On the spin orientation

1. Qualitative rules for predicting preferred spin orientations?

2. Spin orientations of Sr_3NiIrO_6, Sr_2IrO_4, Ba_2NaOsO_6:
Implications on the magnetism of $J_{eff}=1/2$ ions

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Energy Mapping Analysis

1. Model Hamiltonian: Parameters

$$\sum_{i<j} J_{ij} \vec{S}_i \cdot \vec{S}_j$$

$$\sum_{i<j} \vec{D}_{ij} \cdot (\vec{S}_i \times \vec{S}_j)$$

$$\text{"}\sum_i A_i S_{iz}^2\text{"}$$ ← **Qualitative prediction?**

2. DFT calculations

3. Broken-Symmetry Spin States

4. $\Delta E_{spin\text{-}H} \leftrightarrow \Delta E_{DFT}$

Spin orientation ← spin-orbit coupling (SOC)

Large SOC

Topological insulators
Rashba-Dresselhaus effects
Valleytronics
Spin-textured bands
J_{eff}=1/2 states

Coupling between spin and orbital moments

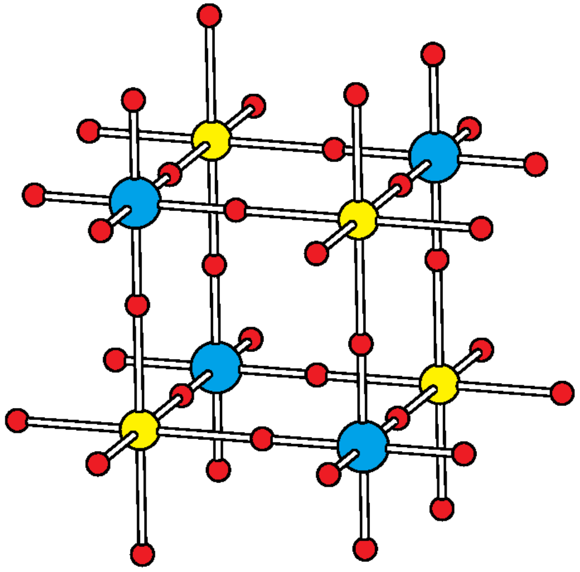
weak → L-S coupling
strong → j-j coupling

