Exotic magnetism in spin-orbit Mott insulators

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- Orbital chemistry
- J=3/2 octupolar order
- J=1/2 magnetism & doping
- J=0 excitonic magnets, etc...
TRANSITION METAL COMPOUNDS

d-electron
f-spins

\( U \gg W \)

localized

p-bands

\( U \ll W \)

\( U \sim W \)

"in-between"

d-electron

1Å
$f$-spins

$d$-electron

$U \sim W$

local moments

bands and chemical bonds

$p$-bands
**Goodenough-Kanamori rules**

**bonding orbitals (hopping $t$):**

*strong AF exchange*  \( J_{AF} = 4t^2/U \)

**non-bonding orbitals (no hopping):**

*weak FM exchange*  \( J_{FM} = -\frac{J_H}{U} \frac{J_{AF}}{U} \)
**Goodenough-Kanamori rules**

- **bonding orbitals (hopping $t$):**
  - strong AF exchange $J_{AF} = 4t^2/U$

- **non-bonding orbitals (no hopping):**
  - weak FM exchange

$$J_{FM} = -\frac{J_H}{U} J_{AF}$$
Lattice geometry dictates orbital overlap & magnetism

180° bonding

\[ \text{Sr}_2\text{CuO}_3 \]

\[ t \neq 0 \]

bonding orbitals

\[ J = 250 \text{ meV} \]

90° bonding

\[ \text{Li}_2\text{CuO}_2 \]

\[ t = 0 \]

non-bonding orbitals

\[ J \sim 0 \]
Lattice geometry dictates orbital overlap & magnetism

180° bonding

$\uparrow \neq 0$

bonding orbitals

$J = 100 \text{ meV}$

$\text{Sr}_2\text{IrO}_4$

90° bonding

$\uparrow = 0$

non-bonding orbitals

$J \approx 0$

$\text{Na}_2\text{IrO}_3$
**d-orbitals**: \( t_{2g} \) versus \( e_g \)

**L-moment** "quenched"

**orbital magnetism** \( L=1 \)

\[
d_1 = z(x + iy)
\]
ORBITAL MAGNETISM

Orbital interactions:

- **bond-dependent**
- **non-Heisenberg**

simple cubic lattice:

\[
\mathcal{H}_{ji}^{(c)} = \left(L_x^i L_x^j\right)^2 + \left(L_y^i L_y^j\right)^2 + L_x^i L_y^i L_y^j L_x^j + L_y^i L_y^i L_x^j L_y^j
\]

non-coplanar  multi-Q  GKh & Okamoto  (2002)
ORBITAL MAGNETISM

Orbital interactions:
- bond-dependent
- non-Heisenberg

Spin-orbit coupling

Orbital $L$ frustration $\Leftrightarrow$ Pseudospin $J$ frustration

Kitaev model

Jackeli & GKh (2009)
Spin-orbit multiplets of TM-ions

A Abragam & Bleaney (1970)

\[ \tilde{J} = \frac{1}{2}, \quad \tilde{l} = 1, \quad \tilde{S} = \frac{1}{2} \]
\[ \tilde{J} = 0, \quad \tilde{l} = 1, \quad \tilde{S} = 1 \]
\[ \tilde{J} = 2, \quad \tilde{l} = 1, \quad \tilde{S} = 1 \]
\[ \tilde{J} = \frac{3}{2}, \quad \tilde{l} = 1, \quad \tilde{S} = \frac{1}{2} \]

\( d^1 \) V, Nb, Mo
\( d^2 \) Mo, Re, Os
\( d^4 \) Ru, Re, Os
\( d^5 \) Co, Rh, Ir

\( g = 0 \quad g = 1/2 \quad g = 0 \quad g = -2 \)

spin - orbit Mott insulators
Spin-orbit multiplets of TM-ions

Aragam & Bleaney (1970)
Phase transition of unknown origin in Sr$_2$VO$_4$

Zhou et al., PRL 2007

Both lattice and magnetism involved

No magnetic Bragg peaks have been detected
Perovskite $\text{Sr}_2\text{VO}_4$

$\text{V}^{4+}$

$S=1/2$, $L=1$

$J=3/2$, $g=0$
Magnetically hidden order in $\text{Sr}_2\text{VO}_4$

Jackeli & GKh (2009)

