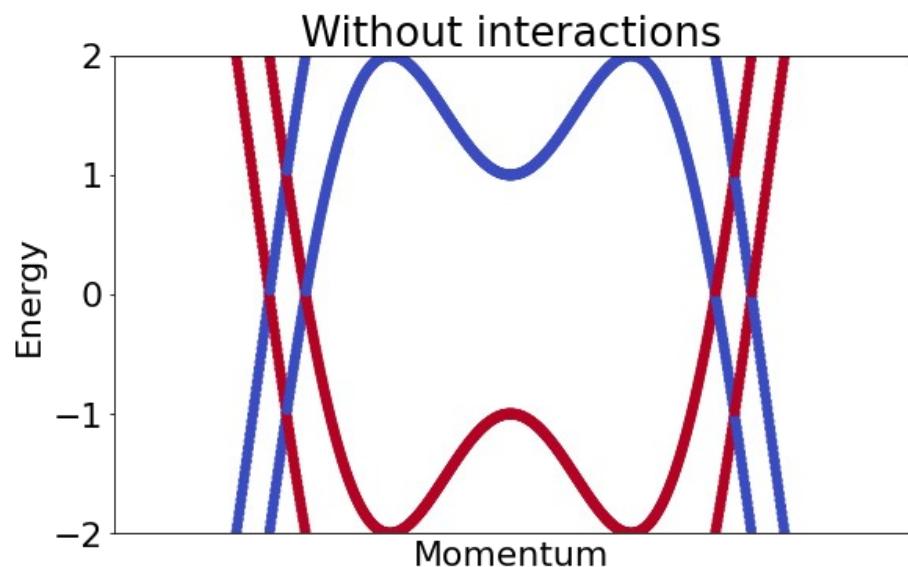
We start with a ferromagnetic 2D material, and see if interactions create superconductivity

$$H = t \sum_{\langle ij \rangle} c_{i,s}^\dagger c_{j,s} + J_z \sum_i \sigma_z^{s,s'} c_{i,s}^\dagger c_{j,s'}$$

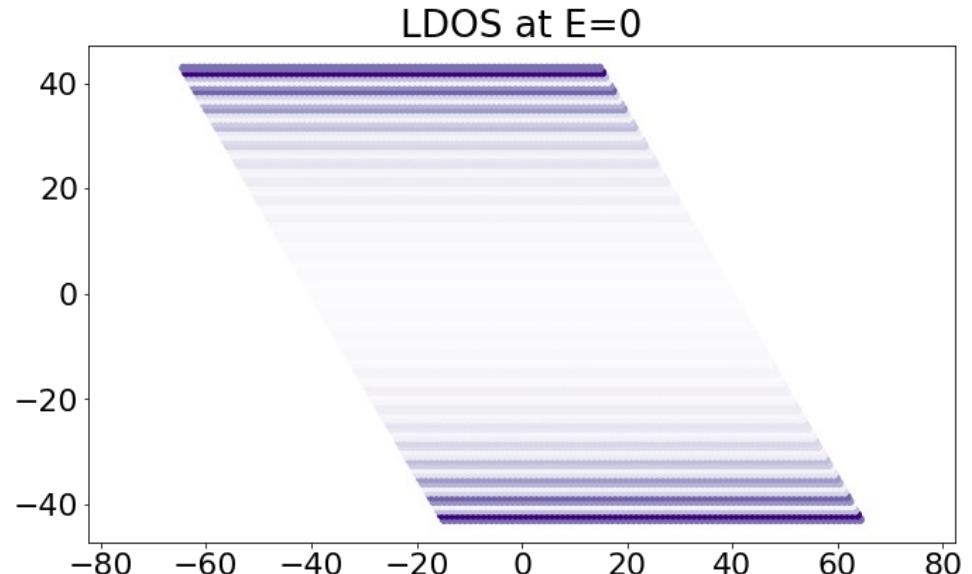
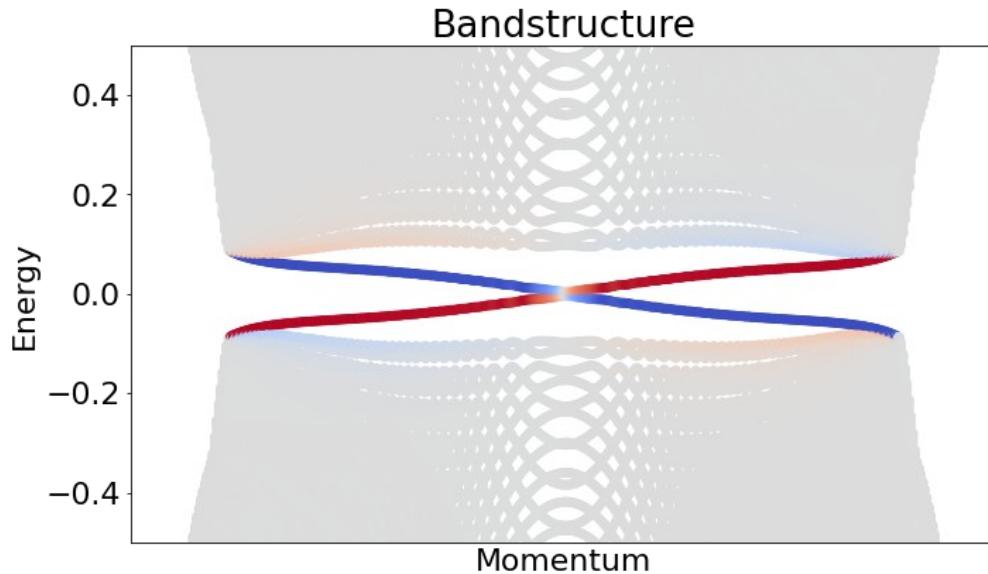


A spin-triplet superconductor

$$H = t \sum_{\langle ij \rangle} c_{i,s}^\dagger c_{j,s} + J_z \sum_i \sigma_z^{s,s'} c_{i,s}^\dagger c_{j,s'} + V_1 \sum_{\langle ij \rangle} \sum_s (c_{i,s}^\dagger c_{i,s}^\dagger) \sum_{s'} (c_{j,s'}^\dagger c_{j,s'}^\dagger)$$



A spin-triplet superconductor, in a strip



A ferromagnetic superconductor can develop edge modes

Gapped and gapless superconductors

Let us focus on the superconducting order $\Delta_{\uparrow\downarrow}(\mathbf{k})$

Fully gapped

$$|\Delta_{\uparrow\downarrow}(\mathbf{k})|^2 > 0$$

Gapless

$$|\Delta_{\uparrow\downarrow}(\mathbf{k}_\alpha)|^2 = 0$$

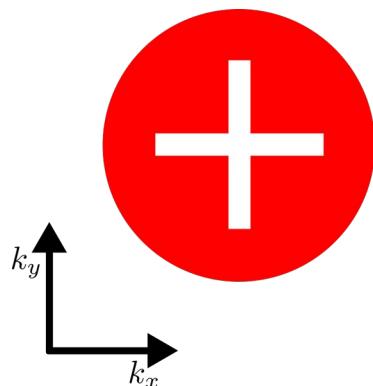
$NbSe_2$

*Twisted trilayer
graphene*

Superconducting momentum symmetries

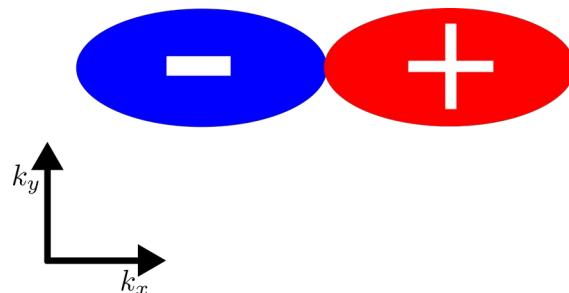
The superconducting state can be characterized by the symmetry of $\Delta(\mathbf{k})$

s-wave



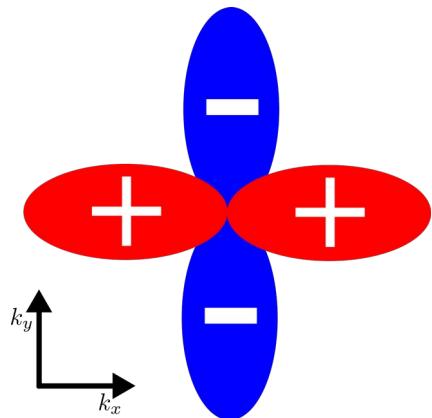
Conventional SC
(driven by phonons)

p-wave



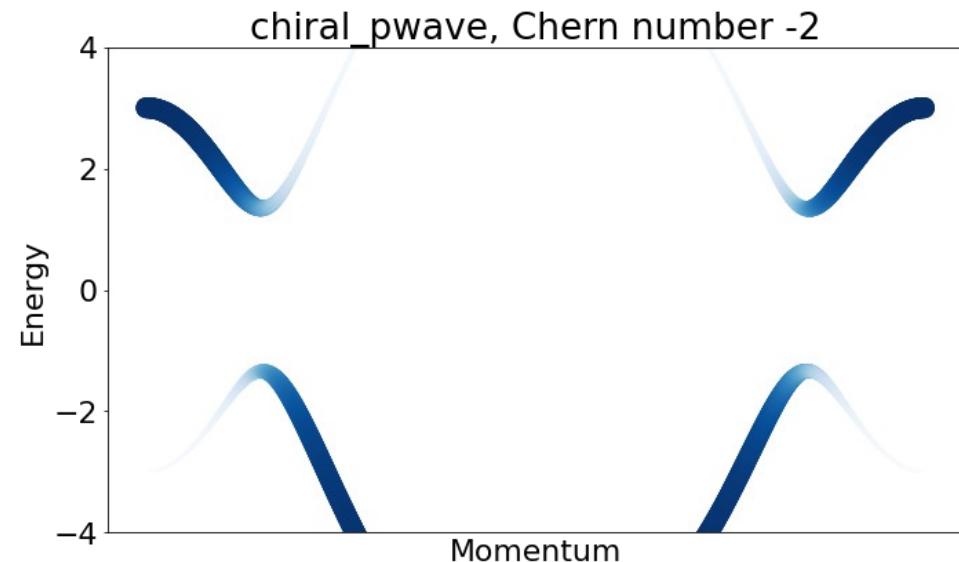
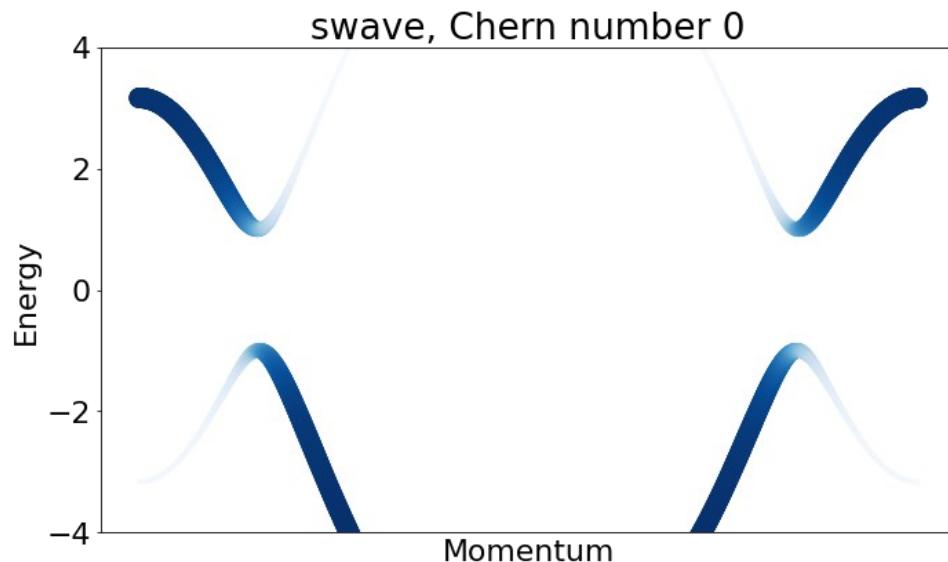
Unconventional SC
(driven by FE magnons)

d-wave



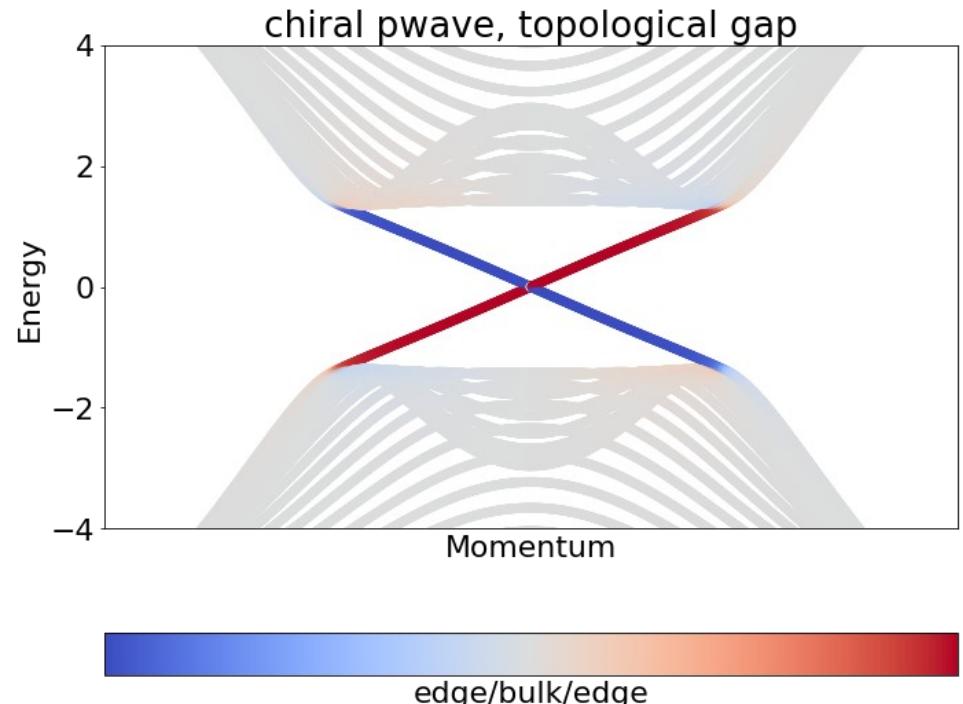
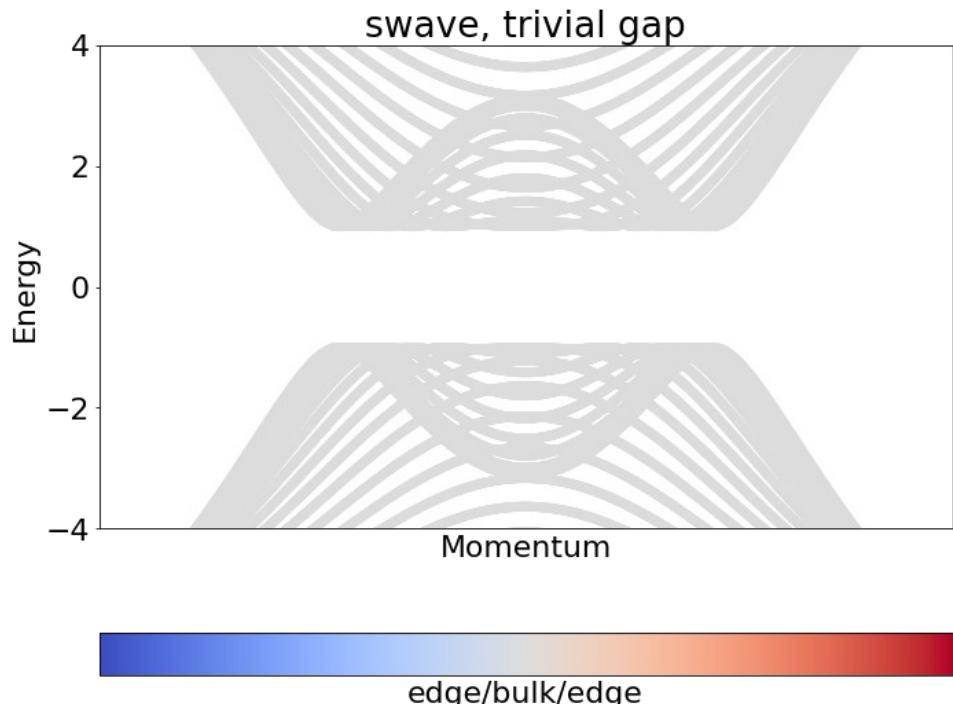
Unconventional SC
(driven by AF magnons)