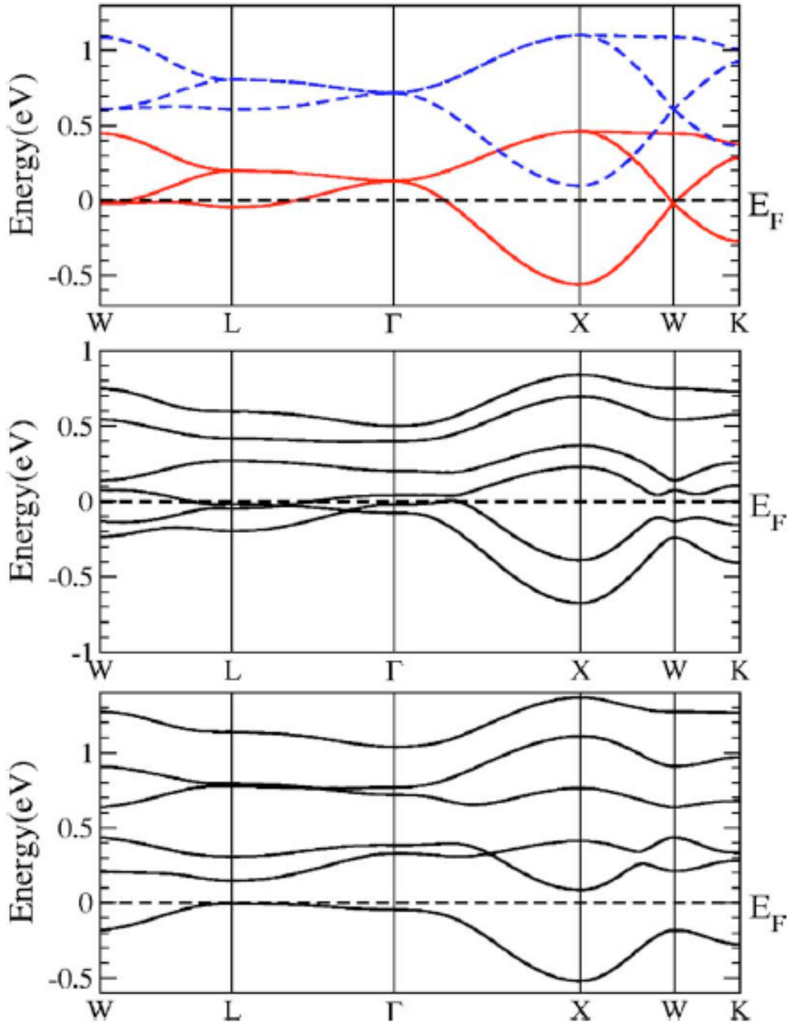
Magnetic insulator of 5d oxides

← **Strong SOC + weak correlation**

PRB 75, 052407 (2007)

Ba_2NaOsO_6: **Os^{7+} ($5d^1$, S = 1/2)**





$\theta_{CW} \approx$ -10 K

FM below $T_C \approx$ 7 K

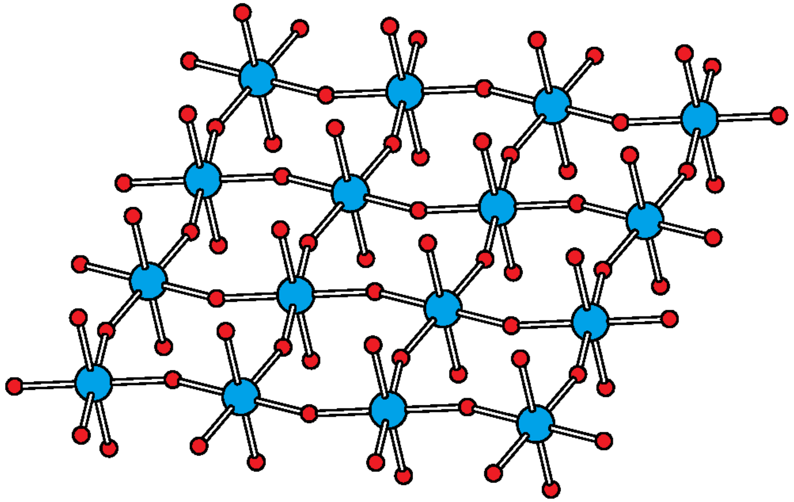
PRB 84, 144416 (2011)

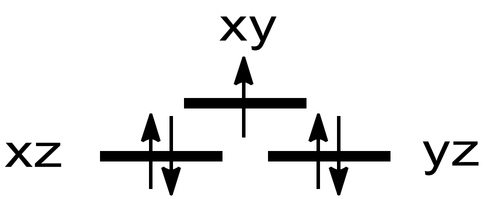
Os^{7+} ($5d^1$, S = 1/2) in Ba_2NaOsO_6