Condensate wave-function

$$|+1, \uparrow\rangle + |-1, \downarrow\rangle$$

$$L^2 \quad S^2$$

$$\langle \hat{S}_i \rangle = \langle \hat{l}_i \rangle = 0 \quad \rightarrow \quad \text{no magnetic moment}$$

Order parameter: octupolar moment

$$S^{x(y)}[(l^x)^2 - (l^y)^2]$$
Elementary Excitations

- Octupolar Bragg peak
- Magnon
- Continuum, $M^x$

X-rays vs. Neutrons
Spin-orbit multiplets of TM-ions

Abragam & Bleaney (1970)

\[ \begin{align*}
  & l = 1, S = \frac{1}{2} \\
  & \tilde{J} = \frac{1}{2}, 3 \frac{\zeta}{2} \\
  & \tilde{J} = \frac{3}{2} \\

  & l = 1, S = 1 \\
  & \tilde{J} = 0, \frac{\zeta}{2} \\
  & \tilde{J} = 1, 2 \\

  & l = 1, S = 1 \\
  & \tilde{J} = 0, \frac{\zeta}{2} \\
  & \tilde{J} = 1, 2 \\

  & l = 1, S = \frac{1}{2} \\
  & \tilde{J} = 0, 3 \frac{\zeta}{2} \\
  & \tilde{J} = \frac{1}{2} \\
\end{align*} \]
Pseudospin 1/2 exchange interactions

$J=1/2$ wavefunction

\[ | \uparrow \rangle = |xy \uparrow \rangle + |yz \downarrow \rangle + i |zx \downarrow \rangle \]

$\tau \neq 0$

bonding orbitals

$\tau = 0$

non-bonding orbitals
Perovskite $\text{Sr}_2\text{IrO}_4$

Two hopping paths A & B: positive interference

Path A: $\textcolor{red}{\text{t/3}}$

Path B: $\textcolor{green}{\text{t/3}}$

$A + B = (2/3) \ t$

Net hopping: large, spin-isotropic

$J (S_i S_j)$

Jackeli & GKh (2009)
Spin-waves: *iridates vs cuprates*

**Sr$_2$IrO$_4$**

\[ T_N \approx 240 \text{ K} \]

*J. Kim et al. (2012)*

**La$_2$CuO$_4$**

\[ T_N \approx 320 \text{ K} \]

*Coldea et al. (2001)*
The key elements in high-$T_c$ cuprates

- quantum spins $1/2$
- single orbital
- strong AF
- two D

*Pseudospin one-half $Sr_2IrO_4$ possesses all these ingredients...*
T-dependent pseudogap in Sr$_2$IrO$_4$

„Fermi-arcs“ at low doping

„normal“ FS

Pseudogap opens at low T

...and closes at 110 K

B.J. Kim et al. (Science 2014)
Two compounds with similar magnetism, fermiology, lattice

\[ \text{Sr}_2\text{IrO}_4 \quad \text{La}_2\text{CuO}_4 \]

Pseudogap: common to both

? superconductivity?
Edge-sharing octahedra

Two hopping paths A & B: *negative interference*

\[ A = i\sigma \frac{t}{3} \quad \text{B} = -i\sigma \frac{t}{3} \]

\[ A + B = 0 \]
no hopping

non-bonding, no exchange
Edge-sharing octahedra

Two hopping paths A & B: negative interference

\[ A = i\sigma \frac{t}{3} \]

\[ B = -i\sigma \frac{t}{3} \]

\[ A + B = 0 \]

non-bonding orbitals

\( \text{weak Ising} = -\left( \frac{J_H}{U} J_{AF} \right) S^z S^z \)

