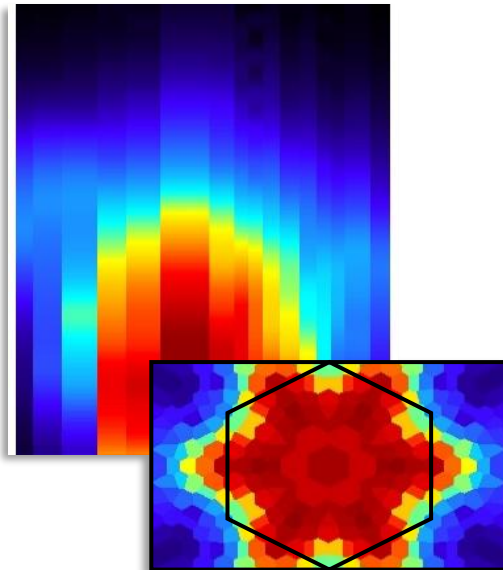
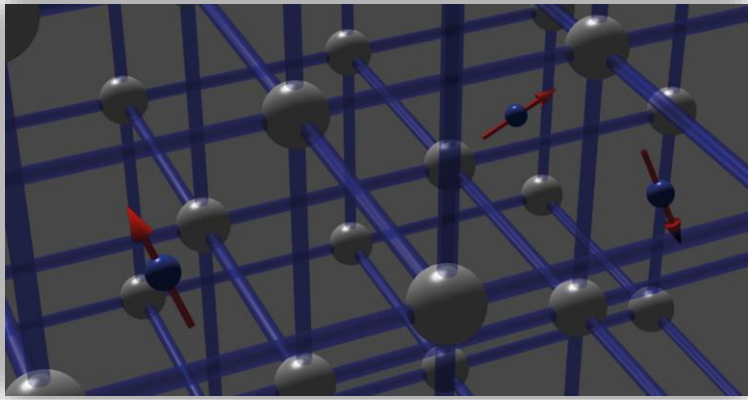


Spin-orbit driven magnetic phenomena: from first principles to experiments I



Roser Valentí
Institute of Theoretical Physics
University of Frankfurt
Germany



European School of Magnetism 2026
Spin-orbit driven magnetic phenomena,
Monday, August 24th 2026

Lecture 1 (DFT and magnetism) techniques

- *Motivation*
 - *Microscopic description of magnetism (frustrated magnetism / spin-orbit coupling)*
- *from first principles to spin model Hamiltonians*

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j + \dots$$

Lecture 2 (Correlated electrons and magnetism) applications

- *Excitations and dynamics in magnetic systems :*

Linear response → INS, ESR, Raman

Non-linear response → non-linear optics (2DCS)

Thermal transport

~100 years ago ...

Ising Model: model for ferromagnetism



The diagram illustrates the Ising model. On the left, a horizontal chain of six spins is shown: two blue upward arrows, followed by three red downward arrows, and finally two blue upward arrows. To the right, a zoomed-in view shows two adjacent spins: a red downward arrow on the left and a blue upward arrow on the right, connected by a dashed line. Below the red arrow is the label $S_j^z = -1$, and below the blue arrow is the label $S_i^z = +1$.

$$H = \sum_{\langle i,j \rangle} J_{ij} S_i^z S_j^z$$

$J_{ij} < 0$ ferromagnetic
 $J_{ij} > 0$ antiferromagnetic

Ising *Beiträge zur Theorie des Ferromagnetismus*,
Zeitschrift für Physik, Band 31, 1925, S. 253–258

Ernst Ising
(1900-1998)

Image: APS

„It is shown that such a model [the one-dimensional] does not yet have ferromagnetic properties and this statement also extends to higher dimensions“

Werner Heisenberg
(1901-1974)

1928: Heisenberg model

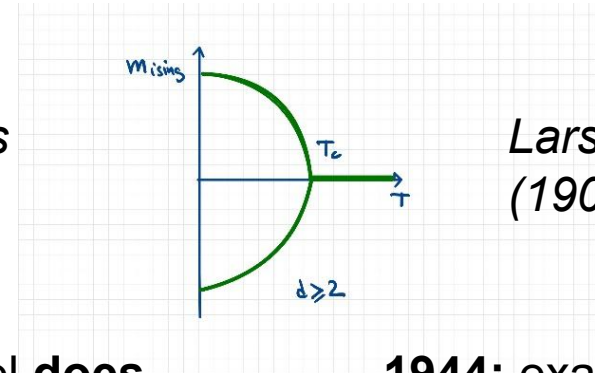
$$H = \sum_{\langle i,j \rangle} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

*Zur Theorie des Ferromagnetismus.
Zeitschrift für Physik, 619 (1928)*

Rudolf Peierls
(1907-1995)

1936: 2D Ising model **does**
have spontaneous
magnetization at low enough
temperatures

*Mathematical Proceedings of the Cambridge
Philosophical Society (1936)*



Lars Onsager
(1903-1976)

1944: exact solution of the 2D
Ising model

Phys. Rev. 65, 117 (1944)

Discovery of the nuclear shell model (1948-49)

Protons and neutrons occupy **quantized energy levels (shells)** inside the nucleus.

→ The crucial insight: **strong spin-orbit coupling** in the nuclear potential.

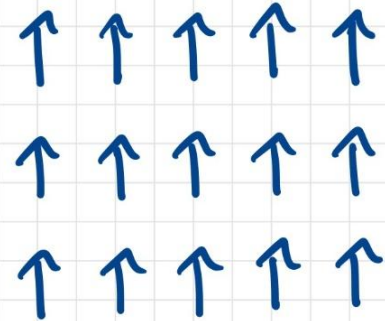
Physics Nobel Prize 1963

*Maria Goeppert Mayer
(1906-1972)*

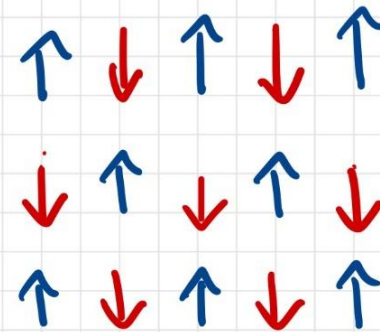
importance of spin-orbit coupling

spin interactions play a fundamental structural role in matter

Magnetism:

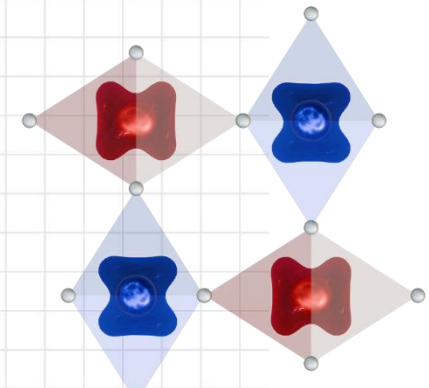


Ferromagnet



Antiferromagnet

(AFM1)



Altermagnet

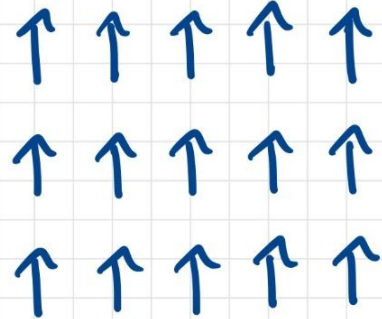
(AFM2)

(Libor Šmejkal's talk)

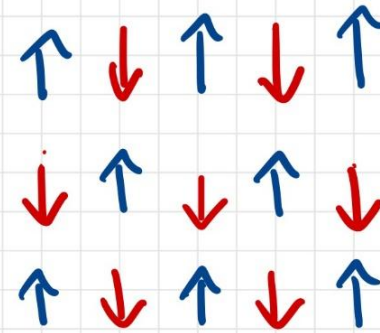
(Landau theory of phase transitions 1937)

Order parameter: magnetization

How to make magnetic states beyond:

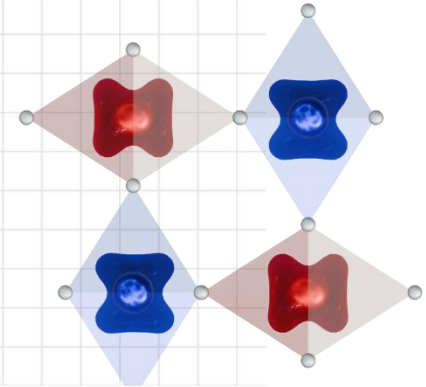


Ferromagnet



Antiferromagnet

(AFM1)



Altermagnet

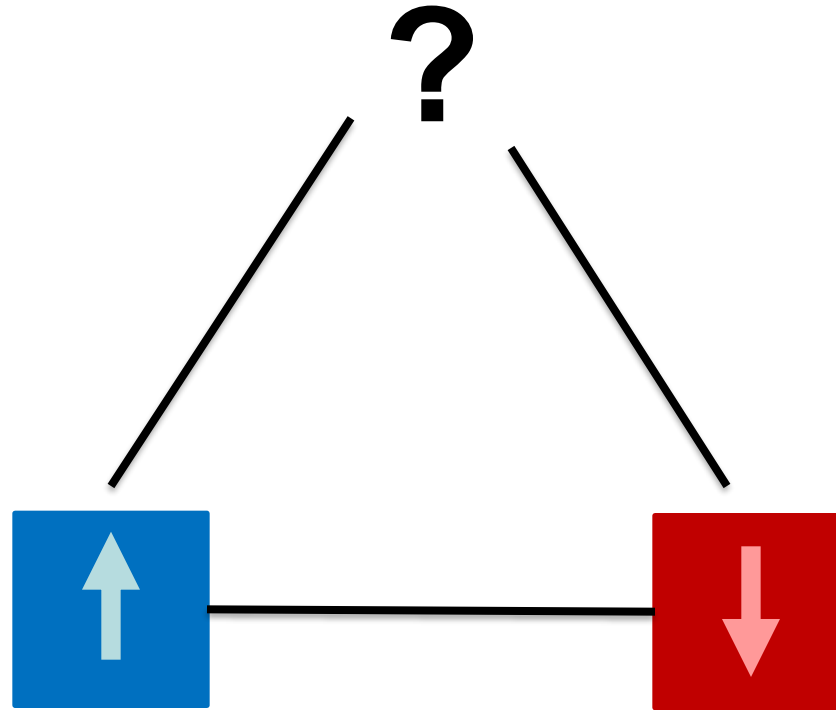
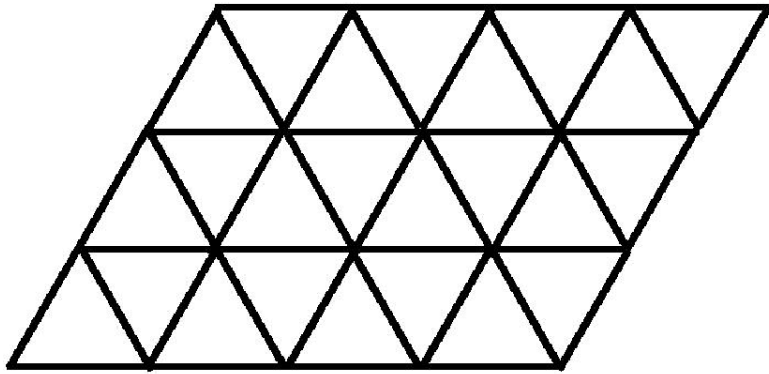
(AFM2)

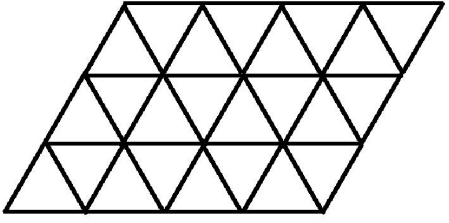
(Landau theory of phase transitions 1937)

