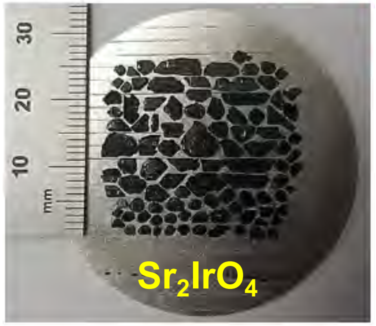
Magnetic insulating states with strong spin-orbit coupling in osmates and iridates

Stuart Calder
Oak Ridge National Laboratory

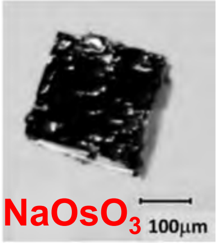
Layout: Investigating the interactions in osmates ($5d^3$) and iridates ($5d^5$) with neutron and resonant x-ray scattering

- Motivation: General interest in 5d oxides and MITs

- Sr_2IrO_4: J_{eff}=1/2 Mott iridate $5d^5$
  - Links to cuprates
  - Doping with Ru and Sn



- $NaOsO_3$: Magnetically driven MIT
  - Following the magnetic interactions through the MIT

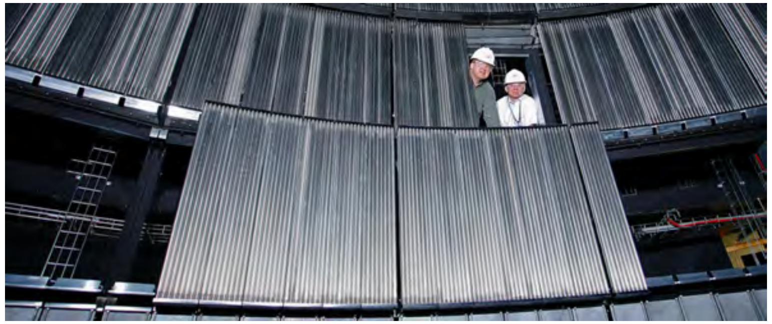


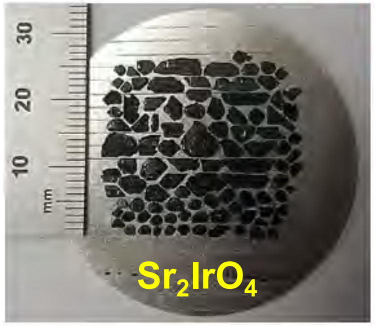
Collaborators

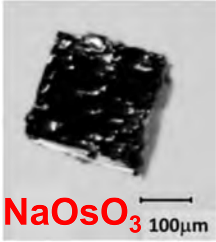
- Neutron scattering
  - D. M. Pajerowski (ORNL)
  - M. B. Stone (ORNL)
  - M. Matsuda (ORNL)
  - A. D. Christianson (ORNL)
  - M. D. Lumsden (ORNL)
  - A. E. Taylor (ORNL)

- Resonant x-ray scattering
  - J. W. Kim (APS)
  - M. Upton (APS)
  - D. Casa (APS)
  - D. Haskel (APS)
  - M. Moretti-Sala (ESRF)
  - D. F. McMorrow (UCL)
  - **J. G. Vale (UCL)**

- Sample synthesis
  - A. F. May (ORNL) [Sr_2IrO_4]
  - G-X. Cao (ORNL) [Sr_2IrO_4]
  - K. Yamaura NIMS [$NaOsO_3$]