Gapped superconducting orders can be radically different



While both orders are gapped, they have different topological properties

Gapped superconducting orders can be radically different

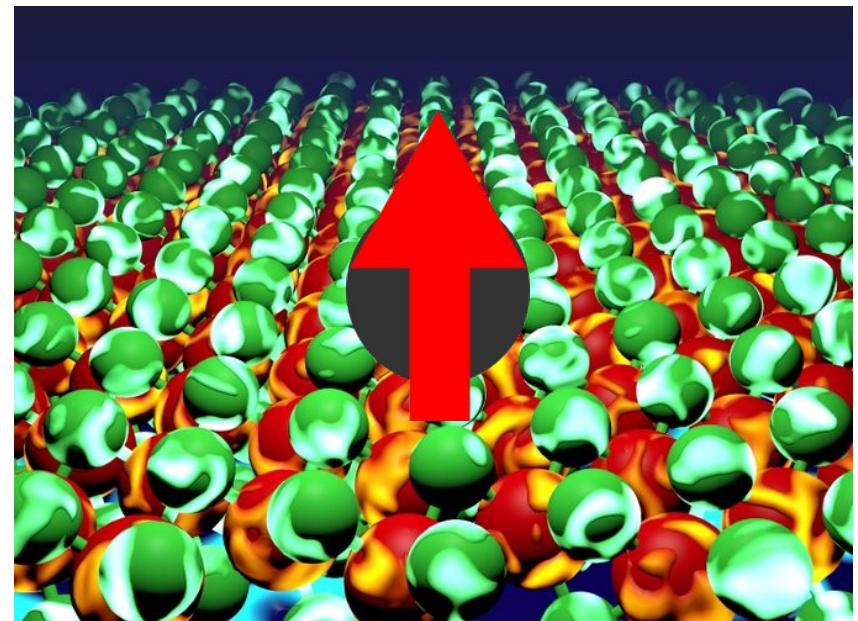
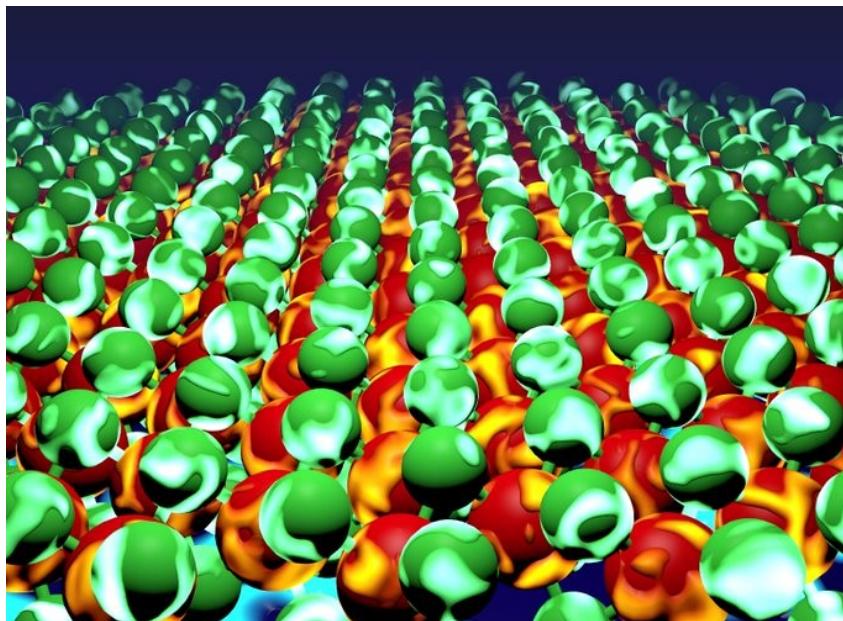


The topological superconducting gap leads to protected edge excitations

Pair breaking effects in superconductors

Impurities in 2D superconductors

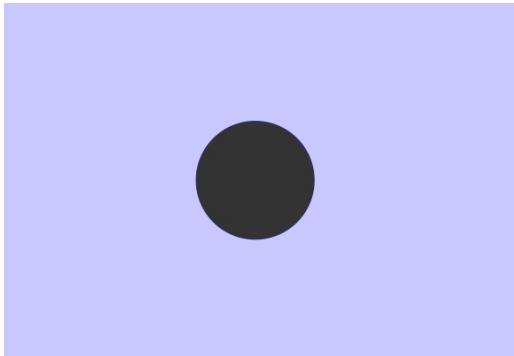
So-far we considered pristine superconductors, but what happens when we put impurities?



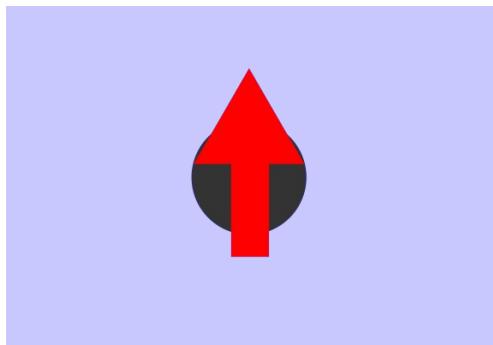
How detrimental are defects in superconductors?

Impurities in 2D superconductors

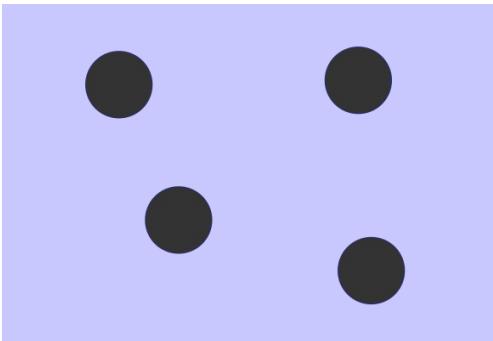
A non-magnetic impurity



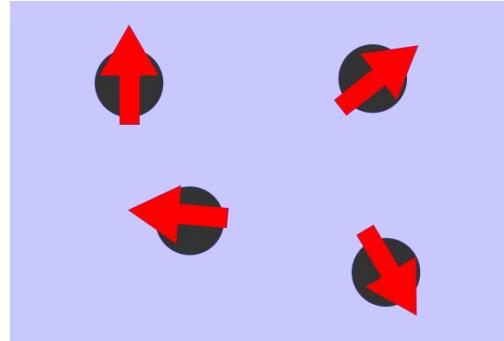
A magnetic impurity



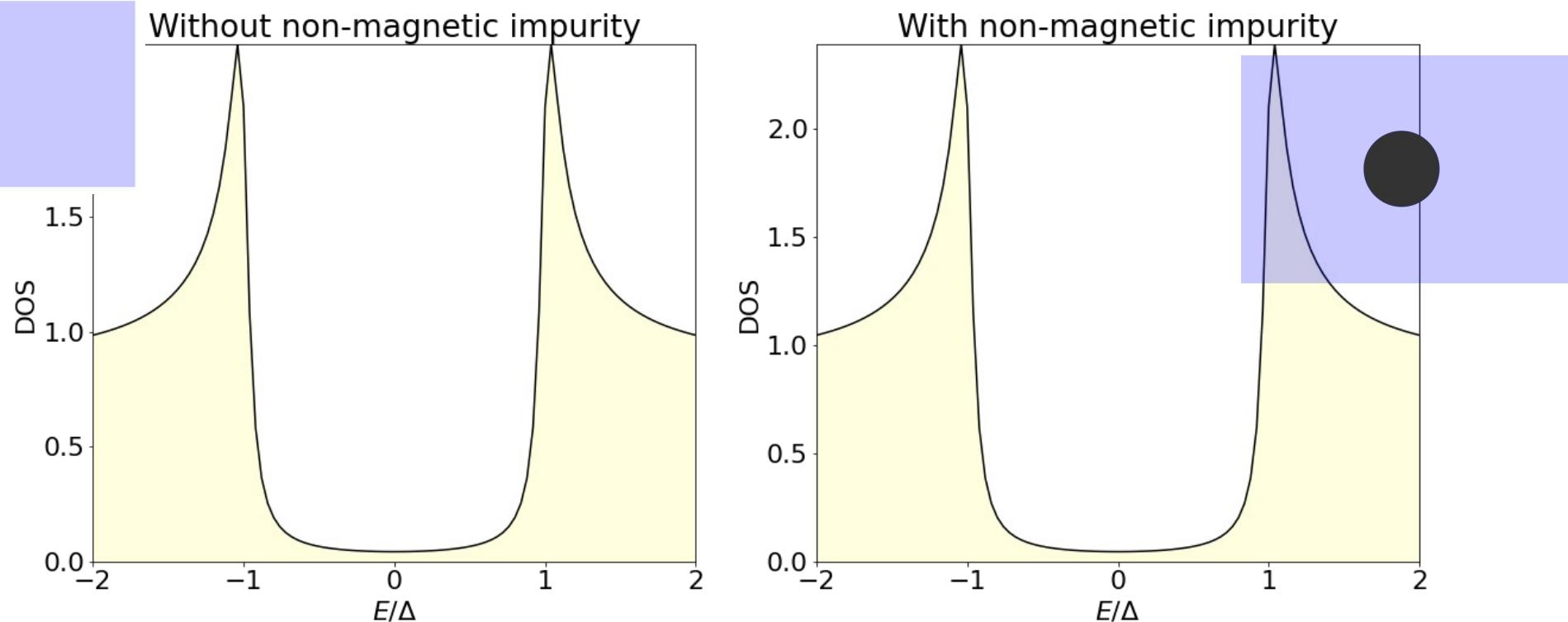
Several non-magnetic impurities



Several magnetic impurities

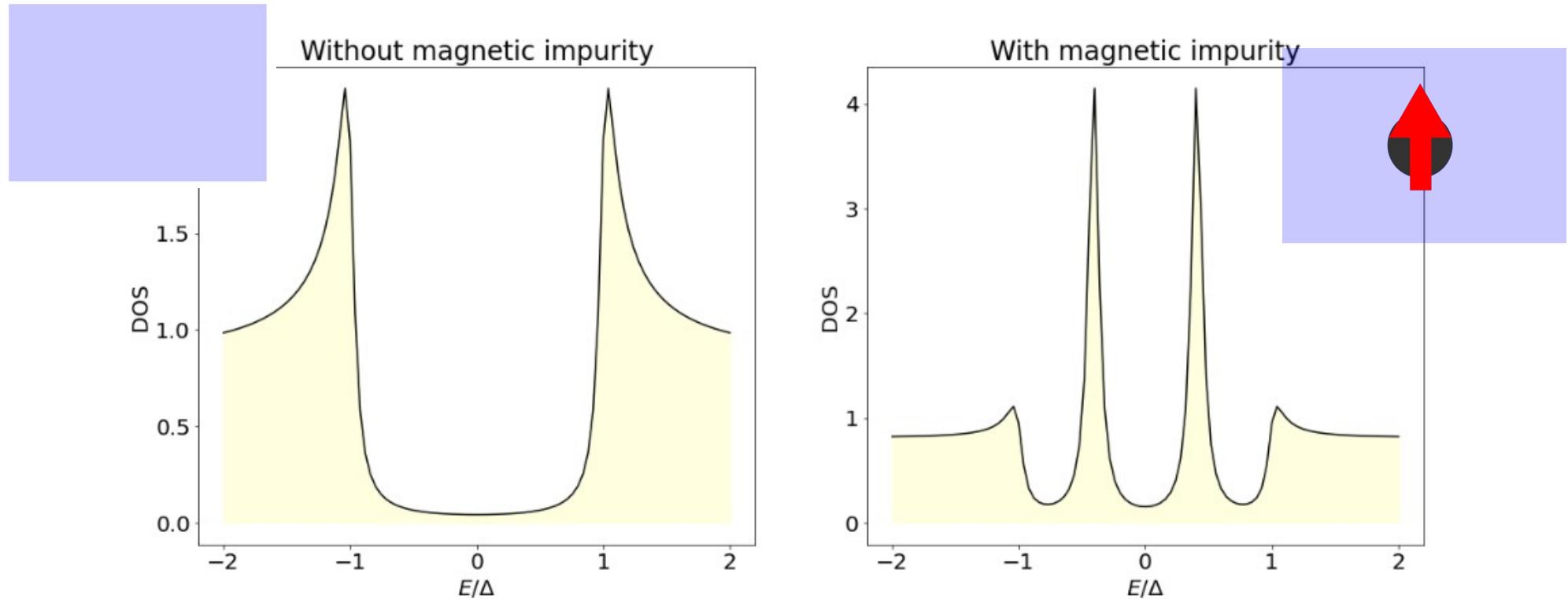


Non-magnetic impurity, conventional S-wave superconductor



A non-magnetic impurity does not affect conventional s-wave superconductors

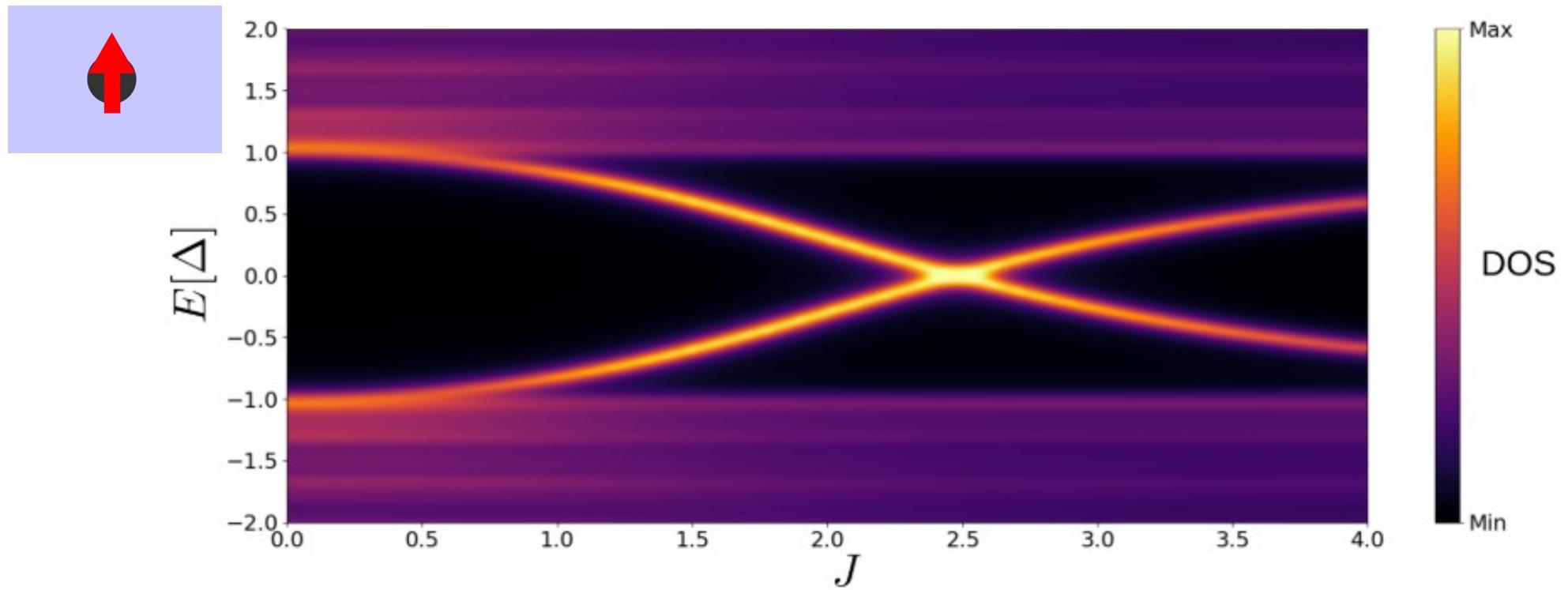
The interplay between magnetism and superconductivity



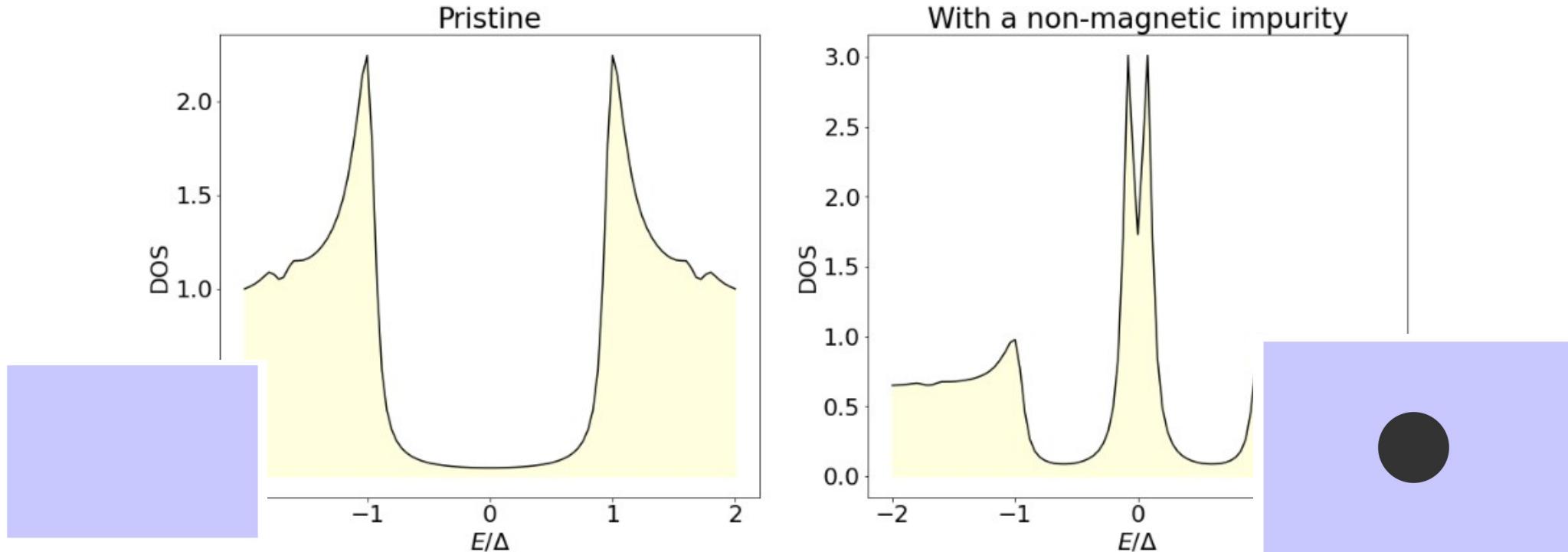
Magnetic impurities create in-gap states in fully gapped superconductors

The interplay between magnetism and superconductivity

The exchange coupling controls the energy of the in-gap state



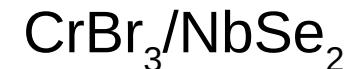
The interplay between impurities and unconventional superconductivity



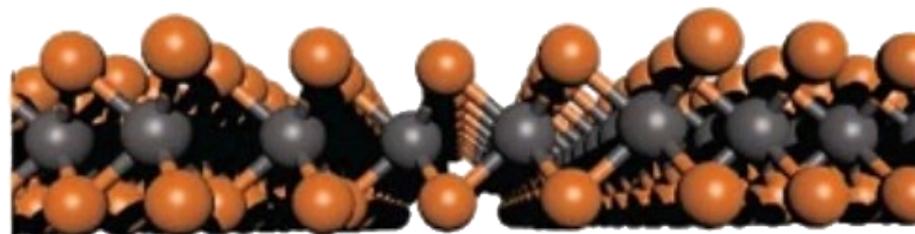
Non-magnetic impurities create in-gap states in fully gapped unconventional superconductors