No preferred spin orientation

Sr_2IrO_4: low-spin Ir^{4+} ($5d^5$, $S = 1/2$) → $(t_{2g})^5$

Corner-sharing IrO_6 → IrO_4 layer, Axially-elongated IrO_6



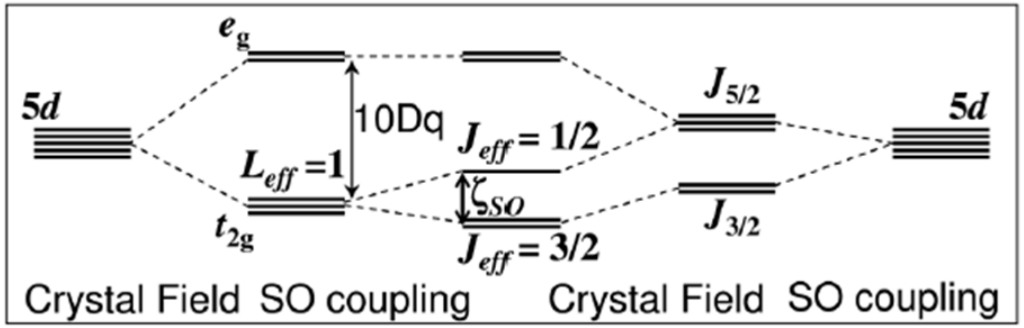


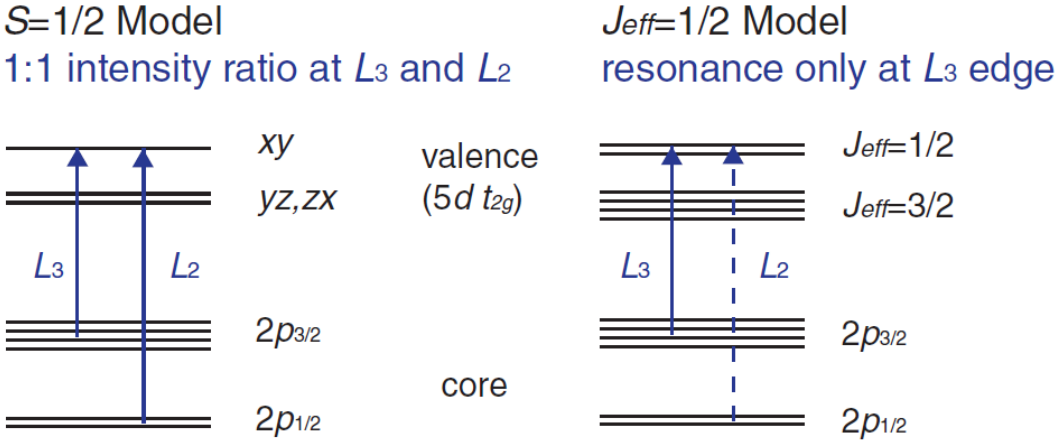
Magnetic insulator

Strong SOC + weak correlation

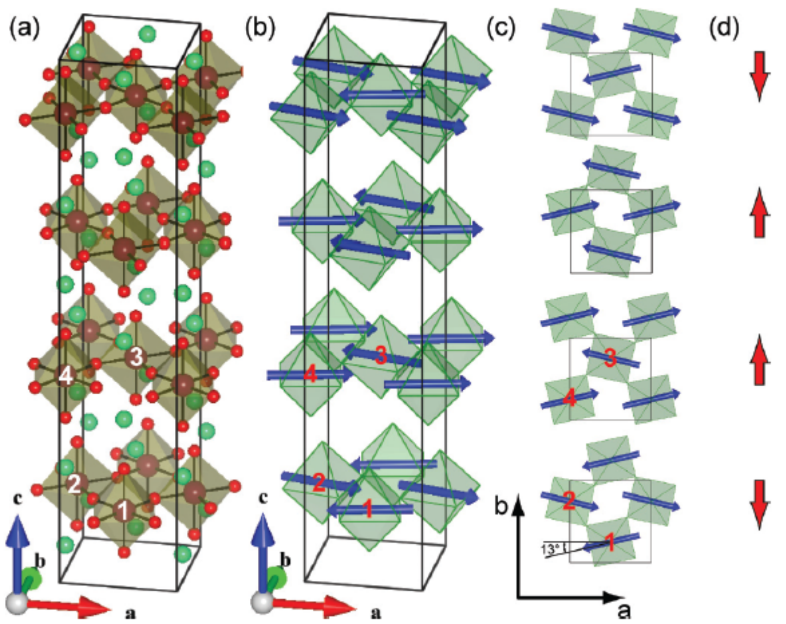
→ split of J_{eff}=1/2

PRL 101, 076402 (2008)
Science 323, 1329 (2009)





Excitation different orbital states



PRB 87, 140406(R) (2013)

Low-spin Ir^{4+} ($5d^5$, S = 1/2) ion in Sr_2IrO_4

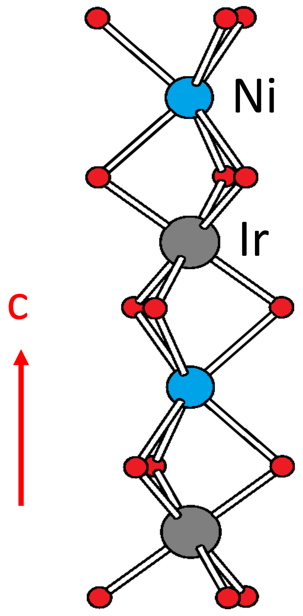
Weakly anisotropic, preferred spin orientation: $\perp$ $Ir\text{-}O_{ax}$