Order parameter: magnetization

Geometric Frustration

magnetism in a
triangular lattice





- **Antiferromagnetism: the triangular Ising net**

Entropy at $T=0$ is finite: the system is disordered at all T

Gregory H. Wannier
Phys. Rev. 79, 357 (1950)
(1911-1983)

- Importance of **quantum effects** to induce new types of nonordered states (**Heisenberg model**)
(quantum spin liquid)

P.W. Anderson
Mat.Res.Bull 8, 153 (1973)
(1923-2020)

Quantum entanglement:

$|\uparrow\uparrow\rangle$ product state, not entangled

$\frac{1}{\sqrt{2}} |\uparrow\downarrow - \downarrow\uparrow\rangle$ entangled state

Nobel Prize 2022

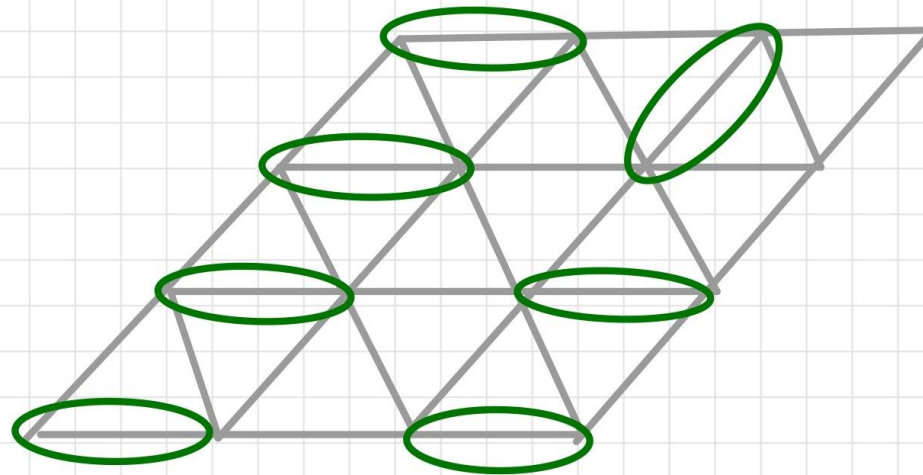
Alain Aspect

John Clauser

Anton Zeilinger

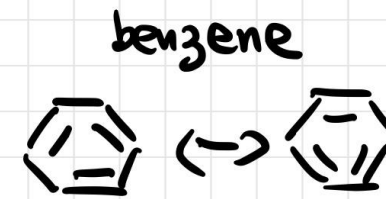
"for experiments with entangled photons, establishing the violation of Bell inequalities and pioneering quantum information science"

wavefunction for the triangular lattice



$$\text{green oval} = \frac{1}{\sqrt{2}} (\uparrow\downarrow - \downarrow\uparrow)$$

"valence bonds"



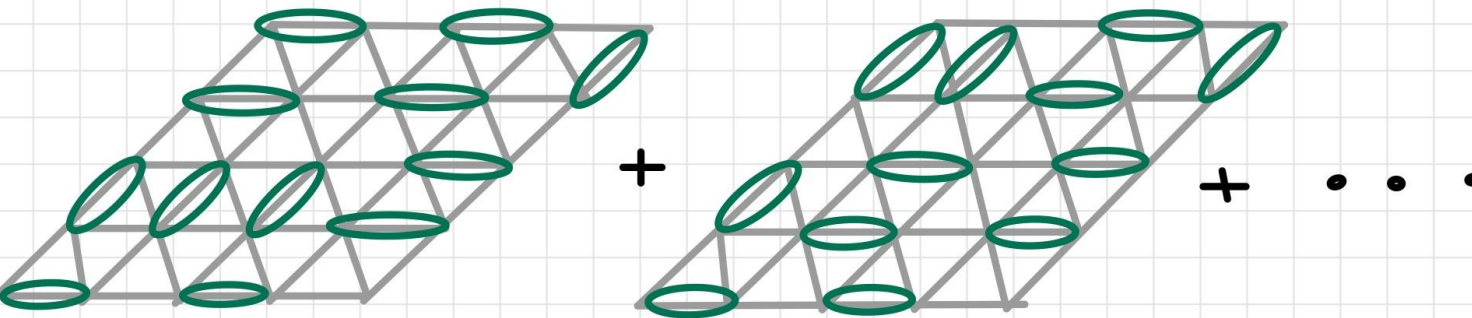
Kekulé valence bonds
Linus Pauling

Quantum spin liquid

triangular lattice

many-body wavefunction:

„linear superposition of quantum states“

$$\Psi =$$


$\frac{1}{\sqrt{2}} (\uparrow\downarrow - \downarrow\uparrow)$

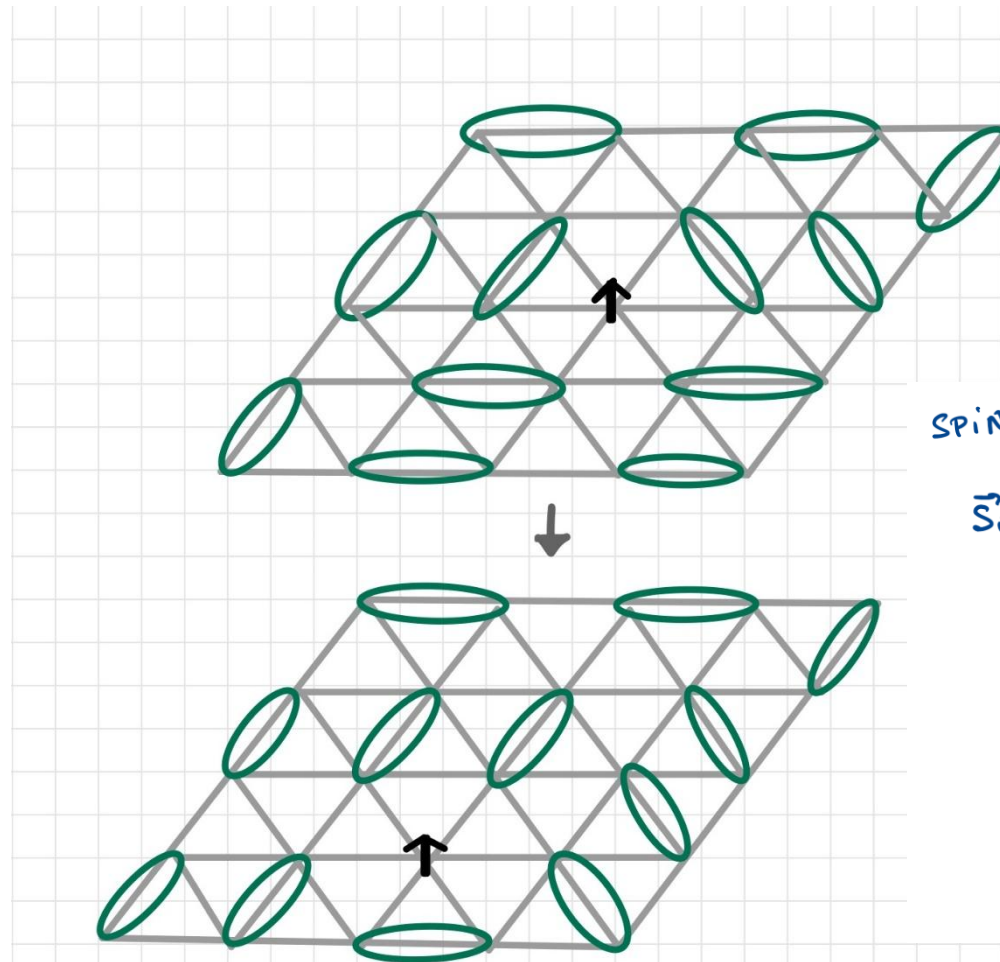
„resonating valence bonds“
long-range entangled state

$$|\Psi_{\text{RVB}}\rangle = \sum_c A_c \prod_{(i,j) \in c} \frac{1}{\sqrt{2}} (\uparrow_i \downarrow_j - \downarrow_i \uparrow_j)$$

Emergence of exotic “quasi-particles”: fractionalization

SPIN

fractionalization



SPIN : fermionic spin representation

SPINON

$$\vec{S}_i = \frac{1}{2} f_{i\alpha}^\dagger \vec{\tau}_{\alpha\beta} f_{i\beta} \quad f_{i\uparrow}^\dagger f_{i\uparrow} + f_{i\downarrow}^\dagger f_{i\downarrow} = 1$$

$$\{f_{i\alpha}, f_{j\alpha'}^\dagger\} = \delta_{ij} \delta_{\alpha\alpha'}$$

* Fermions in the enlarged Hilbert space

$$f_{i\alpha} \rightarrow e^{i\theta_i} f_{i\alpha}$$

* no electric charge, gauge charge 1

* transform as SU(2) spinors

* become physical through gauge invariant transformations

spinon: particle with spin and no charge

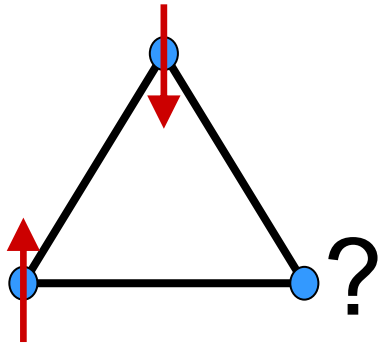
Quantum spin liquid:

- cooperative quantum phase with fluctuating spins down to lowest T and without symmetry breaking
- long-range entangled ground state with fractionalized excitations

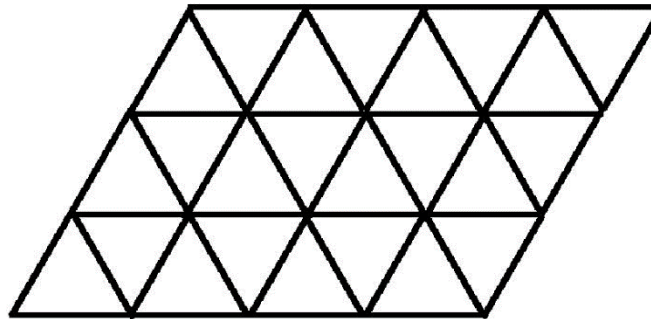
Frustration

Where to look?

