

Russian Science Foundation

Спин-орбитальное взаимодействие, как источник новых эффектов в магнитных системах

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Introduction: Spin-orbit coupling (SOC) in transition metal compounds

$$
\begin{array}{r}\n\hline\n\text{RMSSE} \text{CNNG} \text{4} \text{FSM}^{\text{2}} \\
\hline\n\text{H} \\
\hline\n\text{RDAHTOBA} \\
\text{MEXAHHKA} \\
\text{MEXAHHKA} \\
\text{HHOMOHEA} \\
\text{FROCH}^{\text{2}} \\
\hline\n\end{array}
$$

Spin-orbit coupling:
$$
\hat{H}_{SOC} = \sum_{i} \zeta \hat{\vec{l}}_{i} \hat{\vec{s}}_{i} \rightarrow \lambda \hat{\vec{L}} \hat{\vec{S}}
$$

SOC parameter $\zeta \sim \left(\frac{Ze^2}{\hbar c}\right)^2 \frac{m_e e^4}{\hbar^2}$ is large for heavy elements;

?

SOC for a single electron:

$$
\vec{j} = \vec{l} + \vec{s}
$$

$$
\hat{H}_{SOC} = \zeta \hat{\vec{l}} \hat{\vec{s}} = \zeta \left(\hat{j}^2 - \hat{l}^2 - \hat{s}^2\right) / 2
$$

 $j = 3/2$

 $j = 5/2$

Fixed *s* and *l*; $j=l+s$ or $j=l-s$

Crystal field splitting (CFS)

Introduction: Various energy scales

?

SOC is large in heavy metals: Au, Ir, …, but they have a larger principle number, *n*

Larger $n \Rightarrow$ stronger hybridization \Rightarrow larger the crystal-field field

Introduction: Various energy scales

• Typically all *4d* and *5d* metals are in the **low spin** state (we first fill t_{2g} states)

Typical parameters for transition metals

see e.g. Abragam and Bleaney "EPR of transition ions",

4 • Since $\Delta_{t_{2g}-e_g} \gg \lambda$ we can restrict ourselves to consideration of **SOC** for the *t*₂*g* states only

Introduction: Spin-orbit coupling (SOC) for t_{2g} - states

$$
\hat{H}_{SOC} = \lambda \hat{\vec{l}} \hat{\vec{s}} = \lambda (\hat{l}_x \hat{s}_x + \hat{l}_y \hat{s}_y + \hat{l}_z \hat{s}_z)
$$

Let's calculate this operator! We know how it works with Y_{l,m_l} and transformation rules:

$$
\hat{l}_{z}Y_{l,m_{l}} = m_{l}Y_{l,m_{l}}
$$
\n
$$
D_{x} = \frac{1}{\sqrt{2}}(Y_{1,-1} - Y_{1,1}) \quad D_{xy} = \frac{i}{\sqrt{2}}(Y_{2,-2} - Y_{2,2})
$$
\n
$$
\hat{l}^{-} = \hat{l}^{x} - i\hat{l}^{y} \quad \hat{l}^{+} = \hat{l}^{x} + i\hat{l}^{y}
$$
\n
$$
D_{y} = \frac{i}{\sqrt{2}}(Y_{1,-1} + Y_{1,1}) \quad D_{yz} = \frac{i}{\sqrt{2}}(Y_{2,1} + Y_{2,-1})
$$
\n
$$
\hat{l}^{\pm}Y_{l,m_{l}} = \sqrt{(l \pm m_{l} + 1)(l \mp m_{l})}Y_{l,m_{l} \pm 1}
$$
\n
$$
D_{z} = Y_{1,0}
$$
\n
$$
D_{xz} - \frac{1}{\sqrt{2}}(Y_{2,-1} - Y_{2,1})
$$
\n
$$
D_{3z^{2}-r^{2}} = Y_{2,0}
$$
\n
$$
D_{xz^{2}-y^{2}} = \frac{1}{\sqrt{2}}(Y_{2,2} + Y_{2,-2})
$$

Finally one obtains

$$
\begin{array}{c|c|c|c} \hat{L}_x D_{xz} = -i D_{xy} & \hat{L}_y D_{xz} = i D_{x^2-y^2} - i \sqrt{3} D_{3z^2-r^2} & \hat{L}_z D_{xz} = i D_{yz} \\ \hat{L}_x D_{yz} = i \sqrt{3} D_{3z^2-r^2} + i D_{x^2-y^2} & \hat{L}_y D_{yz} = i D_{xy} & \hat{L}_z D_{yz} = -i D_{xz} \\ \hat{L}_x D_{xy} = i D_{zx} & \hat{L}_y D_{xy} = -i D_{yz} & \hat{L}_z D_{xy} = -2i D_{x^2-y^2} \\ \hat{L}_x D_{x^2-y^2} = -i D_{yz} & \hat{L}_y D_{x^2-y^2} = -i D_{xz} & \hat{L}_z D_{x^2-y^2} = 2i D_{xy} \\ \hat{L}_x D_{3z^2-r^2} = -i \sqrt{3} D_{yz} & \hat{L}_y D_{3z^2-r^2} = i \sqrt{3} D_{xz} & \hat{L}_z D_{3z^2-r^2} = 0 \end{array}
$$

Spin operators are even simpler (just use s^+ and s^-)…

ENERGY

BBBBBBBBBBBBBBBBBBBBBBBBBBB@ **Introduction:** \bigcap for t_2 – states 0 0 0 00 *I* 0 00 0 BBBBBBBBBBBBBBBBBBBBBBBBBBB@ <u>lo se especiento de la provincia de la provin</u> \bigcap for t_2 \bigcap states 0 0 0 00 *I* 0 00 0 on:
C) for t_{2g} - states duction:
g (SOC) for t_{2g} - states on:
C) for t_{2g} - states Print["Hamiltonian="*,* HSOCptsk//MatrixForm] HSOCptsk//MatrixForm] Spin-orbit coupling (SOC) for t_{2g} - states