Two-dimensional artificial topological superconductivity

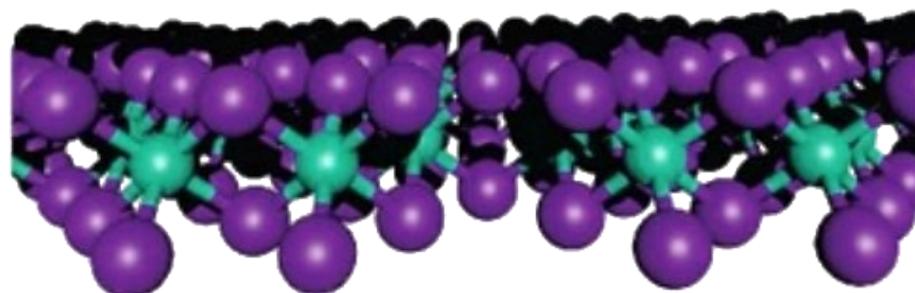
Engineering unconventional superconductors with conventional ones



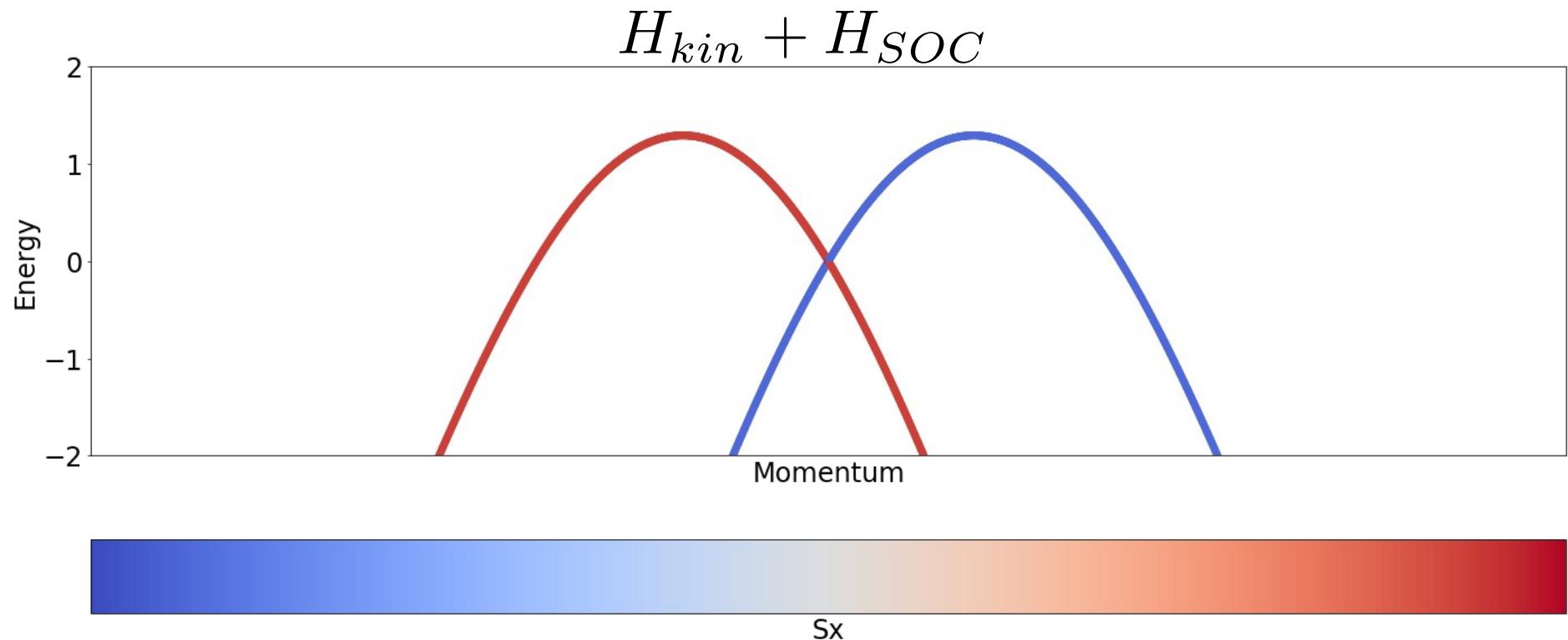
Van der Waals superconductor



Van der Waals ferromagnet



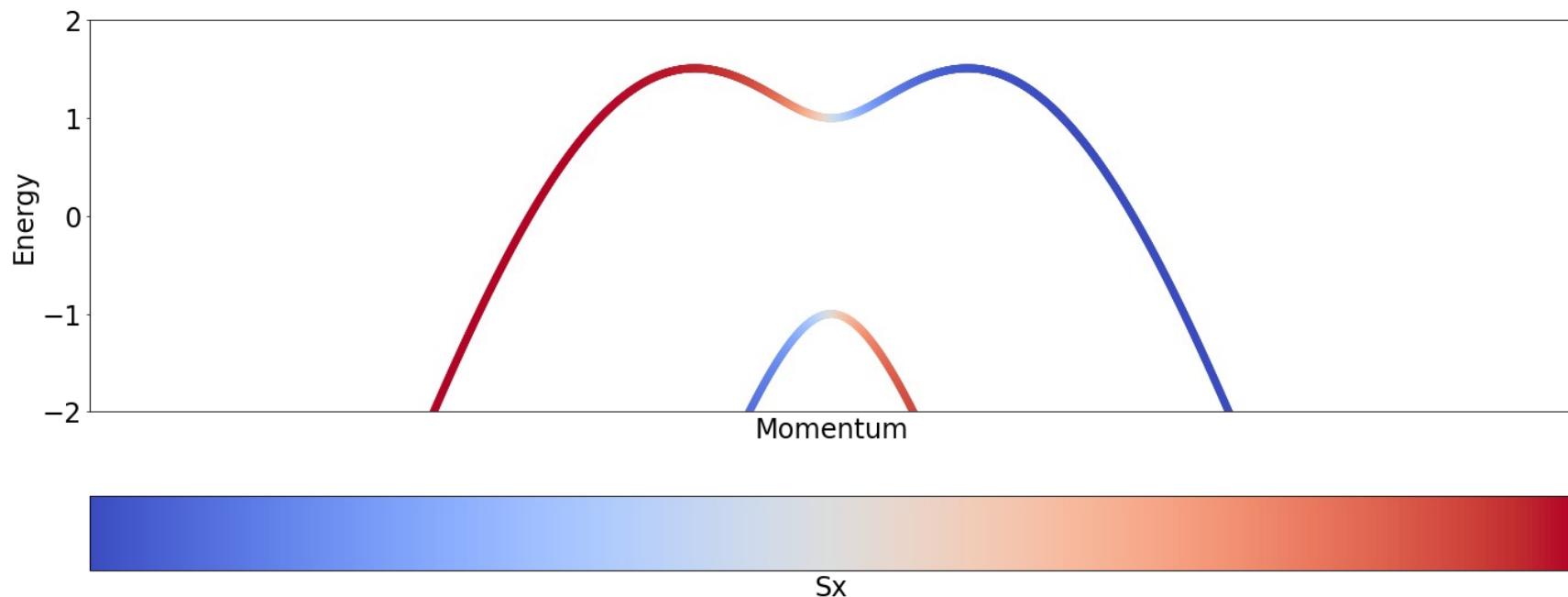
Engineering helical states



With Rashba SOC, a spin-dependent spin splitting appears

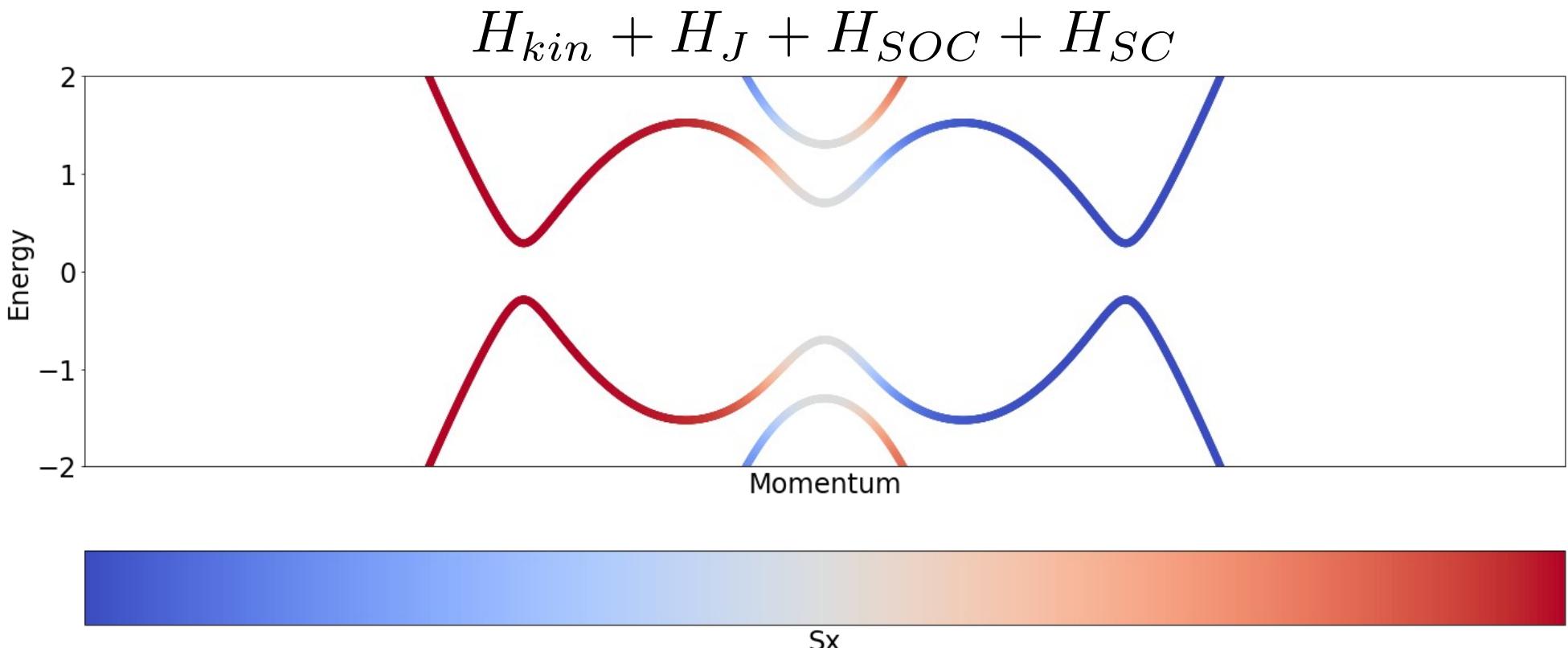
Engineering helical states

$$H_{kin} + H_J + H_{SOC}$$



With Rashba SOC and exchange, helical states appear

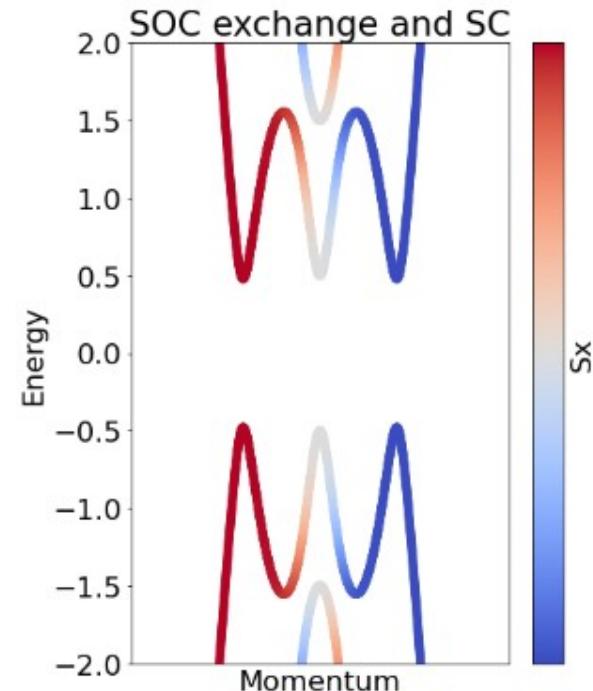
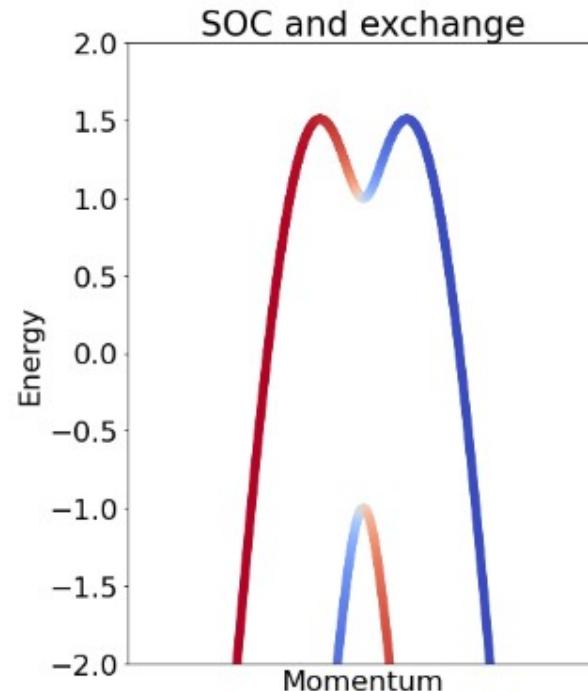
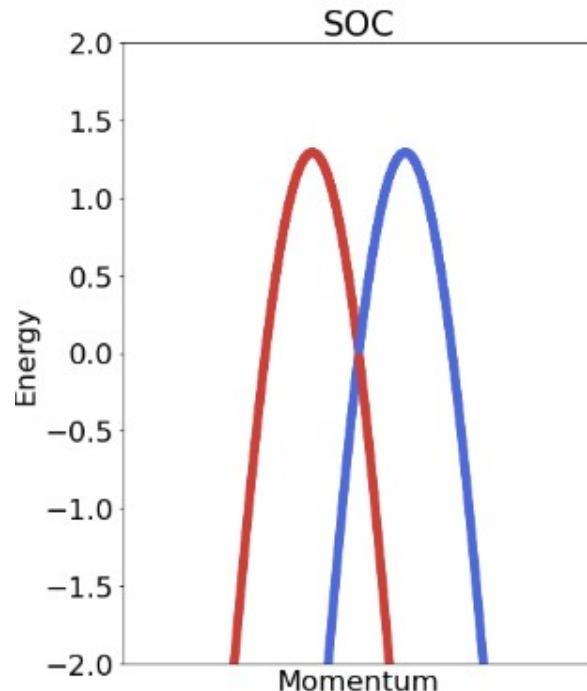
Engineering a topological superconductor



An s-wave superconducting gap opens up a topological gap in the heterostructure

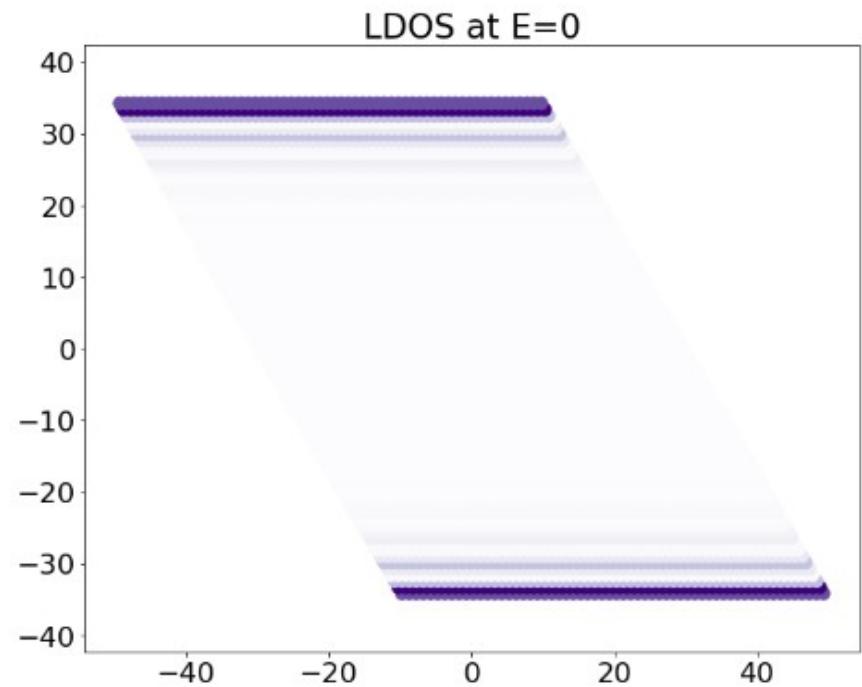
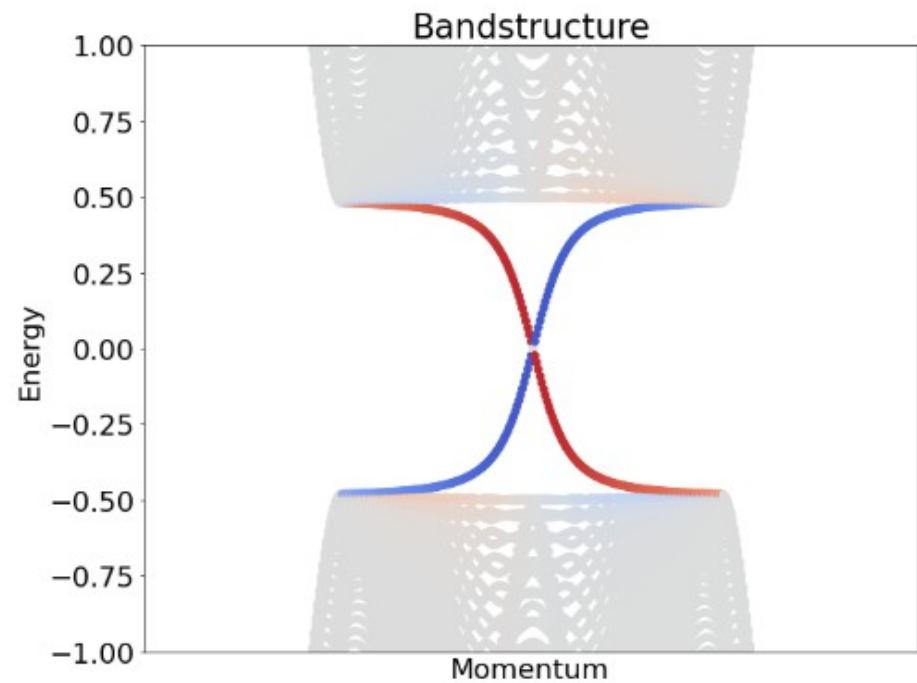
Artificial topological superconductivity C=1, bulk electronic structure