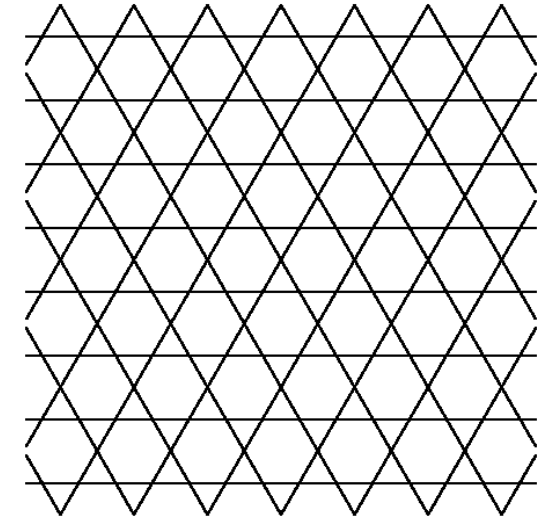
1. Geometric frustration



Triangular lattice



kagomé lattice

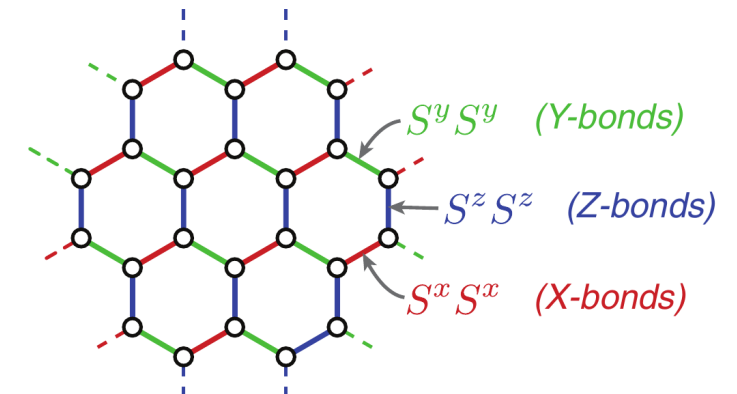


Bauer et al. Nat. Comm. (2014)

2. Hamiltonian engineering

(Hamiltonian with only non-commuting terms.
Anisotropic interactions)

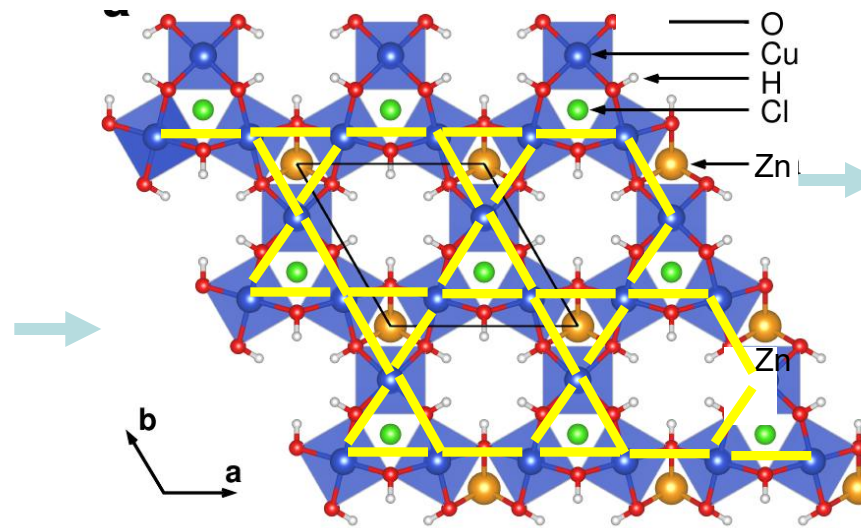
$$[S^y, S^z] = i \hbar/2\pi S^x$$



Kitaev hamiltonian

1. Geometric frustration

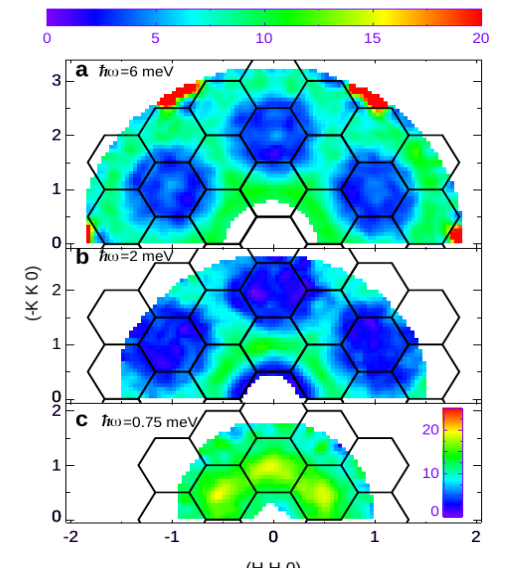
Herbertsmithite : spin $\frac{1}{2}$ kagome lattice
 $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$



candidate for an RVB state

spin liquid?

(disorder effects ...)



Inelastic Neutron Scattering

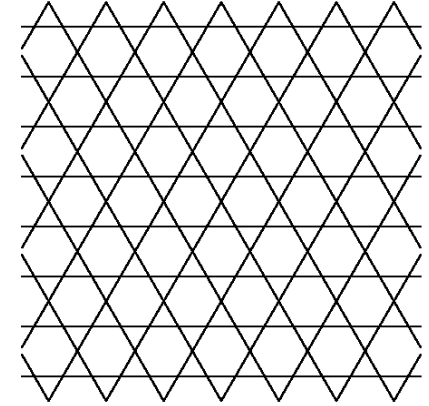
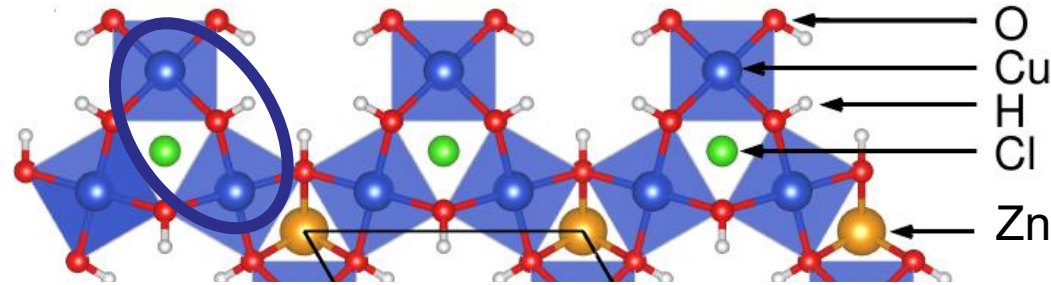
F. Bert, P. Mendels JPSJ (2010)

J.S. Helton et al. PRL (2010)

P. Khuntia et al. Nat. Phys. (2020)

Spin Hamiltonian

(Cu: 3d electrons
spin 1/2)



$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

(Isotropic Heisenberg
Exchange)

(Anisotropic Term)

strongest interaction

Geometric Frustration

2. Hamiltonian engineering

Kitaev Honeycomb Model

Alexei Kitaev, *Annals of Physics* 321, 2 (2006)

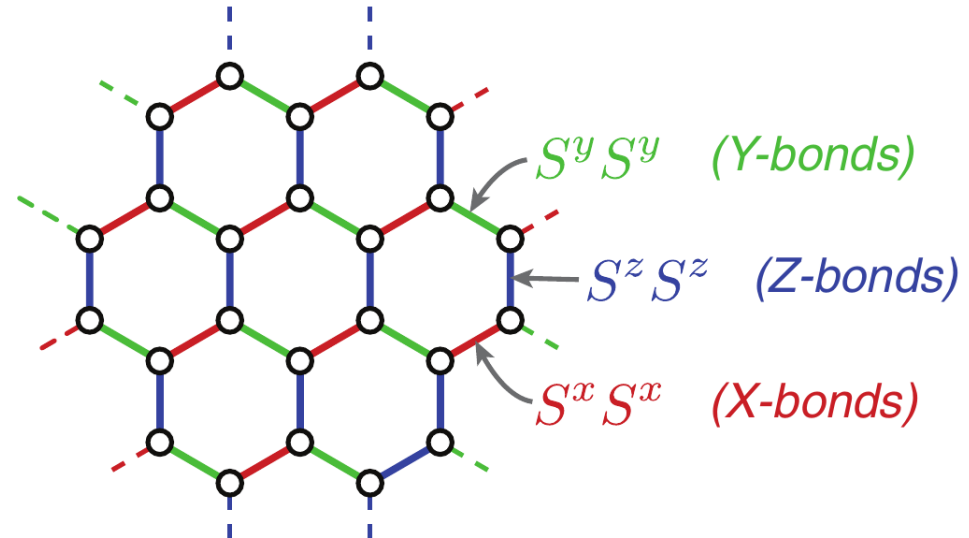
$$\mathcal{H} = K S_i^\gamma S_j^\gamma$$

$$\gamma = x, y, z$$

precursor model:

„compass model“

K.I. Kugel, D.I. Khomskii *Usp. Fiz. Nauk* **136**, 621 (1982)



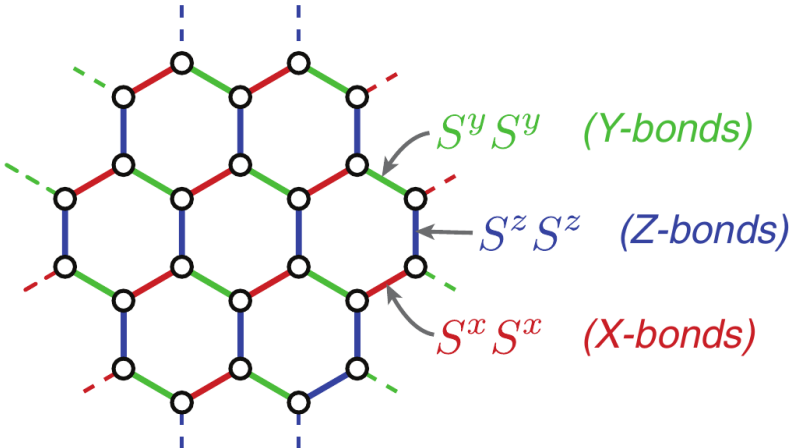
Ising interactions

$$[S^y, S^z] = i \hbar/2\pi S^x$$

Kitaev Honeycomb Model

Alexei Kitaev, *Annals of Physics* 321, 2 (2006)

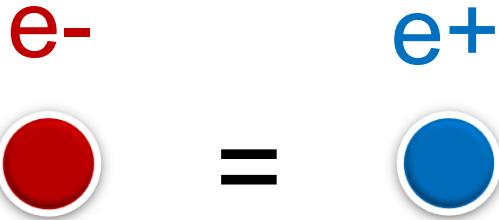
$$\mathcal{H} = K \left[\sum_{\langle i,j \rangle \in Z} S_i^z S_j^z + \sum_{\langle i,j \rangle \in Y} S_i^y S_j^y + \sum_{\langle i,j \rangle \in X} S_i^x S_j^x \right]$$



Majorana Fermions

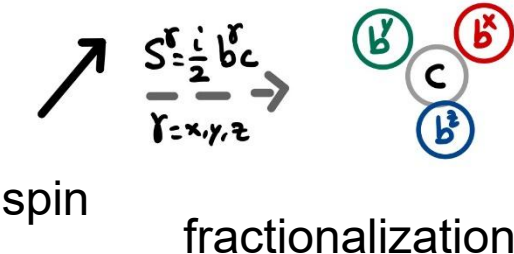


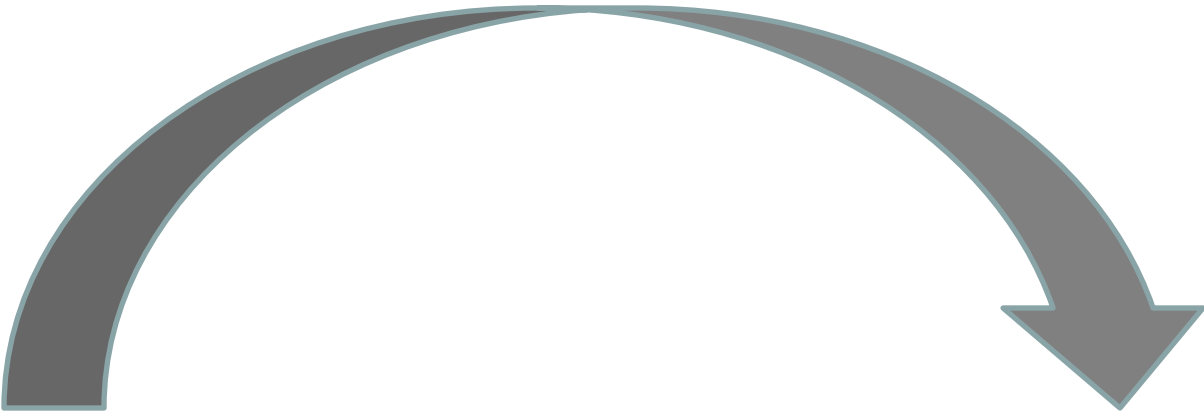
Ettore Majorana
(1906-1938)



$$c_i^\dagger = c_i \quad , \quad c_i^2 = 1 \quad , \quad \{c_i, c_j\} = 0$$

$$\begin{aligned} S_i^x &= ib_i^x c_i \\ S_i^y &= ib_i^y c_i \\ S_i^z &= ib_i^z c_i \end{aligned}$$





model

materials

Kitaev Honeycomb Model

◆ Conventional to write: $\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j$