6

Introduction: Spin-orbit coupling and crystal-field splitting

*4d***-***5d* **transition metal compounds**

$$
l_{\text{eff}} = -1 \quad j_{\text{eff}} = \{1/2, 3/2\} \tag{7}
$$

Electronic properties: Spin-orbit assisted Mott transition

Mott-Hubbard transition in a nutshell

Spin-orbit assisted Mott insulator oln-orbit assisted $\frac{1}{2}$ \mathbf{t} of \mathbf{t} the contractors remarks \mathbf{t} narrow bands crossing EF are split of the rest of Γ partially filled wide t2^g band [Fig. 1(a)]. An unrealistically hin_orbit assisted l Jiil-VI dil abdibleu to an insulating state as seen from the fact that Sr2RhO4 \mathbf{F}_{α} \mathbf{A} , \mathbf{F}_{α} and \mathbf{F}_{α} is nearly identical to \mathbf{F}_{α} tou insulator **the F** \overline{p} and \overline{p} and \overline{p} and \overline{p} represented by holelike by holes. partially filled wide t2^g band [Fig. 1(a)]. An unrealistically large u Bronne Bronne II
Bronne de Vilhelm insulator [Fig. 1(b)]. However, a reasonable U cannot lead to an insulating state as seen from the fact that Sr2RhO4 $\sqrt{644}$ in and $\sqrt{640}$ tou insulatol nin qubit ground anii-Ohan assisted are split intot2^g and eg orbital states by the crystal field **Tott insulator** ing while Sr2RhO4 ($\frac{1}{\sqrt{3}}$

 \mathbf{F} and \mathbf{F} and \mathbf{F} and \mathbf{F} and \mathbf{F} pill-of dit assisted fyfolt st (2 eV), and (d) LDA þ U. In (c), the left panel shows topology rin orbit ossisted Mott st pin-of dit assisted friott st **Spin-orbit assisted Mott state!**

10 $\rm nCl_2$ Other SO-assisted Mott insulators: α –RuCl₃, Ba₂NaOsO₆, Ca₂MnReO₆ etc.

Spin-orbit assisted Mott state: What exactly SOC does

1. SOC lifts orbital degeneracy / induces additional splitting

 $E_{gap} \approx U - W\sqrt{N}$ *PRB 54, R11026 (1996) U_{c1}* ~ \sqrt{N}

POTERYAEV *et al.* PHYSICAL REVIEW B **78**, 045115 !2008"

and thus stimulates metal-insulator transition

Compare with the crystal-field splitting (Δ) :

Crystal-field splitting () helps Δ **metal-insulator transition!**

1/4-filled two-band Hubbard model on square lattice Poteryaev et al., PRB 78, 045115 (2008)

Spin-orbit assisted Mott state: What exactly SOC does

2. Coulomb correlations effectively increase SOC

Correlation effects make electrons more localized, $\lambda(r) \sim -\frac{1}{r}$ *λ*(*r*) ∼ − $\frac{1}{r}$

$$
Sr2RhO4: \quad \lambda = 0.13 \, eV
$$

\n
$$
\lambda_{eff} = 2.15\lambda = 0.28 \, eV
$$

\n*G. Liu et al., PRL 101, 26408 (2008)*

Variational Monte Carlo (VMC)

- $4.4.$ Variational Wave \mathcal{A} and \mathcal{A} and \mathcal{A} and \mathcal{A} • Two-orbital model (yz/zx)
	- Square lattice
- quater filling $(n = 1)$ $\mathcal{P}(\mathcal{P})$ is a one-body wave function and $\mathcal{P}(\mathcal{P})$ is a one-body wave function and $\mathcal{P}(\mathcal{P})$
- *I* square width $t = dd\pi$ *L* - square width; $t_1 = dd\pi$

K. Kubo J. Phys. Soc. Jpn. 91, 124707 (2022)

r ∂*V* ∂*r* $V \sim -\frac{e^{-r}}{r}$ *r* -0.1 $^{−0.2}$ -0.3 Potential energy

0

Spin-orbit assisted Mott state: Model results 0*.*6 corresponding *Z* of the *dxz* orbital in the case of an infinite tetragonal crystal-field splitting. scenario that pictures Hund's metals as doped Mott insulators at half filling [33–36]. Figure 7 presents the values of *U* where \mathcal{A} Motor occurs. Let \mathcal{A} is first discuss the case without \mathcal{A}

L. Du et al., Eur. Phys. J. B. 86, 94 (2013) al.. Eur. We turn now to the small-*U* regime where the SOC reduces the *T_{righ}l* at all DDD 00 205120 (201 *R. Triebl et al., PRB 98, 205128 (2018)* **Structural properties:** Jahn-Teller effect and Spin-orbit coupling

Jahn-Teller effect in a nutshell

Jahn-Teller effect vs. Spin-orbit coupling general idea (on example of t_{2g}^1 configuration)

The Jahn-Teller effect and Spin-orbit coupling may compete! But… Hund's exchange also compete with spin-orbit coupling!

