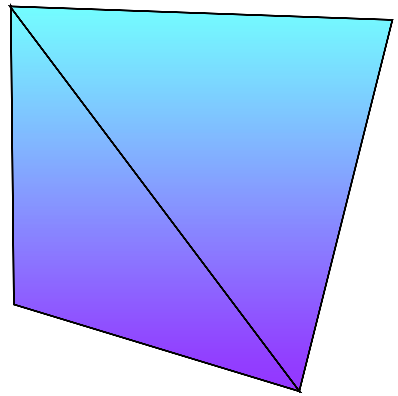
幾何学的電子相関がもたらす異常金属相の解明
―金属絶縁体転移の周辺―

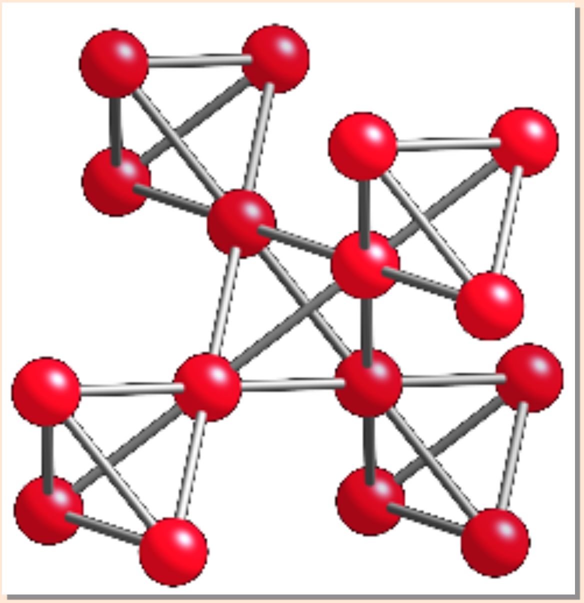


Ryosuke Kadono
IMSS-KEK/CMRC/Sokendai

Introduction

Issue: Geometrical Correlation of Itinerant Electrons

Stage of itinerancy = highly symmetric crystal



Pyrochlore lattice (Spinel)

Pyrochlore oxides: $A_2B_2O_7$

Spinels: AB_2O_4

◆Problem

Structural phase transition that accompanies metal-insulator (MI) transition.

◆Solution

Mixed valence state may suppress MI transition by charge frustration(?).

e.g., LiV_2O_4

◆Issues

Is there any sign of geometrical frustration in the insulator phase associated with MI transition?

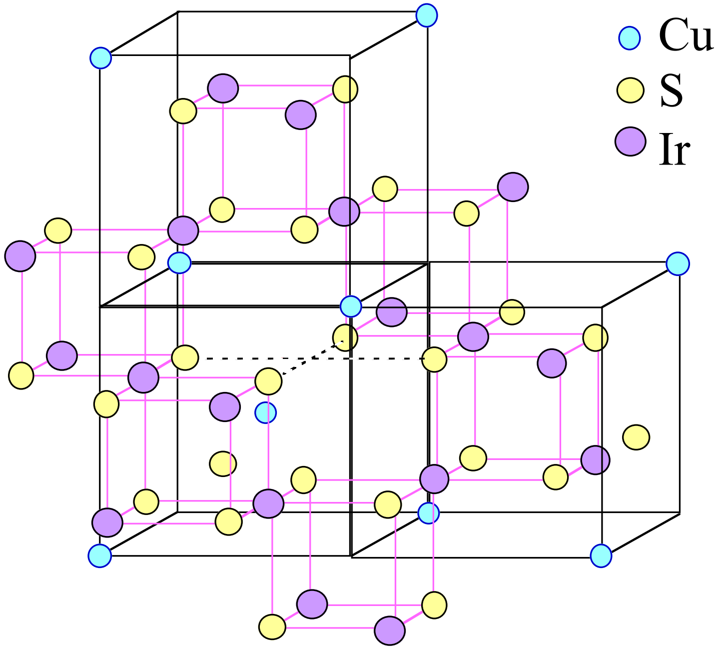
Insulator phase of $CuIr_2S_4$ revisited

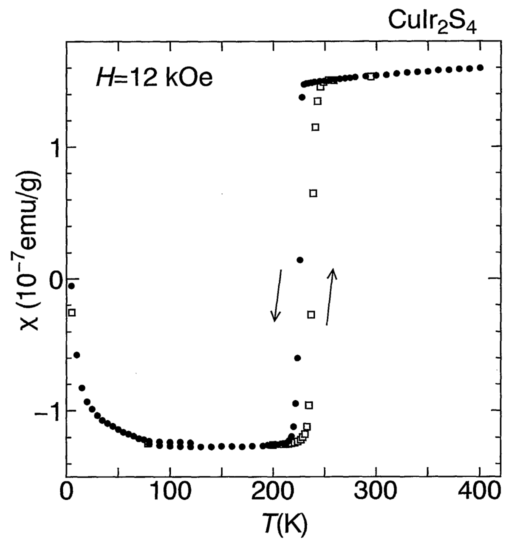
Kenji M. Kojima[A,B], Masanori Miyazaki[A], Masatoshi Hiraishi[A], Ichihiro Yamauchi[A], Akihiro Koda[A,B], Ryosuke Kadono[A,B], Yoshinori Tsuchiya[C], Hiroyuki S. Suzuki[C], Hideaki Kitazawa[C]

[A]KEK/J-PARC, [B]SOKENDAI,[C]NIMS

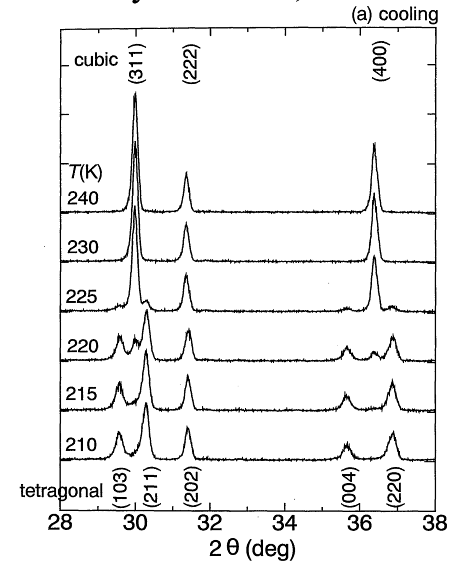
Metal-insulator transition in $CuIr_2S_4$

Structural phase transition accompanied by MI transition at ~230 K:

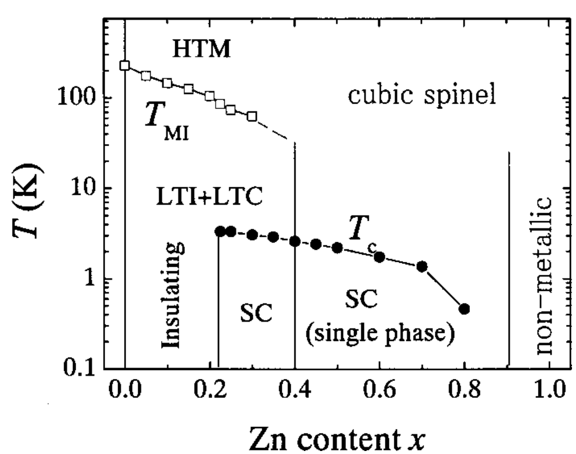


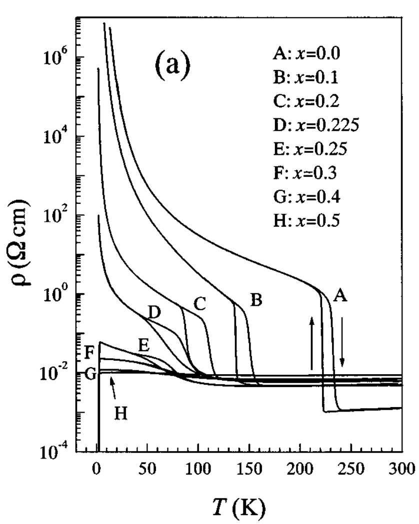


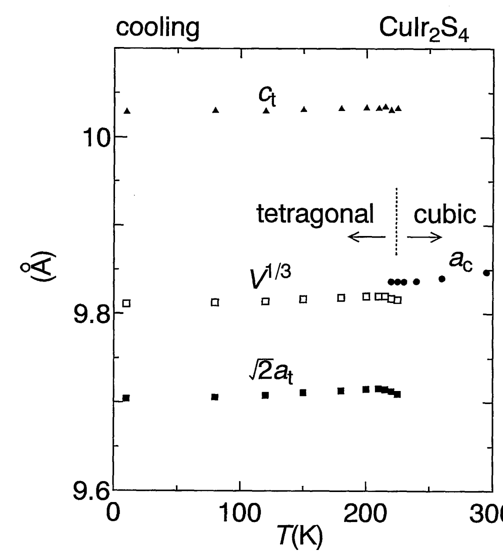
Furubayashi *et al.*, '94



Superconductivity in $Cu_{1-x}Zn_xIr_2S_4$



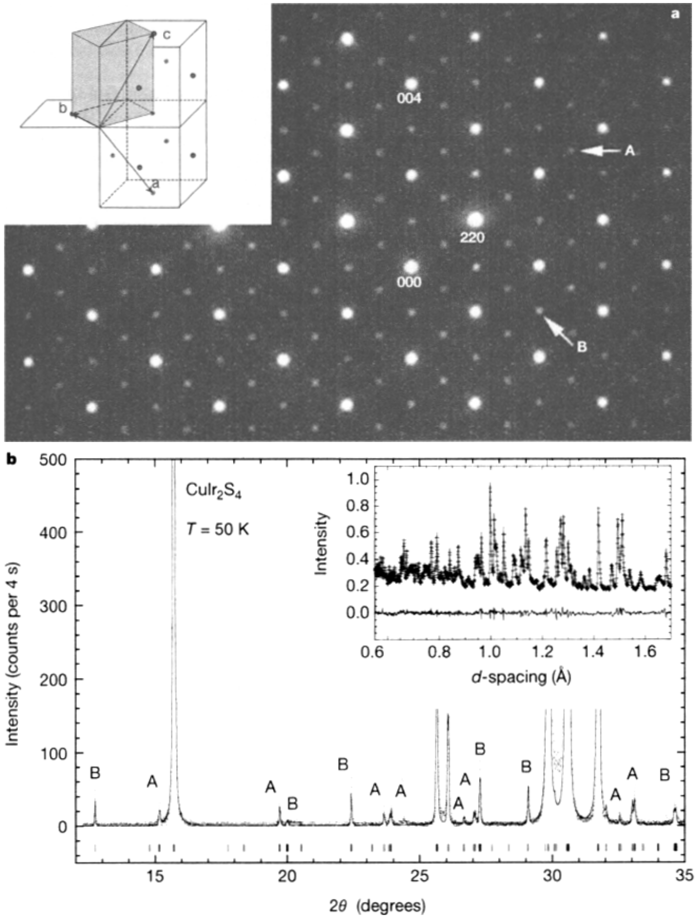




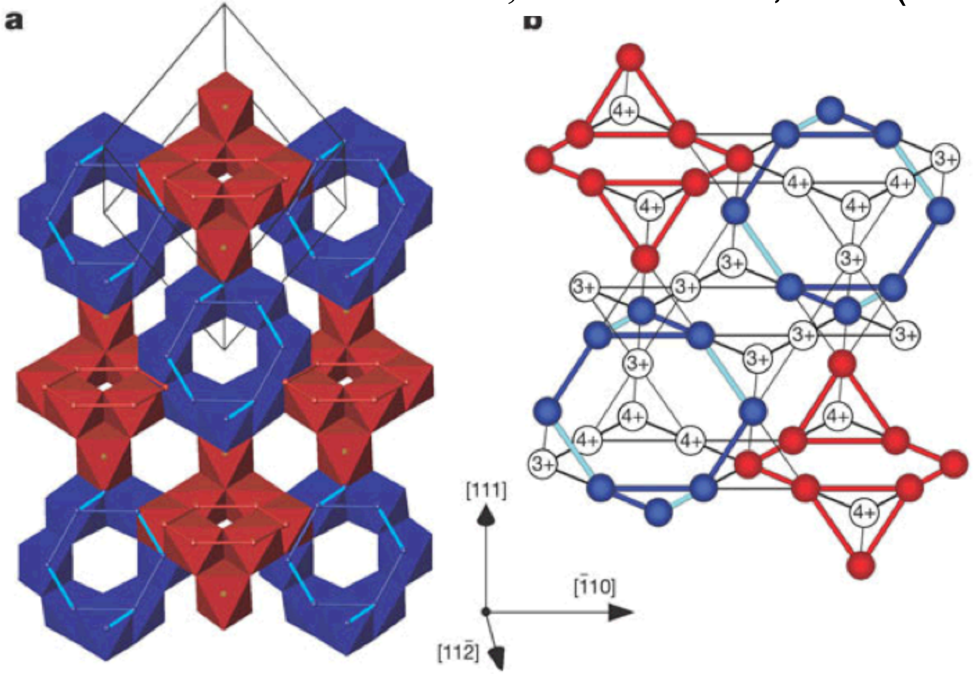
Metal-insulator transition in $CuIr_2S_4$

Formation of isomorphic Ir^{3+} and Ir^{4+} octamers and spin dimerization:

Radaelli *et al.*, Nature **416**, 155 (2002)



a) Electron diffraction pattern of $CuIr_2O_4$ at 93 K with incident beam parallel to the [110] cubic axis. b) SR X-ray diffraction pattern with Rietveld fit.



a) Red=Ir^{3+}, Blue=Ir^{4+}, with spin dimers shown by light blue lines.

b) Same as a), showing Ir skeleton of octamers.

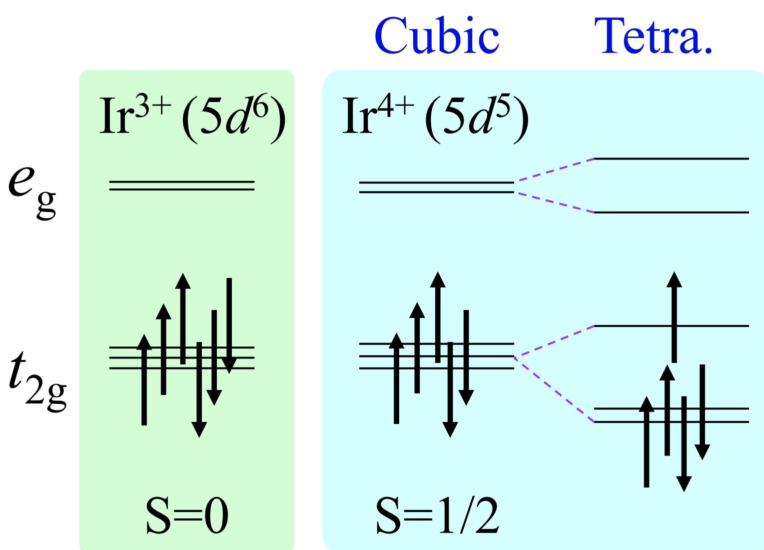
…suggesting the effect of charge/spin frustration

cf. Fe_3O_4 (magnetite: Fe^{2+} & Fe^{3+})

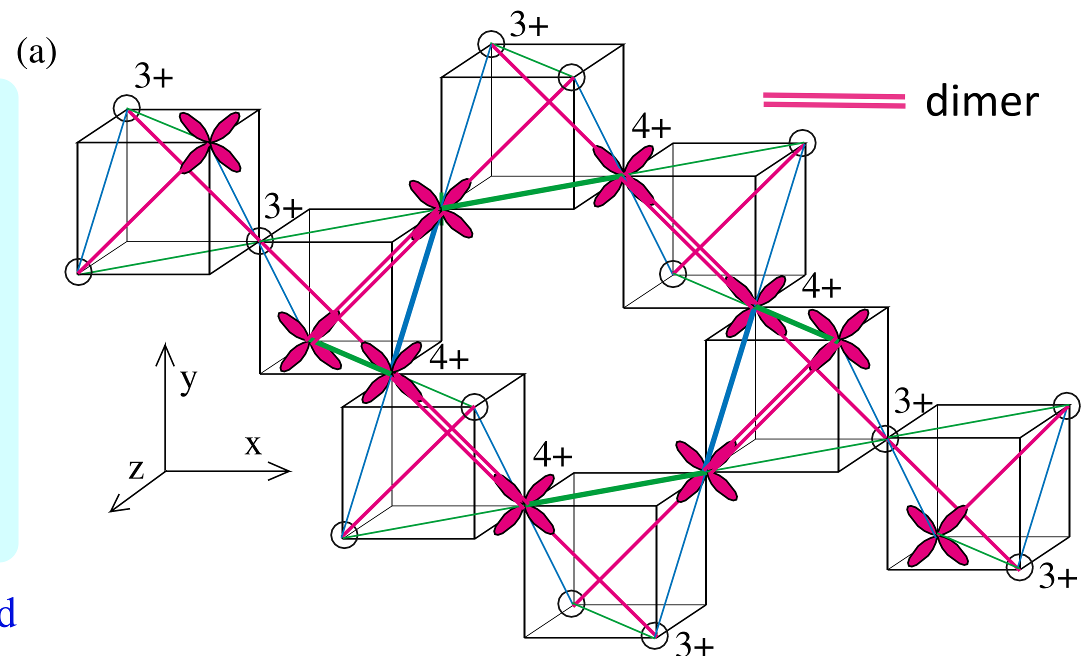
Metal-insulator transition in $CuIr_2S_4$

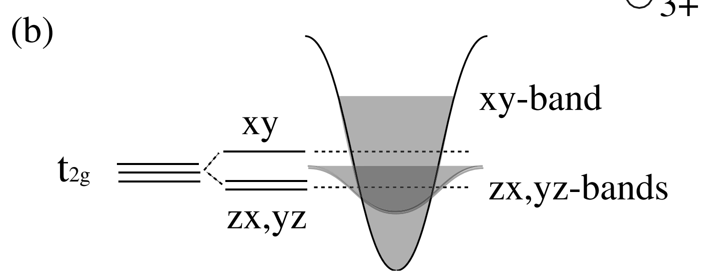
Theoretical model on t_{2g} manifold: Dimerization of Ir^{4+} on intersecting Ir chains:

Khomskii and Mizokawa, PRL **94**, 156402 (2005)



low spin state…relatively large crystal field



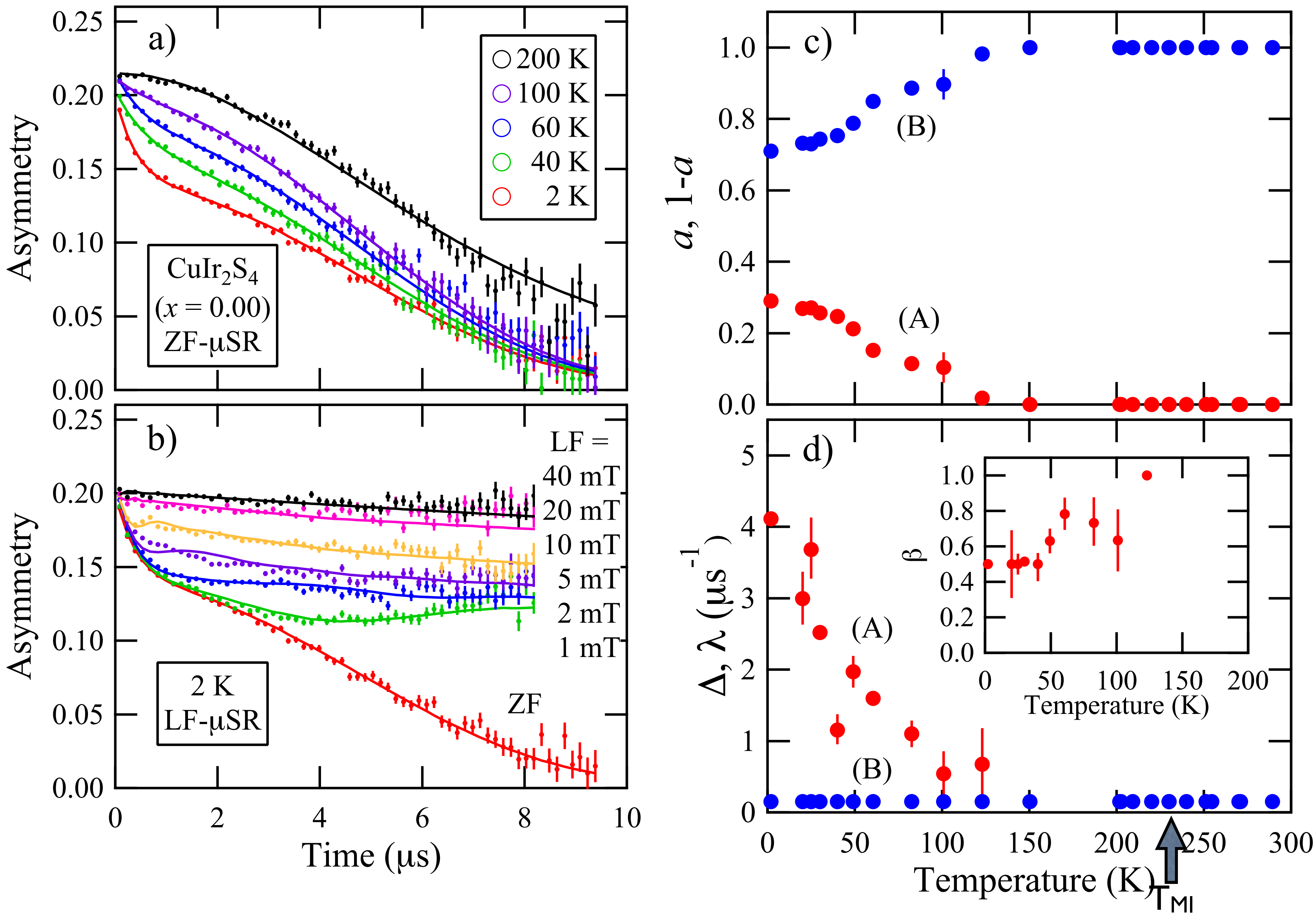


Presence of orbital degeneracy for Ir^{4+} → Jahn-Teller instability

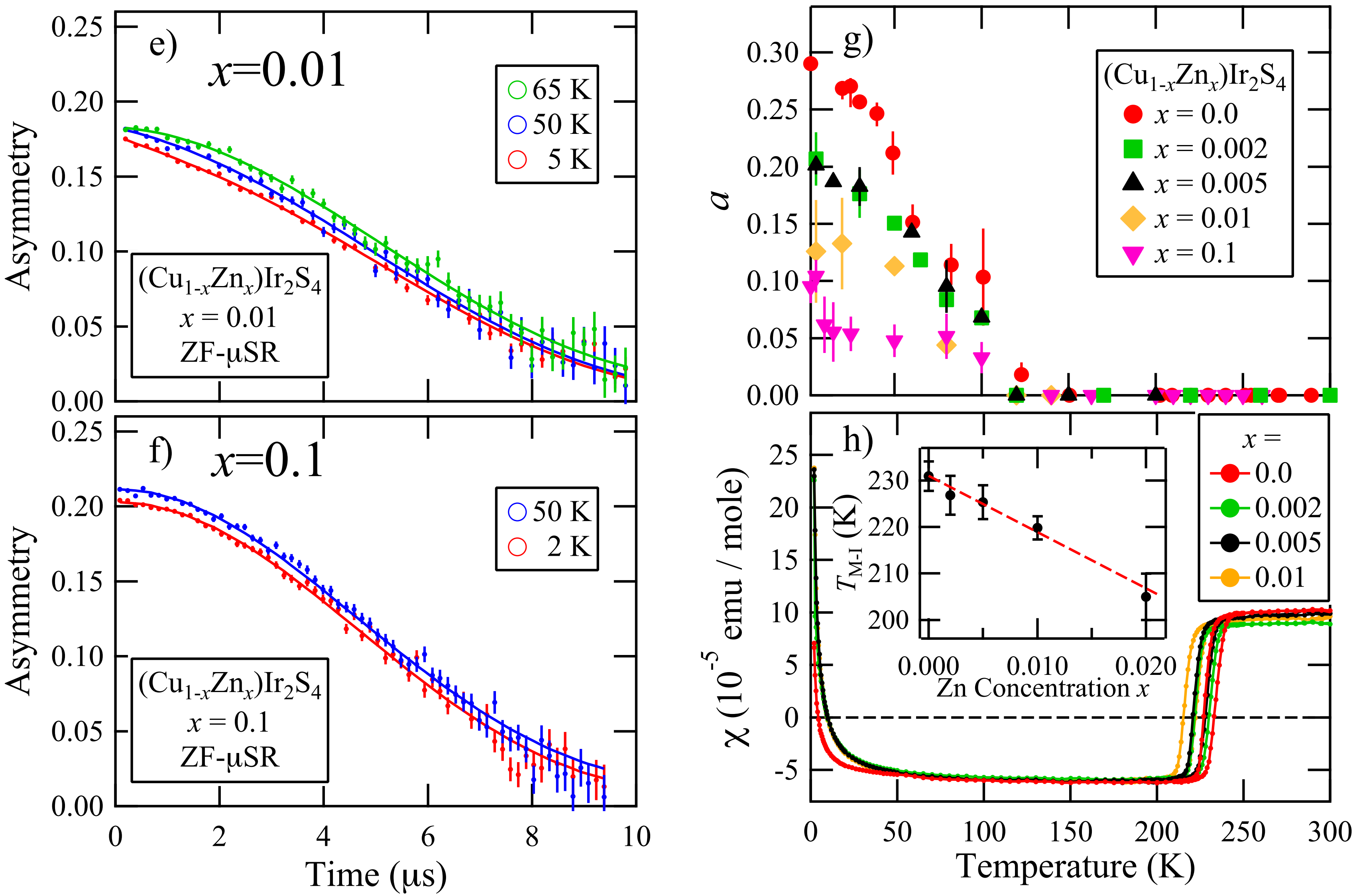
Cu^{1+}: non-magnetic ($3d^{10}$)

…but no reason to take octamer arrangement.

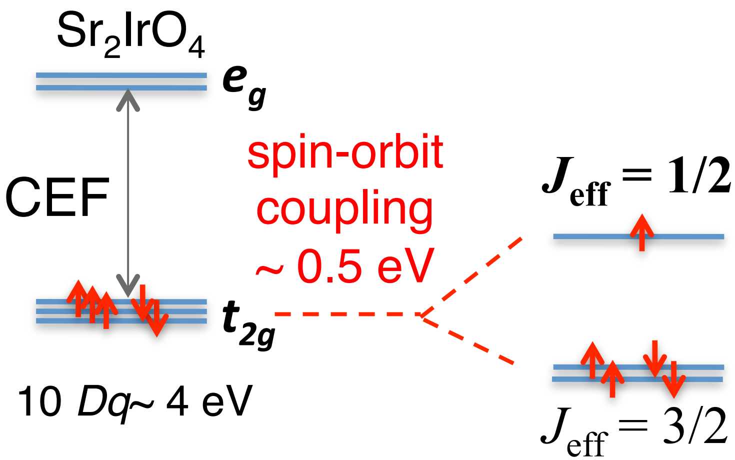
μSR in $CuIr_2S_4$



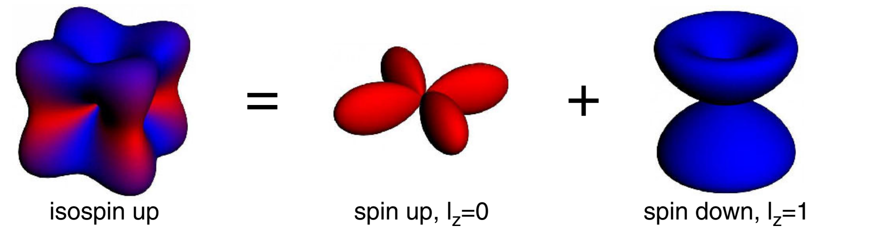
μSR in $Cu_{1-x}Zn_xIr_2S_4$

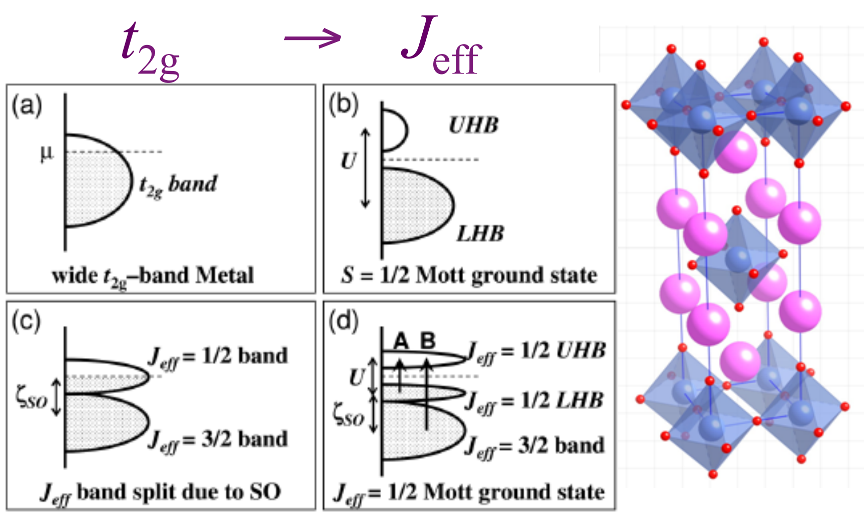


Physics of Ir^{4+} ($5d^5$) ions:

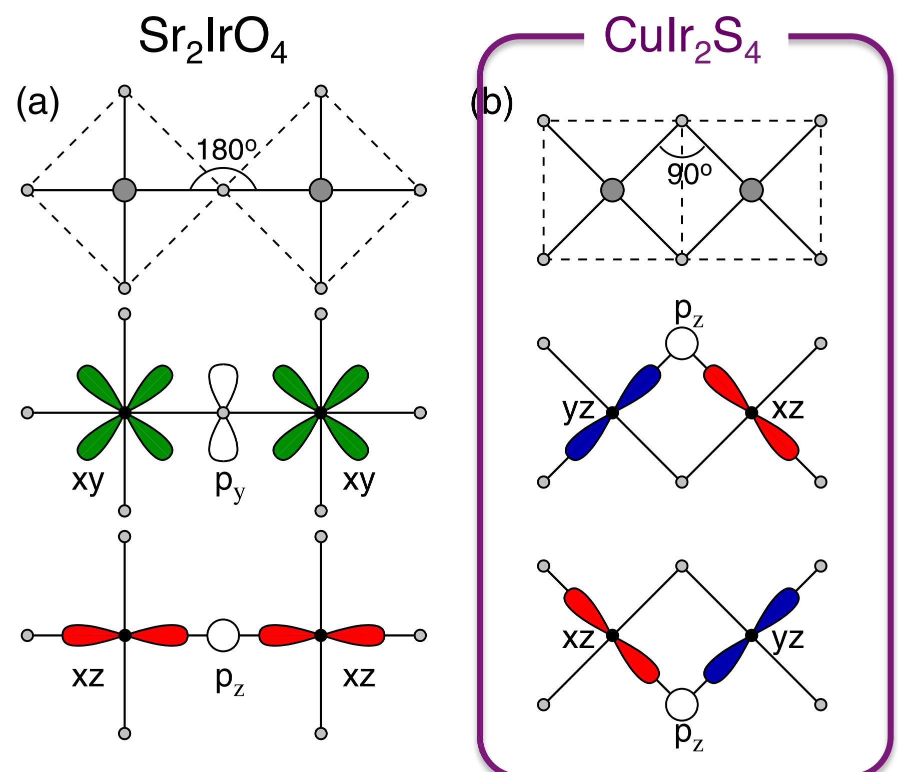


Jackeli and Khaliullin, PRL **102**, 017205 (2009)





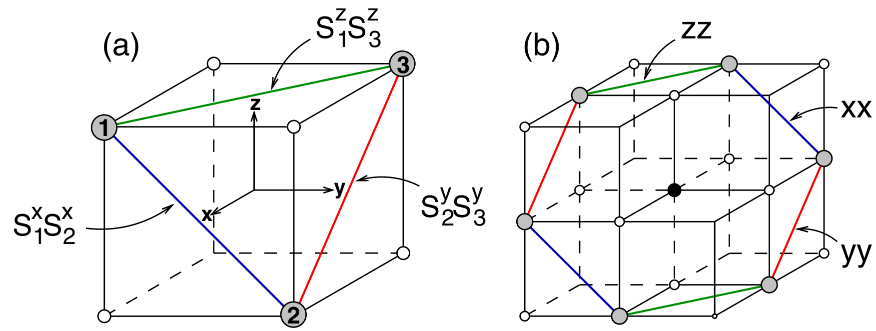
B.J. Kim *et al.*, PRL **101**, 76402 (2008)
Science 323, 1329 (2009)



Physics of Ir^{4+} ($5d^5$) ions:

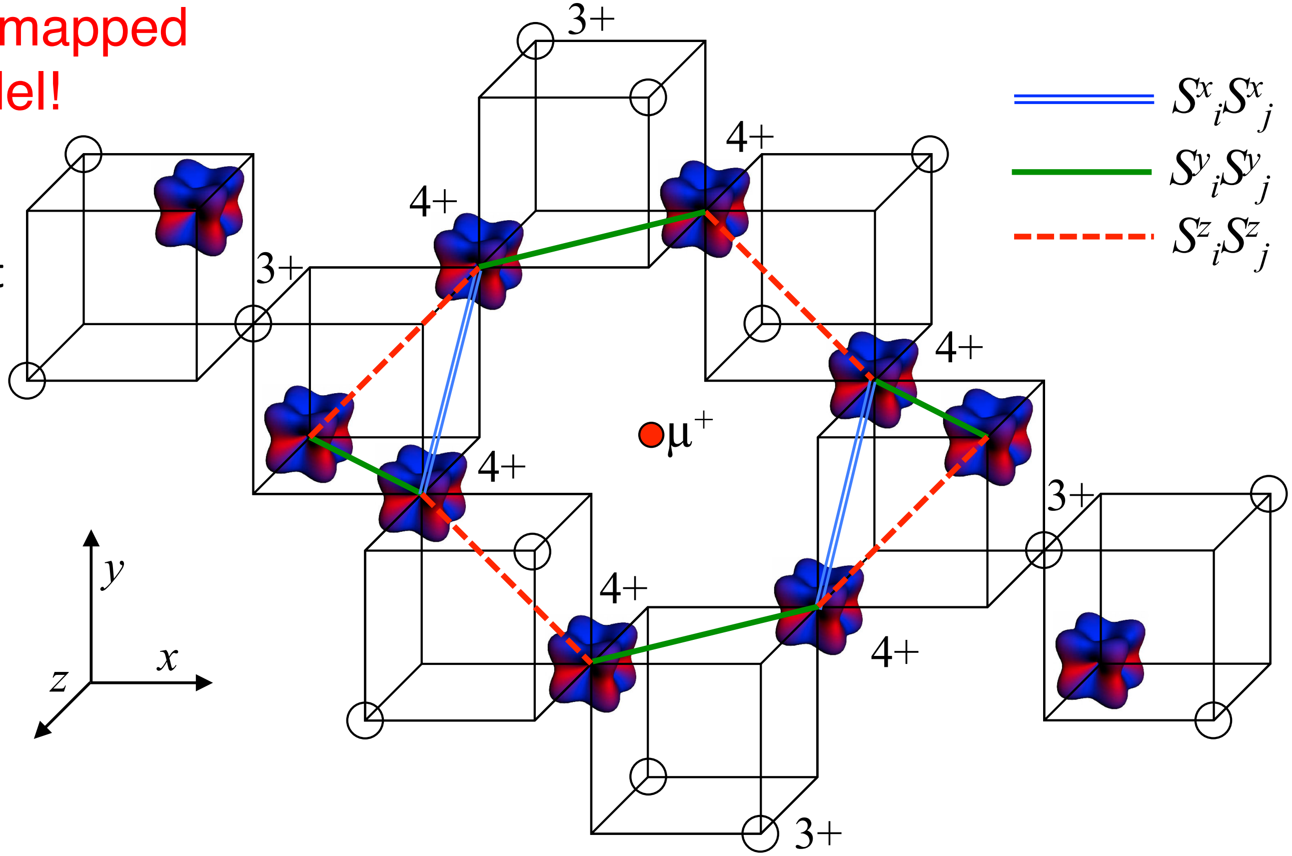
Effective Hamiltonian:

$H^{(\gamma)}_{ij} = -JS^{\gamma}_{i}S^{\gamma}_{j}, \quad \gamma = x,y,z$



Compass model on triangle/honeycomb
= Kitaev model

Ir^{4+} octamer can be mapped onto the Kitaev model!



- While single (isolated) octamer would not exhibit local magnetism*, weak inter-octamer correlation might lead to freezing of spin-liquid.

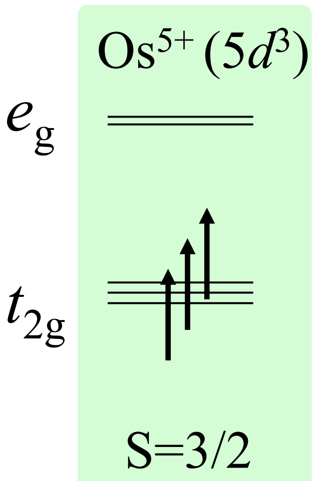
*Y. Motome, private communication.