Bulk electronic structure



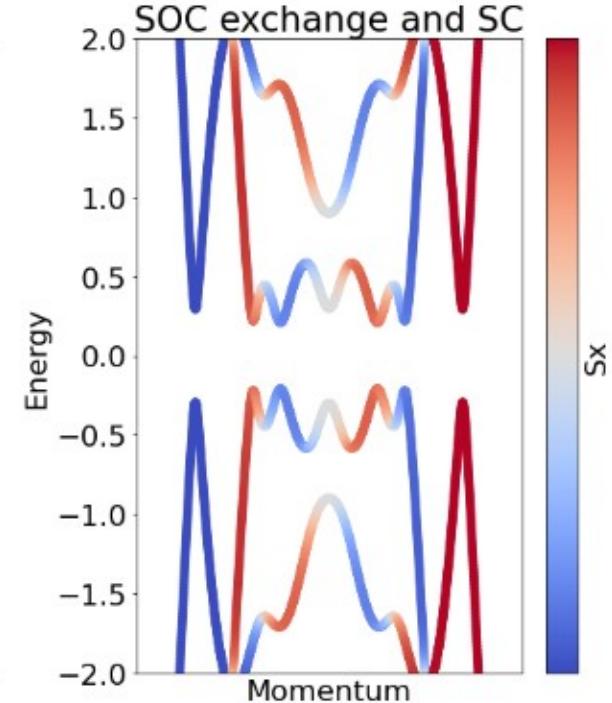
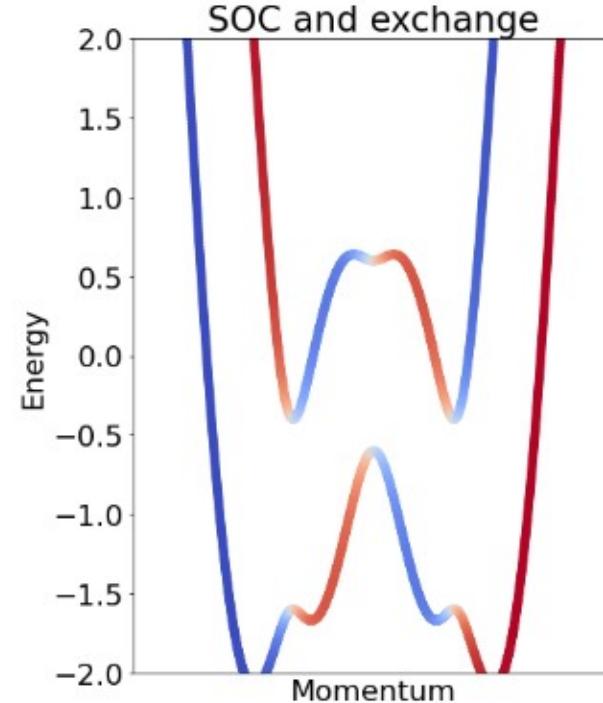
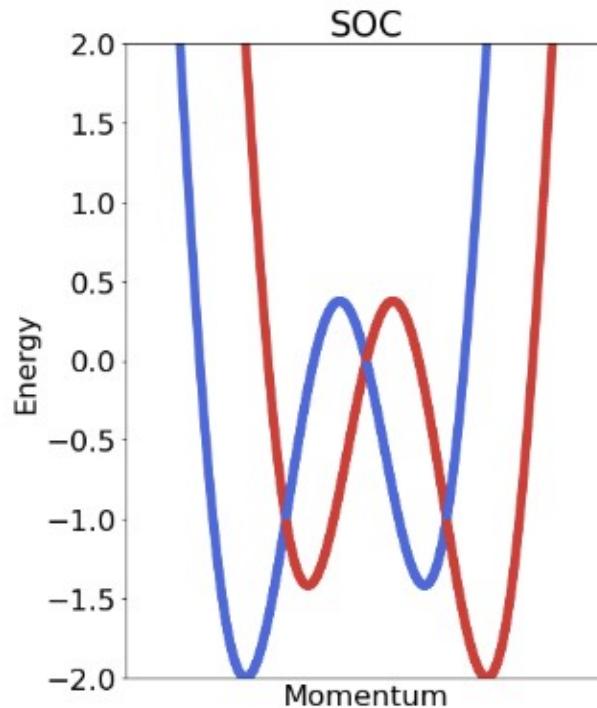
The combination of SOC and exchange creates helical states
Superconductivity gaps out the helical states in a non-trivial way

Artificial topological superconductivity C=1, ribbon electronic structure



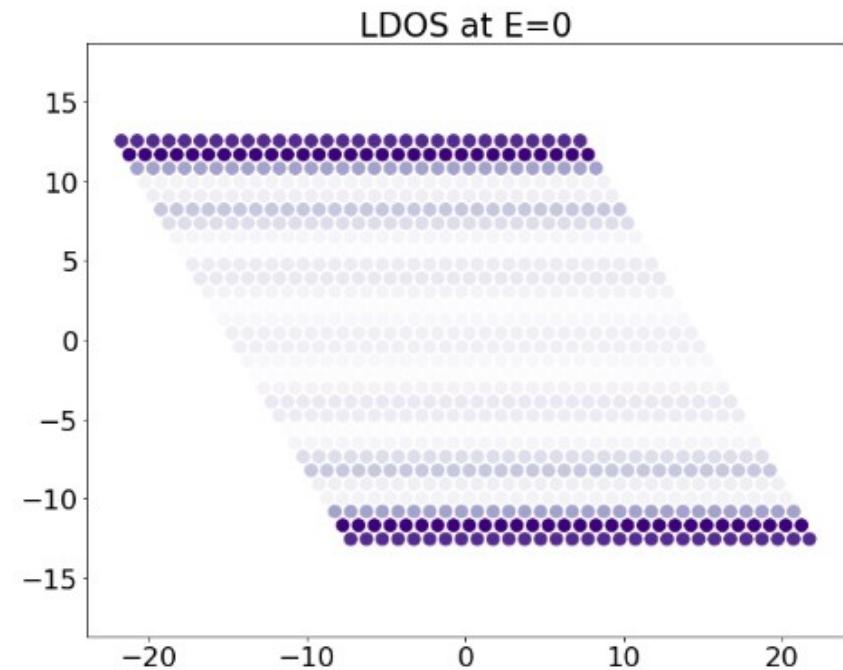
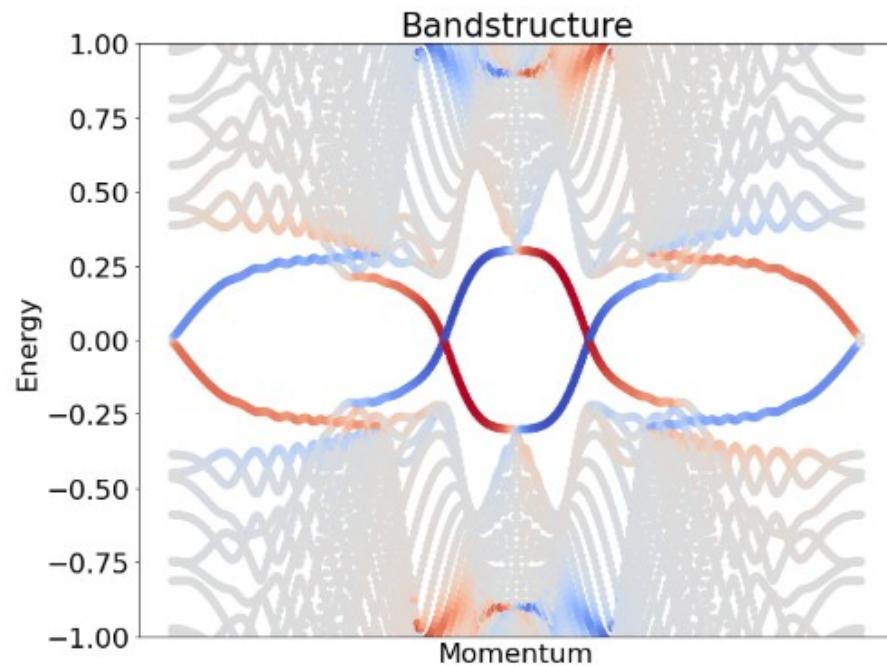
Artificial topological superconductivity C=3, bulk electronic structure

Bulk electronic structure



The combination of SOC and exchange creates helical states
Superconductivity gaps out the helical states in a non-trivial way

Artificial topological superconductivity C=3, ribbon electronic structure



Break

5 min break

(optional) to discuss during the break

What is the correct sign for this equality, and why?

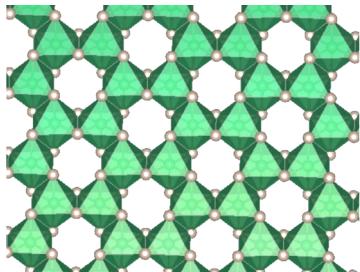
$$\langle c_{k,\uparrow}^\dagger c_{-k,\uparrow}^\dagger \rangle = \langle c_{-k,\uparrow}^\dagger c_{k,\uparrow}^\dagger \rangle$$

$$\langle c_{k,\uparrow}^\dagger c_{-k,\uparrow}^\dagger \rangle = -\langle c_{-k,\uparrow}^\dagger c_{k,\uparrow}^\dagger \rangle$$

Van der Waals magnetism

Van der Waals magnetic materials

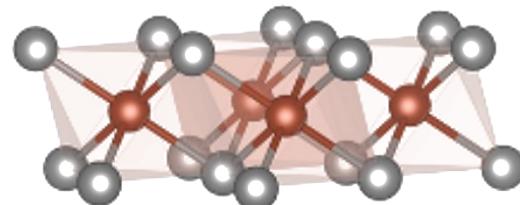
Ferromagnet,
antiferromagnets



CrI_3 , CrCl_3 , CrBr_3

Break time-reversal

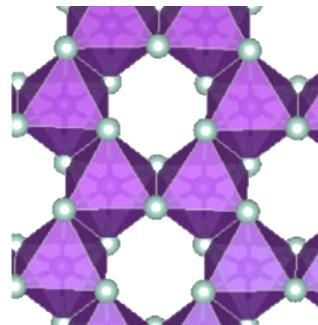
Multiferroics



NiI_2

Break time-reversal
and inversion symmetry

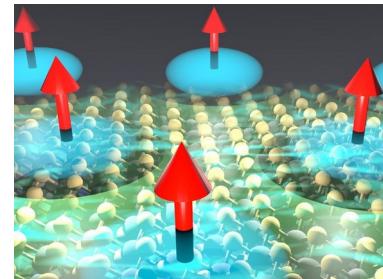
(proximal)
Quantum
spin-liquids



RuCl_3 , 1T-TaS₂

Do not break time-reversal

Heavy-fermion
Kondo insulators



1T-TaS₂/1H-TaS₂

A simple interacting Hamiltonian

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

Free Hamiltonian

*Interactions
(Hubbard term)*

What is the ground state of this Hamiltonian?

$U < 0$ Superconductivity

$U > 0$ Magnetism

The mean-field approximation

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

**Four fermions
(not exactly solvable)**

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

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

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

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

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

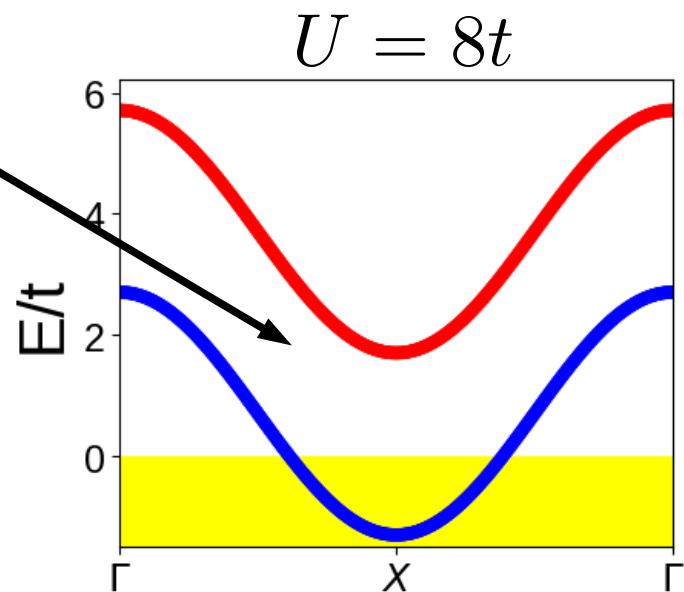
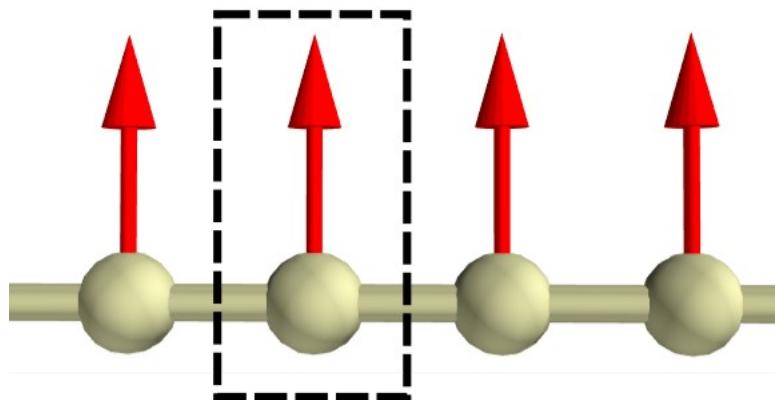
What if interactions are much stronger than the kinetic energy?

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

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

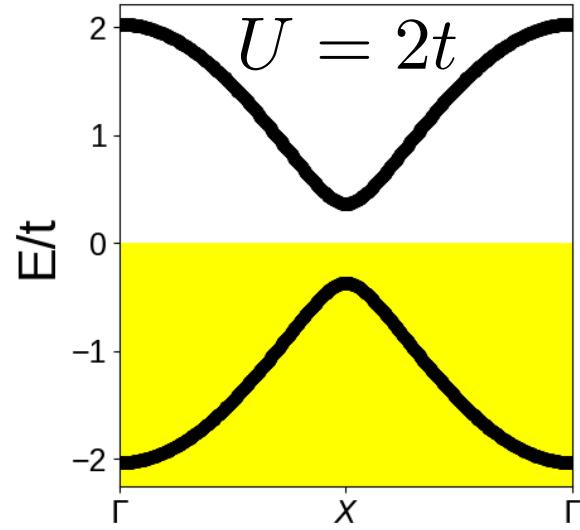
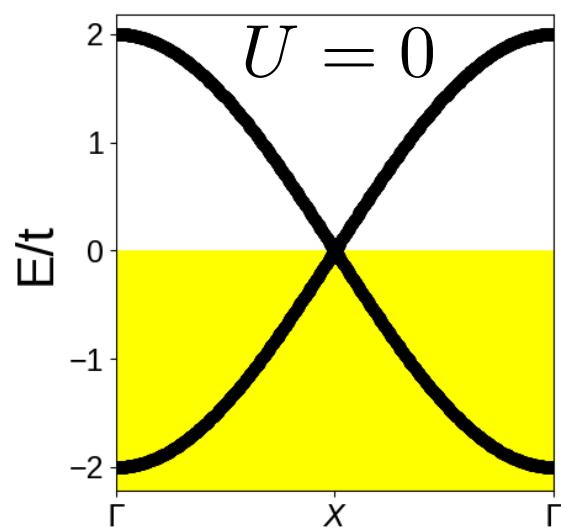
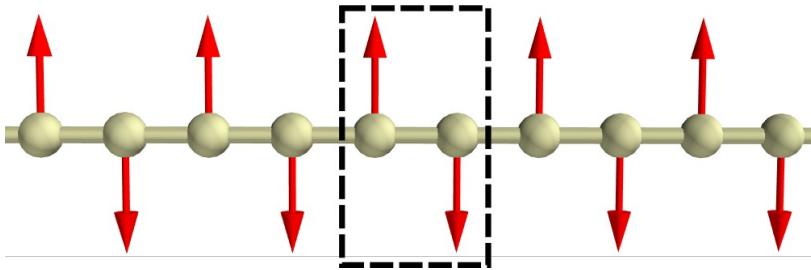
Filling 0.2 (full would be 1)
Interaction-induced splitting



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

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



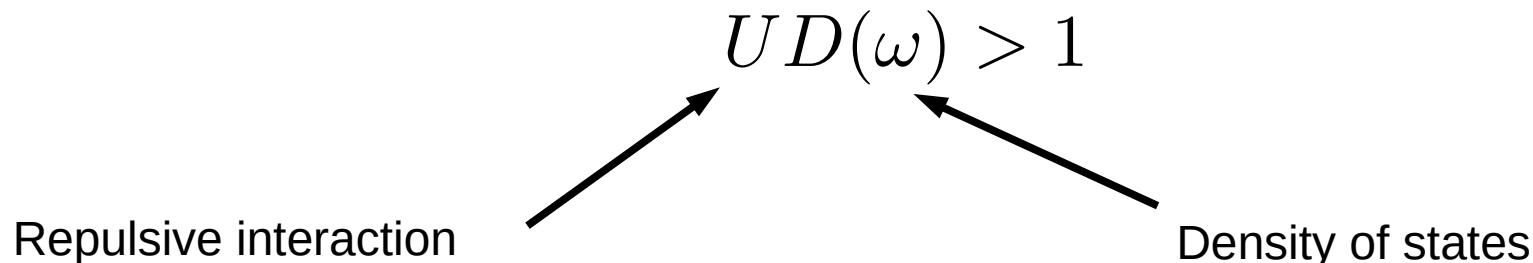
The critical interaction for magnetic ordering