How to solve the Jahn-Teller problem in practice?

$$
\hat{H} = \hat{H}_{SOC} + \hat{H}_{elast} + \hat{H}_{JT} + \hat{H}_U
$$

⃗*li* ̂*s*⃗ *Spin-orbit coupling ⁱ*

^N̂ *Interaction between electrons*

Elastic term classical vibrations Ĥ

Coupling to lattice static + classics

³ *dynamic+quantum*)

$$
\hat{H}_{SOC} = -\zeta \sum_{i} \hat{\vec{l}}_{i} \hat{\vec{s}}_{i}
$$
\n
$$
\hat{H}_{U} = (U - 3J_{H}) \frac{\hat{N}(\hat{N} - 1)}{2} - 2J_{H} \hat{S}^{2} - \frac{J_{H}}{2} \hat{L}^{2} + \frac{5}{2} \hat{N}
$$
\n
$$
\hat{H}_{elast} = \frac{B}{2} \sum_{j} Q_{j}^{2} \qquad dynamic + quantum: \ \hat{H}_{elast} = \sum_{j} \hbar \omega_{j} (a_{j}^{\dagger} a_{j} + \frac{1}{2})
$$
\n
$$
\hat{H}_{JT}^{t\otimes E} = -g \left(\hat{I}_{x}^{2} - \hat{I}_{y}^{2}\right) Q_{2} - g \left(\hat{I}_{z}^{2} - 2J_{x}^{2}\right) Q_{3}
$$
\n
$$
\hat{H}_{JT}^{t\otimes E} = -\frac{g}{\sqrt{2\hbar \omega}} \left(\hat{I}_{x}^{2} - \hat{I}_{y}^{2}\right) (a_{2} + a_{2}^{\dagger}) - \frac{g}{\sqrt{2\hbar \omega}} \left(\hat{I}_{z}^{2} - 2J_{x}^{2}\right) (a_{3} + a_{3}^{\dagger})
$$

ERT 1

Technique: Exact diagonalization

The Jahn-Teller effect vs. SOC: *d1* suppression of JT distortions Jahn-Teller e↵ect and spin-orbit coupling: friends of foes?

betails: **Details:** Exact diagonalization, *T* ⊗ *e*problem (no dynamic effects)

Examples: problem (no dynamic effects)

Compression is tiny:

Cs₂TaCl₆, Rb₂TaCl₆ etc. $Cs₂TaCl₆, Rb₂TaCl₆ etc.$ Seems undistorted: Ba2NaOsO6, Ba2MgReO6 etc

resolution neutron powder diffraction neutron powder diffraction (NPD) data; we show that the show of the show that Γ \blacksquare the symmetry for both persons in both persons is compatible with \blacksquare the *P2*1/*n* space group; (2) in addition, we report on the magnetism and the magnetic structure from low-tempera- Γ_{α} llow offect ye ΩC_{α} d The Jahn-Teller effect vs. SOC: d^3 $\cos \alpha$ of IT distantions increase of JT distortions

JT effect
$$
(\lambda = 0)
$$

 \mathcal{L} subsequently, there have been explorations on the have been exploratory studies on the studies on the studies on the studies of the studi

ture NPD data, and (3) we describe the results of an Xray absorption spectroscopy investigation, which gives an

insight into the Ir oxidation states.

No Jahn-Teller distortions Jahn-Teller active! No Jahn-Teller distortions and Bastien Dalla Piazza, **No. Iohn Tollor distortions** magnetism and the magnetic structure from $\frac{1}{2}$

Iohn Tollor gatival $varin \mathbf{r}$ reflective.

Spin-orbit coupling induces Jahn-Teller distortions (compression)! R_{\Box} Spin-orbit coupling matters URL http://rspa.royalsocietypublishing.org/cgi/ σ Sr2Mg_{ir}of– σ ing original sites in the spin-orbit coupling induces n-Taller distortions (compression)! portant atomic parameters of the structure references of the structure references \mathbf{r}_i

(*x*, *y*, *z*) sites. The Rietveld analysis resulted in a satisfactory

 $D_{3.4}$ 1.1π $R_8A1_2IrO_{14}$ Sr₂MgIrO₆ Sr₂Ca $Ba_8Al_2IrO_{14}$ S

the Sr2CaIrO6 sample and the presence of oxygen vacancies for \overline{a} at \overline{b} at \overline{b} at \overline{b} at \overline{b} at \overline{b} positions. The remain-

Botany of the Jahn-Teller effect

Magnetic properties: Higher order multipoles

O

Spin-orbit coupling and d^1 configuration

Weak SOC

Orbital moment is quenched, $S = 1/2 \implies M = 2S = 1 \mu_B$ Example: YTiO₃ ($M = 0.84\mu_B$) t_{2g} \equiv \equiv $\sum_{x=0}^{\infty}$ $\sum_{x=0}^{\infty}$ $\sum_{x=0}^{\infty}$

Strong SOC (wrt non-cubic field)

$$
p - t_{2g}
$$
 equivalence: $p \rightarrow t_{2g}$, $l \rightarrow -l_{eff}$
\n $\frac{j_{eff}}{2g}$ $\frac{j_{eff}}{2g}$ $\frac{j_{eff}}{2g}$ $\frac{1}{2g}$ $\frac{1}{2g}$ $\frac{j_{eff}}{2g}$ $\frac{1}{2}$ $\frac{1}{2g}$ $\frac{S = 1/2, l_{eff} = 1 \Longrightarrow M}{N_0 \text{ local magnetic magnetic field}}$

Mag. mom: *M* = 2*S* − *l eff* $M = 0$

No local magnetic moment!

TOM Œ

E la T

Any examples (materials)?