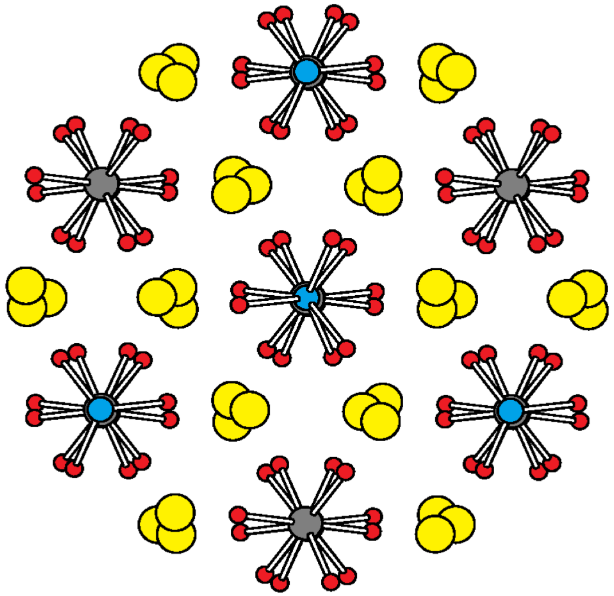
ESR study of Sr_2IrO_4

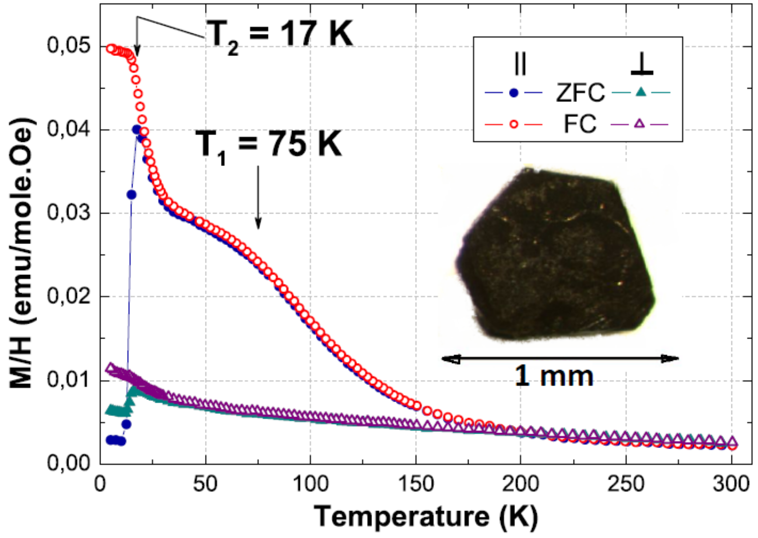
Isotropic Heisenberg interactions between Ir^{4+} spins

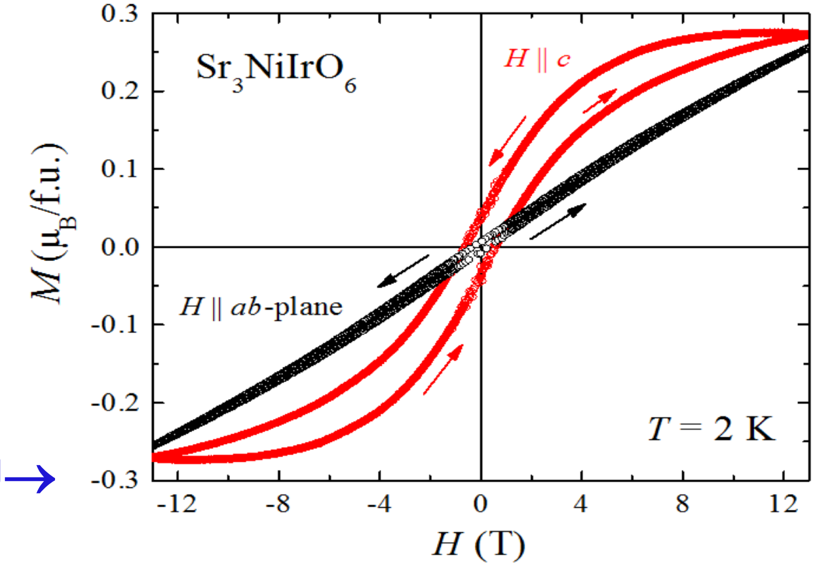
PRB 89, 180401(R) (2014)

