Contrasting electronic states of RuI_3 and $RuCl_3$

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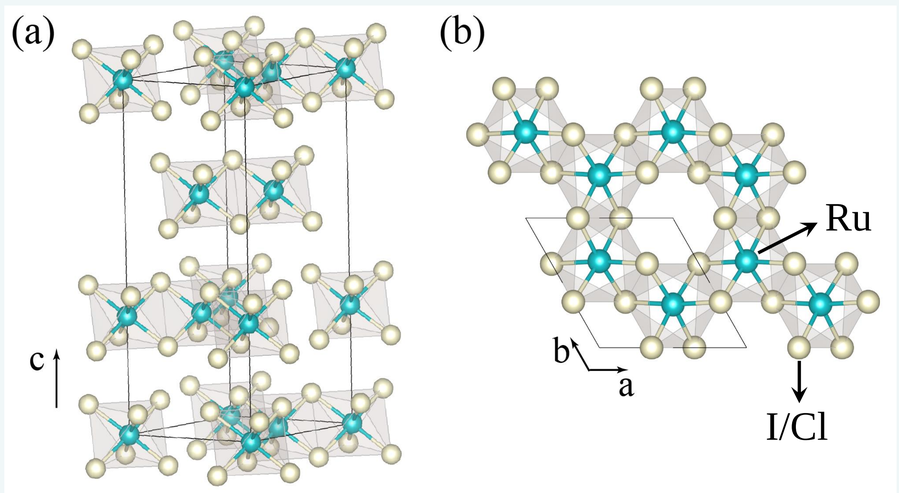
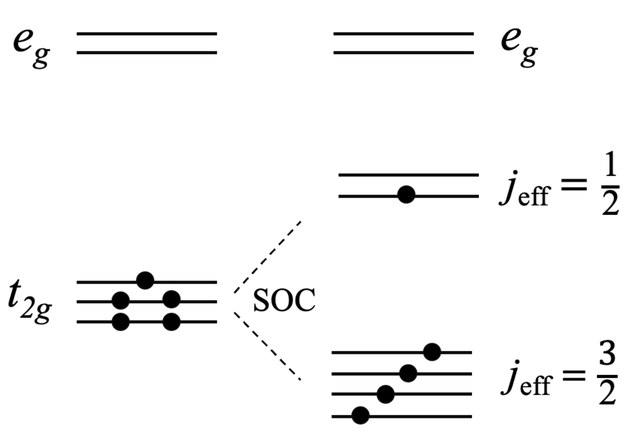


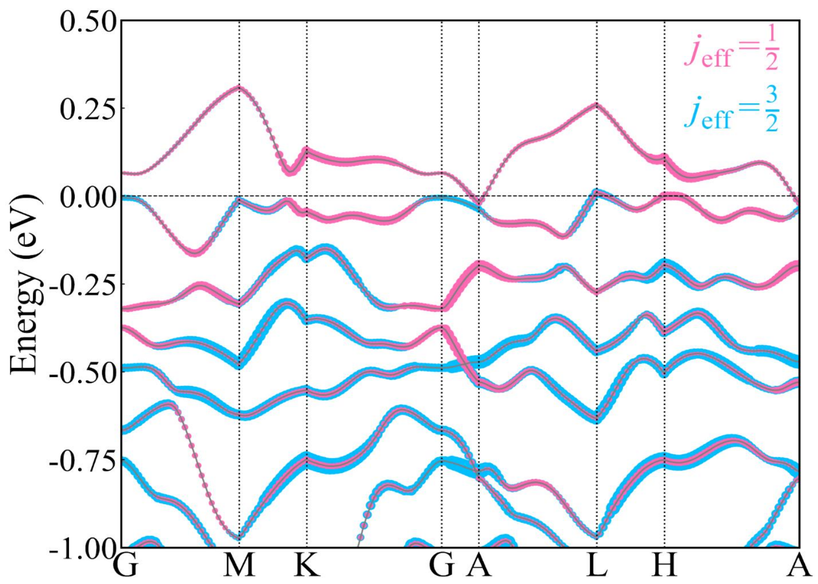
The spin-orbital entangled states are of great interest as they hold exotic phases and intriguing properties. Here we investigate the electronic and magnetic properties of RuI_3 and $RuCl_3$. We find that the Ru^{3+} ion of **RuI_3** is in the spin-orbital entangled **j_{eff} = 1/2 paramagnetic state** owing to the strong Ru 4d-I 5p hybridization, weak spin polarization, and strong SOC. More interestingly, a metal-insulator transition occurs from RuI_3 bulk to monolayer. In contrast, **$RuCl_3$** bulk and monolayer both show Mott-insulating behavior, the Ru^{3+} ion is in the **S = 1/2 and L_x = 1 zigzag AFM state** due to considerable Hund exchange and trigonal crystal field splitting. This result well explains the experimental electronic and magnetic behavior of bulk $RuCl_3$. The present work demonstrates the contrasting spin-orbital states and the varying properties of RuI_3 and $RuCl_3$.