Strong spin-orbit coupling and d^1 -0 za podruženi programa za programa za programa za programa za predstavanja za predstavanja za programa za progr
Dogodki \blacksquare

Sr2VO4: V4+ (3*d*) 1

PRB 89, 020402 (2014)

JUN SUGIYAMA *et al.* PHYSICAL REVIEW B **89**, 020402(R) (2014)

 \overline{Q} lin
?0 0.2 *PRL 103, 067205 (2009) Jackeli and Khaliullin*

A 0.1 A *PRB 84, 212407 (2011)Eremin et al.,*

u $\frac{1}{N}$ K $\frac{1}{N}$ some AFM for $T < 8K$ \overline{a} 0.1 A $\tilde{\mathcal{L}}$ solid line represents a Curie-Weiss fit in the *T* range between 110 *PRB 92, 064408 (2015)*

λ for *3d* ions is typically small…

Fig. 1. (Color of The United States of Galaxie of $\frac{1}{2}$ $M = 0$ ² **What is being ordered if magnetic** $M = 0$? In the reduce to random *H*₂ k_O extends the moment is zero? distribution of internal fields (*H*int). Since the LF clearly decoupled the **What is going on at 100K?** $a_{\text{eff}} = 3/2$ **moment is zero?** *moment* is zero?

 $\overline{}$

A

3-band Hubbard model with 1 electron on the square lattice (= Sr2VO4)

P. Igoshev, V. Irkhin, S.S. arXiv:2406.07386

xz/*yz*

Strong spin-orbit coupling and d^2 appears to be ∼R lnð2Þ per mole, as explicitly shown for times more sensitivity to magnetic α unling and d^2 surements the C₂ instruments the C₂ instruments of The entropy release of the αποτελεί προσελεί με το αναπτύξεται στη αναπτύξεται στη αναπτύξεται στη αναπτύξεται appears to be γr lnd be γr ln times more sensitivity to magnetic Bragg scattering as corbit counling and d^2 sure to a plant of the C

Magnetic properties: Kitaev model and Kitaev materials

Kitaev interaction: all new is well-forgotten old

(simplified) Heisenberg model:

$$
\hat{H} = \sum_{i>j} J_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j
$$

Heisenberg model:

A. 2

$$
\widehat{H} = \sum_{i>j} \widehat{\vec{S}}_i \begin{pmatrix} J^{xx} & J^{xy} & J^{xz} \\ J^{yx} & J^{yy} & J^{yz} \\ J^{zx} & J^{zy} & J^{zz} \end{pmatrix}_{ij} \widehat{\vec{S}}_j
$$

Spin-orbit coupling

- Symmetric anisotropic exchange
- Dzyaloshinskii-Morya
- Single-ion anisotropy

Ising model, Kitaev model …

Alexei Kitaev = Алексей Китаев

$j_{\text{eff}} = 1/2 + \text{Common edge} + \text{ligand hopping}$ **= Kitaev exchange**

G. Jackeli, G. Khaliullin, PRL 102, 17205 (2009)

- **Let** 1) Configuration t_{2}^5 2*g*
	- 2) SOC is strong
	- 3) Common edge geometry
	- 4) Ligand-assisted hoppings **only**

Superexchange:

$$
H = \sum_{i>j} J_{ij} \hat{\vec{S}}_i \hat{\vec{S}}_j
$$

$$
\frac{y}{\sqrt{\frac{y}{x}}}
$$

$$
|j_{1/2}^{z}\rangle = -\frac{1}{\sqrt{3}} (|xy \uparrow \rangle + i|xz \downarrow \rangle + |yz \downarrow \rangle)
$$

$$
|j_{-1/2}^{z}\rangle = \frac{1}{\sqrt{3}} (|xy \downarrow \rangle + i|xz \uparrow \rangle - |yz \uparrow \rangle)
$$

but
$$
\langle j_{1/2,a}^z | \hat{t} | j_{-1/2,b}^z \rangle = \langle j_{-1/2,a}^z | \hat{t} | j_{1/2,b}^z \rangle = 0
$$

 $\langle j_{1/2,a}^z | \hat{t} | j_{1/2,b}^z \rangle = \frac{1}{3} (i \langle yz \downarrow_a | \hat{t} | xz \downarrow_b \rangle - i \langle xz \downarrow_a | \hat{t} | yz \downarrow_b \rangle = (it - it) = 0$

NO conventional AFM superexchange!

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

U

$j_{\text{eff}} = 1/2 + \text{Common edge} + \text{ligand hopping}$ **= Kitaev exchange**

G. Jackeli, G. Khaliullin, PRL 102, 17205 (2009)

- **Let** 1) Configuration t_{2}^5 2*g*
	- 2) SOC is strong
	- 3) Common edge geometry
	- 4) Ligand-assisted hoppings **only**

Analogue of

Exchange between f -filled/empty orbitals $K \sim -\frac{t_{dd}^2 J_H}{U^2} < 0$ half-filled/empty orbitals

$$
K \sim -\frac{\tilde{t}_{dd}^2 J_H}{U^2} < 0
$$

$$
\hat{H}_{ij} = K J_i^z J_j^z \rightarrow J S_i^z S_j^z
$$

z ⊥ basal plane

29 **Bond-depended anisotropic exchange**

Possible candidates for Kitaev physics (1st generation)

- 1) Configuration *t* 5 2*g*
- Common edge geometry
- $2)$ Common edge geometry
3) Ligand-assisted hoppings $N a_2 I r O_3$
- SOC is strong

G. Jackeli, G. Khaliullin, PRL 102, 17205 (2009)

Kitaev model
$$
\hat{H} = -\sum_{\langle ij \rangle_{\gamma}} K_{\gamma} \hat{S}_{i}^{\gamma} \hat{S}_{j}^{\gamma}
$$

Kitaev Ann. Phys. 321, 2 (2006)