Sr_3NiIrO_6: low-spin Ir^{4+} ($5d^5$, $S = 1/2$), high-spin Ni^{2+} ($3d^8$, $S = 1$)









~1/3 μ_B/FU→

Strongly anisotropic

M-H hysteresis → FM?

Ferrimagnetic?

PRB 89, 180401(R) (2014)

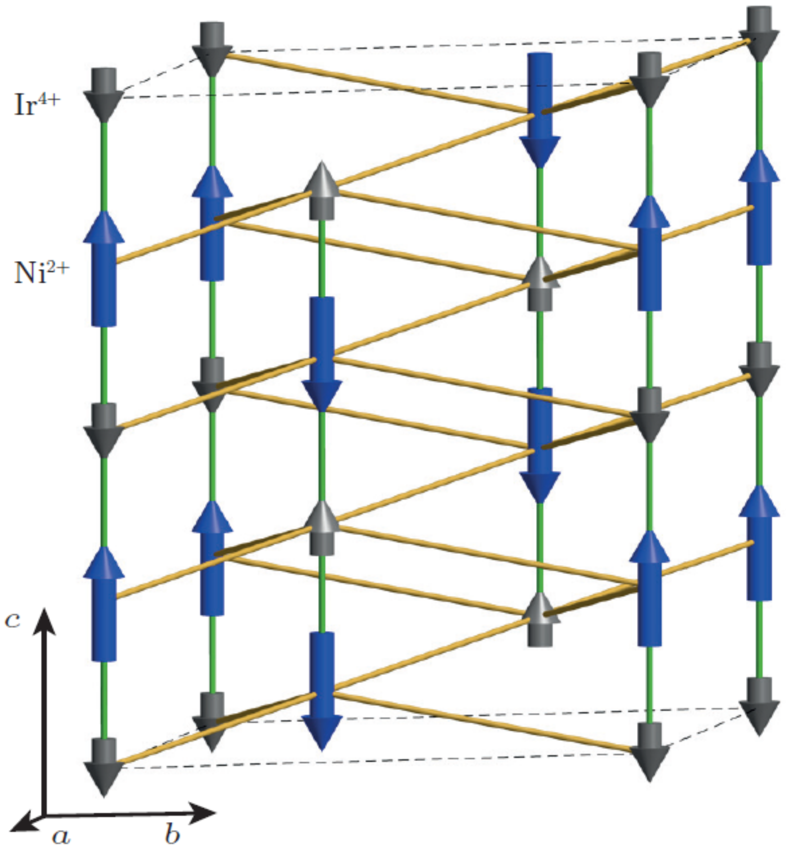
PRL, under review

Ni^{2+} and Ir^{4+} spins

both ||c

nn AFM → Ferrimagnetic

PRB 90, 014408 (2014)
arXiv:1501.05735 [cond-mat.str-el]

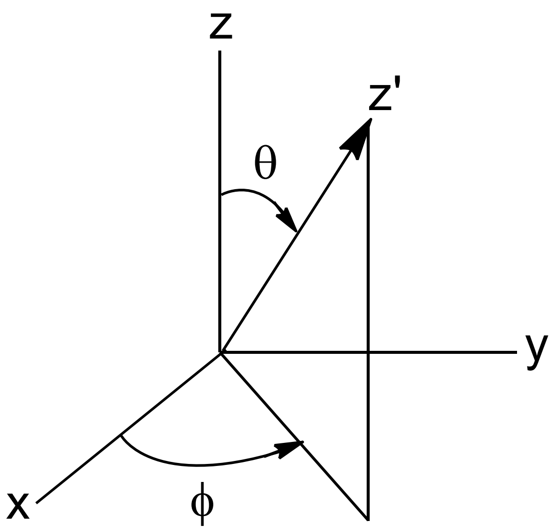


Partially disordered AFM (PDA)