Jackeli, GKh (2009)
$t_{2g}$-hopping: 180° versus 90° bonding

orbital conserved

$\Delta M_L = 0$

isotropic exchange

orbital unconserved

$\Delta M_L = \pm 2$

anisotropic exchange

$$t(d_{1,\sigma}^\dagger d_{1,\sigma} + d_{-1,\sigma}^\dagger d_{-1,\sigma})_{ij}$$

$$it(d_{1,\sigma}^\dagger d_{-1,\sigma} - d_{-1,\sigma}^\dagger d_{1,\sigma})_{ij}$$
Honeycomb lattice $\Rightarrow$ Kitaev model

$\Delta M = \pm 2$

no spin-flip possible

\[ Ising = -\left( \frac{J_H}{U} J_{AF} \right) S^z S^z \]

Jackeli, GKh (2009)
The Kitaev model

- Exactly solvable, QSL
- Free Majorana fermions
- Relevant for quantum computing

\[ S_\alpha = c f_\alpha \]

local

Itinerant

\[ E_F \]

Dirac cones
- interactions other than Kitaev one…
- spin-lattice coupling…
- dirt (nobody is perfect)…
Exotic magnetism $\rightarrow$ exotic pairing
Superconductivity in two-dimensional CoO$_2$ layers

Kazunori Takada*, Hiroya Sakurai†, Eiji Takayama-Muromachi†
Fujio Izumi*, Ruben A. Dilanian* & Takayoshi Sasaki*†


+ water

edge-shared octahedra

SC below 5 K

RVB-theories: chiral d+id pairing, orbital currents
Spin-orbit coupling $\Delta (T/5) \sim 80 \text{ meV for } \text{Co}^{2+}$

$|\uparrow\rangle = \alpha |E_g, \uparrow\rangle + \beta |A_g, \downarrow\rangle$

Pseudospin, mixed spin/orbital particle
Gutzwiller bands with SOC

$J=1/2$ band

$J=3/2$ bands

Fermi level

$GKh$, Koshibae, Maekawa (2004)
Pseudospins $J=1/2$ on a triangular lattice

Hamiltonian = Heisenberg + Ising$^{(x,y,z)}$

$H_J(ij) = -J_f \kappa_s s^\dagger_{ij} s_{ij} - J_f \kappa_t T^\dagger_{ij} T_{ij}$

singlet  triplet

GKh, Koshiba, Maekawa (2004)
GKh (2005)
S=1/2 versus J=1/2 pairing

Na$_{1-x}$CoO$_2$

Spin-singlet:

| $\uparrow \downarrow - \downarrow \uparrow$ |

chiral d+id

Baskaran (2003)
Kumar, Shastry (2003)
Ogata (2003)
P. Lee et al. (2004)

Pseudospin-triplet:

$\Delta_{tr} = \alpha_0 \langle \tilde{r}^i + i \tilde{r}^j \rangle_j + \alpha_i e^{i\phi_j} \langle \tilde{r}^i \rangle_j + \alpha_i e^{-i\phi_j} \langle \tilde{r}^j \rangle_i$

$d$ -vector pattern on FS:

$p + f$ symmetry

GKh, Koshibae, Maekawa (2004)
GKh (2005)
Competition between $d$-wave and topological $p$-wave superconducting phases in the doped Kitaev-Heisenberg model

Timo Hyart, Anthony R. Wright, Giniyat Khaliullin, and Bernd Rosenow

Doping a spin-orbit Mott insulator: Topological superconductivity from the Kitaev-Heisenberg model and possible application to (Na$_2$/Li$_2$)IrO$_3$

Yi-Zhuang You, Itamar Kimchi, and Ashvin Vishwanath
Spin-orbit multiplets of TM-ions

Abragam & Bleaney (1970)

$d^1$  

$d^2$  

$d^4$ Ru, Re, Os  

$d^5$

$L \leftrightarrow S$
$J=0$ Mott insulators

Interacting singlet-triplet models
Why bother about $J=0$?