J_{ij}
(Isotropic Exchange)

$\mathbf{D}_{ij}$
(Dzyaloshinskii-Moriya Term)

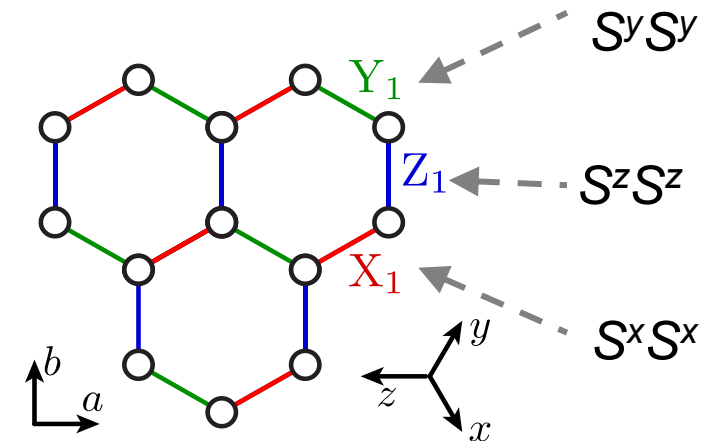
$\boldsymbol{\Gamma}_{ij}$
(Symmetric Term)

General Interactions

$$J_{ij} = 0 \quad \begin{matrix} D_{ij}^a = 0 & \Gamma_{ij}^{ab} = 0 \\ D_{ij}^b = 0 & \Gamma_{ij}^{ac} = 0 \\ D_{ij}^c = 0 & \Gamma_{ij}^{bc} = 0 \end{matrix} \quad \Gamma_{ij}^{aa}, \Gamma_{ij}^{bb}, \Gamma_{ij}^{cc} = \begin{cases} K \\ 0 \end{cases} \quad \begin{matrix} \text{(Depending on} \\ \text{orientation of the bond.)} \end{matrix}$$

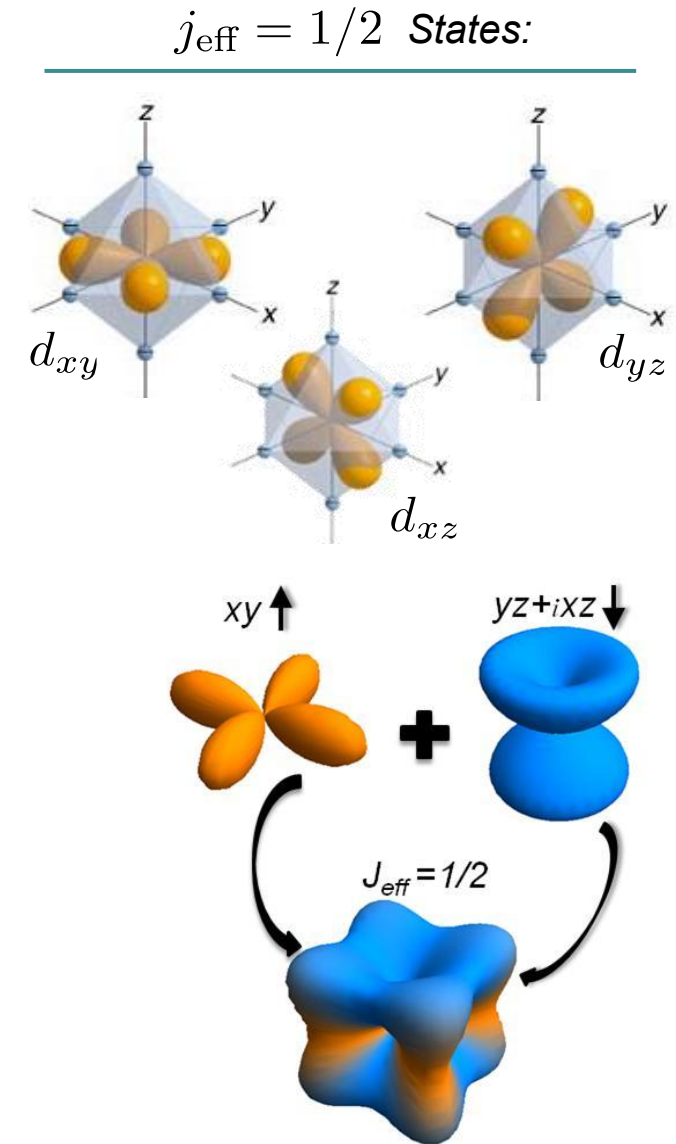
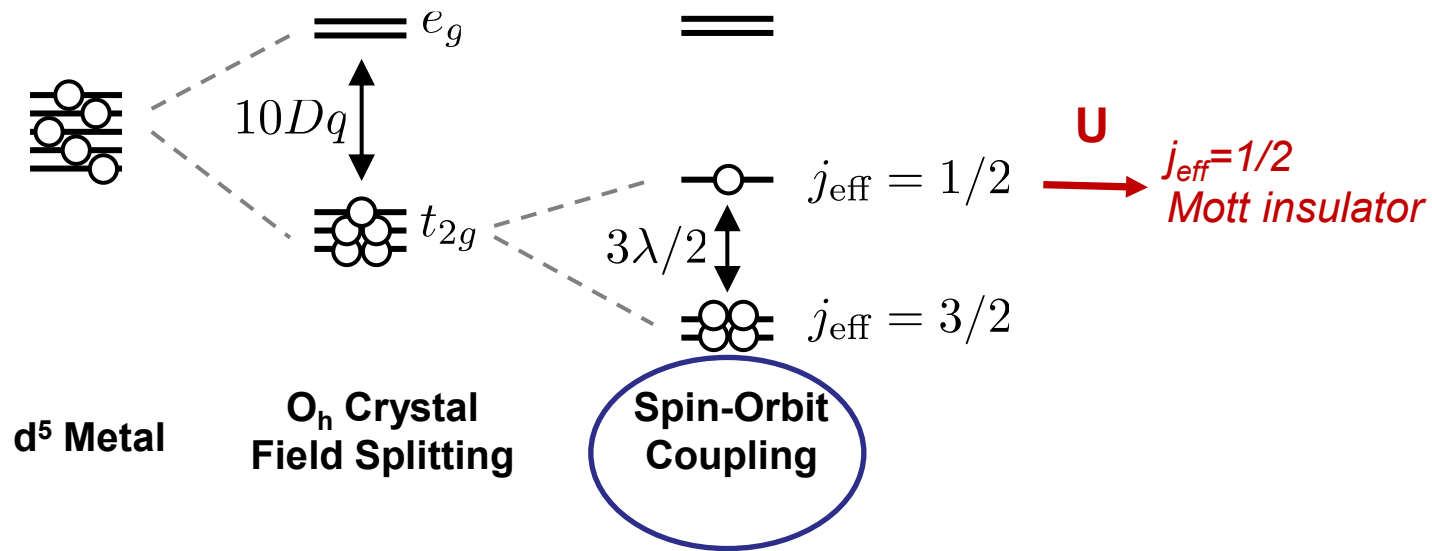
Kitaev Model

$$\mathcal{H} = \begin{pmatrix} S_i^x & S_i^y & S_i^z \end{pmatrix} \begin{pmatrix} J & \Gamma & \Gamma' \\ \Gamma & J & \Gamma' \\ \Gamma' & \Gamma' & J + K \end{pmatrix} \begin{pmatrix} S_j^x \\ S_j^y \\ S_j^z \end{pmatrix}$$



Kitaev Honeycomb Model – Relation to Real Materials

How to engineer dominant Kitaev terms?

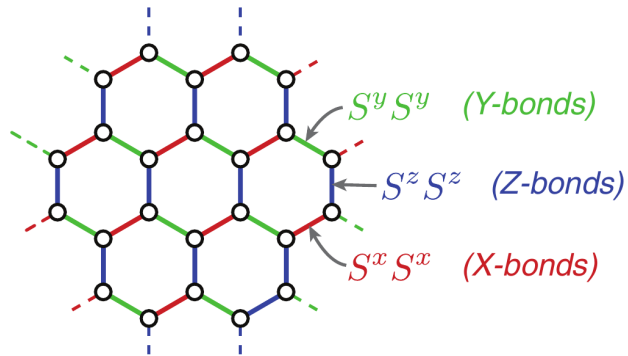


Khaliullin, Maekawa PRL 85, 3950 (2000),
 Khaliullin, Horsch, Oles PRL 86, 3879 (2001)

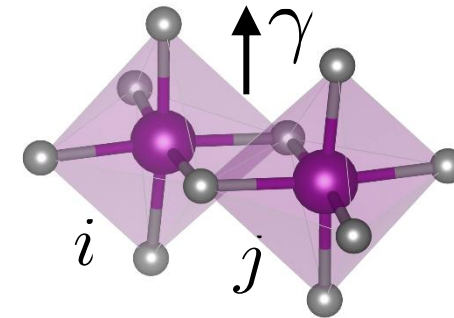
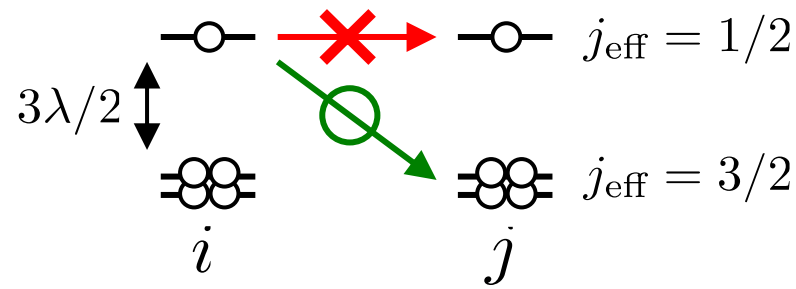
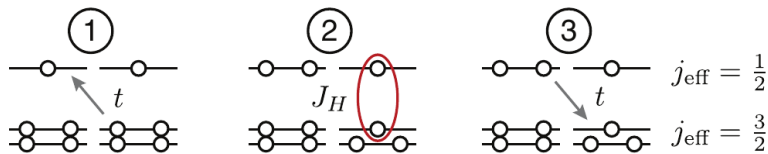
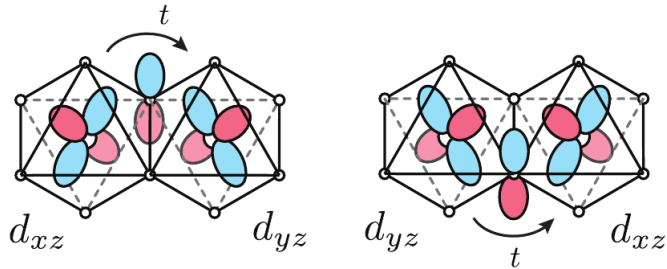
G. Jackeli, G. Khaliullin PRL 102, 017205 (2009)

Kitaev Honeycomb Model – Relation to Real Materials

How to engineer dominant Kitaev terms?