RuI_3 — strong Ru 4d-I 5p hybridization & weak spin polarization ⟶ SOC-dominant

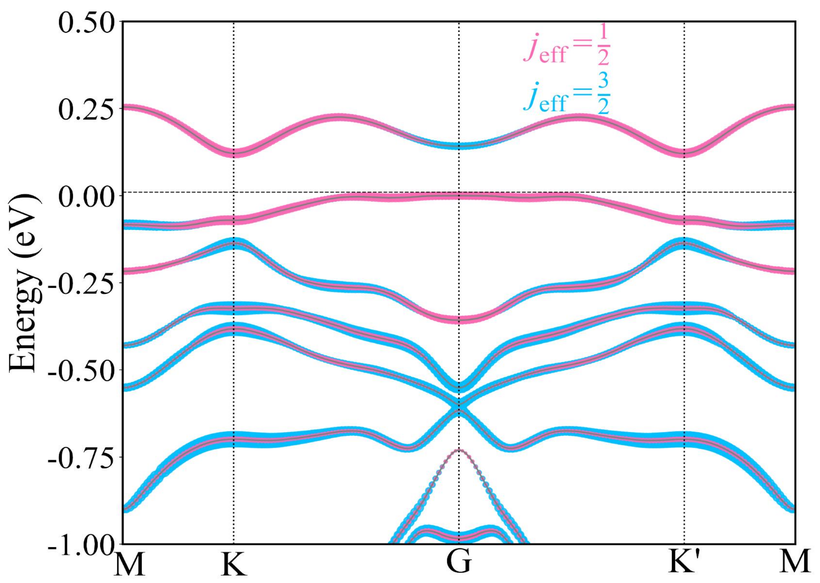
crystal field level diagram



bulk: j_{eff} = 1/2 NM metallic state



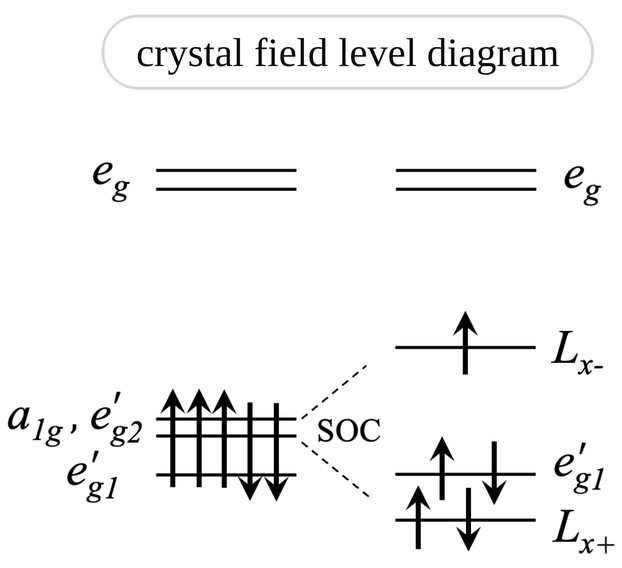
monolayer: j_{eff} = 1/2 NM insulating state