Lets take the Hamiltonian

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

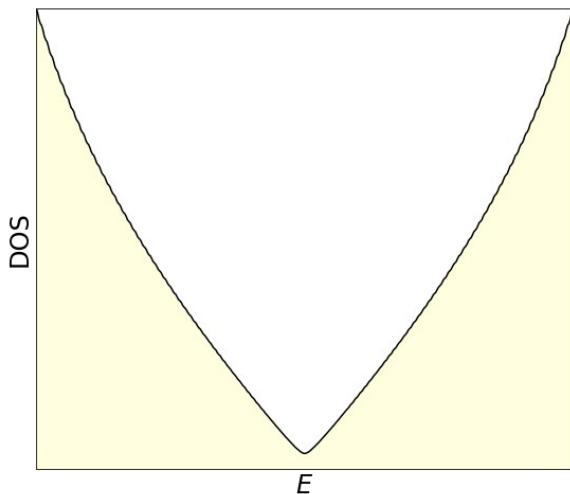
Do we have magnetism for any value of U ? $\langle S_z \rangle \neq 0$

In general, in the weak coupling limit magnetism appears when



The critical interaction for magnetic ordering

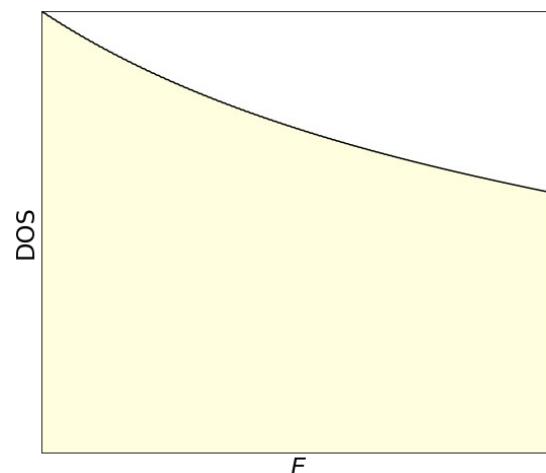
Semimetals



No low coupling instability

$$U_C \gg t$$

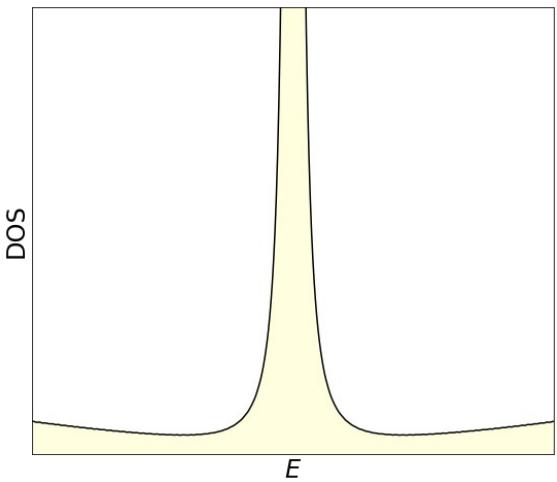
Metals



Controlled by DOS

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

Flat bands

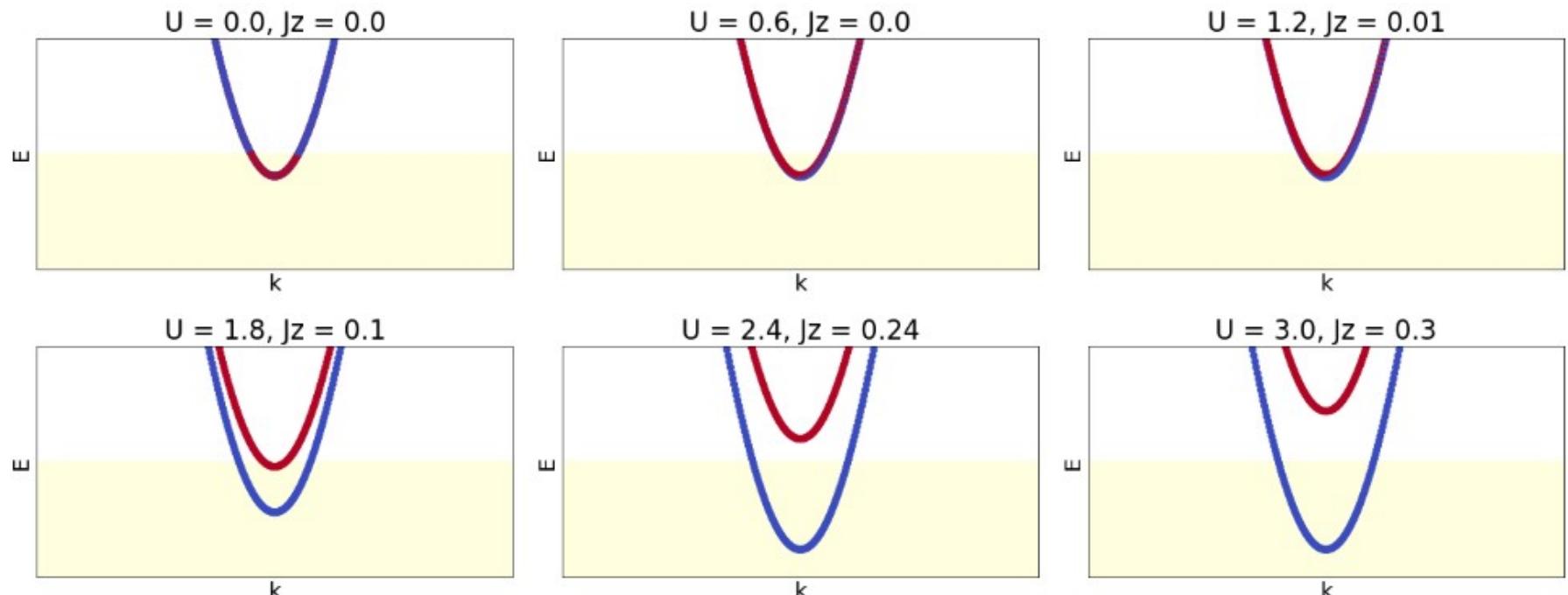


Arbitrarily small interactions

$$U_C \rightarrow 0$$

The critical interaction for magnetic ordering

Magnetic instabilities occur once interactions are strong enough



For interactions below a threshold, no magnetic order occurs

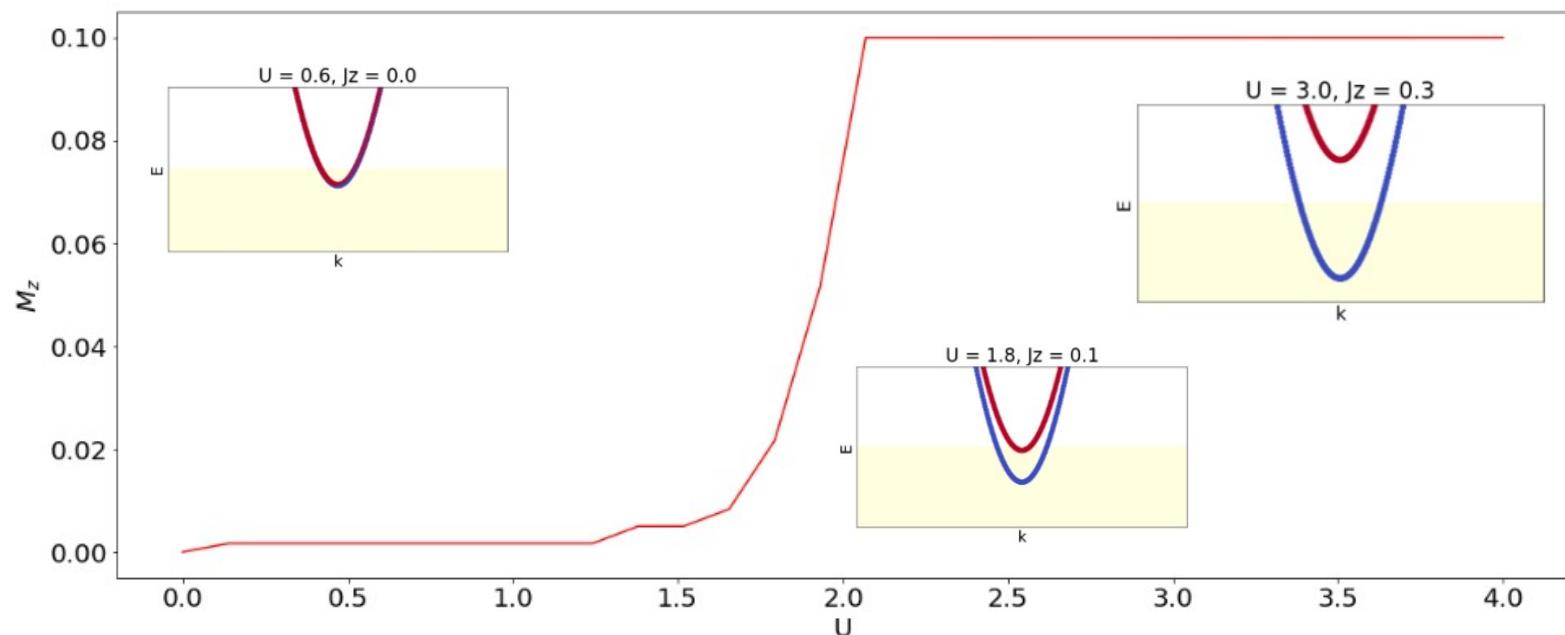
The critical interaction for magnetic ordering

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

No magnetism

Non-saturated magnetization

Saturated magnetization (half metal)



The strongly localized limit
and the Heisenberg model

The strongly localized limit

Let us start with a Hubbard model dimer

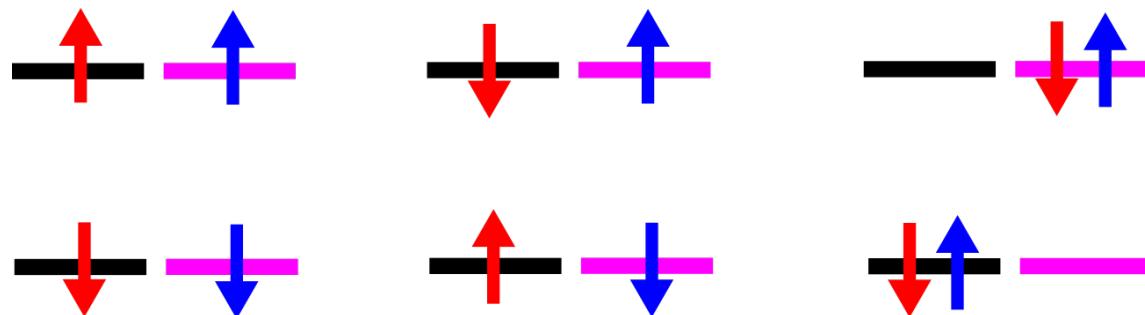
$$H = t[c_{0\uparrow}^\dagger c_{1\uparrow} + c_{0\downarrow}^\dagger c_{1\downarrow}] + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

Now in the limit

$$U \gg t$$



The full Hilbert space at half filling is

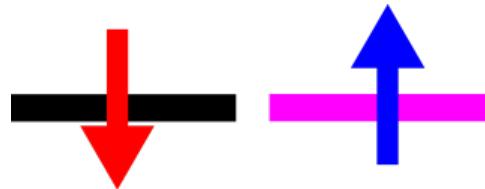


The strongly localized limit

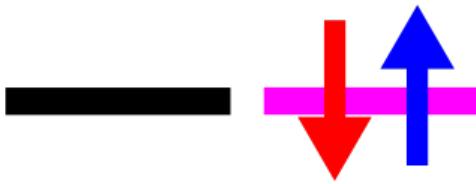
Let us start with a Hubbard model dimer

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

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



~ 0

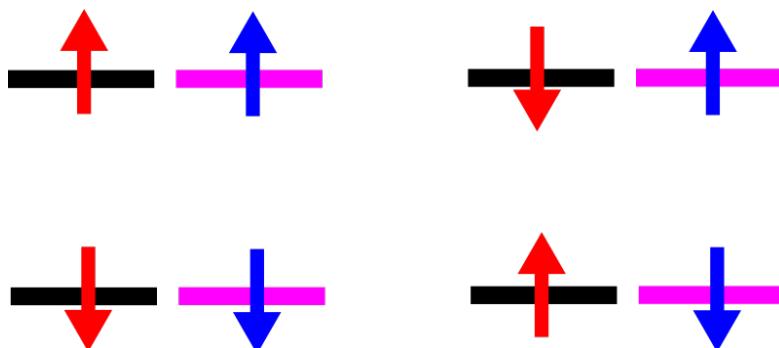


$\sim U$

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

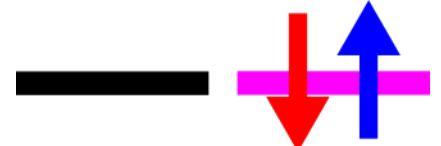
$$H = H_0 + V$$

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

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

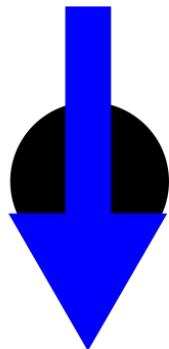
Spin-orbit coupling introduces anisotropic couplings

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

The Heisenberg model

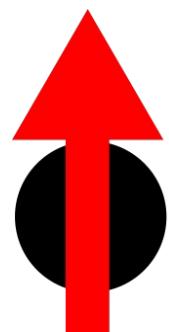
$$J_{ij} > 0$$

Antiferromagnetic coupling



$$J_{ij} < 0$$

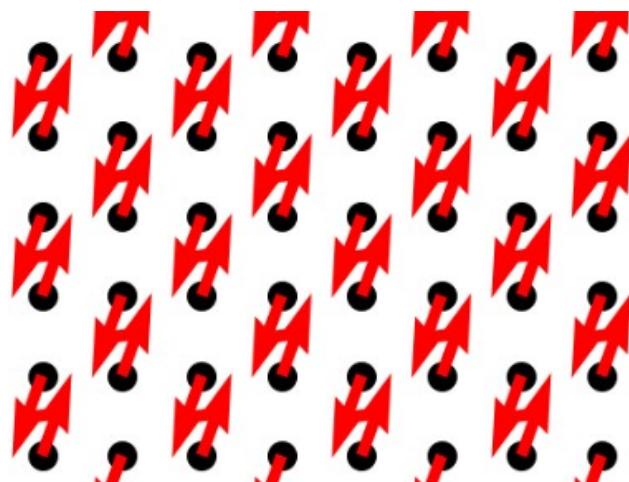
Ferromagnetic coupling



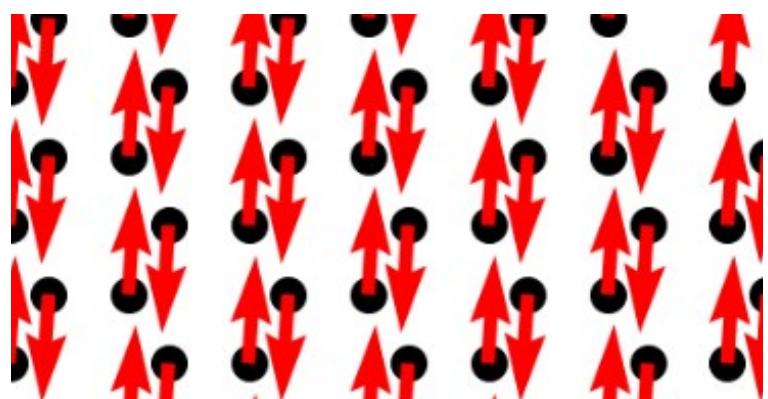
Classical ground states

Antiferromagnetism driven by superexchange

In the square lattice



In the honeycomb lattice

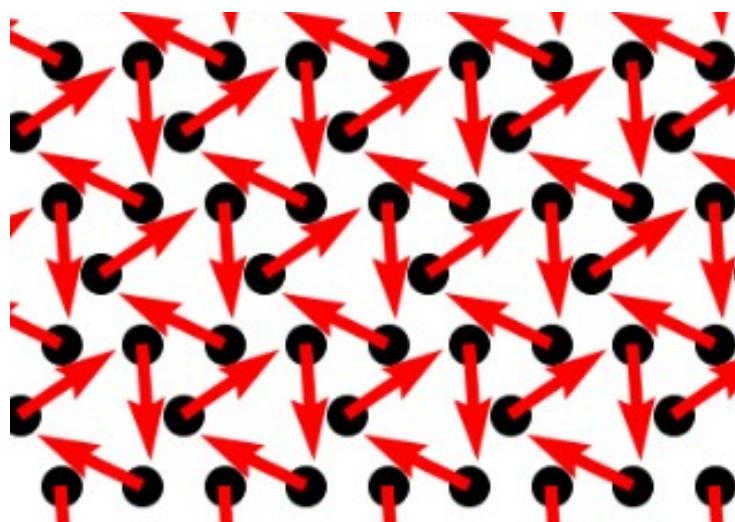


$$H = \sum_{ij,s} t_{ij} c_{i,s}^\dagger c_{j,s} + \sum_i U c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} + h.c.$$

In bipartite lattices, the magnetization is collinear

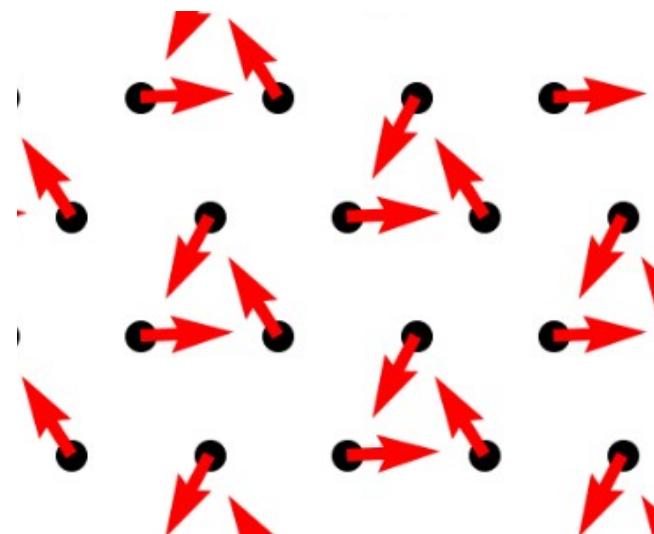
Antiferromagnetism driven by superexchange