Only off-diagonal terms



◆ When such metals are **edge-sharing**

$$J_{ij} \sim 0$$

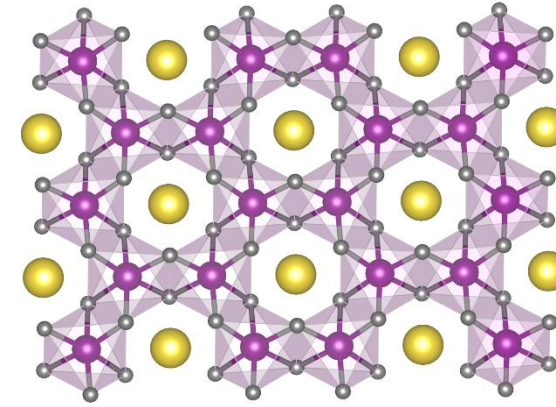
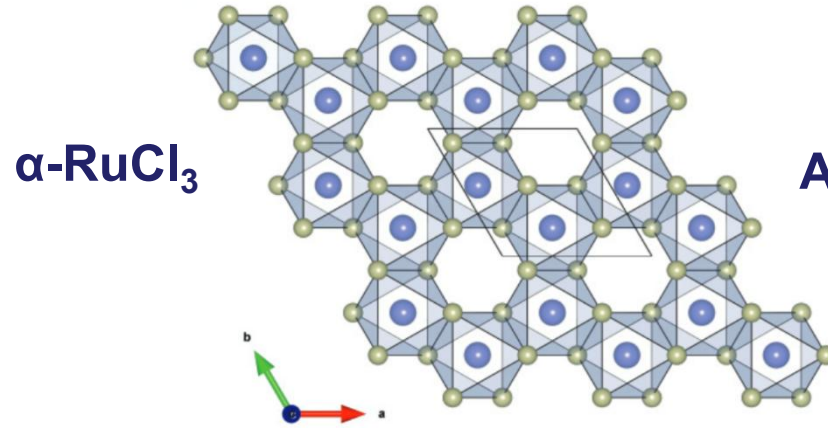
◆ When such metals have **inversion symmetry**,

$$\mathbf{D}_{ij} = 0 \quad \mathbf{\Gamma}_{ij} = 0 + \mathcal{O}(J_H t^2)$$

◆ Subleading term from effective **Hund's coupling** between $j_{\text{eff}} = \frac{1}{2}$ states to the $m_j = \pm 3/2$ states that is Ising-like.

$$\mathcal{H} = K S_i^\gamma S_j^\gamma \quad K \sim \frac{J_H t^2}{(2U + 3\lambda)^2}$$

Kitaev Materials



$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

(Isotropic Heisenberg
Exchange)

(Dzyaloshinskii-Moriya Term)

(Symmetric Term)

Hamiltonian engineering

strongly spin-orbit coupled systems

bond-dependent

strongest interaction

$$\mathcal{H} = K S_i^\gamma S_j^\gamma$$

**How do we extract the spin model Hamiltonian
from first principles?**

Material $\leftrightarrow$ Model

**... and calculate properties to compare with
experiments?**

Ab initio

Solve the Schrödinger /Dirac equation for interacting electrons and ions:

$$i\hbar \frac{\partial}{\partial t} \Psi(\mathbf{r}, t) = H \Psi(\mathbf{r}, t)$$

- 10^{23} coupled equations!!!

Density Functional Theory

Walter Kohn
Nobel Prize in Chemistry 1998

Hohenberg-Kohn Theorem:

The groundstate energy of a system of interacting particles is a functional of the groundstate density $\rho(r)$: $E[\rho(r)]$

Kohn-Sham Ansatz:

Replace interacting many-body problem:

$$E = E[\rho]$$

with a non-interacting problem with all many-body effects in a XC functional

Replace interacting many-body problem:

$$E = E[\rho]$$

with a non-interacting problem with all many-body effects in a XC functional:

$$E[\rho] = T_S[\rho] + E_{ext}[\rho] + E_H[\rho] + E_{xc}[\rho]$$

$$(T + V_{ei}(\mathbf{r}) + V_H(\mathbf{r}) + V_{xc}(\mathbf{r}))\psi_i(\mathbf{r}) = \epsilon_i \psi_i(\mathbf{r})$$

$$\rho(\mathbf{r}) = \sum_{occ} \psi_i^*(\mathbf{r}) \psi_i(\mathbf{r})$$

$$V_H = e^2 \int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}$$

$$V_{xc}(\mathbf{r}) = \frac{\delta E_{xc}[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r})}$$

$$\psi_{\vec{k}}(\vec{r}) = e^{i\vec{k} \cdot \vec{r}} u_{\vec{k}}(\vec{r})$$

Variational principle !

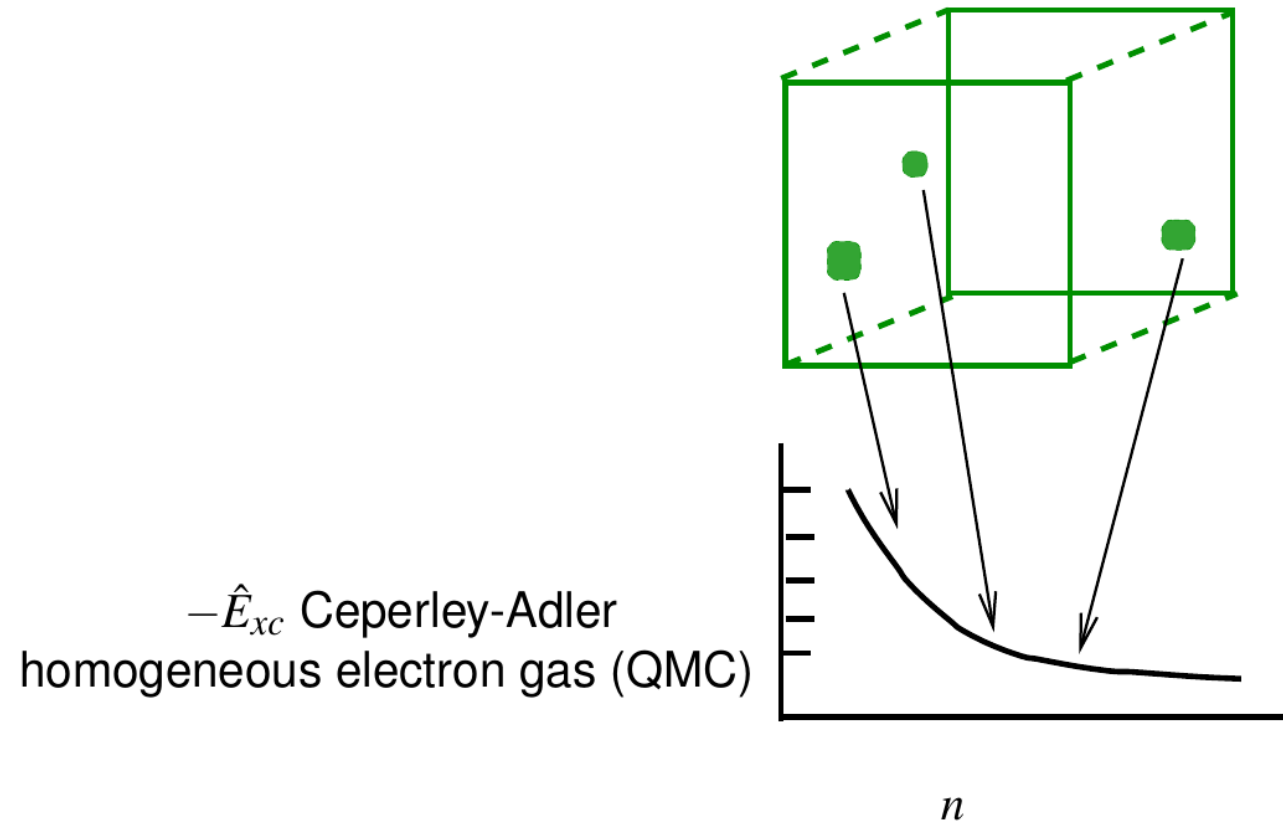
Bloch wavefunctions

Local Density Approximation (LDA)

$$E_{xc}[n] = \int d\mathbf{r} n(\mathbf{r}) \hat{E}_{xc}(n(\mathbf{r}))$$

Kohn, Sham (1964)

$\hat{E}_{xc}(n(\mathbf{r}))$ is approximated by a local function of the density.



$$(T + V_{ei}(\mathbf{r}) + V_H(\mathbf{r}) + V_{xc}(\mathbf{r}))\psi_i(\mathbf{r}) = \varepsilon_i\psi_i(\mathbf{r})$$

$$V_H = e^2 \int d\mathbf{r}' \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} \quad V_{xc}(\mathbf{r}) = \frac{\delta E_{xc}[\rho(\mathbf{r})]}{\delta \rho(\mathbf{r})} \quad \rho(\mathbf{r}) = \sum_{occ} \psi_i^*(\mathbf{r})\psi_i(\mathbf{r})$$

- **Plane wave methods :**

use of plane waves to solve the Kohn-Sham equations

- **Localized atomic(-like) orbitals (LCAO):**

basis that captures the atomic-like features of solids and molecules

- **Atomic sphere methods:**

efficient representation of atomic-like features that are rapidly varying near each nucleus and smoothly varying functions between the atoms

→ **APW.-** Augmented Planewave

$$\psi_k(\mathbf{r}, E) = \frac{1}{\Omega^{1/2}} \sum_G c_G e^{i(\mathbf{G}+\mathbf{k})\mathbf{r}} \quad r \in I$$

$$\sum_{lm} A_{lm}^{\mathbf{k}} u_l(r, E) Y_{lm}(\mathbf{r}) \quad r \in S$$

$$\left\{ -\frac{d^2}{dr^2} + \frac{l(l+1)}{r^2} + V(r) - E_l \right\} r u_l(r) = 0$$

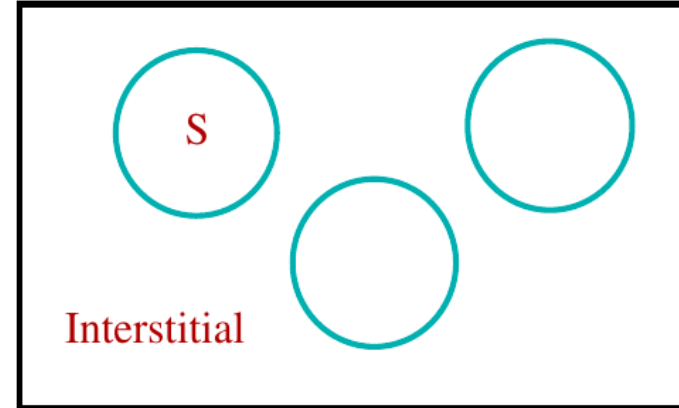
→ **MTO.-** Muffin Tin Orbitals

$$K_l(r) Y_L(\mathbf{r}) \quad r \in I$$

$$\psi_L(\mathbf{r}, E) = N_L(E) u_l(r, E) Y_L(\mathbf{r}) + P_L(E) J_l(r) Y_L(\mathbf{r}) \quad r \in S$$