- **• Exactly solvable**
- **• Highly frustrated model**
- Quantum spin-liquid (based on Ising model)
- t is discussed in section II \mathbb{R}^3 . ences.
The first terial is discussed in section II B. See text for relevant reference in section III B. See text for relevant re ence en terial is discussed in section III B. See text for relevant references in section III B. See text for relevant references in the section in the section in the section of the section in the section in the section in the sec vi) and **• Fractionalized excitations (Majoranas)**

Kitaev materials (1st generation): Na_2IrO_3 α -Li₂IrO₃ Li_2RhO_3 α -RuCl₃ $Na₂IrO₃$ α -Li₂IrO₃ Li₂RhO₃ α -RuCl₃

Sy

Kitaev model: classical variant **1210007**

)— , .

$$
H = -K_x \sum_{x \text{ bonds}} S_i^x S_j^x - K_y \sum_{y \text{ bonds}}
$$

Features of classical Kitaev model \sim

 $S_i^y S_j^y - K_z \sum S_i^z S_j^z = - \sum K_y S_i^y S_j^y$

z bonds

- Spins are strongly frustrated
	- Spins can't order even at $T = 0$

 1982 *i. Anpeab* T *on 136, 66tn.* 4

 $\boldsymbol{y}\boldsymbol{C}\boldsymbol{\varPi}\boldsymbol{E}\boldsymbol{X}\boldsymbol{H}\boldsymbol{\varPhi}\boldsymbol{H}\boldsymbol{3}\boldsymbol{H}\boldsymbol{q}\boldsymbol{E}\boldsymbol{C}\boldsymbol{K}\boldsymbol{H}\boldsymbol{X}\boldsymbol{H}\boldsymbol{A}\boldsymbol{y}\boldsymbol{K}$

: , ЭФФЕКТ ЯНА—ТЕЛ
пері . . 4,

⟨*ij*⟩*^γ*

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

 $\sum K_{\nu} S_{i}^{\gamma} S_{i}^{\gamma}$

К. И. Кугель, Д. И. Хомский

$$
H = J\left(\sum_{\langle \mathbf{i},\mathbf{j}\rangle} \tau_{\mathbf{i}}^x \tau_{\mathbf{j}}^x + \sum_{\langle \mathbf{i},\mathbf{j}\rangle} \tau_{\mathbf{i}}^y \tau_{\mathbf{j}}^y + \sum_{\langle \mathbf{i},\mathbf{i}\rangle} \tau_{\mathbf{i}}^z \tau_{\mathbf{j}}^z\right),\tag{34}
$$

31 т пары *i*, *j*, расположенные по осям x, y) (633). (6 $\langle i,\ j\rangle_{x,\ y,\ z}$ обозначает пары $i,\ j,\ \text{pa$ **сположенные по осям** $$x,$$

Majorana fermions

(Dirac) fermions: *a* and *a*†

Majorana fermions (majoranas) $c_1 = a + a^{\dagger}$ $c_2 = -i(a - a^{\dagger})$ $c_1^{\dagger} = a^{\dagger} + a = c_1$ $c_2^{\dagger} = i (a - a^{\dagger})$ $i^{\dagger} = -i(a - a^{\dagger}) = c_2$ Majoranas: (1) $c^{\dagger} = c$ (3) $c_j c_i = -c_i c_j$ if $j \neq i$ (2) $c^2 = 1$

Spins can be expressed be expressed via two (conventional) fermions: *a*, *a*†

$$
\hat{\vec{S}}_i = \frac{1}{2} \sum_{\sigma \sigma'} a_{i\sigma}^{\dagger} \vec{\sigma}_{\sigma \sigma'} a_{i\sigma'}, \quad \sigma, \sigma' = \uparrow, \downarrow
$$

W=±1 *W*=±1

Spin operators can be expressed* via four majoranas: b^x , b^y , b^z , c

32 *commutation relations for Pauli matrixes are conserved, if additional constraint $b^xb^yb^zc=1$ is applied 32 dependent Ising interactions. The green bonds have easy axes parallel to the *x* axis,

Kitaev model: quantum case 3 5

Kitaev model via majoranas $\hat{H} = -\sum$

Quadratic form! Readily diagonizable if u_{ij}^{γ} were numbers. Non-interacting c majoranas?

 $K_{\gamma}b_i^{\gamma}b_j^{\gamma}c_ic_j$

 μ_{ij}

(*y*)

Sz $W_i U_i U_i = \frac{1}{4} L$

=

1

 $\frac{1}{4}$ $\frac{1}{4}$ ⟨*ij*⟩*^γ*

 $K_{\gamma} u_{ij}^{\gamma} c_i c_j$

6

$$
u_{ij}^{\gamma}u_{ij}^{\gamma} = b_i^{\gamma}b_j^{\gamma}b_i^{\gamma}b_j^{\gamma} = -1 \Rightarrow u_{ij}^{\gamma} = \pm i
$$

⟨*ij*⟩*^γ*

 $K_{\gamma}\hat{S}_{i}^{\gamma}$

i Ŝ *γ* $\frac{\gamma}{j} =$ 1

 $\frac{1}{4}$ $\frac{2}{111}$ ⟨*ij*⟩*^γ*

Flux: $W_p = u_{12}u_{23}u_{34}u_{45}u_{56}u_{61} = \pm 1$

the ground state corresponds to $W_p = +1$ on all hexagons One can show numerically or analytically that

conservation but control the sign of hopping of the

Possible candidates for Kitaev physics *(1st generation)* schemes to parametrize this tensor, which are appropricondition increases the sensitivity of *ab-initio* estimates ssible candidates for Kita experiments 161 on $(1^{31}$ generation) of the interactions to methodological details. As *L* Ossibic Candidates for ϵ and ϵ analyzed in terms of ϵ and ϵ analyzed in terms of ϵ a Heisenberg-Kitaev model with *K*¹ *>* 0 and *J*¹ *<* 0, as condition increases the sensitivity of *ab-initio* estimates of the interactions to methodological details. \overline{A} s with \overline{A} experiments (1) $(1st$ conoration (e.g. *for dominant* k *x* + *Ky*) into a gapped spin line k $T_{\rm tot}$ is a $T_{\rm tot}$ generation ($T_{\rm tot}$ generation)