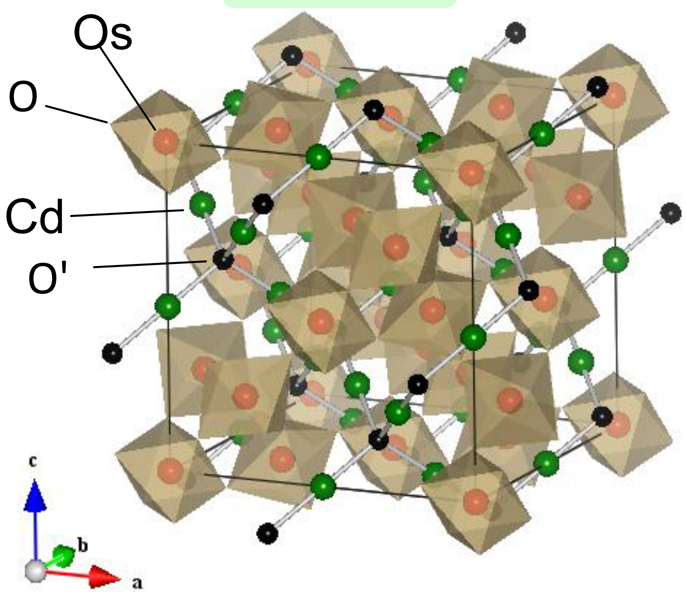
Quest for the origin of MI transition in $Cd_2Os_2O_7$

Akihiro Koda[A,B], Kenji M. Kojima[A,B], Masanori Miyazaki[A], Masatoshi Hiraishi[A], Ichihiro Yamauchi[A], Ryosuke Kadono[A,B], Jun-ichi Yamaura[C], Zenji Hiroi[C]
[A]KEK/J-PARC, [B]SOKENDAI,[C]ISSP-U.Tokyo

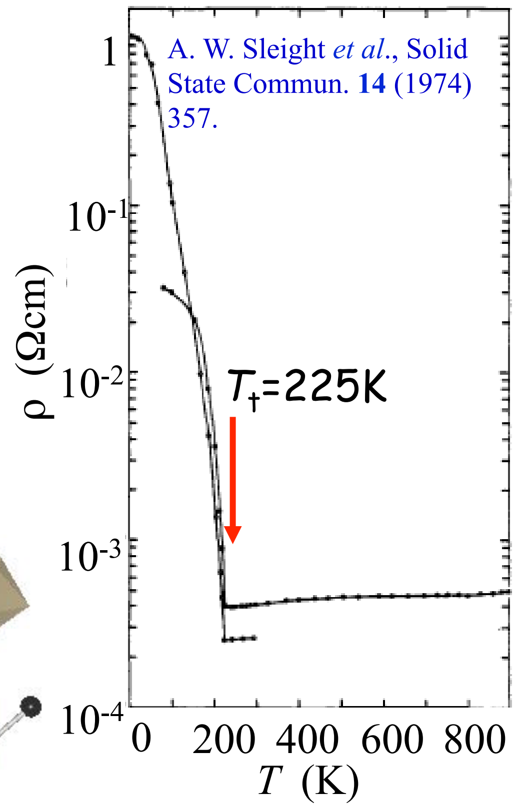
Metal-insulator (MI) transition in $Cd_2Os_2O_7$

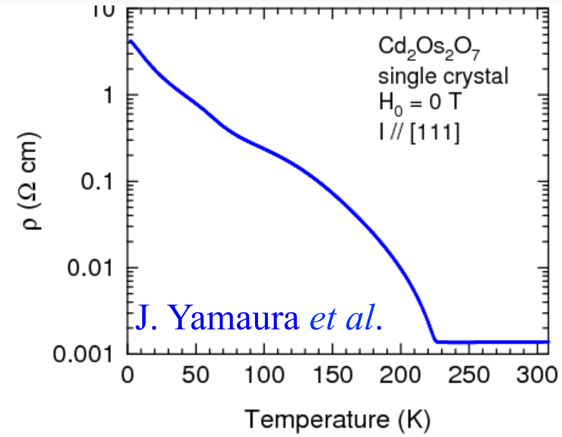
Half-filled t_{2g} band?

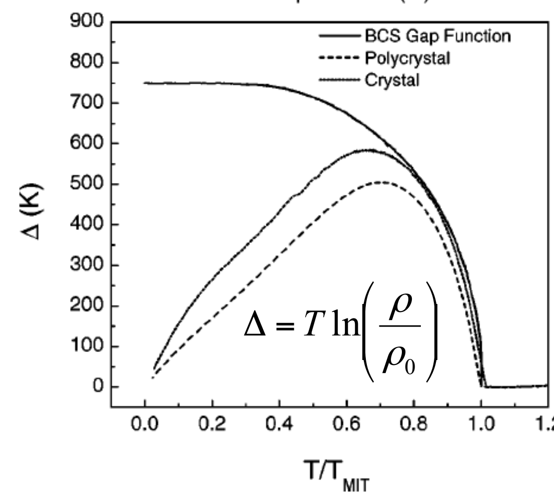




Resistivity:







D. Mandrus et al., PRB **63** (2001) 195104.

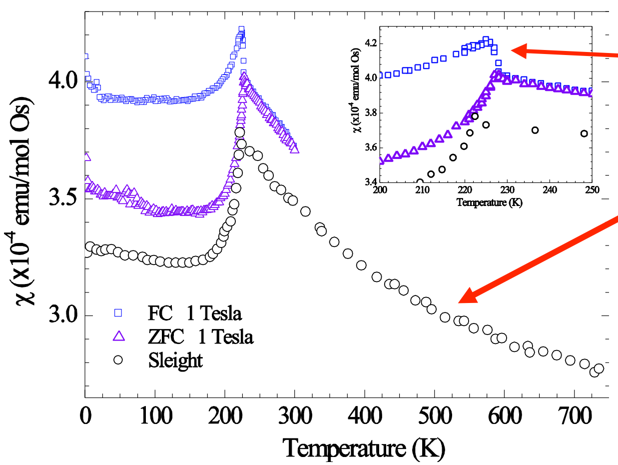
Much smaller than a typical Mott gap.

Fermi surface instability ?

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

Bulk magnetic property:

D. Mandrus *et al.*



Parasitic ferromagnetism

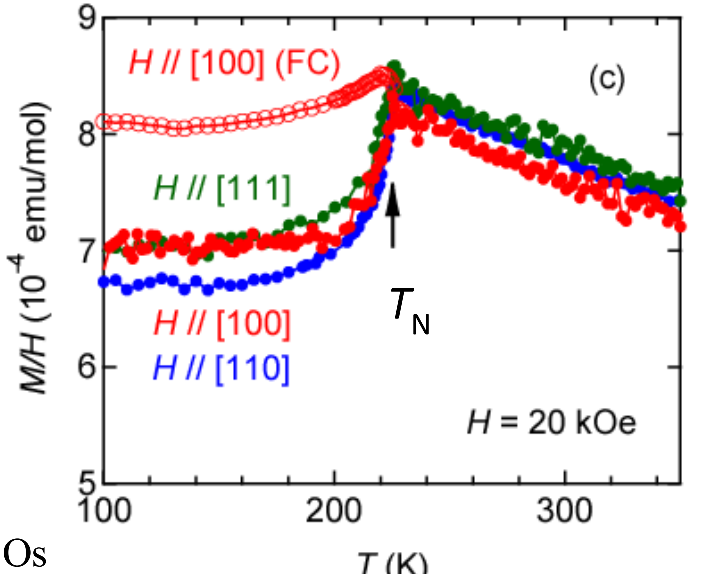
Not Curie-Weiss

Moderate enhancement

$$N(E_F) = 4.62 \ \text{eV}^{-1}/\text{Os}$$

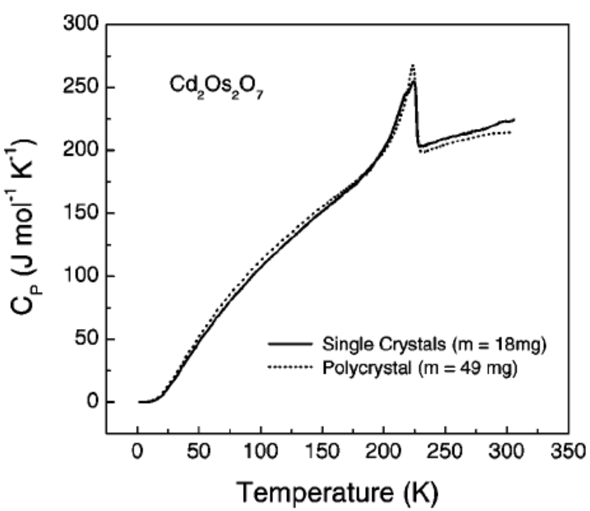
$$\chi_0 = 1.5\times10^{-4} \ \text{emu/mole Os}$$

Singh *et al.* Phys. Rev. B65, 155109 (2002).



J. Yamaura *et al.*

Specific heat:



2^{nd} order transition

Gap opening due to SDW order ?

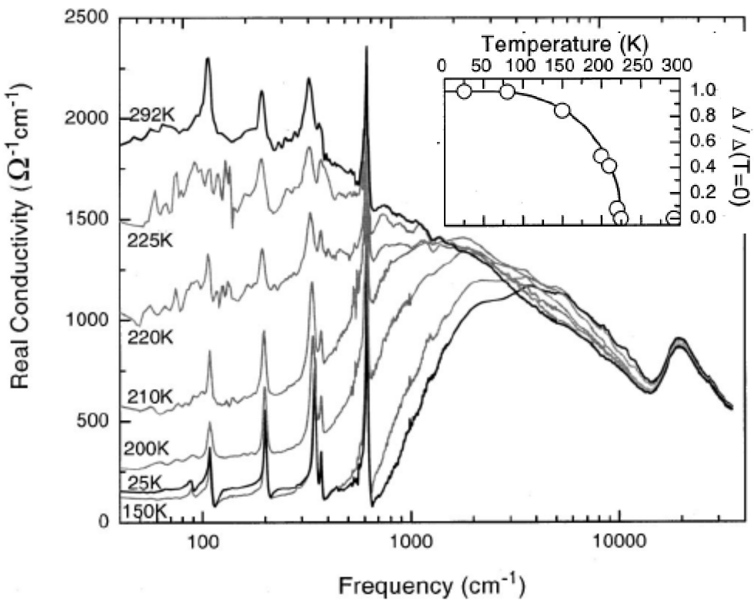
(Slater transition)

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

Structural property:

Optical conductivity on a small single crystal

W. J. Padilla, D. Mandrus, D. N. Basov, PRB **66** (2002) 035120.



BSC-like gap: $2\Delta/kT_c$~5.2

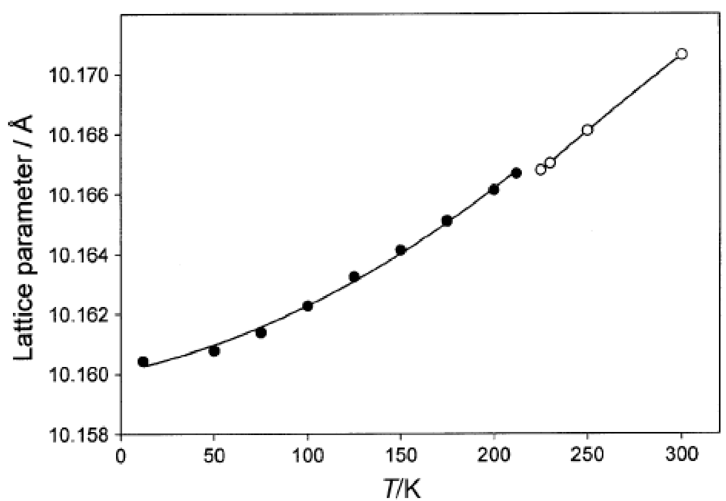
All IR active phonons preserved.
No new mode below T_{MI}

Not CDW
No change of crystal symmetry

Neutron diffraction $^{114}Cd_2Os_2O_7$

Weak lattice anomaly
No symmetry change
No magnetic Bragg peak

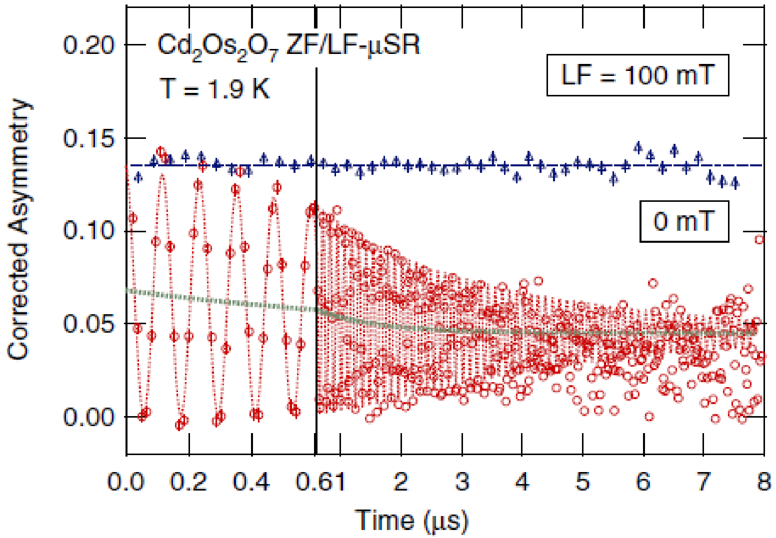
J. Reading and M. T. Weller, J. Mater. Chem. **11** (2001) 2373.

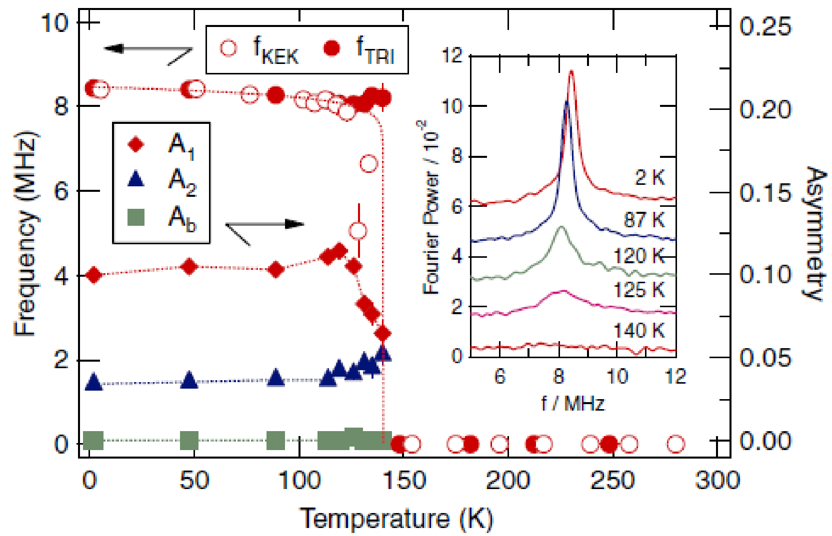


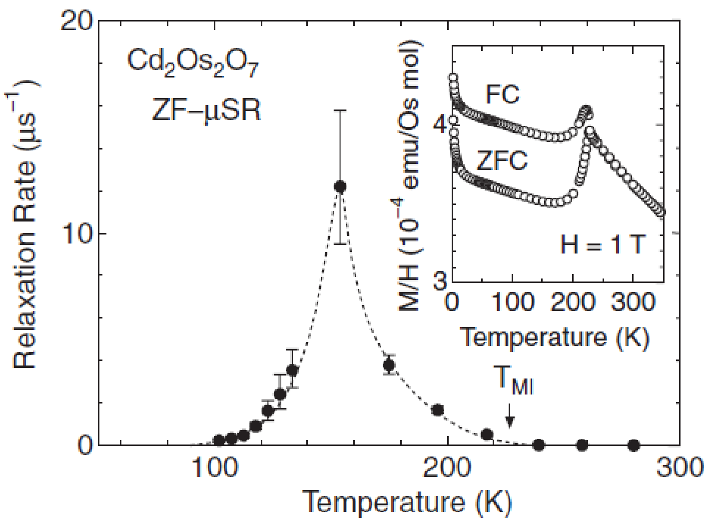
Metal-insulator (MI) transition in $Cd_2Os_2O_7$

μSR on powder sample:

A. Koda *et al.*, JPSJ 76 (2007) 063703.







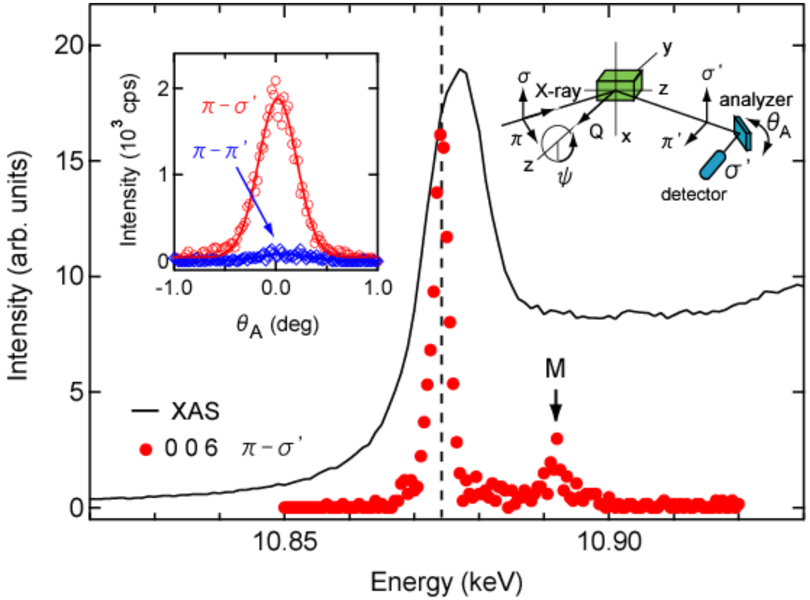
- Static magnetic order appears not at T_{MI} = 230 K but at T = 150 K.
- Distribution of internal field →SDW?

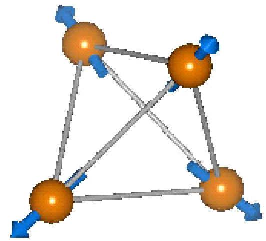
Spin fluctuations observed between T_{MI} and 150 K.

However, no anomaly at 150 K detected by other experiments.

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

Res. X-ray Scattering on single crystal: J. Yamaura *et al.* : PRL, **108,** 247205 (2012).





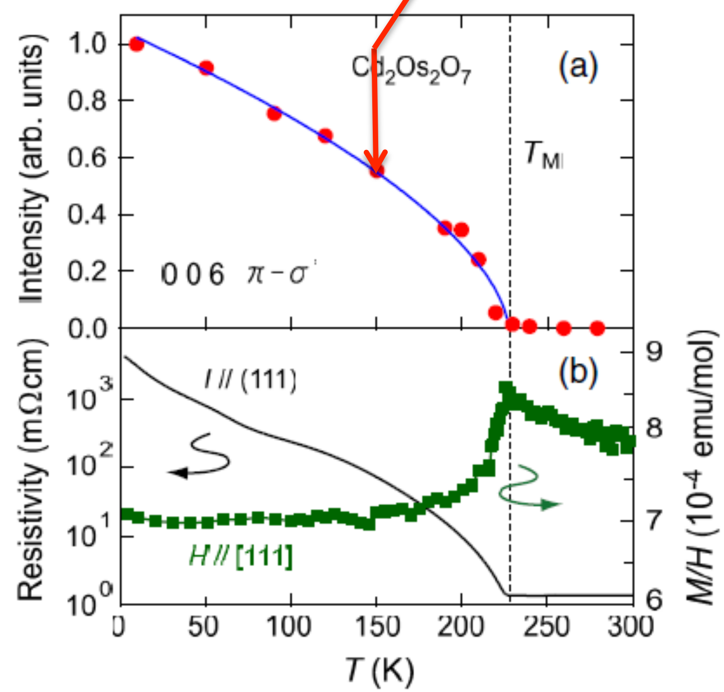
ψ_1 (Γ_3)