$J_l(r), K_l(r)$ Bessel Functions $L = lm$

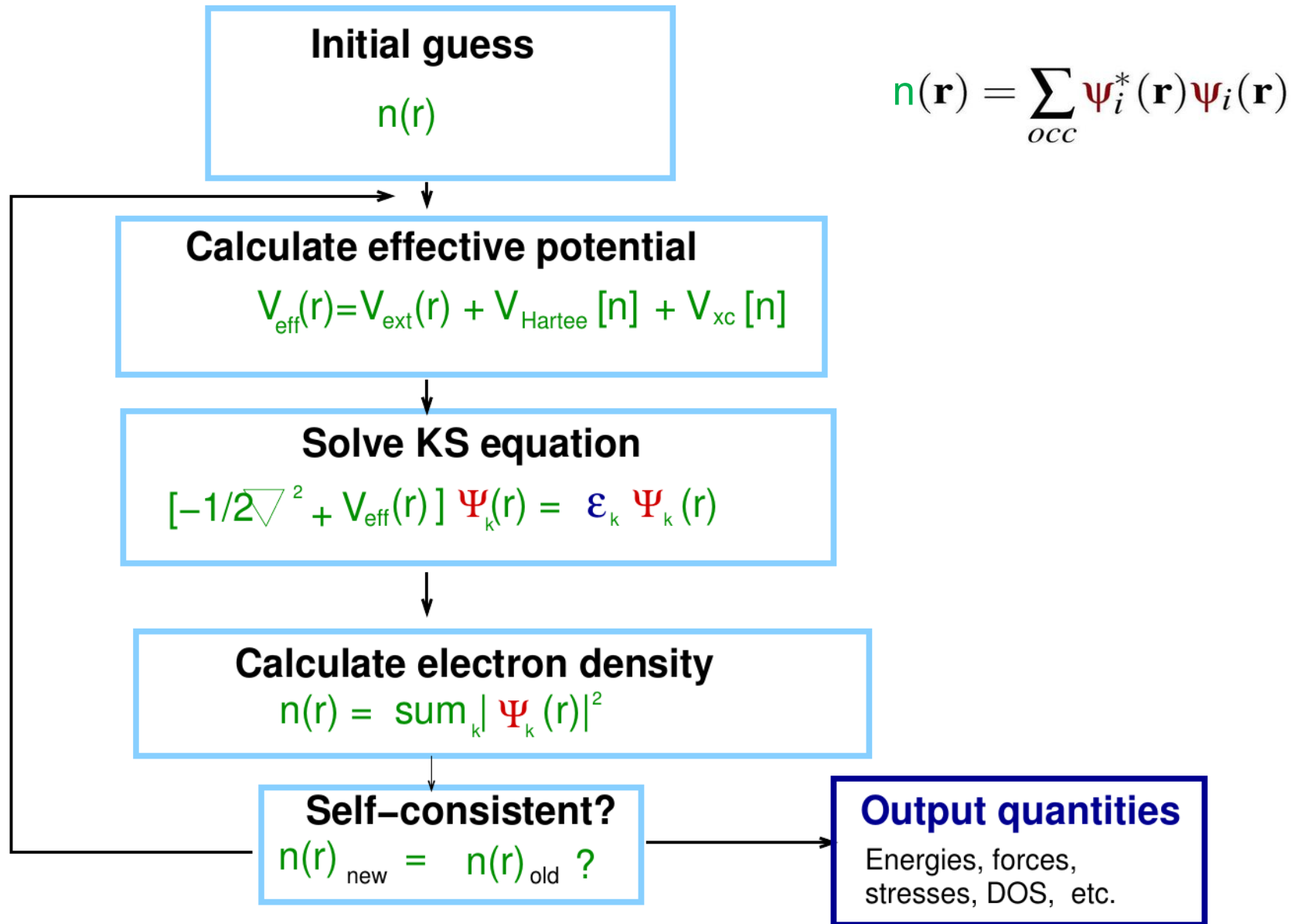
- calculation of the Energy bands through the KKR - equation (Tail-cancellation theorem).



Codes: Wien2k, LMTO, VASP, FPLO, QUANTUM ESPRESSO, ...

Korringa-Kohn-Rostoker method:
Multiple scattering theory and variational method

Self-Consistent Kohn-Sham Equations

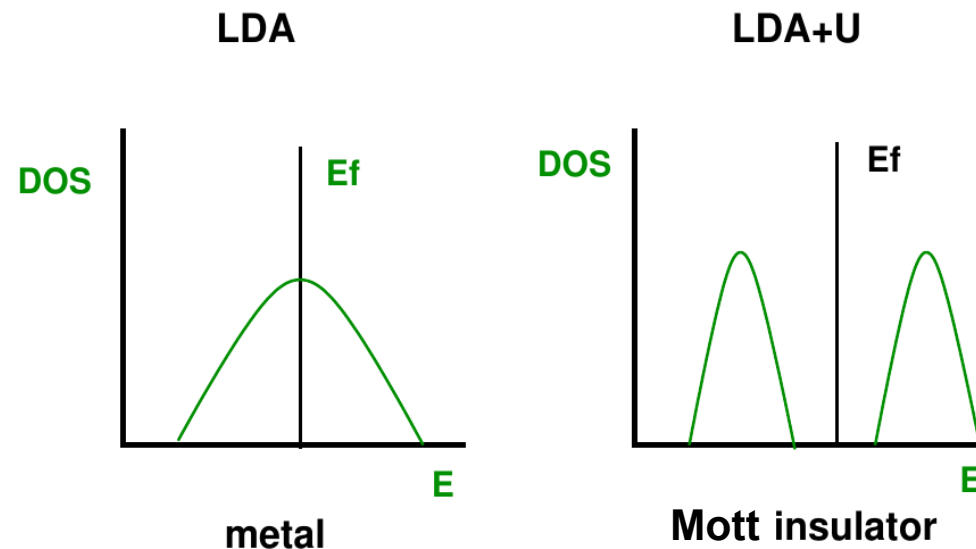


Anisimov, Zaanen, Andersen, Phys. Rev. B **44**,943 (1991)

- **Inclusion of exchange correlation J (Hund's exchange):**

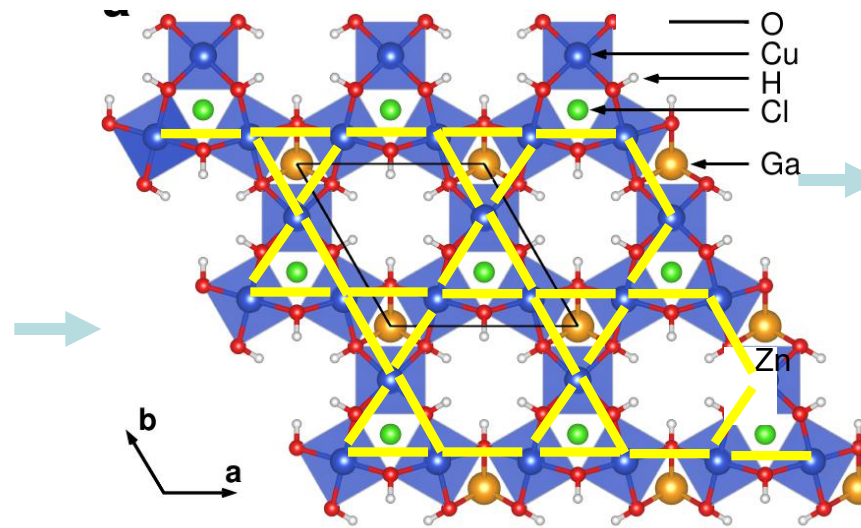
$$E = E^{LDA} - \frac{U}{2}N(N-1) + \frac{J}{2}[N^{\uparrow}(N^{\uparrow}-1) + N^{\downarrow}(N^{\downarrow}-1)] + \frac{U}{2} \sum_{j,k,\sigma} n_{j,\sigma} n_{k,-\sigma} + \frac{1}{2}(U-J) \sum_{i \neq j, \sigma} n_{i,\sigma} n_{j,\sigma}$$

- ▶ electrons with the same spin feel interaction $U - J$
- ▶ electrons with different spin feel interaction U



1. Geometric frustration

Herbertsmithite : spin $\frac{1}{2}$ kagome lattice
 $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$



Mott insulator

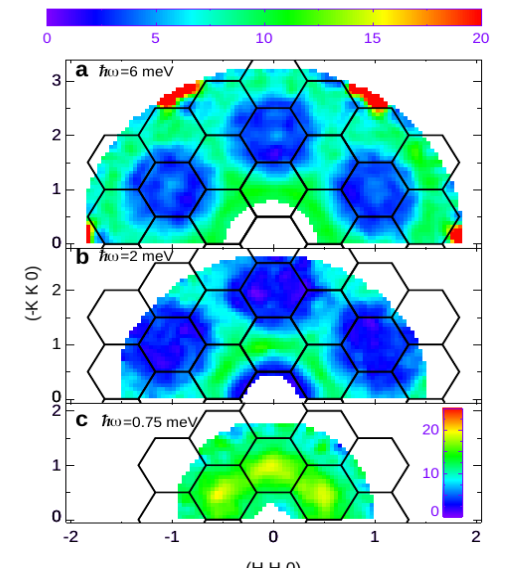
spin liquid?

(disorder effects ...)

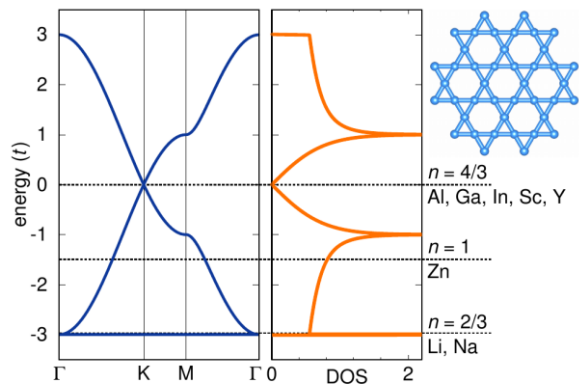
F. Bert, P. Mendels JPSJ (2010)

J.S. Helton et al. PRL (2010)

P. Khuntia et al. Nat. Phys. (2020)



Inelastic Neutron Scattering

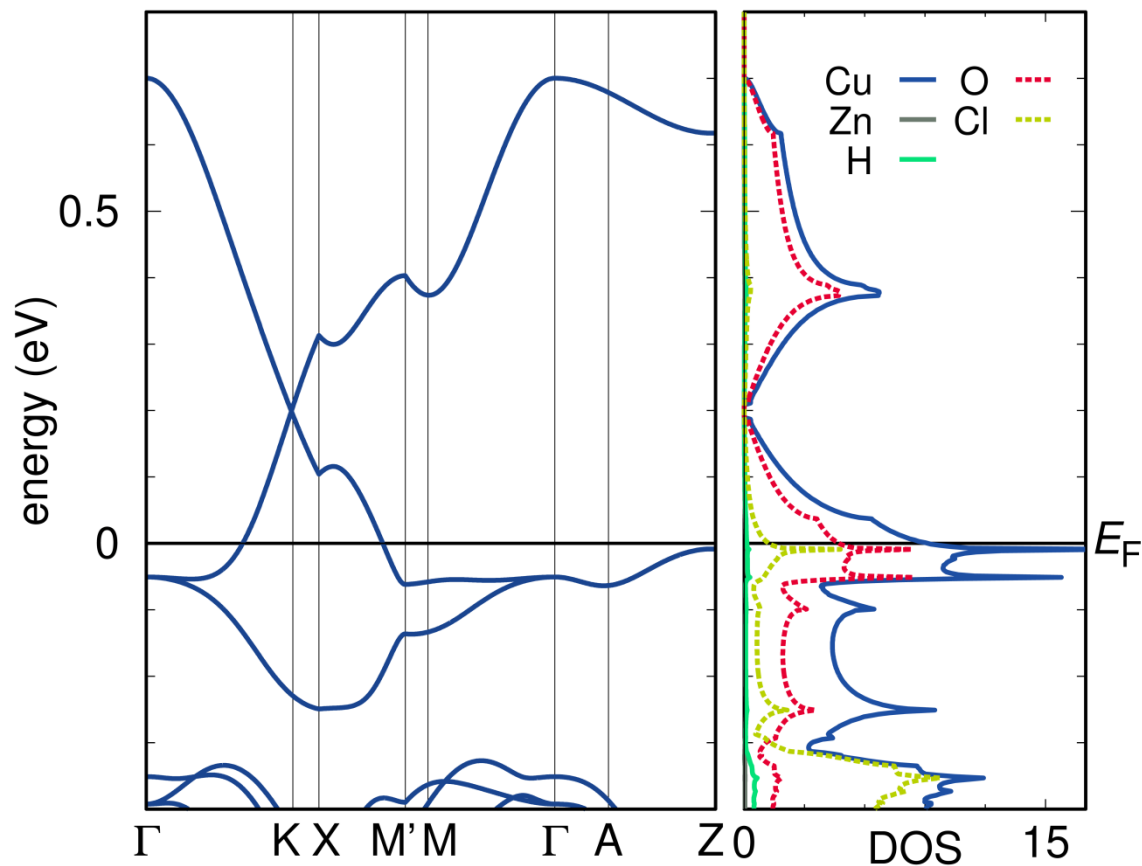


Herbertsmithite

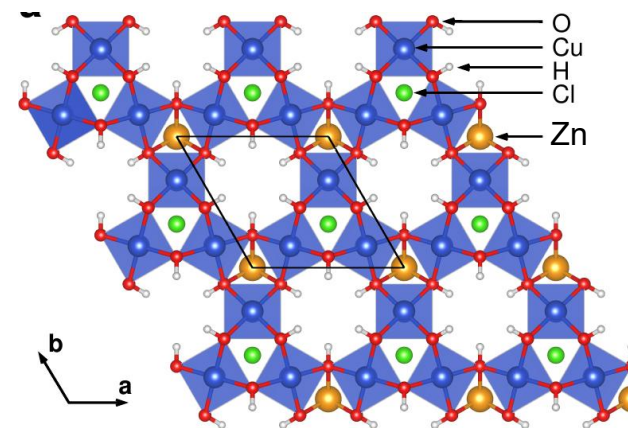
$\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