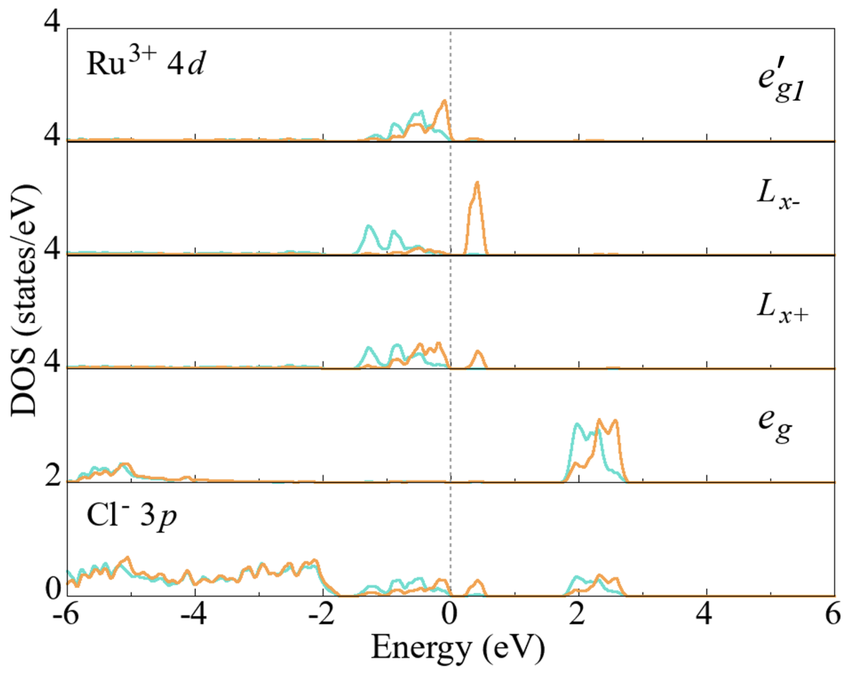


$RuCl_3$ — weak Ru 4d-Cl 3p hybridization & strong spin polarization ⟶ Hund exchange & trigonal splitting-dominant

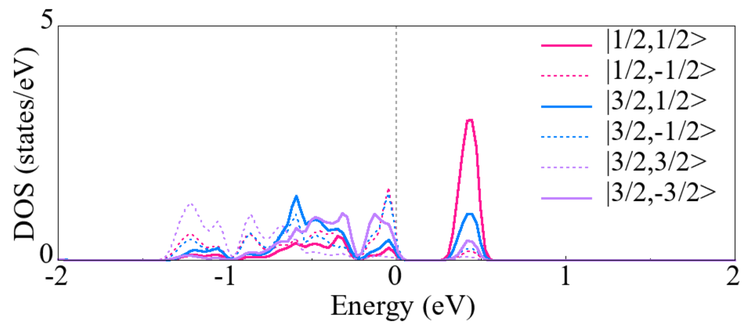
bulk & monolayer: S = 1/2 and L_x = 1 Mott-insulating state

crystal field level diagram





project onto j_{eff} = 1/2 basis



j_{eff} = 1/2 is not suitable for $RuCl_3$!

Experiments:

- zigzag AFM insulator
- g factors: $g_{ab} > g_c$
- $M_{in\text{-}plane} \sim 1.2\ \mu_B/Ru^{3+}$
- $\mu_{eff} = 2.0-2.4\ \mu_B/Ru^{3+}$

Our work:

- zigzag AFM insulator
- in-plane magnetic anisotropy energy = 26.7 meV/Ru^{3+}
- $M_{spin} + M_{orb} = 0.70 + 0.47 = 1.17\ \mu_B/Ru^{3+}$
- $\mu_{eff} = \sqrt{g_s^2 S(S+1) + g_l^2 L(L+1)} = 2.24\ \mu_B/Ru^{3+}$