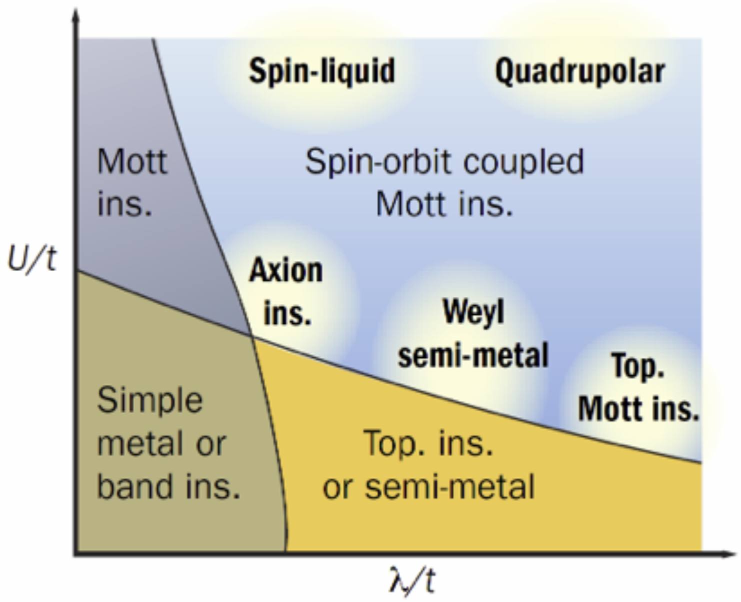




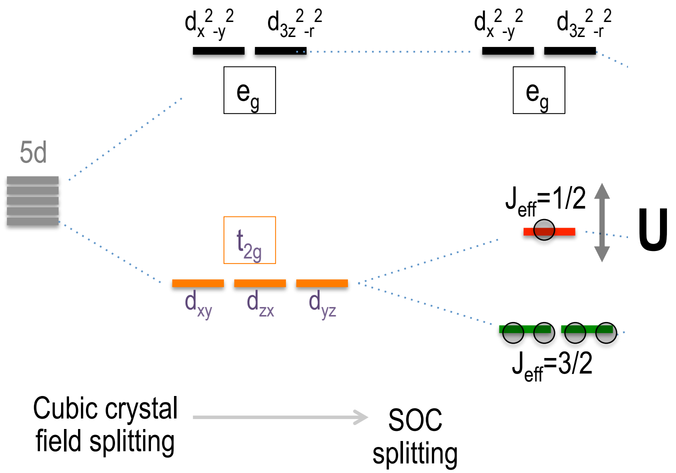
General Motivation: probing magnetic interactions in $5d^3$ and $5d^5$ compounds with MITs

Interaction	*3d* (e.g. Cu)	*5d* (e.g Ir)
Coulomb interaction, *U*	3-5 eV	1-2 eV
Spin-orbit coupling (λ)	0.01 eV	0.5 eV
Crystal field splitting	1-2 eV	1-4 eV

5*d*: U~λ



Witczak-Krempa, *et al.*, *An. Rev. Cond. Mat.* (2014)



Ir^{4+} ($5d^5$): SOC J_{eff}=1/2 Mott insulator → Sr_2IrO_4

Os^{5+} ($5d^3$): Magnetically driven MIT → $NaOsO_3$

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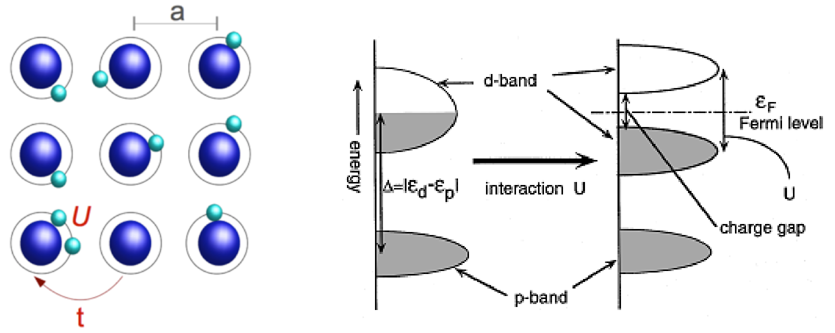
Mott MIT (1949)

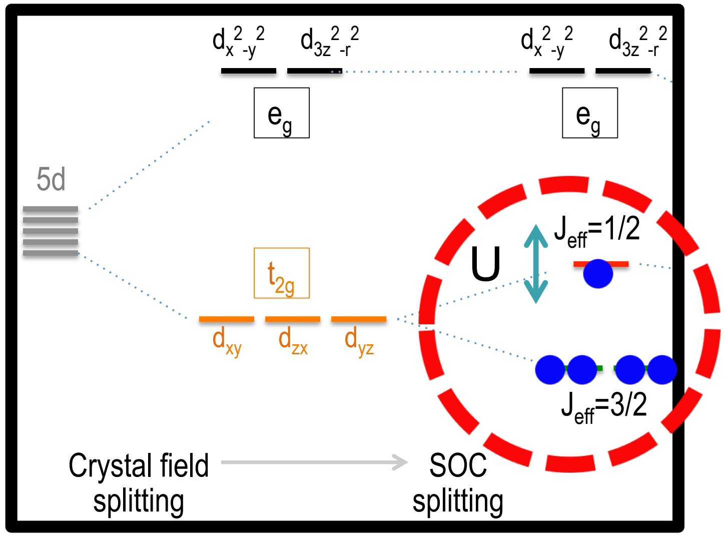
Coulomb interactions, *U*, open gap at Fermi energy resulting in a MIT. Discontinuous.