→ 1/3 μ_B/FU

Low-spin Ir^{4+} ($5d^5$, S = 1/2) in Sr_3NiIrO_6
Strongly anisotropic

Weak single-ion isotropy:



$$\Delta E_{SOC} = -\frac{\left|\langle \psi_o | \hat{H}^0_{SO} | \psi_u \rangle\right|^2}{|e_o - e_u|}$$

SOC from a trace orbital moment $\delta\vec{L}$

(x, y, z) for orbital, (x’, y’, z’) for spin

$$\hat{H}_{SO} = \lambda \hat{S} \cdot \hat{L} \approx H^0_{SO}$$
$$= \lambda \hat{S}_{z'} (\hat{L}_z \cos\theta + \frac{1}{2} \hat{L}_+ e^{-i\phi} \sin\theta + \frac{1}{2} \hat{L}_- e^{i\phi} \sin\theta)$$

θ = 0°: easy-axis anisotropy

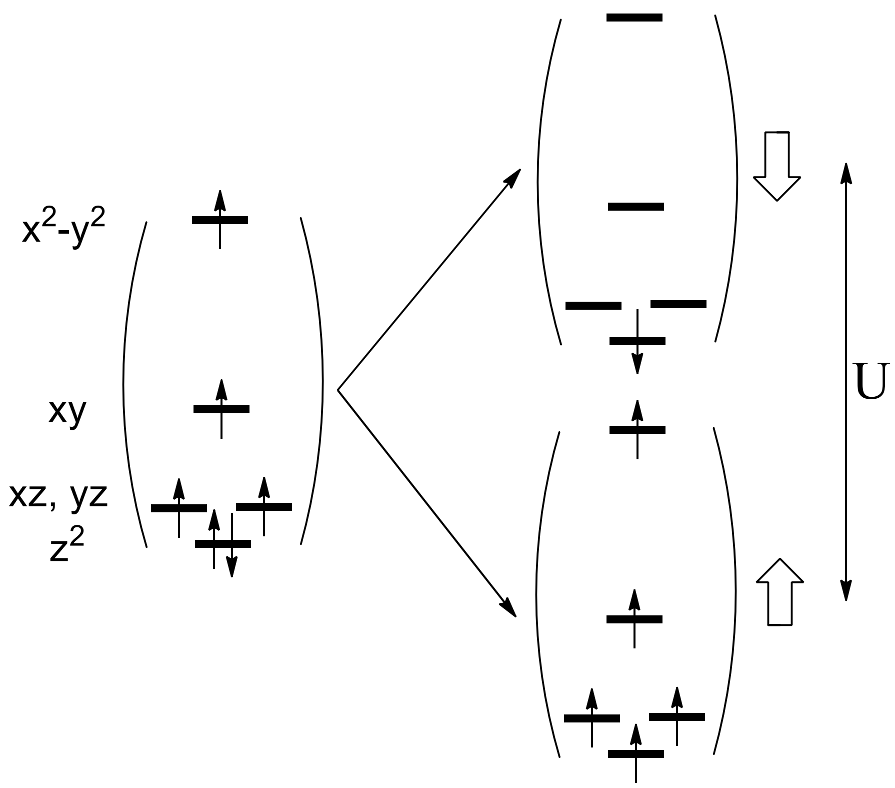
θ = 90°: easy-plane anisotropy

J. Comput. Chem. **29**, 2187 (2008)

Most important,
between the HO and the LU states

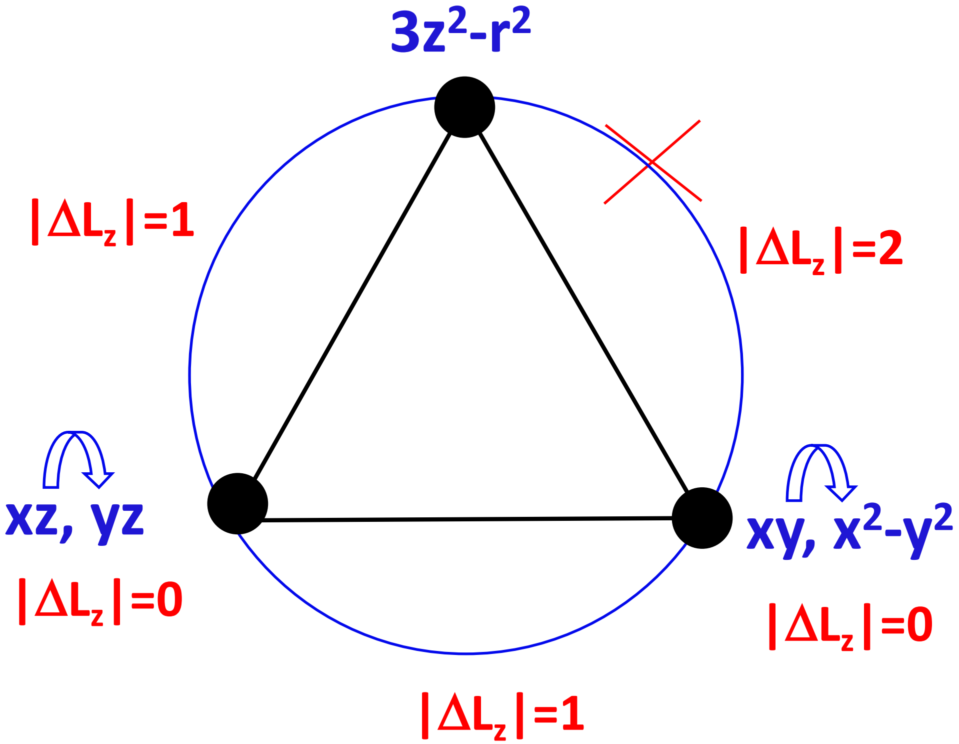
Dalton Trans. **42**, 823 (2013)

Spin-polarized d-states