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

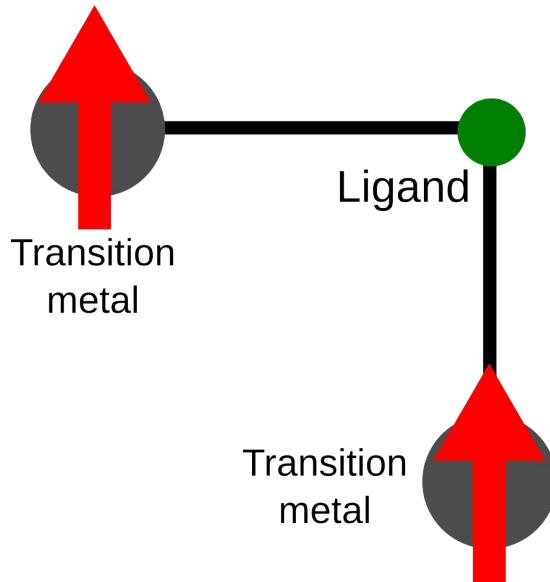


Geometric frustration promotes non-collinear order at the mean-field level

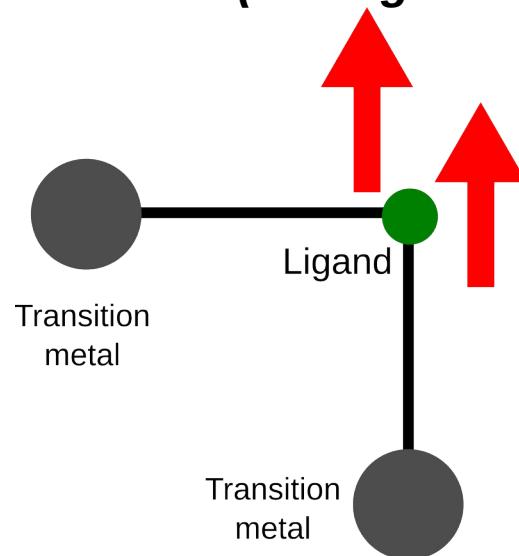
The origin of ferromagnetic coupling

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

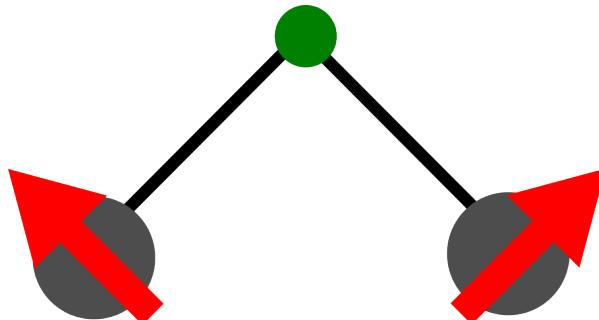
Goodenough-Kanamori rules

Non-isotropic exchange coupling

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

Antisymmetric exchange

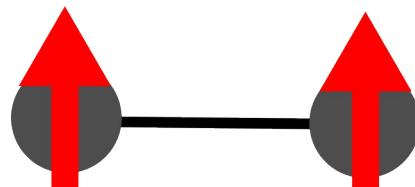
$$(\mathbf{r}_{ik} \times \mathbf{r}_{kj}) \cdot \vec{S}_i \times \vec{S}_j$$



Promotes
non-collinear order

Anisotropic exchange

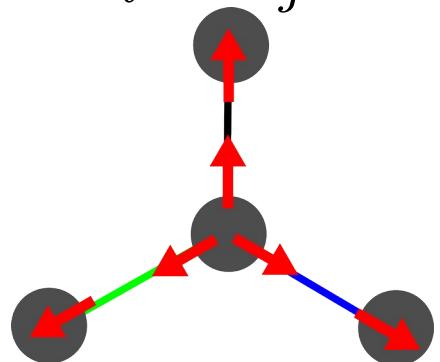
$$S_i^z S_j^z$$



Promotes
easy axis/plane

Kitaev interaction

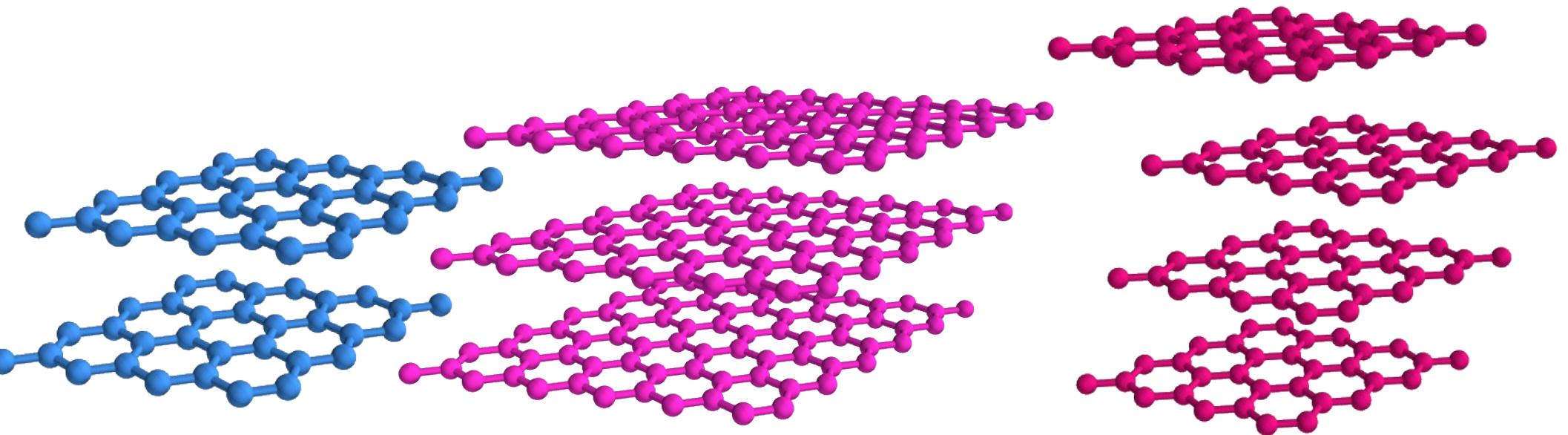
$$S_i^{\alpha(i)} S_j^{\alpha(j)}$$



Promotes
frustration

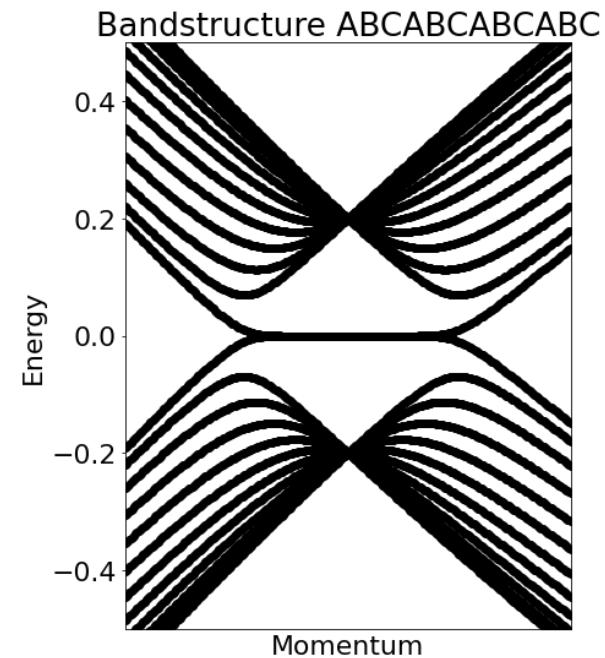
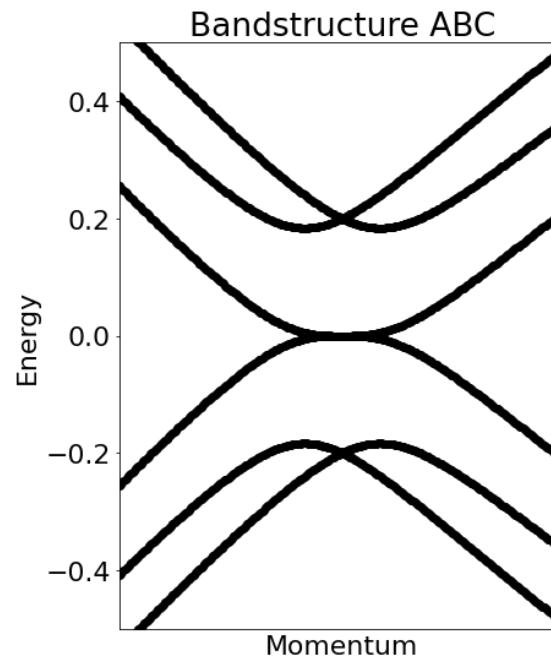
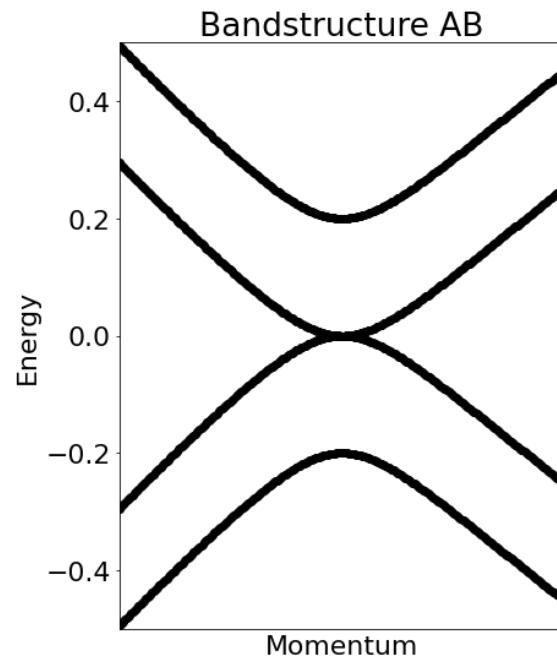
Symmetry breaking in graphene multilayers

Magnetic symmetry breaking in graphene multilayers



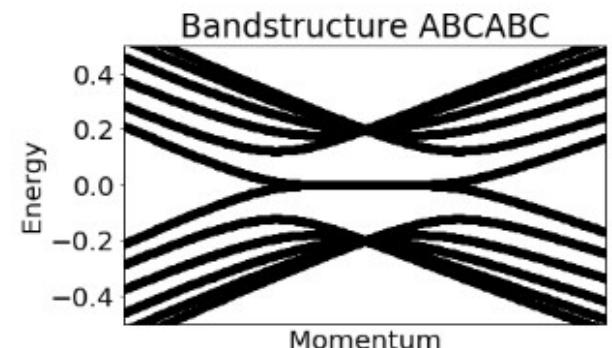
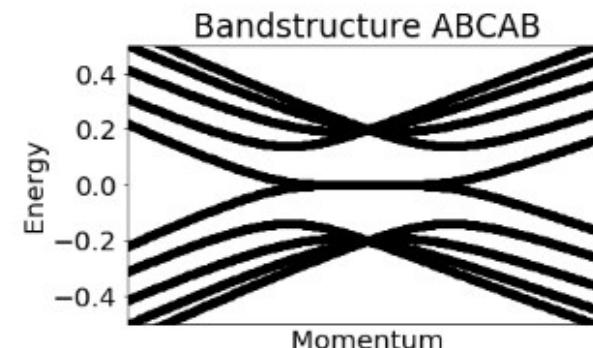
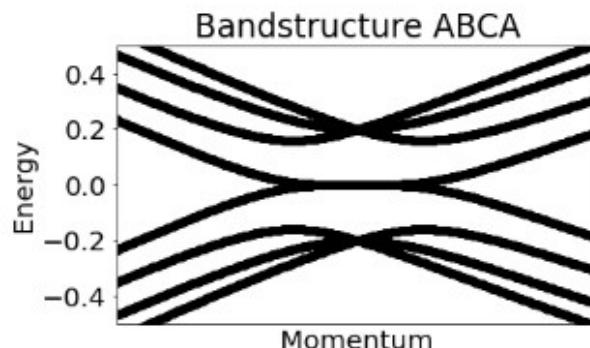
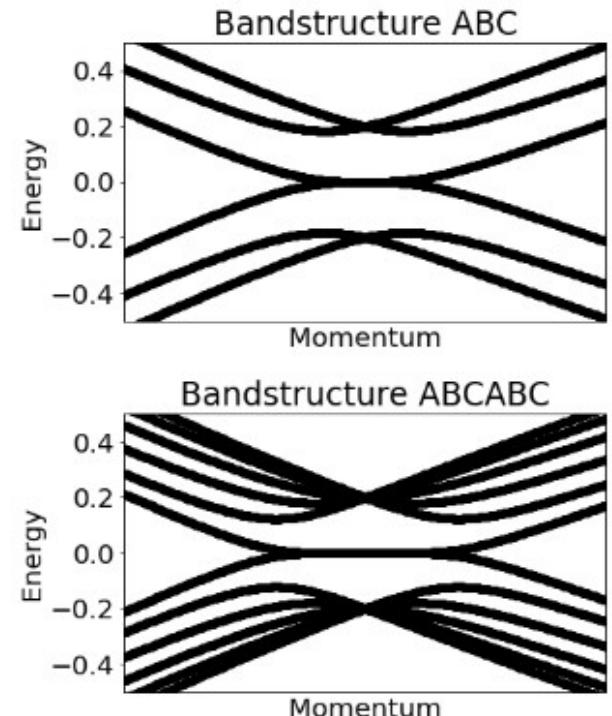
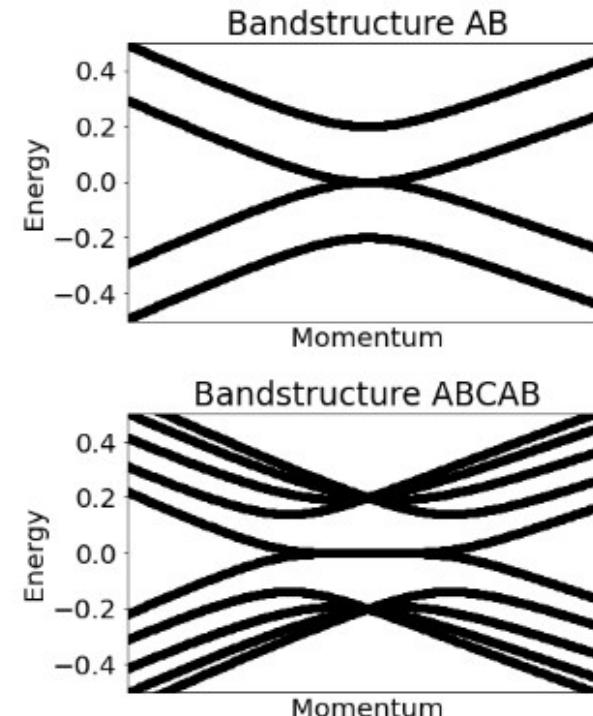
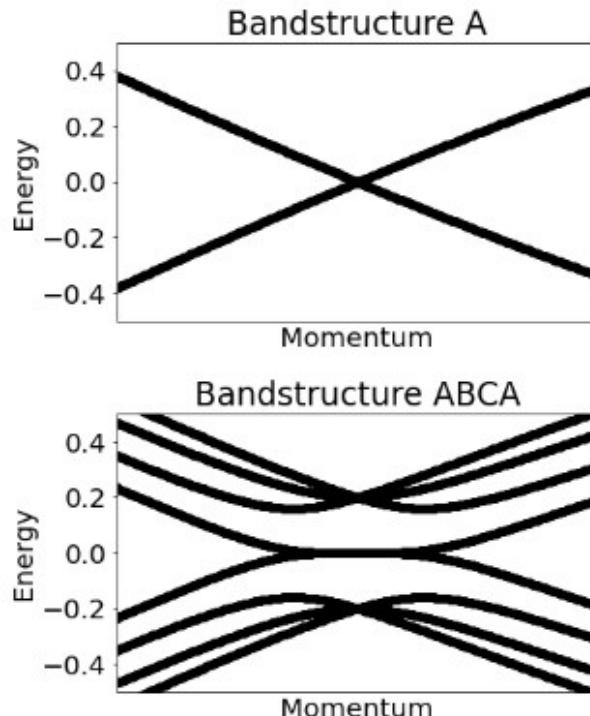
Magnetism in graphene multilayers

Graphene trilayers can have magnetic instabilities driven by repulsive interactions