Preformed moments add to model, but do not drive MIT.

$5d^5$ Iridates (SOC driven Mott MIT - 2009)

Sr_2IrO_4



L-S coupling

Magnetic MIT (1951)

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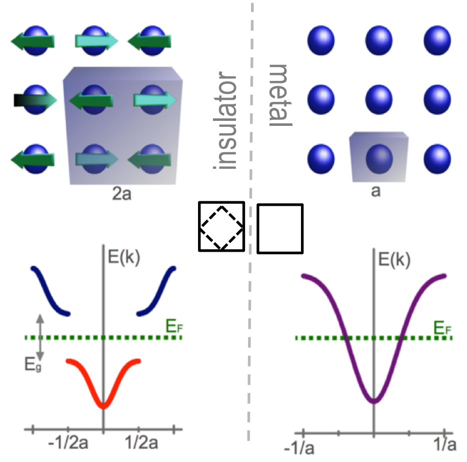
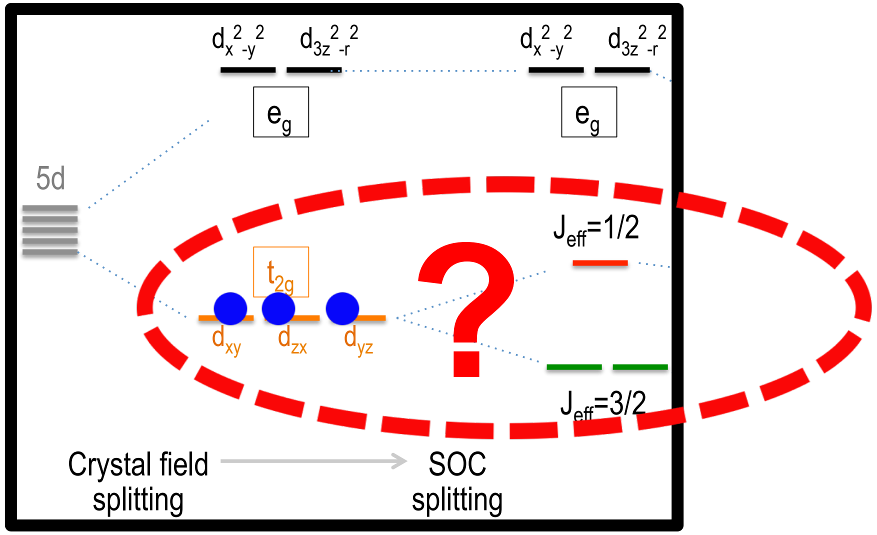


Slater: Ordered moments create mean field confining electrons.

Lifshitz: Small *U* and large hybridization drive renormalization of the Fermi surface

$5d^3$ Osmates (Magnetic MIT: Slater/Lifshitz - 2009)

$NaOsO_3$ (2009)



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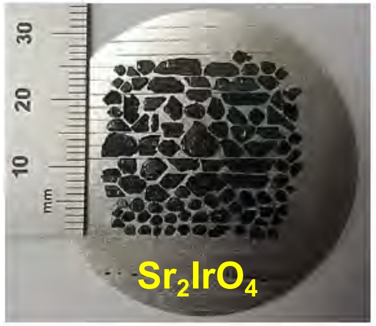

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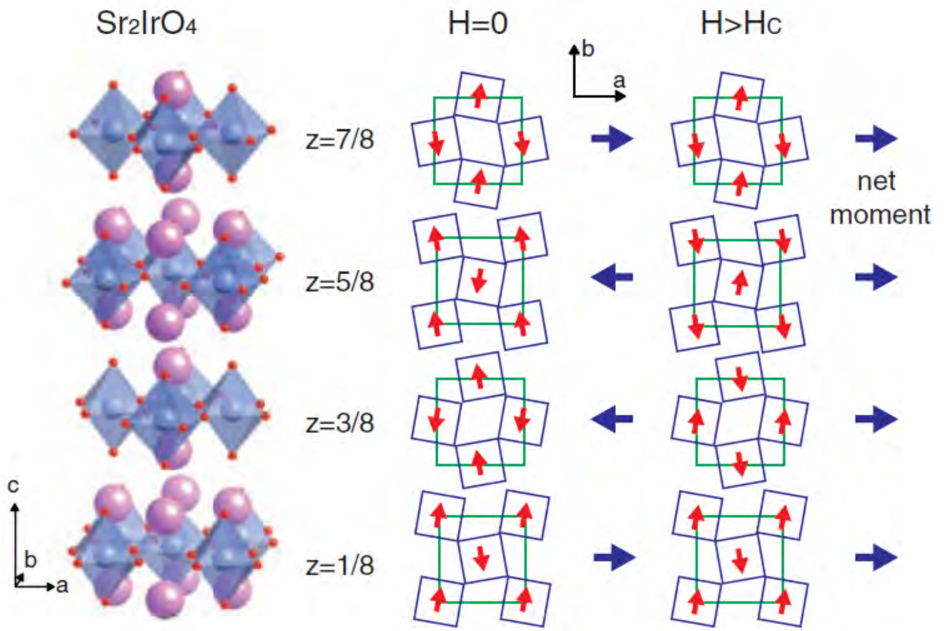
Sr_2IrO_4: J_{eff}=1/2 Mott insulator