DFT-GGA calculation/FPLO basis

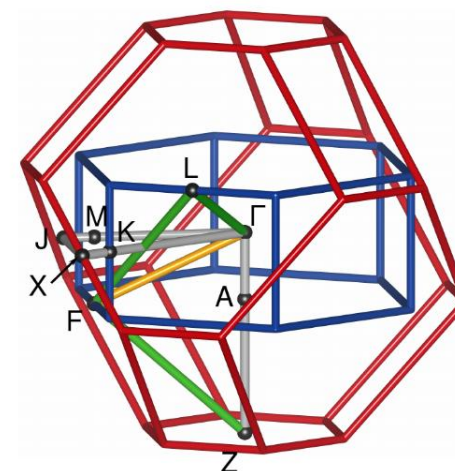
nn tight-binding



Kagome lattice of spin $\frac{1}{2}$ Cu



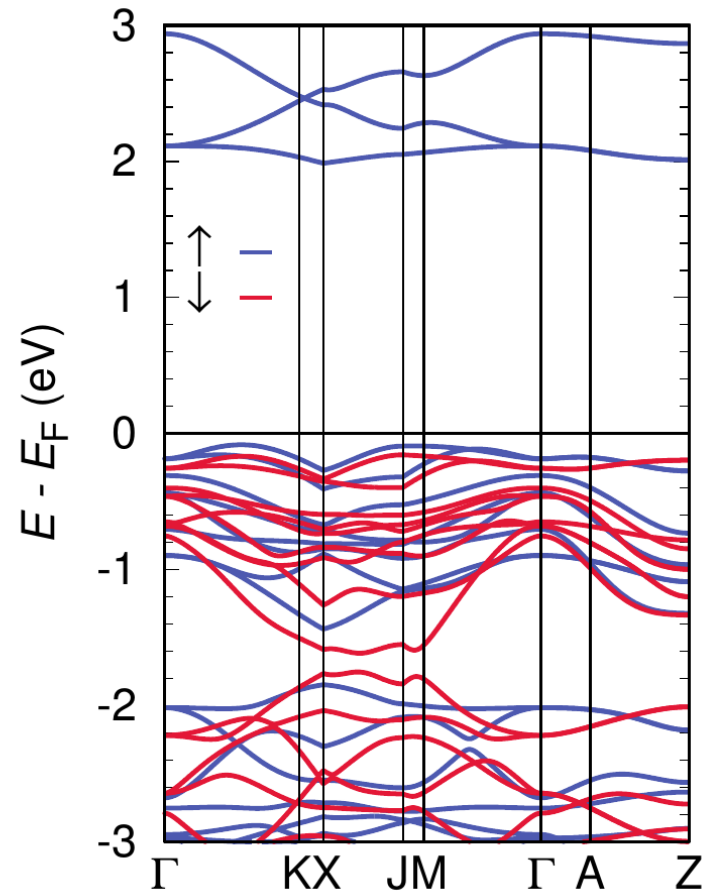
Cu $d_{x^2-y^2}$ bands



Herbertsmithite

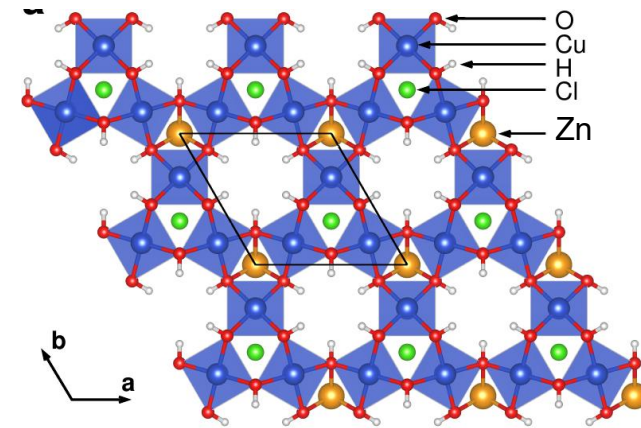
$\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

DFT-GGA+U



PRB **88**, 075106 (2013)
PRB **97**, 020104(R) (2018)

Kagome lattice of spin $\frac{1}{2}$ Cu
 $n=1$



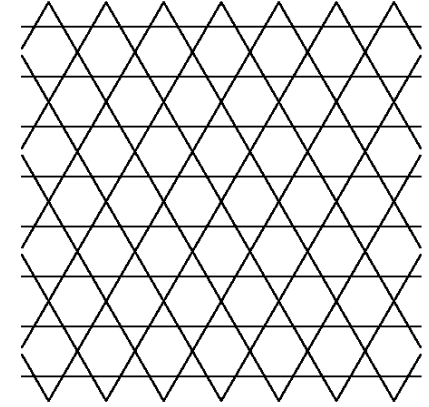
Cu $d_{x^2-y^2}$ bands

Mott insulator

nature of spin liquid?

Spin Hamiltonian

(Cu: 3d electrons)



$$\mathcal{H} = \underbrace{J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j}_{\text{(Isotropic Heisenberg Exchange)}} + \underbrace{\mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j}_{\text{(Dzyaloshinskii-Moriya Term)}} + \underbrace{\mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j}_{\text{(Symmetric Term)}}$$

*(Isotropic Heisenberg
Exchange)*

(Dzyaloshinskii-Moriya Term)

(Symmetric Term)



strongest interaction

Geometric Frustration

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

Total energy calculations (isotropic case):

- Map the energy differences of frozen collinear spin configurations in DFT supercell calculations onto a spin-1/2 Heisenberg model and evaluate J in the dimer approximation

$$H = -2J\mathbf{S}_i\mathbf{S}_j$$

$$E_{FM} = -J[S_T(S_T + 1) - 2S(S + 1)]$$

$$E_{AFM} = J[2S(S + 1)]$$

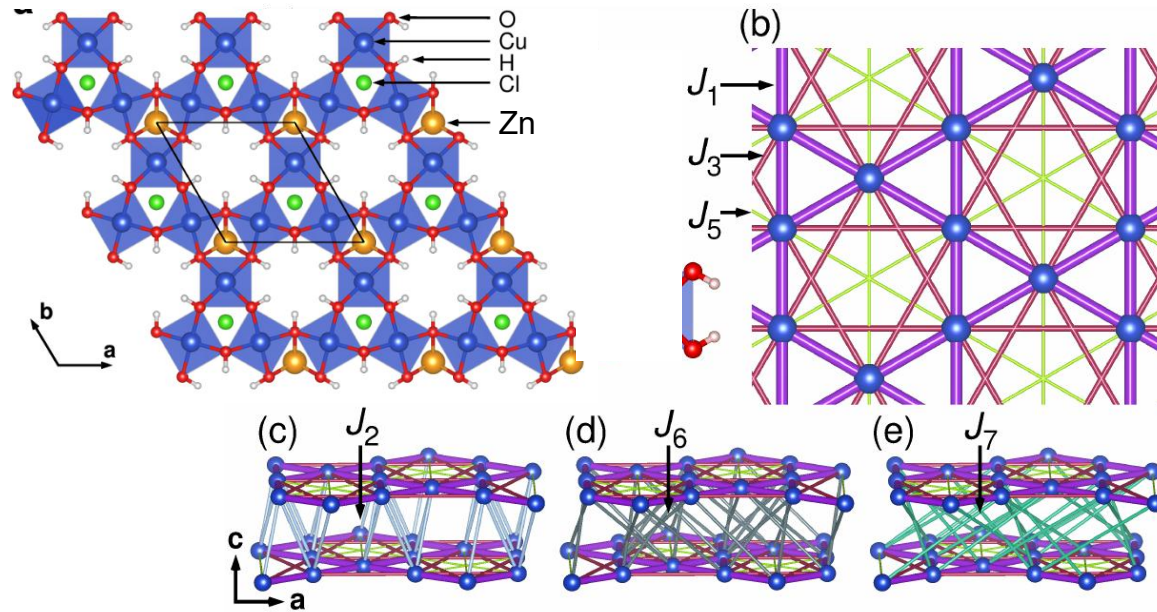
$$\Delta E = \frac{E_{FM} - E_{AFM}}{2} = -\frac{J}{2}S_T(S_T + 1)$$

mind the neighbors' scaling factors!

Total energy calculations (isotropic case):

Herbertsmithite
 $\text{ZnCu}_3(\text{OH})_6\text{Cl}_2$

PRB 88, 075106 (2013)