The more layers a stacking has, the flatter the dispersion

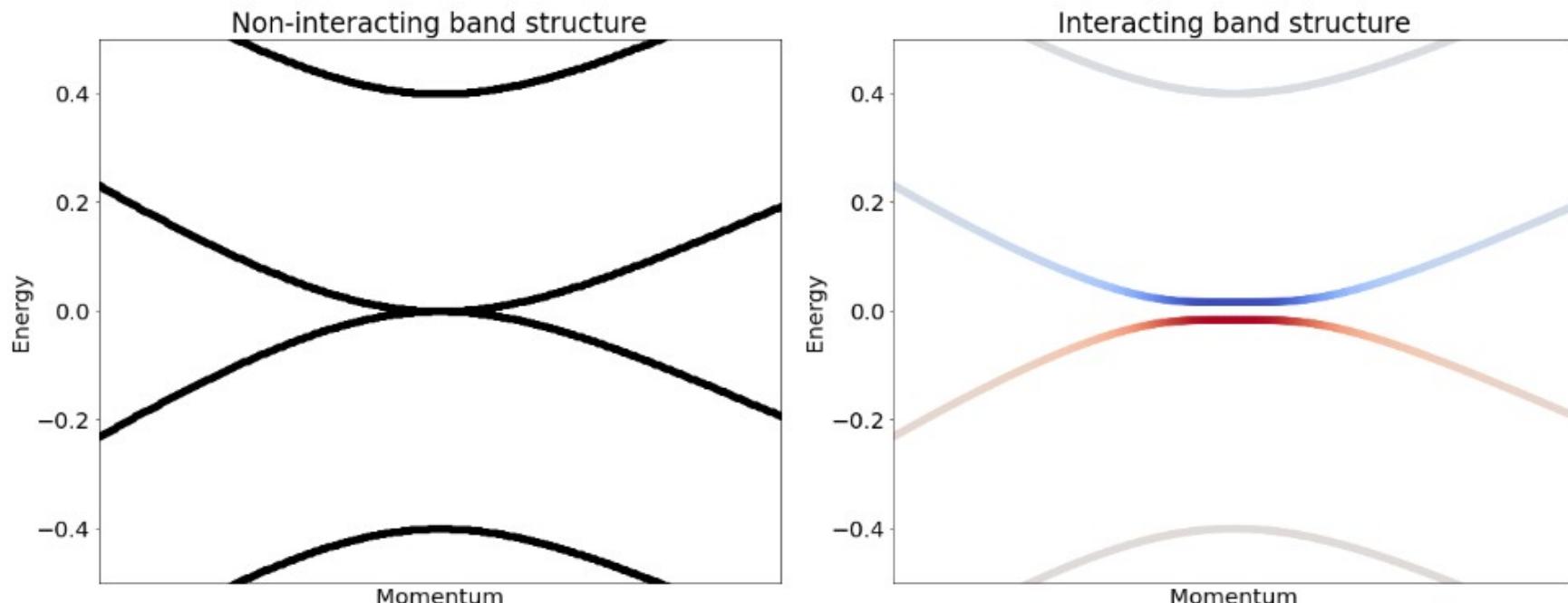
Magnetism in graphene multilayers



The more layers a stacking has, the flatter the dispersion

Magnetism in graphene bilayers

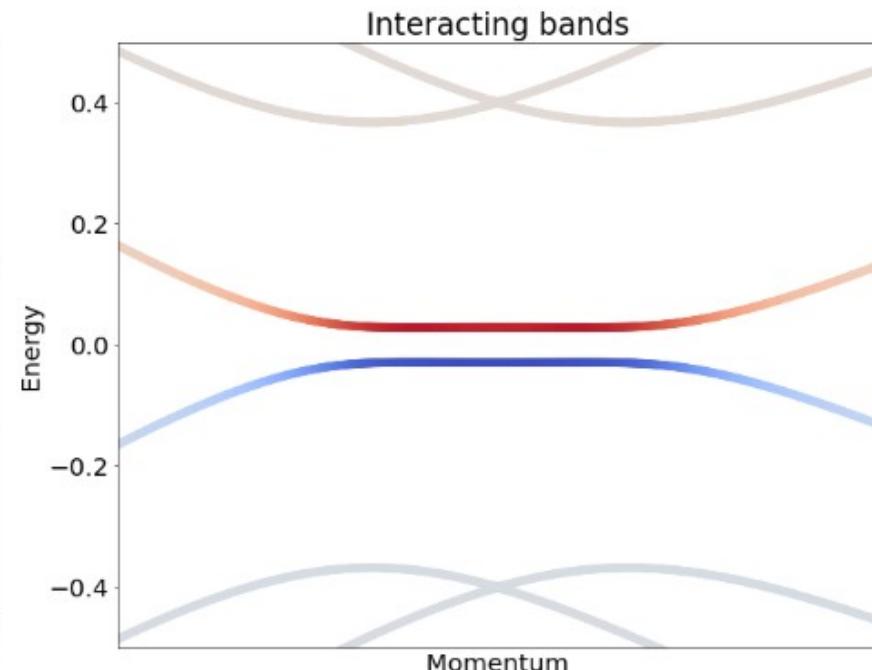
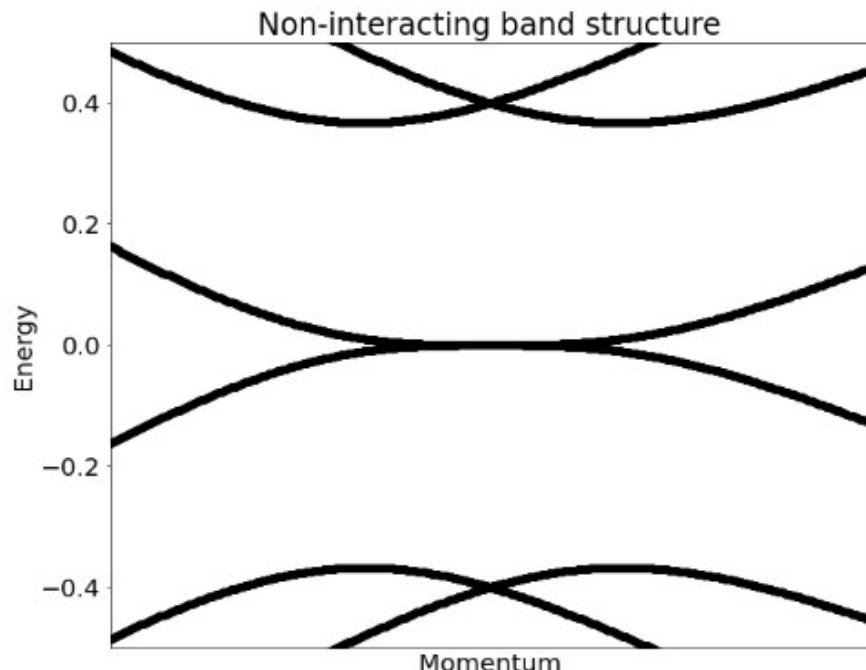
Graphene bilayers can have magnetic instabilities driven by repulsive interactions



AB graphene bilayer

Magnetism in graphene trilayers

Graphene trilayers can have magnetic instabilities driven by repulsive interactions

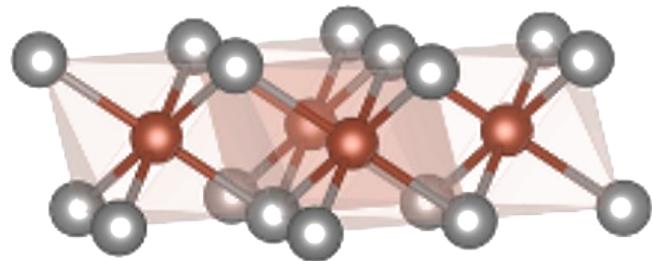


ABC graphene trilayer

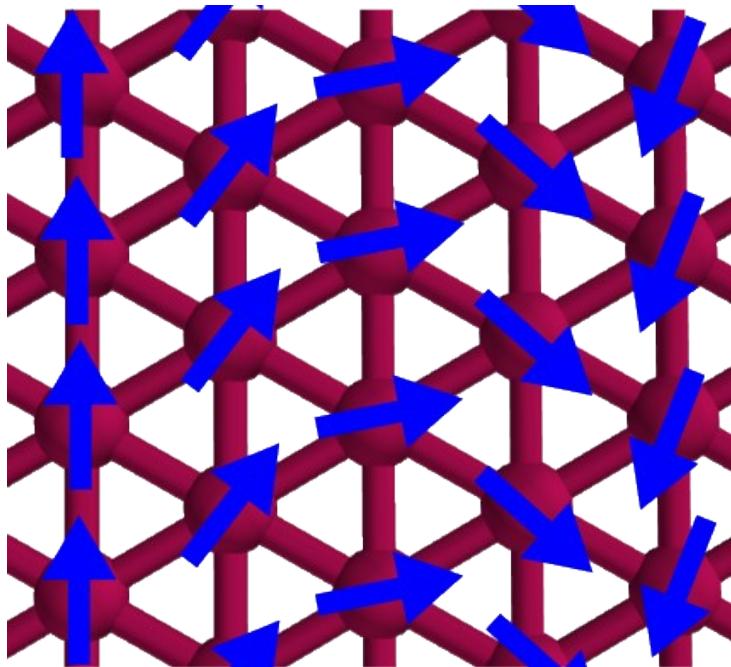
Multiferroic van der Waals materials

Electric polarization in a magnetic material

Multiferroics host, simultaneously, magnetism and electric polarization



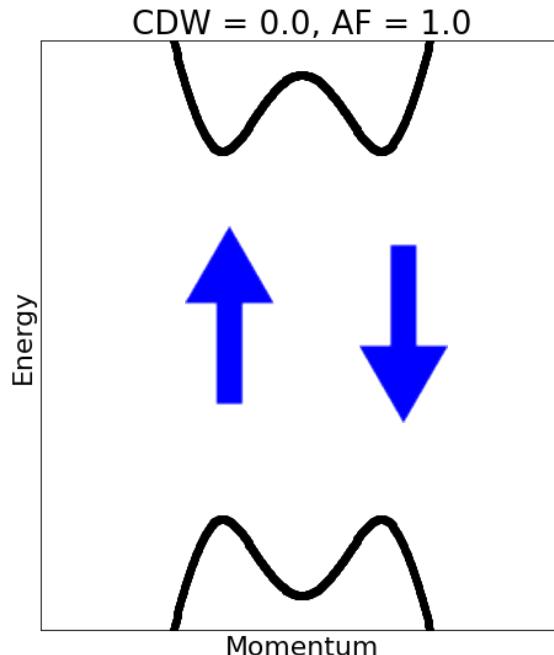
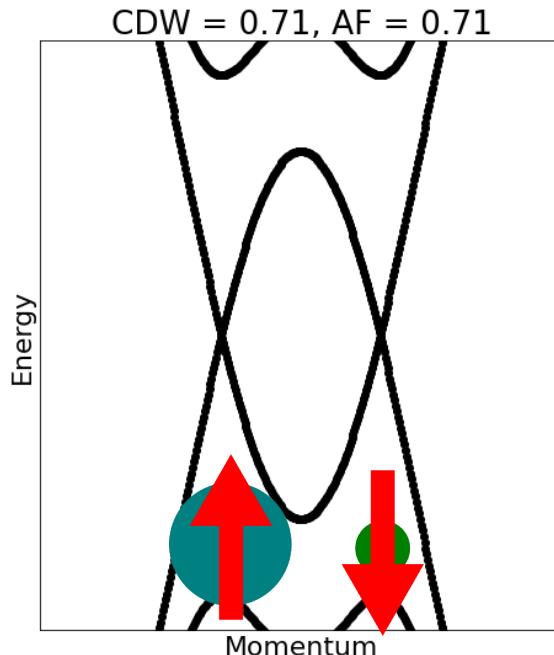
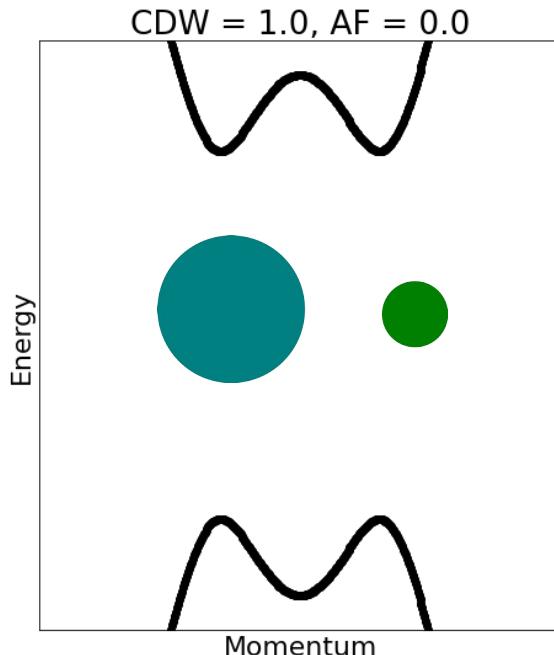
NiI_2



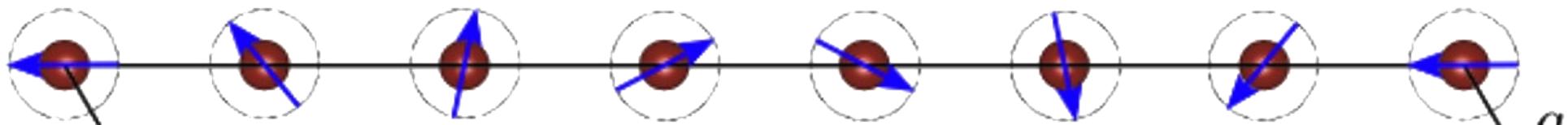
Why are multiferroics rare

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

However, magnetic and charge order often compete to open gaps



Coupling between magnetism and polarization



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

Electric polarization

SOC driven

Spin rotation axis

Wavevector of the spin spiral

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

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

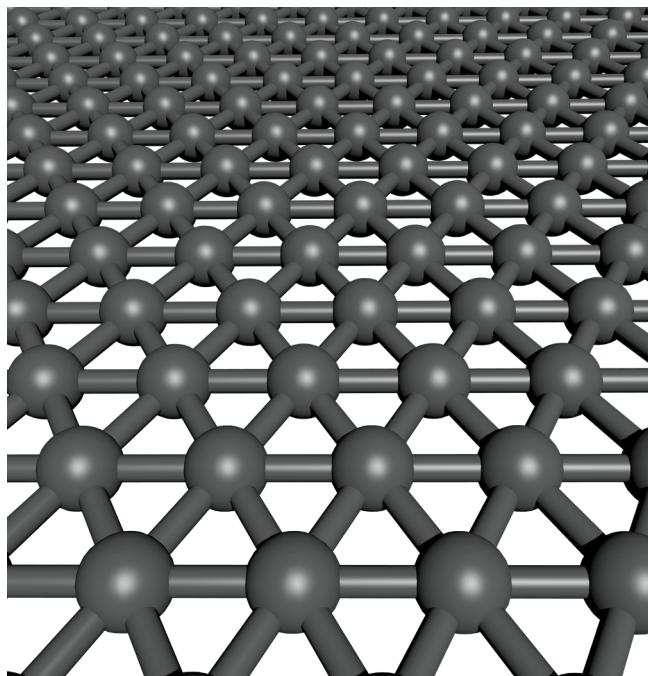
The state is maximally entangled

Can we have a macroscopic version of this ground state?

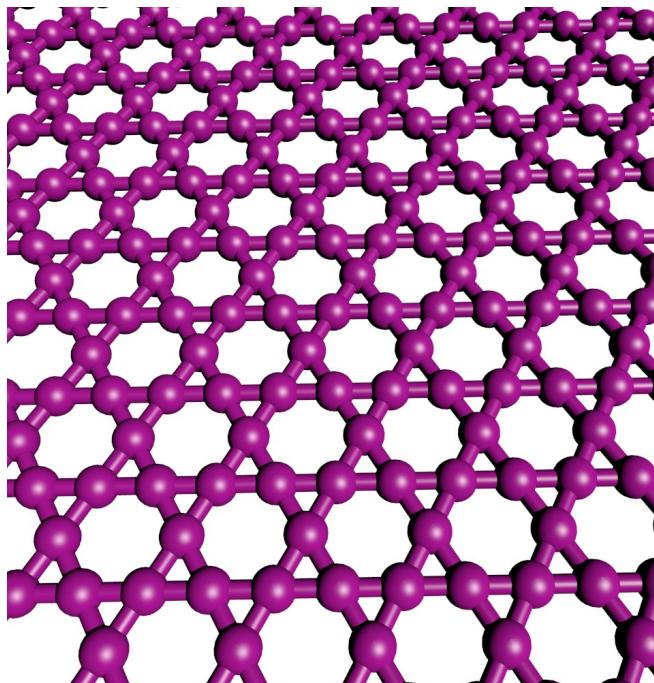
$$\langle \vec{S}_i \rangle = 0$$

Frustrated lattices

Triangular



Kagome



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)

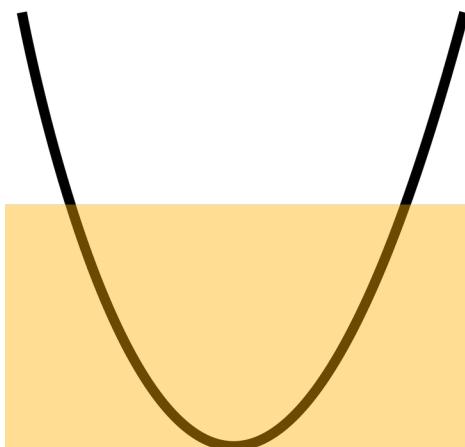
$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \longrightarrow \mathcal{H} = \sum_{ij,s} \chi_{ij} f_{i,s}^\dagger f_{j,s}$$

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

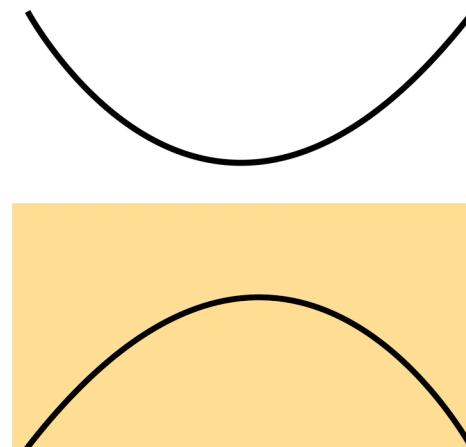
The excitations of the QSL are described by a single particle spinon Hamiltonian