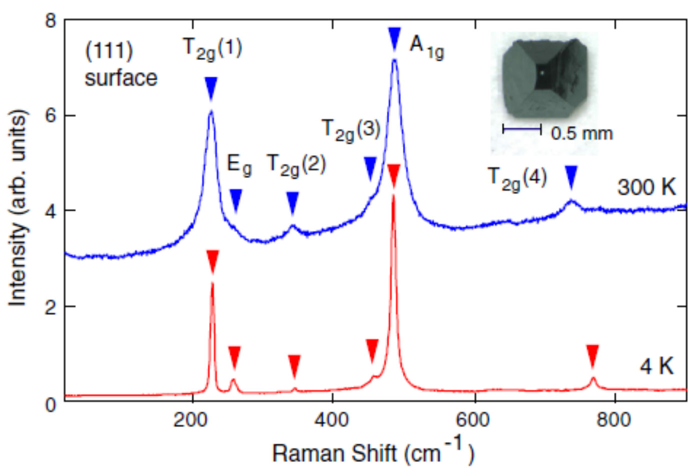
***q* = 0** spin structure with preserved cubic symmetry

(Ising-like all-in-all-out spin structure)

⇒Not the Slater transition

No anomaly at 150 K

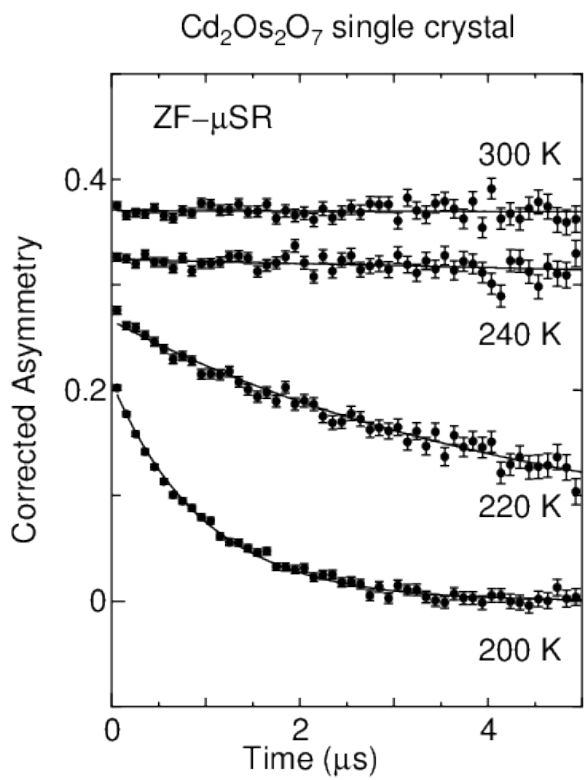


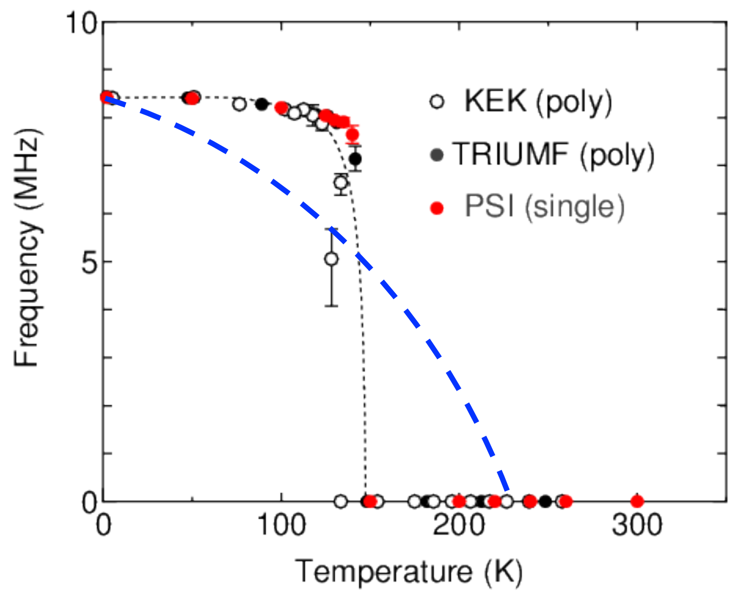


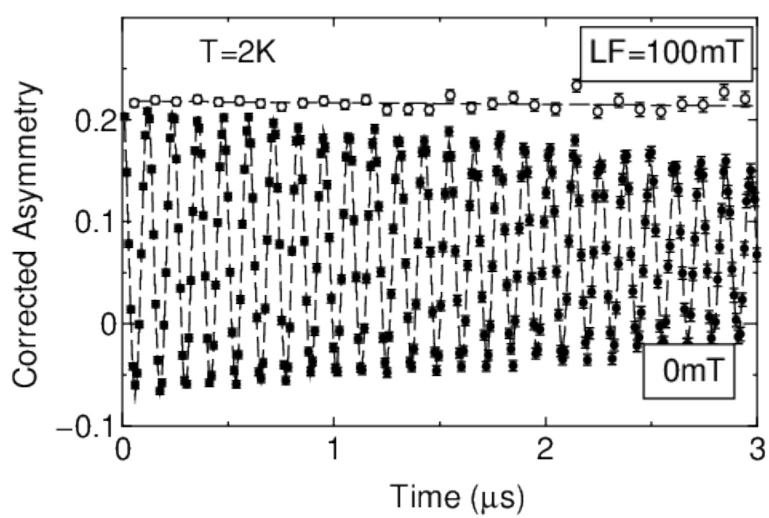
Metal-insulator (MI) transition in $Cd_2Os_2O_7$

μSR on single crystal:

A. Koda *et al.*







- Strong spin fluctuation below T_{MI}=226 K was confirmed.
- Truly static order establishes below ~150 K.

(Not SDW, but comm. AF)

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

μSR on single crystal: **Issue: Spin fluctuation rate**

A. Koda *et al.*

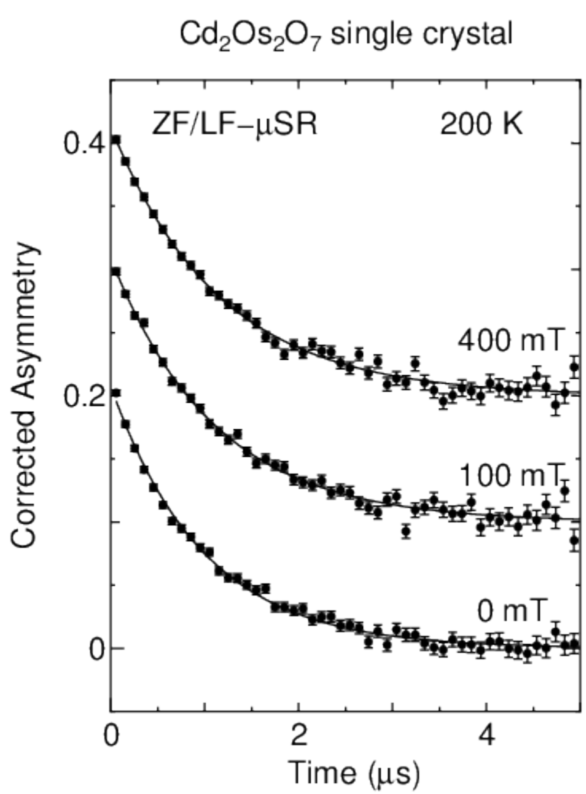
Longitudinal depolarization
$\lambda(\omega)$ [$=1/T_1$]

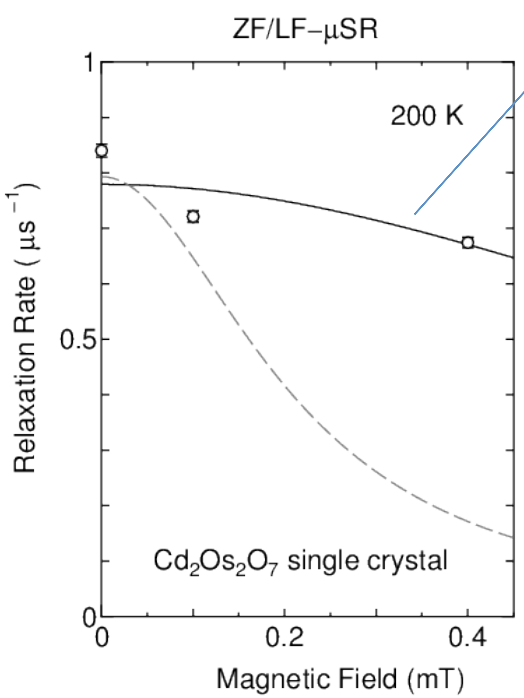
Spin fluctuation rate ν

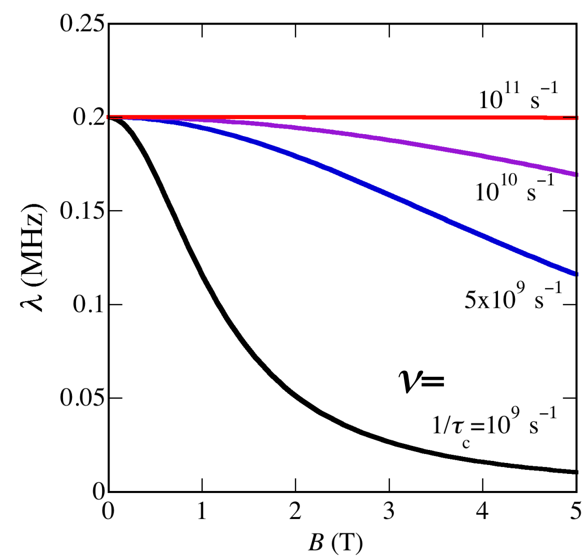
$$\lambda(\omega) = \frac{2\delta^2 \nu}{\omega^2 + \nu^2},$$
$$\omega \equiv \gamma_\mu B$$

δ : Hyperfine field

e.g., at 200 K
δ = 18.2(4) MHz
ν = 846(34) MHz





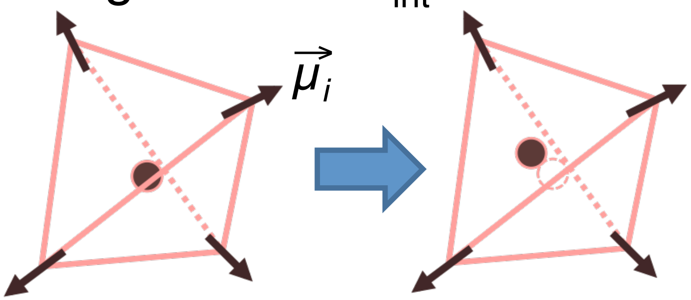


Metal-insulator (MI) transition in $Cd_2Os_2O_7$

μSR on single crystal: **Issue: Muon site and δ**

A. Koda *et al.*

$T < 150$ K: Muon site may be slightly off-center in the Os tetrahedron cage so that H_{int} does not cancel out.