$$\lambda \hat{S}_{z'}(\hat{L}_z \cos\theta + \frac{1}{2}\hat{L}_+ e^{-i\phi} \sin\theta + \frac{1}{2}\hat{L}_- e^{i\phi} \sin\theta)$$

Selection rules for SOC

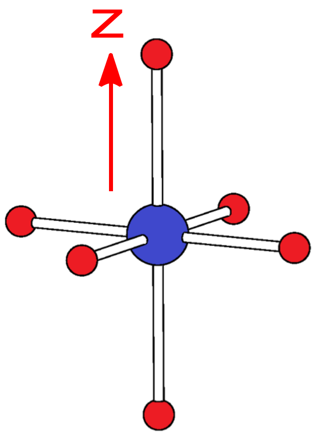


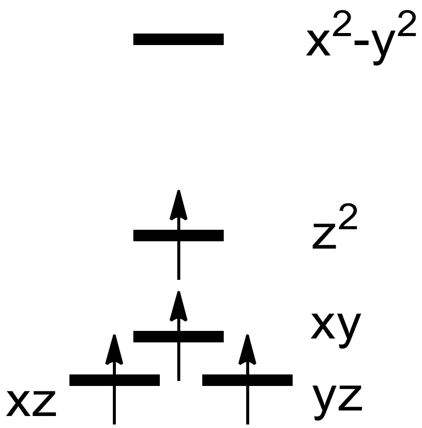
$$\langle L_z | \hat{L}_z \cos\theta | L_{z'} \rangle \rightarrow |\Delta L_z| = 0 \rightarrow \text{easy-axis}$$

$$\langle L_z | \hat{L}_+ \sin\theta | L_{z'} \rangle \rightarrow |\Delta L_z| = 1 \rightarrow \text{easy-plane}$$

$$\langle L_z | \hat{L}_- \sin\theta | L_{z'} \rangle \rightarrow |\Delta L_z| = 1 \rightarrow \text{easy-plane}$$

High-spin Mn^{3+} in $TbMnO_3$ & Ag_2MnO_2
Axially-elongated MnO_6 octahedron





$(xy\uparrow)^1 < (z^2\uparrow)^1 < (x^2-y^2\uparrow)^0$

||z, easy-axis anisotropy

PRL **101**, 037209 (2008)
PRB **81**, 094421 (2010)

Concluding remarks

The preferred spin orientations of magnetic ions, predicted by
uniaxial magnetism, or
weak single-ion anisotropy.