Spinon dispersions

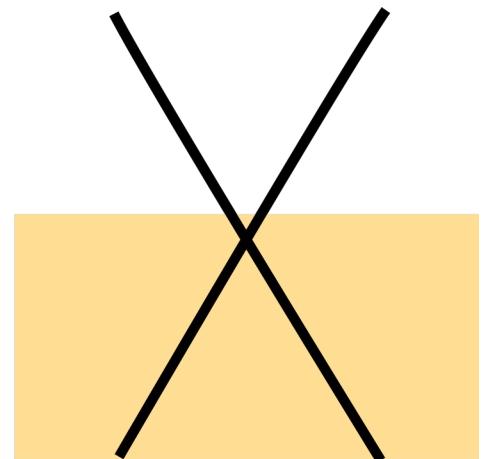
Gapless spinons



Gapped spinons



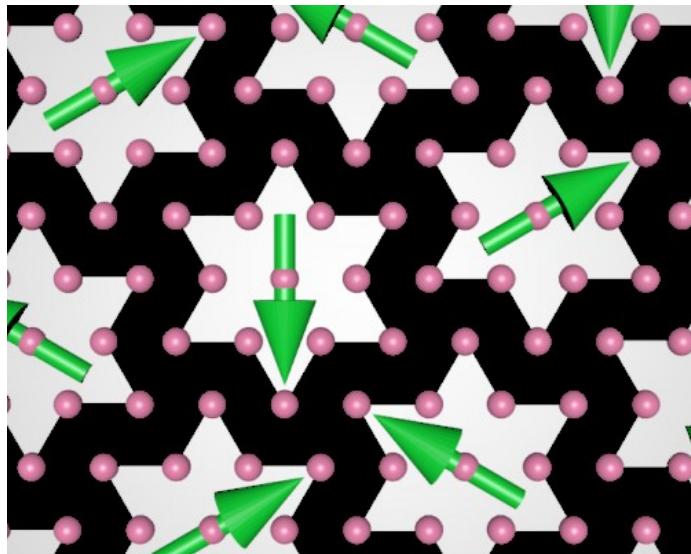
Dirac spinons



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

Frustrated magnetism in 1T-TaS₂

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



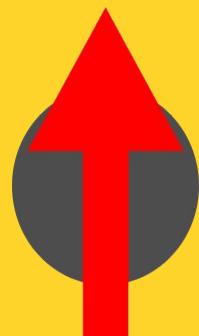
$$\mathcal{H} = \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

Strong interactions give rise to local moment formation

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

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

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

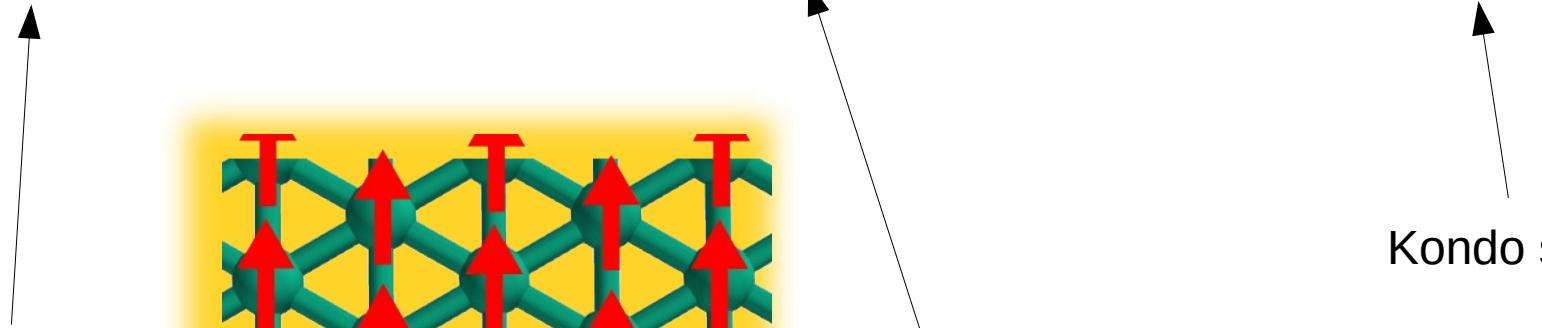
We now take a quantum spin $S=1/2$

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

The Kondo lattice problem

The Kondo lattice problem

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



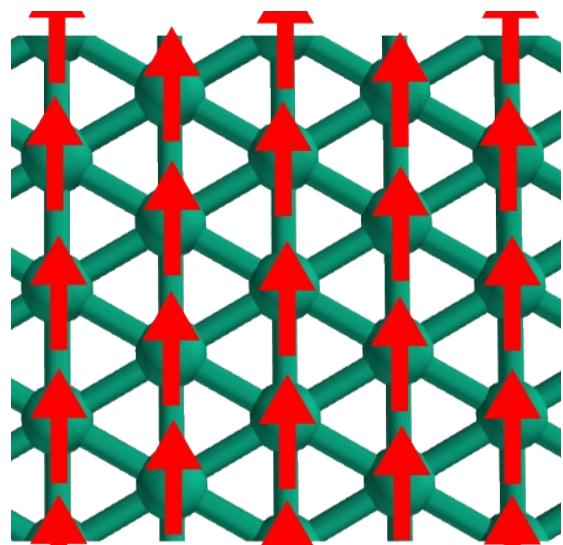
Conduction electrons

Kondo coupling

Kondo sites

Building an artificial heavy fermion state

Lattice of Kondo impurities



Dispersive electron gas

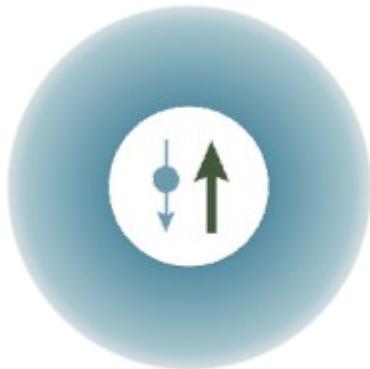
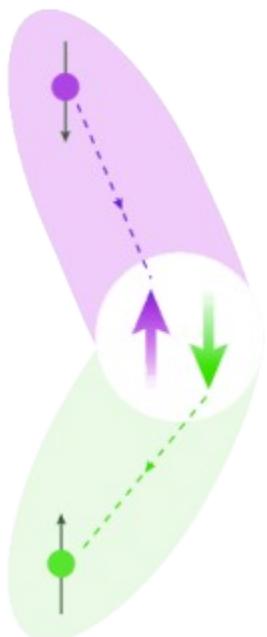
$$J_K \longleftrightarrow$$



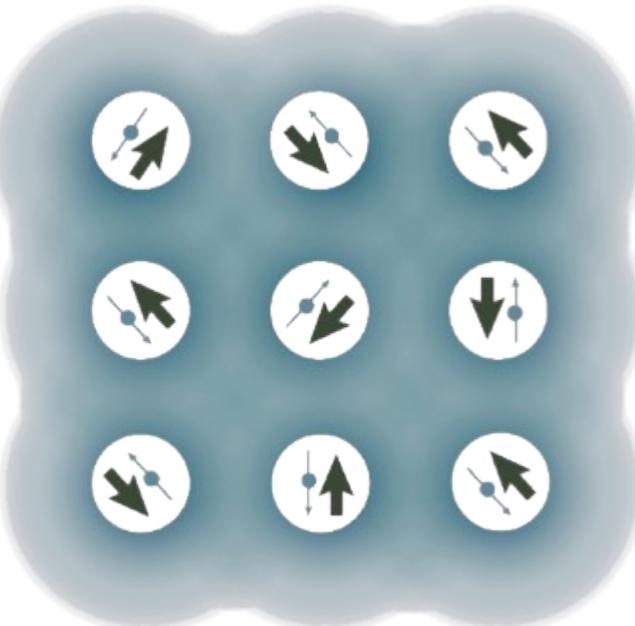
Both ingredients coupled through Kondo coupling

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

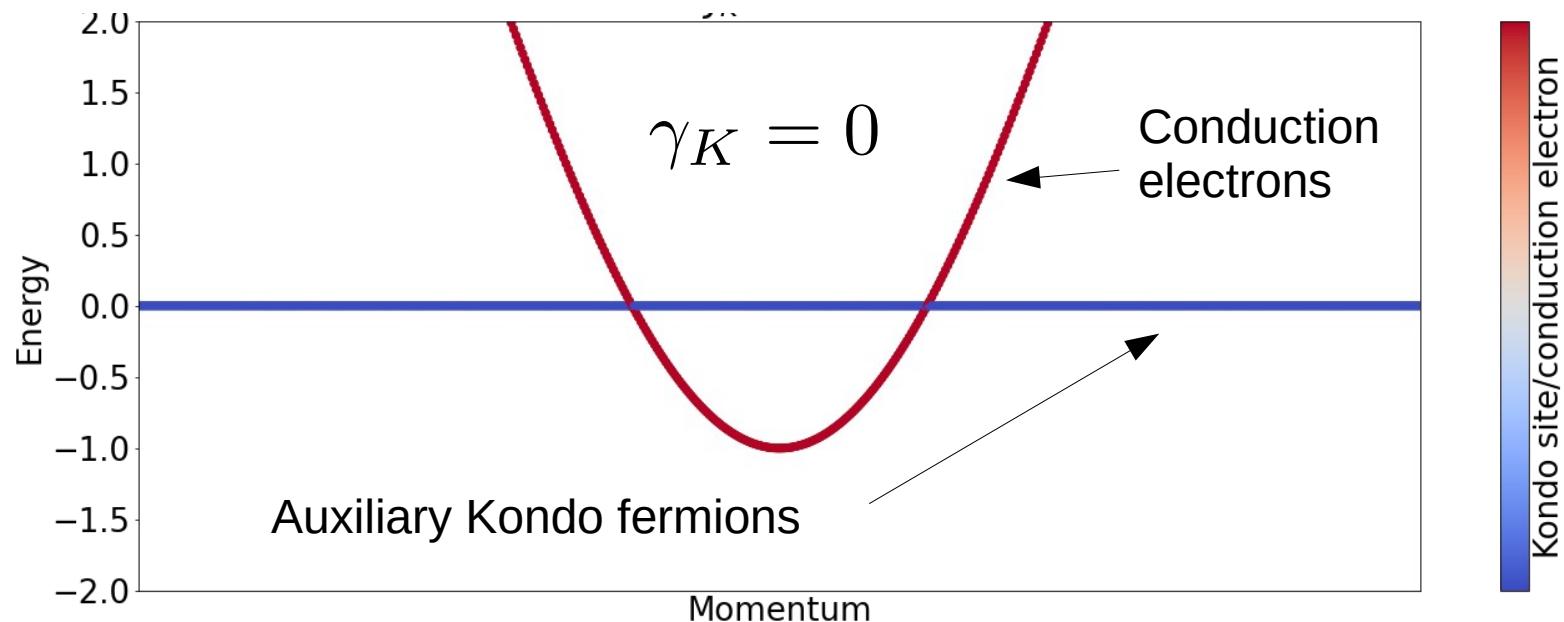
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} + h.c.$$

Conduction band dispersion

Kondo hybridization

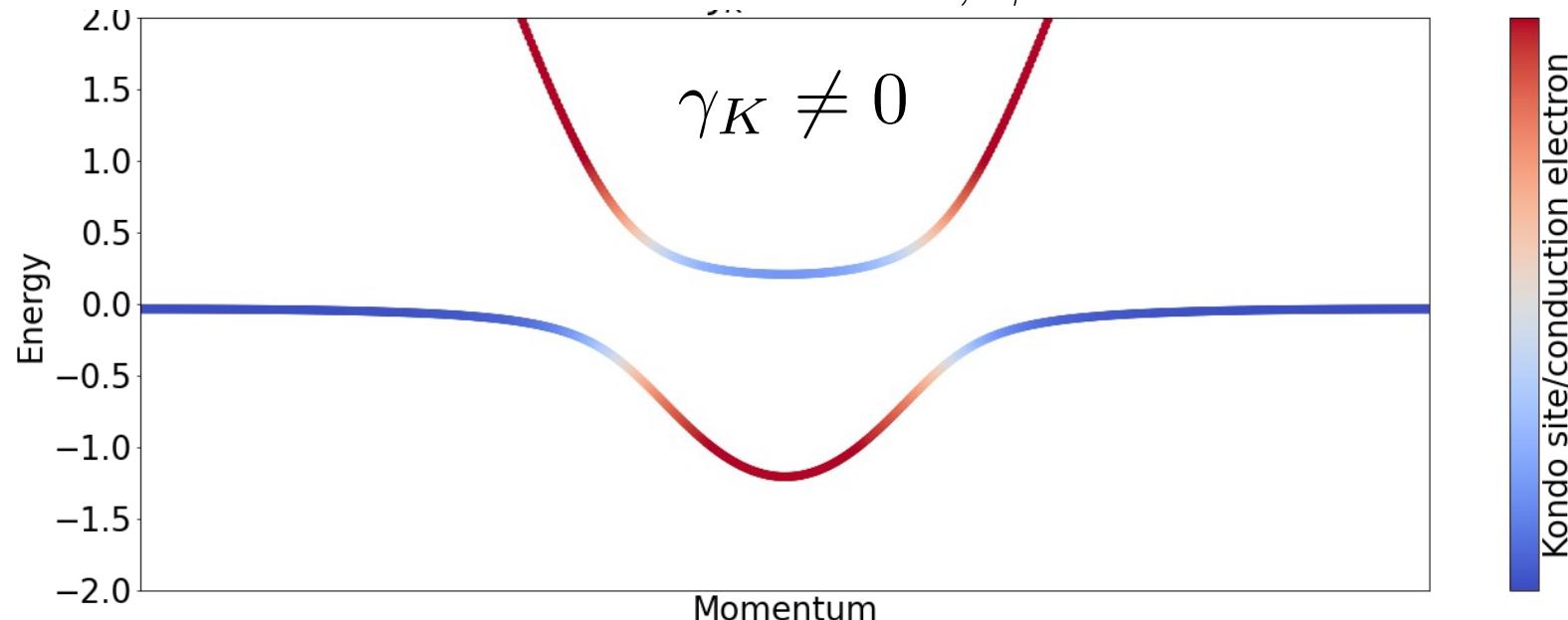
Electronic structure of the Kondo lattice problem

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



Electronic structure of the Kondo lattice problem

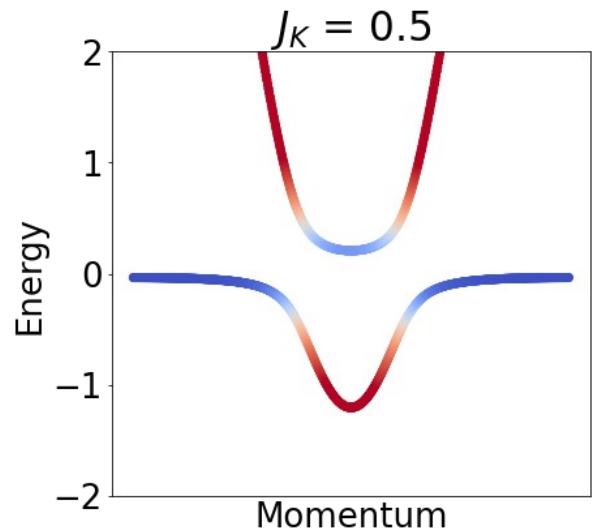
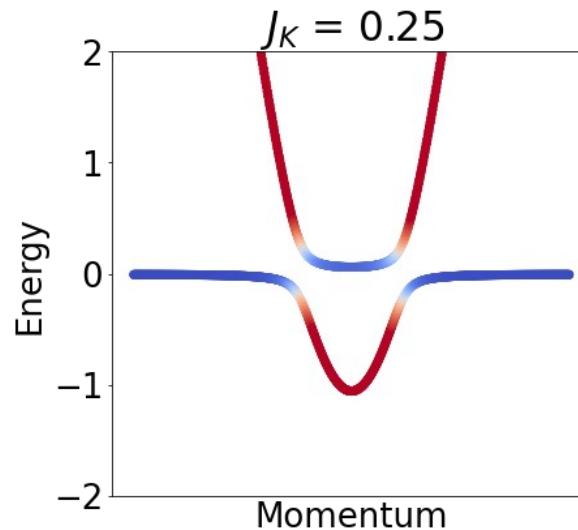
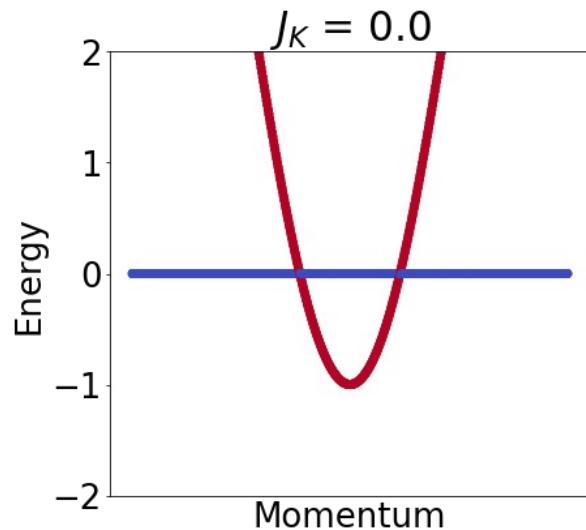
$$H \sim \sum_{\mathbf{k}\alpha} \epsilon_{\mathbf{k}} c_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} - \gamma_K \sum_{\mathbf{k},\alpha\beta} f_{\mathbf{k}\alpha}^\dagger c_{\mathbf{k}\alpha} + h.c.$$



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

Heterostructures of 1H-TaS₂/1T-TaS₂

(Optional) exercise session

Download the Jupyter-notebook from

https://github.com/joselado/emergent_phenomena_in_van_der_Waals_school_tifr_2023

The tasks during the exercise session

You will see examples with the code

```
from pyula import geometry
g = geometry.triangular_lattice() # generate a chain
g = g.get_supercell((3,3)) # generate a supercell
hs = g.get_supercell((1,1)) # generate the Hamiltonian
```

generate the SCF Hamiltonian

```
U = 10 # strong Hubbard interaction
h = hs.get_mean_field_hamiltonian(U=U,mf="XY",mix=0.9) # solve the interacting problem with a mean-field guess
```

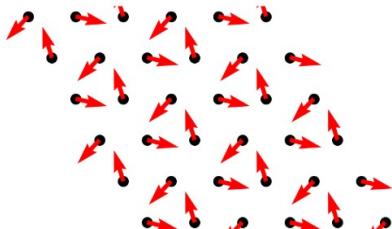
hs = h.get_supercell((2,2)) # generate a supercell

```
mx,my,mn = hs.get_magnetic_structures(); 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.scatter(x,y,c="black",s=400); plt.axis("off")
```

```
(-4.124999999999999, 4.124999999999999, -2.3815698604072066, 2.381569860407206)
```



You can 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?