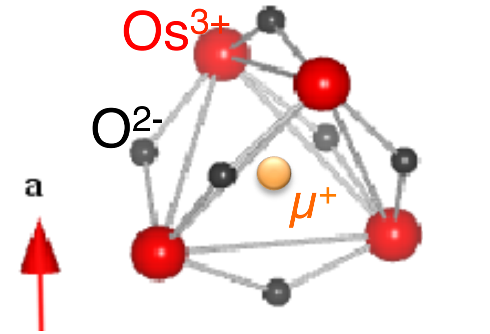


$H_{\text{int}} = 0$ $\quad$ $H_{\text{int}} > 0$

$$H_{\text{int}} = \sum_i \hat{A}_i \vec{\mu}_i$$

i.e. H_{int} is a vector sum.

$$\hat{A}_i^{\alpha\beta} = \frac{1}{r_i^3}\left(\delta_{\alpha\beta} - \frac{3r_i^\alpha r_i^\beta}{r_i^2}\right)$$



…μ^+ is attracted by negative charge of oxygen?

$T > 150$ K: Hyperfine interaction:

$$2\delta^2 \propto \sum_{i,\alpha,\beta} \left(A_i^{\alpha\beta}\right)^2 \quad (\alpha = x, y; \beta = x, y, z)$$

i.e. δ is a rms value of dipole tensor.

$A_\mu/\gamma_\mu = 0.21$ T/μ_B (Calculated for 4 nn Os ions)

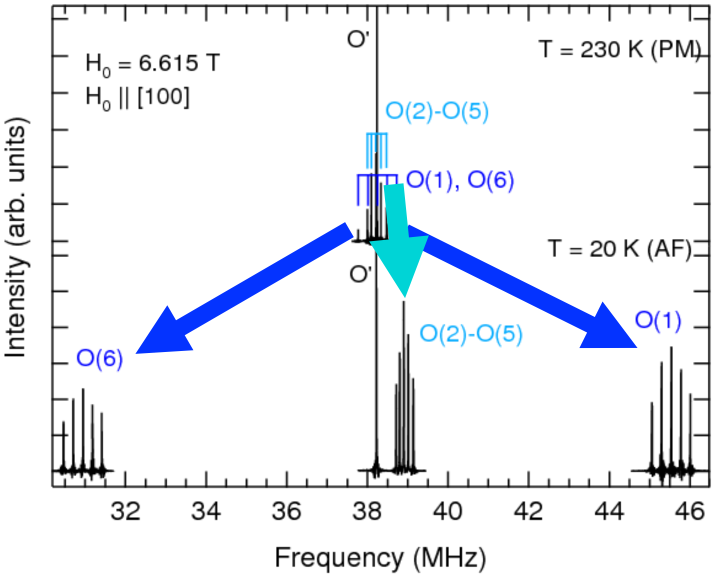
$\delta = 27.8(9)$ MHz (experiment) $\longrightarrow$ $|\mu'_i| = \delta/\gamma_\mu A_\mu \doteqdot 0.15(1)\mu_B$

…having no direct relationship with H_{int}.

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

NMR on single crystal:

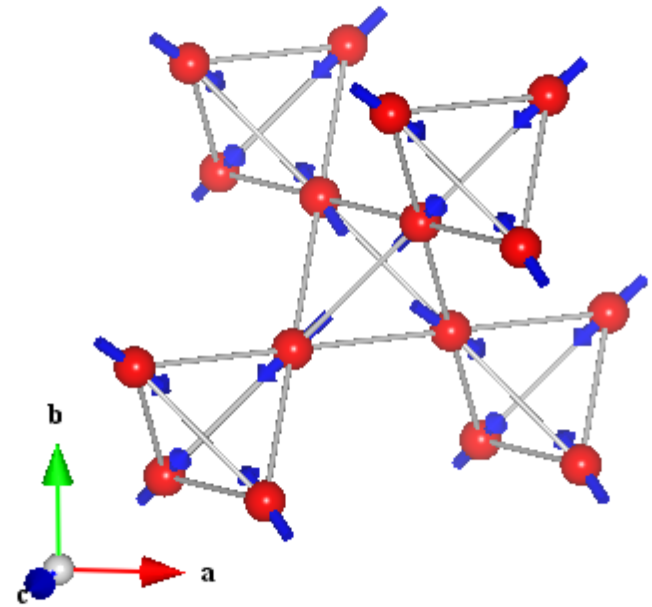
I. Yamauchi *et al.*

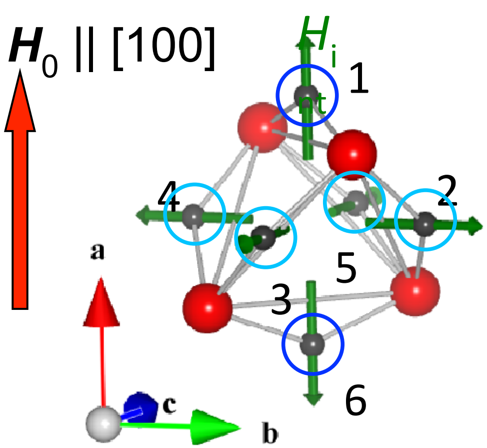


FT ^{17}O NMR spectrum

$\Psi_1(\Gamma_3)$

all-in/all-out spin structure





Estimated ordered moment is
1.0~1.5 μ_B/Os
(from HF parameter for $T > T_{MI}$)

Metal-insulator (MI) transition in $Cd_2Os_2O_7$

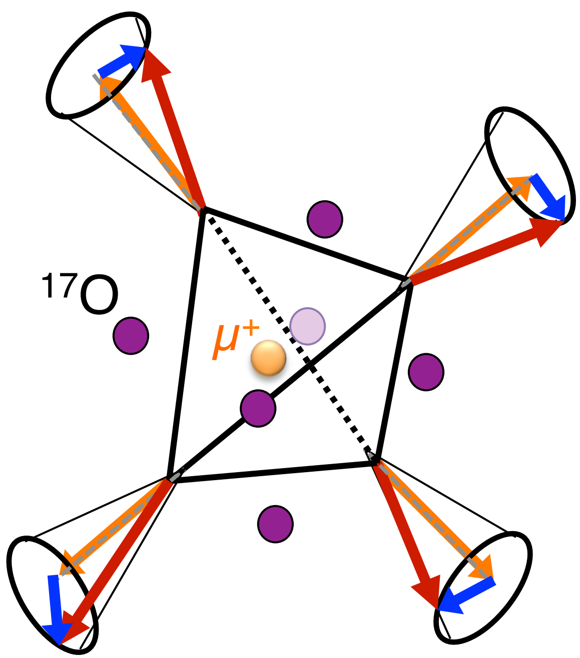
μSR vs NMR:

A. Koda *et al.*

Muon probes fluctuation of the Os moments perpendicular to [111] axis?

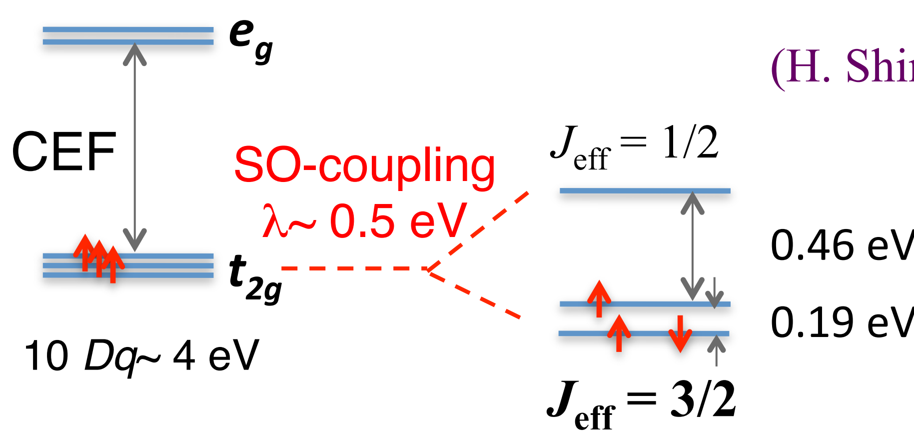
Os moment parallel to [111] : 1-1.5μ_B

Os moment perpendicular to [111] : 0.15μ_B



Origin: anisotropic exchange interaction under relatively large spin-orbit interaction for *5d* electrons?

$Cd_2Os_2O_7$ ($5d^3$)



(H. Shinaoka)

Tetragonal splitting Δ~ 0.29 eV

Summary

- The ground state property of 5d electron compounds is subject to strong influence of spin-orbit interaction whose energy scale is comparable with that of the Hund coupling and/or tetragonal CF.

- In an iridium spinel $CuIr_2S_4$, it is likely that Ir^{4+} ions are in $J_{eff}=1/2$ state and are mapped onto the Kitaev model due to strong anisotropy of exchange interaction, where the observed random magnetism may be interpreted as a frozen spin liquid state due to inter-octamer interaction.

- In an osmium pyrochlore $Cd_2Os_2O_7$, anomalous enhancement of spin fluctuation is observed over a wide temperature range below MI transition. While theoretical study based on LSDA+U suggests Mott transition as possible origin of MI transition, the strong residual fluctuation suggests important role of geometrical frustration.