 \setminus

 \overline{K} itoov \overline{H} **Kitaev-Heisenberg model** ruclasses in this sense, in the specific magnetic m **Kitaev-Heisenberg model** RuCl3. In this sense, identifying the specific magnetic iaev-Heisenberg model

a Heisenberg-Kitaev model with *K*¹ *>* 0 and *J*¹ *<* 0, as required to stabilize zigzag order in the absence of other

 \mathcal{L}_{max} . However, such a combination of interactions is interactions in

$$
\mathcal{H}_{ij} = J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + K_{ij} S_i^{\gamma} S_j^{\gamma} + \Gamma_{ij} \left(S_i^{\alpha} S_j^{\beta} + S_i^{\beta} S_j^{\alpha} \right)
$$
\n
$$
+ \Gamma'_{ij} \left(S_i^{\gamma} S_j^{\alpha} + S_i^{\gamma} S_j^{\beta} + S_i^{\alpha} S_j^{\gamma} + S_i^{\beta} S_j^{\gamma} \right)
$$
\n
$$
\phi = \pi
$$
\n
$$
(J_1 < 0)
$$
\n
$$
\sum_{\text{FM of } \text{N}} \sum_{\text{P} \in \text{N}} \sum_{\text{N}} \
$$

 $X_{\rm eff}$, and $Z_{\rm eff}$ and $Z_{\rm eff}$ and $Z_{\rm eff}$ symmetry. For lower symmetry, respectively. For lower symmetry

 δ – δ

 $rac{1}{\sqrt{2\pi}}$

the availability of the updated *C*2*/m* or *R*¯3 structures,

 $u_{\rm{max}}$

ate for di↵erent local symmetries. Assuming local *C*2*^h* symmetry of the *ij*-bond, the convention is to write the

into α into α and α and α and α are level for α α

required to stabilize zigzag order in the absence of other $t_{\rm eff}$ the combination of interaction of interactions is interactions in the combination of interactions is in impossible from a microscopic perspective; and an antiferro- $\mathsf{Spin}(n)$

into question the relevance of the Kitaev model for ↵-

local environments, further terms may also be required by \mathcal{L}_max Experimental results. For example, and interactions. For example, and interactions. For example, and interactions. For example, and in the set of of a small parameter, i.e. when \mathbf{I} **Experimental results** and **above in section III B. See text for relevant relevant relevant references** ences. all *ab-initio* estimates have been in line with the origi- \blacksquare xperimental results ing to *K*¹ *>* 0, and *|*1*|* ⇠ *|J*1*|* ⇠ *|K*1*|*. However, since **Experimental results** condition increases the sensitivity of *ab-initio* estimates

 \sim \sim \sim

Possible candidates for Kitaev physics where $(1st generation)$ schemes to parametrize this tensor, which are appropricondition increases the sensitivity of *ab-initio* estimates ssible candidates for Kita $(1st$ conoration experiments 161 on $(1^{31}$ generation) of the interactions to methodological details. As *L* Ossibic Candidates for ϵ and ϵ analyzed in terms of ϵ and ϵ analyzed in terms of ϵ a Heisenberg-Kitaev model with *K*¹ *>* 0 and *J*¹ *<* 0, as condition increases the sensitivity of *ab-initio* estimates of the interactions to methodological details. \overline{A} s with \overline{A} experiments (1)

Kitaev-Heisenberg model <u>interactions in the sense, in the specific magnetic magneti</u> **Kitaev-Heisenberg model**

a Heisenberg-Kitaev model with *K*¹ *>* 0 and *J*¹ *<* 0, as required to stabilize zigzag order in the absence of other

 \mathcal{L}_{max} . However, such a combination of interactions is interactions in

$$
\mathcal{H}_{ij}=J_{ij}\;\mathbf{S}_{i}\cdot\mathbf{S}_{j}+K_{ij}\;S_{i}^{\gamma}S_{j}^{\gamma}+\Gamma_{ij}\left(S_{i}^{\alpha}S_{j}^{\beta}+S_{i}^{\beta}S_{j}^{\alpha}\right)\\+\Gamma_{ij}^{\prime}\left(S_{i}^{\gamma}S_{j}^{\alpha}+S_{i}^{\gamma}S_{j}^{\beta}+S_{i}^{\alpha}S_{j}^{\gamma}+S_{i}^{\beta}S_{j}^{\gamma}\right)\\ \phi=\pi\left(\left(\begin{array}{c}120^{3}\\\text{Zigzag}\end{array}\right)\right)
$$

into question the relevance of the Kitaev model for ↵-

$$
H_{ij} = \vec{S}_i \begin{pmatrix} x & y & z \\ J & \Gamma & \Gamma' \\ \Gamma & J & \Gamma' \\ \Gamma' & \Gamma' & J + K \end{pmatrix}_{ij} \vec{S}_j
$$