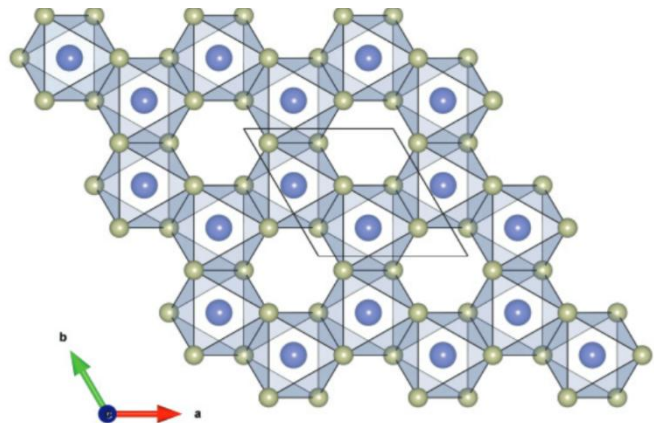
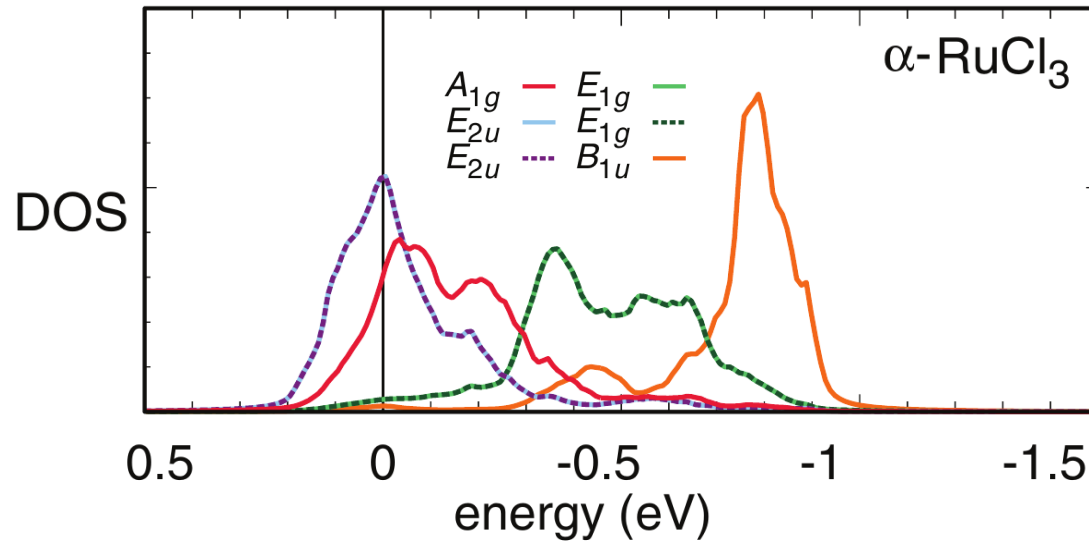
name	$d_{\text{Cu}-\text{Cu}}$	type	J_i (K) $U = 6 \text{ eV}$
kagomé layer couplings			
J_1	3.4171	kagomé nn	182.4
J_3	5.91859	kagomé 2nd nn	3.4
J_5	6.8342	kagomé 3rd nn	-0.4
interlayer couplings			
J_2	5.07638	interlayer 1st nn	5.3
J_4	6.11933	interlayer 2nd nn	-1.5
J_6	7.00876	interlayer 3rd nn	-6.4
J_7	8.51328	interlayer 4th nn	3.0
J_9	9.17347	interlayer 6th nn	2.5

J_i determined from total GGA+U energies of 9 spin configurations in 2x1x2 supercells

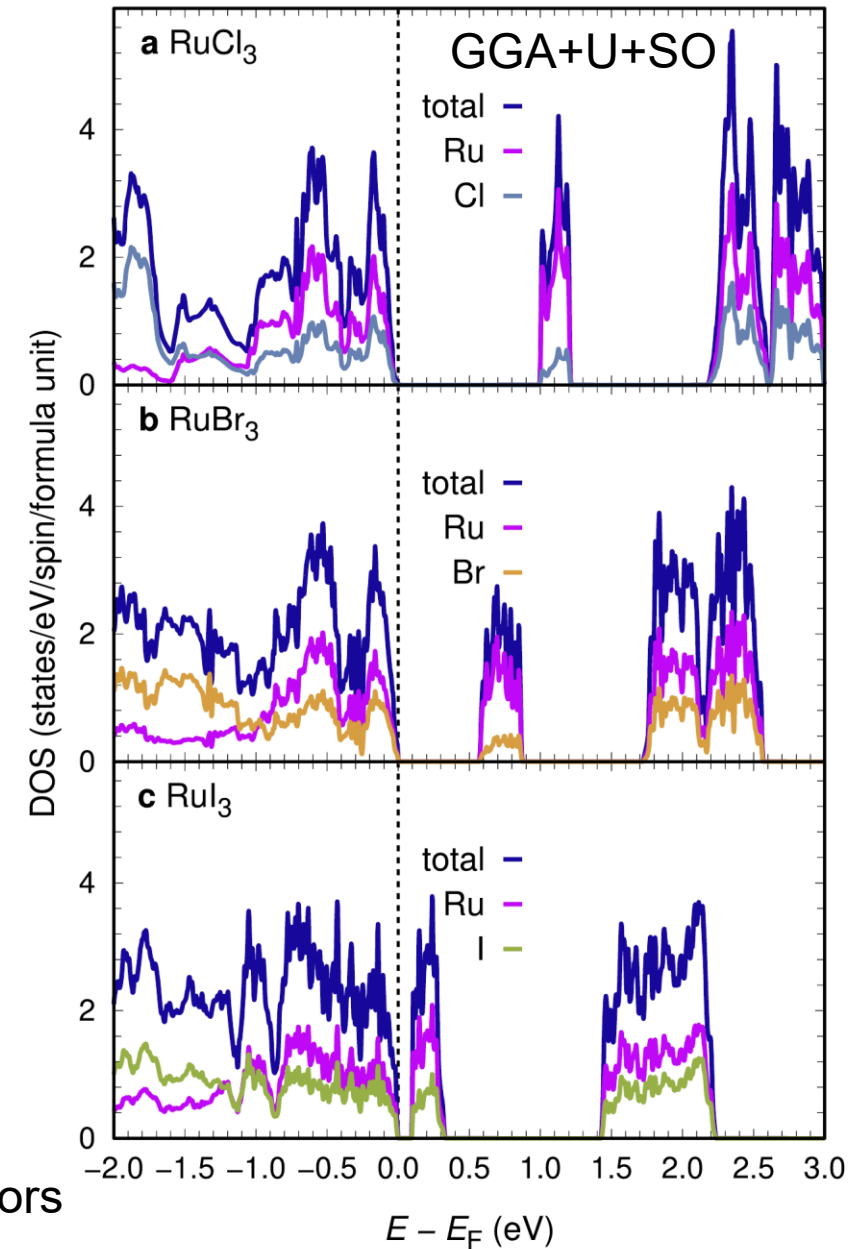
→ agreement with experiment

Kitaev materials

DFT-GGA calculation/FPLO basis



(Spin-orbit assisted) Mott insulators



What about anisotropic spin hamiltonians?

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

spin-orbit coupling

(relevant for non-centrosymmetric systems, Kitaev, ...)

- The method of total energy calculations requires **relativistic non-collinear spin configurations**

What about anisotropic spin hamiltonians?

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \mathbf{\Gamma}_{ij} \cdot \mathbf{S}_j$$

spin-orbit coupling

(relevant for non-centrosymmetric systems, Kitaev, ...)

→ we will consider an alternative method →
obtain the low-energy spin Hamiltonian
from **numerical projection from DFT**

Low-energy spin-Hamiltonian from numerical projection

1) An effective Hubbard-like hamiltonian $\mathcal{H}_{\text{eff}}$ is derived from DFT

2) $\mathcal{H}_{\text{eff}}$ is exactly diagonalized for finite clusters

Low-energy eigenstates $\{|\{\psi_n\}\rangle\}$ E_n

3) Choose a suitable low-energy Hilbert space of pure spin states to map the low-energy values of $\mathcal{H}_{\text{eff}}$ to an effective spin Hamiltonian

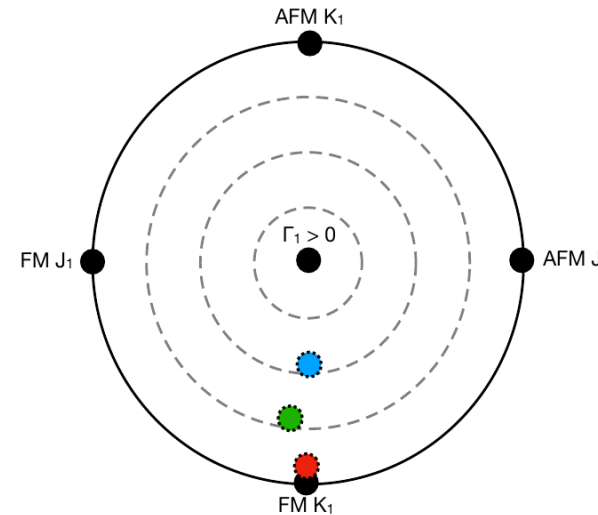
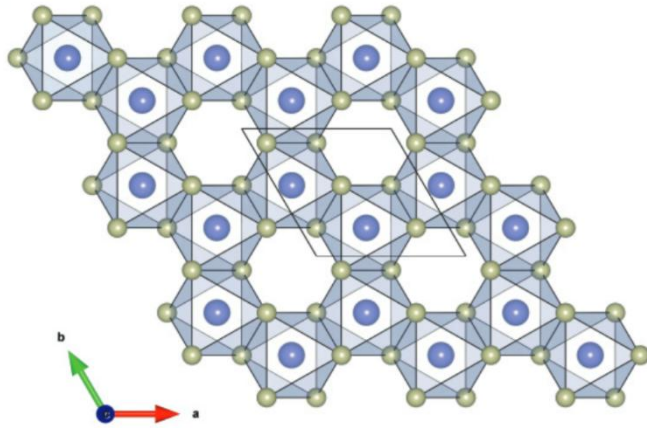
$$\mathcal{S}_{\text{low}} = \{|\phi_n\rangle\} \quad |\xi_n\rangle \in \mathcal{S}_{\text{low}} \quad |\xi_n\rangle = \mathcal{P}|\psi_n\rangle = \sum |\phi_m\rangle \langle \phi_m | \psi_n \rangle$$

4) Orthonormalize $[\mathbb{S}]_{nm} = \langle \xi_n | \xi_m \rangle \quad |\xi'_n\rangle = \mathbb{S}^{-1/2} \mathcal{P} |\psi_n\rangle$

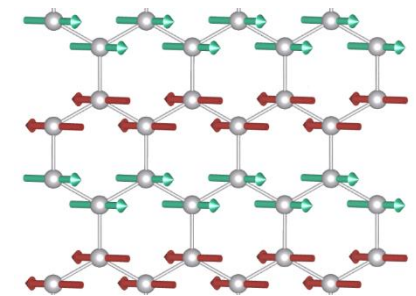
$$\mathcal{H}_{\text{spin}} |\xi'_n\rangle = E_n |\xi'_n\rangle$$

Materials: *Ab initio*-based Kitaev Honeycomb Model

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_1 \mathbf{S}_i \cdot \mathbf{S}_j + \boxed{K_1 S_i^\gamma S_j^\gamma} + \Gamma_1 (S_i^\alpha S_j^\beta + S_i^\beta S_j^\alpha) \\ + \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} J_3 \mathbf{S}_i \cdot \mathbf{S}_j + \Gamma'_1 (S_i^\alpha S_j^\gamma + S_i^\beta S_j^\gamma + S_i^\gamma S_j^\alpha + S_i^\gamma S_j^\beta)$$



Dominant interactions:
Anisotropic and
Low Symmetry



◆ Zigzag magnetic order state

(in meV)

- α -RuCl₃: $K_1 \approx -5$, $\Gamma_1 \approx +2.5$, $J_1 \approx -0.5$, $J_3 \approx +0.5$
- Na₂IrO₃: $K_1 \sim -15$, $J_3 \sim +7$, $\Gamma_1 \sim +2$, $\Gamma'_1 \sim -2$
- α -Li₂IrO₃: $K_1 \sim -13$, $\Gamma_1 \sim +10$, $DM_2 > 0$, $J_3 \sim +4$, $\Gamma'_1 \sim -2$

Winter...,RV PRB 93, 214431 (2016)

Kaib,...,RV npj Quantum Materials (2022)

Yadav et al. Sci. Rep. 6 (2016)

Lecture 1 (DFT and magnetism) techniques

- *Motivation*
- *Microscopic description of magnetism (frustrated magnetism / spin-orbit coupling) → Kitaev frustration*

→ from first principles to spin model Hamiltonians

$$\mathcal{H} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \mathbf{D}_{ij} \cdot \mathbf{S}_i \times \mathbf{S}_j + \mathbf{S}_i \cdot \boldsymbol{\Gamma}_{ij} \cdot \mathbf{S}_j + \dots$$

Lecture 2 (Correlated electrons and magnetism) applications

- *Excitations and dynamics in magnetic systems :*

Linear response → INS, ESR, Raman

Non-linear response → non-linear optics (2DCS)