A) Compounds of $\text{Re}^{3+}, \text{Ru}^{4+}, \text{Os}^{4+}, \text{Ir}^{5+}$

B) Magnetic QCP is granted

C) Orbital frustration near QCP
   - exotic condensates?
Singlet-triplet model, 180° bonding

\[ H = \lambda \sum_i n_i + J \sum_{ij} [T_i^\dagger \cdot T_j - \frac{1}{2}(T_i \cdot T_j + H.c.)] \]

- symmetry: O(3)
- no frustration, magnetic condensate

GKh (2013)
phenomenology:

**Soft-spin $O(N)$ model**

\[
S[\varphi] = \int_x \left\{ \frac{1}{2} (\nabla \varphi)^2 + \frac{1}{2c^2} (\partial_\tau \varphi)^2 + \frac{r_0}{2} \varphi^2 + \frac{u_0}{4!N} (\varphi^2)^2 \right\},
\]

- J-dispersion dynamics
- magnetization
- spin gap
- „soft“ constraint

Diagram:
- Disorder and order phases
- QCP transition
- $\varphi = 0$ and $\varphi \neq 0$
Soft-spin magnets: excitations

\[ \varphi = 0 \]

QCP

disorder

spin gap

two normal modes

phase

density

Goldstone

„Higgs“
J=0 spin-orbit Mott insulator

Candidate material: Ca$_2$RuO$_4$

- MIT at ~360 K
- AF below 110 K
Ca$_2$RuO$_4$ time-of-flight INS

Jain et al. (Nat.Phys.2017)

unusual AF-magnon: max at $\Gamma$-point

excitonic AF

Heisenberg AF ruled out
SPIN-POLARIZED, purely magnetic

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**Figure a**

$q = (0,0)$

```
\chi'' (arb. units)
```

- **ab**
- **c**

exp. vs theory

well defined "Higgs" mode

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**Figure b**

$q = (\pi,\pi)$

```
\chi'' (arb. units)
```

Energy (meV)

broad continuum
Electron doped $J=0$ Mott insulators

$J=0$ Mott

$J=1/2$ Mott

Naive expectation
Doping-induced FERROMAGNETISM

Chaloupka, GKh (2016)

$doped \ \text{Ca}_2\text{RuO}_4$ FM observed:
- Cao $et$ $al.$: La-doping
- Maeno $et$ $al.$: E-field induced
- Hughes $et$ $al.$: STO/ RuO$_2$/STO
Doped J=0 Mott insulators, SC pairing

triplet pairing, non-collinear d-vector

\[ d = -i\Delta(\sin \phi_k, \cos \phi_k, 0) \]

Chaloupka, GKh (2016)
Frustrated singlet-triplet models

90° bonding geometry
(triangular, honeycomb, etc)
90° bonding geometry
(triangular, honeycomb, etc)

Triplons are „orbitally-colored“

bond-selective dynamics expected
Honeycomb lattice

(A) hopping via anions

bond-dependent „XY“

GKh (PRL 2013)

(B) direct hopping

bond-dependent „ISING“

GKh (KITP talk 2015)
Each boson $T_x, T_y, T_z$ has its own zigzag

Emergent 1D physics: **FLAT BANDS**

$J=J_{\text{crit}}$: **zero-energy lines**

Compass model behavior, partial magnetic order
Kitaev-like bosonic model

x-bond: \( T_{ix}^T T_{jx} + T_{ix} T_{jx} \)

- hopping
- pair-generation

Number of x-bosons on each x-type bond is even / odd conserved:

\[ n_x = (0, 2) \] and \[ n_x = 1 \] sectors are separated

Bond parity \( P_{ij} = (1 - 2n_{\alpha i})(1 - 2n_{\alpha j}) \) is conserved
1) Extensive number of conserved quantities

2) Nonmagnetic ground state

3) Short-range only spin correlations, finite spin gap

4) However, no gapless Majoranas, no entanglement (unlike spin-Kitaev)

Chaloupka, GKh (unpub.)
Bosonic Kitaev model

„Strongly correlated“ paramagnet

Dynamical structure factor

Chaloupka, GKh (unpub.)
Spin–Orbit Mott Insulators

- Orbital chemistry
- $J=3/2$ octupolar order
- $J=1/2$ „cuprates“, Kitaev, exotic pairings
- $J=0$ excitonic magnets, bosonic Kitaev model