 α -RuCl₃ $T_N = 7K$ $= 0.5$ and $= 0$ α -Ru \mathcal{C} ₁₁ \mathcal{C} ₁₂ $T_{\alpha} = 7K$ $\frac{1}{2}$ functional theory total energy, $\frac{1}{2}$ α -Ru \mathcal{C} ₂ $T_{\alpha} = 7K$ $\frac{1}{s}$ functional theory total energy, $\frac{1}{s}$ α -Ru \mathcal{C} ₁₁ \mathcal{C} ₁₂ $T_{\alpha} = 7K$ $\frac{1}{\sqrt{N}}$ functional theory total energy, $\frac{1}{\sqrt{N}}$ from various methods. For Ref. 146, the two numbers represent the range of values found in various relaxed structures. "Pert. α -Ru \mathcal{C} ₁₁ \mathcal{C} ₁₂ $T_{\alpha} = 7K$ $\frac{1}{2}$ density functional theory total energy, $\frac{1}{2}$ from various methods. For Ref. 146, the two numbers represent the range of values found in various relaxed structures. "Pert. α -Ru \mathcal{C} ₁₁ \mathcal{C} ₁₂ $T_{\alpha} = 7K$ $\frac{1}{2}$ density functional $\frac{1}{2}$ $\frac{1}{2}$ $\Omega_{\text{in}}\Omega_{\text{in}}\Omega=\frac{1}{2}V$ α -ita α ₁₃ $I_N - I_N$ $\sum_{N}=7K$ Γ_{LZ} the T_{LZ} is the Hamiltonian. The Hamiltonian supplementary supp α -RuCl₃ $T_N = 'K$ nal Jackeli-Khaliullin mechanism.39,43,146,175 That is, *K*¹

 $\frac{1}{2}$ is resented $\frac{1}{2}$ (isotropic exenting with for \mathbf{H} signals we \mathbf{I} (i.e. the ziggag order at roughly \mathbf{H}) $\frac{1}{\sqrt{1-\frac{1$ $\frac{1}{\sqrt{3}}$

Field-induced phenomena in *α***-RuCl3**

AFM is destroyed by in-plane magnetic field

J. Zheng et al., PRL 119, 227208 (2017)

S.-H. Baek et al., PRL 119, 37201 (2017)

A. Banerjee et al., NPJ Quantum Mater. 3, 8 (2018)

and n

Two possible scenarios: highly debated; no final answer yet

2nd generation of Kitaev materials defeat of happy *a b c*

a b

α-Li $\overline{}$

α-Li \bullet

b d'an deus de la commune de la commune

NO magnetic order due to H zero-point motion order due to H zero-point motion
(**dynamic disorder**) *Y. Li et al., PRL 121, 247202 (2018)* ar, and a honey combined a horizon and the spin motion \mathbb{R}^2 such that $\begin{array}{ccc} \mathbf{b} & \mathbf{c} & \mathbf$ dependent ferromagnetic Ising interactions. The three 120° bonds with interactions. The three 120° bonds with α \sim thogonal Ising axes compete with each other, giving rise to strong **Figure 1** | **Crystal structure and basic physical properties of H3LiIr2O6.** <u>aetic</u> order due to H zero-point motion $(d_{\text{vnomio}} \text{ diagonal on})$ α and α is interactions. The three 120° bonds with α

Li et al., PRL 121, 24/202 (2018) magnetic defects (see also Extended Data Fig. 2a). The inset shows 1/*χ* assumptions in section 1/*χ* $V_I: I = I$ (dotted 101 0.47000 (0.010) l. Li et al., PRL 121, 24/202 (2010)

a b

a b c

 \sim

H₃Linux

i.

PRL 123, 237203 (2019), PRB 103, 094427 (2021), JAC the presumed intrinsic *χ* (dotted line) after numerically subtracting a 20.200 139 15371 (2017) is observed. The sizable anisotropy is observed. The sizable anisotropy is 37 $\frac{1}{\sqrt{2}}$ $\begin{bmatrix} 2 & 227202 & 0.010 \\ 0 & 0 & 0 & 0 \end{bmatrix}$ magnetization magnetiza 3, 23/203 (2019), I KD 103, 09442/ (2021*)*, *JA* P_{C} (120 15271 (2017) provided and note that $\sqrt{27}$ *PRL 123, 237203 (2019), PRB 103, 094427 (2021), JACS 139, 15371 (2017)* 37

What about other geometries? or anisotropic exchange on FCC lattice

What about other geometries? or anisotropic exchange on FCC lattice

FIG. 4. (a) Phase diagram for the *J*1-*J*2-*K* model. We find ordered phases with ordering wave vectors (100) (red), (¹ **Other materials with Kitaev-like interactions**: Ba₃IrTi₂O₉ (triangular), Na₄Ir₃O₈ (hyperkagome)

Kitaev materials, 3rd generation: Cobaltites ???

Kitaev materials, 3rd generation: Cobaltites ???

Strong anisotropy of exchange interaction or *jeff=1/2* physics is important not only for t_{2g}^5 configuration (i.e. Ru^{3+} , Ir⁴⁺ ions) and honeycomb geometry

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Spin liquid in BaCo₂(AsO₄)₂?

BaCo₂(AsO₄)₂ is not Kitaev material (but still extremely interesting) *Trigonal elongation*: For ² *>* 0, we find that *K* is less θ and θ ¹. In the sign(*J*₎. Interactions θ *Jxy*. In the hypothetical ligand-assisted hopping region, we find that *Jz* may be almost completely suppressed In this work, we have considered the magnetic cou-plings in edge-sharing *d*⁷ compounds. On this basis, we \mathbb{C}_{α} ($\Lambda_{\alpha} \Omega$) is not V^{\prime} to an material CO2(ASO4*)*2 is <mark>Hot K</mark>itaev IIIatei exchange was chosen to be *JH*=0.9 eV. Conventional DFT calculations underestimate the e↵ect of strong Coulomb correlations, which must be taken into $\mathbf{D}_e \mathbf{C}_e$ $(\Lambda_e \mathbf{Q})$ is not V^2 to prove on-bac 02(ASO4*)*2 is not mitaev h energy in the extended Kitaev model. In t exchange was chosen to be **JH** conventional DFT calculations under of strong Coulomb correlations, which must be taken into σ account for extraction in the extraction in σ

U= 5 eV *U*= 6 eV *U*= 7 eV

J Γ Γ′

Γ *J* Γ′

Γ′ Γ′ *J* + *K ij*

Exchange DFT+U+SOC Why there is no extensor $(U=6 \text{ eV})$ *U*= 5 eV *U*= 6 eV *U*= 7 eV

 K_1 2.2 K K_3

 $\begin{array}{ccc} \n\mathbf{K} & \Gamma_1 & -1.7 \text{ K} & \Gamma_3 \n\end{array}$

 γ ij Γ'_1 4.0 K Γ'_3

Table I: The dependence of exchange interaction parameters \mathcal{L} on on-site Coulomb *U* computed from DFT+SOC+U total

 Γ_1'

 K_1 2.2 K K_3

Table I: The dependence of exchange interaction parameters in the dependence of exchange interaction parameters

U= 5 eV *U*= 6 eV *U*= 7 eV

 J_3 24.6 K

 $0.2 K$

 Γ'_{3}

\cdots \cdots Why there is no a snin lio $\mathbf C$ Why there is no a spin liquid of strong Coulomb correlations, which must be taken into of strong Coulomb correlations, which must be taken into $\frac{1}{2}$ n_{initial}

 \overline{f} = 0.05 \overline{f} = 0.05 meV for significant transmitted transmitted to \overline{f} = 0.05 meV for significant transmitted transmitted to \overline{f} = 0.05 meV for significant transmitted transmitted to \overline{f} = 0.05 tion of = */*2. . As a result, we expect such materials

ically relevant region of large direct hopping (*t*³ *t*2), $\frac{1}{2}$ $\frac{1}{2}$ induced. The interpreted interpreted interpreted in the more equal to Γ 1. Isotropic exchange (J_1) much larger than Kitaev (K_1) J_1 -40.9 K J_3 24.6 K $^{-1}$ er than Kitaev (K_1) J_3 24.6 K 1.1 Solid processes K_3 0.2 K \blacksquare **much larger the** $\overline{1}$ $\overline{1}$

the region of large ligand-assisted hopping. For the phys-

eV, which is compatible with the *ab-initio* estimates. In Fig. 7(erv. efficient very efficient Γ_1 -1.7 K Γ_3 -6.0 K I_1' 4.0 K Γ_3' -2.3 K 2.3 K $\mathbf{V}^{\mathbf{3}}$ PAVEL A. MAKSIMOV *et al.* PHYSICAL REVIEW B **106**, 165131 (2022) PAVEL A. MAKSIMOV *et al.* PHYSICAL REVIEW B **106**, 165131 (2022) $\left(\begin{array}{ccc} \n\begin{array}{ccc} \n\text{P}_1 & \text{P}_2 & \text{P}_1 & \text{P}_2 & \text{P}_3 & \text{P}_2 & \text{P}_3 & \text{P}_4 & \text{P}_5 \n\end{array} & \begin{array}{ccc} \n\text{P}_2 & \text{P}_3 & \text{P}_2 & \text{P}_3 & \text{P}_4 & \text{P}_5 & \text{P}_6 & \text{P}_6 & \text{P}_7 & \text$

This is the only major that the only major so there are no additional terms to compete, and a rela-Conventional DFT calculations under the electromagnetic structure the electromagnetic structure the electromagnetic structure the e
Conventions under the e⊿ectromagnetic structure the electromagnetic structure that the e⊿e

approaches to estimate the magnetic exchange couplings.

K3 K3

magnetic interactions based on \mathbb{R}^n

magnetic interactions based on DFT(GGA)+U+SOC

netic configurations.

calculation of total energies of four non-collinear mag-calculation of total energies of four non-collinear mag-

approaches to estimate the magnetic exchange couplings.

P. Maksimov et al, PRB 106, PRR 106 with Slater parameters *F*4*/F*² = 0*.*625, and *F*⁰ ⌘ *U* and *F*² ⌘ 14*J/*(1+0*.*625) set according to *U* = 5 to 7 eV, and *P. Maksimov et al, PRB 106, F*₂ (2022) *F*² ⌘ 14*J/*(1+0*.*625) set according to *U* = 5 to 7 eV, and

Co-based pyroxenes ABCo2O6: novel (1D?) materials with strong Kitaev

Table 1. Comparison of the exchanges from ab initio (GGA+U+SOC) Kitaev **calculations and neutron scattering fit (LSWT).** Method and notified bottomy in (2011)

data. This characteristic is reminiscent of the behavior observed in other honeycomb lattice magnets with zigzag AFM order [43, 44]. The exchanges are in units of meV. *J*^{*x*} *J*^{*zz} <i>J*^{*zz*} *J*^{*zz*} *J</sup>*

Kitaev is large, $K/|J| = 0.96!$ **The system is not 1D!**

What about ACoSi2O6?

, *n* answe *Maksimov et al., arXiv:2401.13550*

Take-home messages

- Mott-Hubbard transition is affected by
- **E** spin-orbit coupling (U_c is typically decreased)

S

- Spin-orbit coupling strongly affects the Jahn-Teller effect (result depends on elec. number)
- There can hidden (magnetic) orders in spin-orbit materials **M**
	- Kitaey materials: Iridates/ruthenates: Kitaev can be large; Field-dependence? Cobaltites no solid evidence for Kitaev physics yet

D. Khomskii, S. Streltsov Chem. Rev. 121, 2992 (2021) T. Takayama et al., JPSJ 90, 062001 (2021)