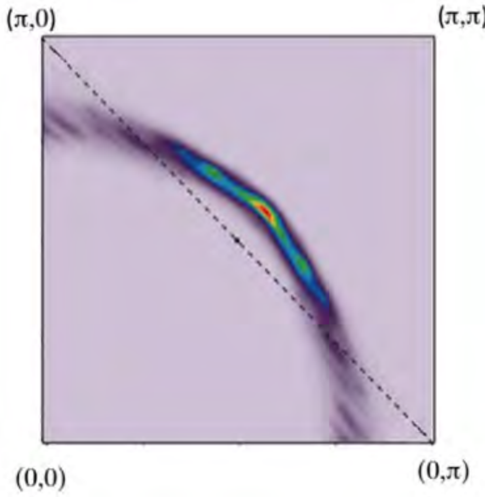
- Layered perovskite structure ($I4_1/a$ #88)
- T_N=240 K
- Spins follow in-plane IrO_6 canting (~11°) due to SOC
- H_c>0.3 T → spins reorder within *ab*-plane



Kim, Science 323, 6 (2009)

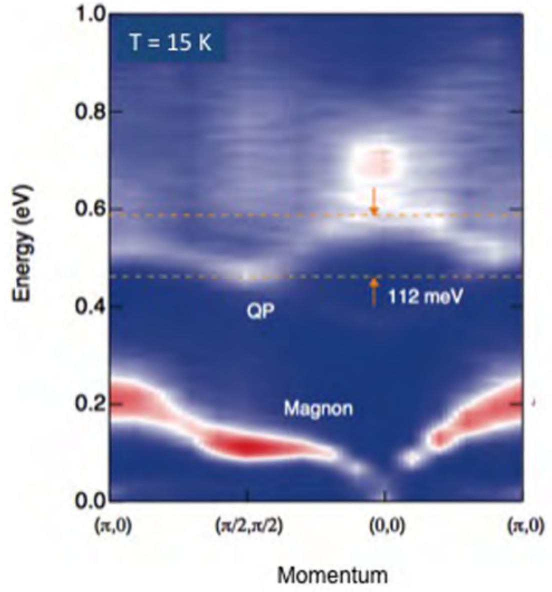
Fermi arcs

BJ Kim et al., Science (2014)



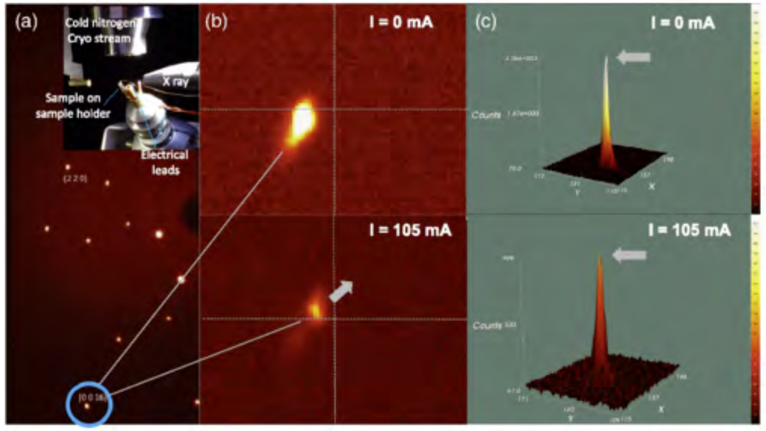
Spin-orbit exciton quasi particle

JH Kim Nat. Commun. 5, 1-6 (2014)



Electric control of lattice due to SOC

Cao, PRL 120, 017201 (2018)



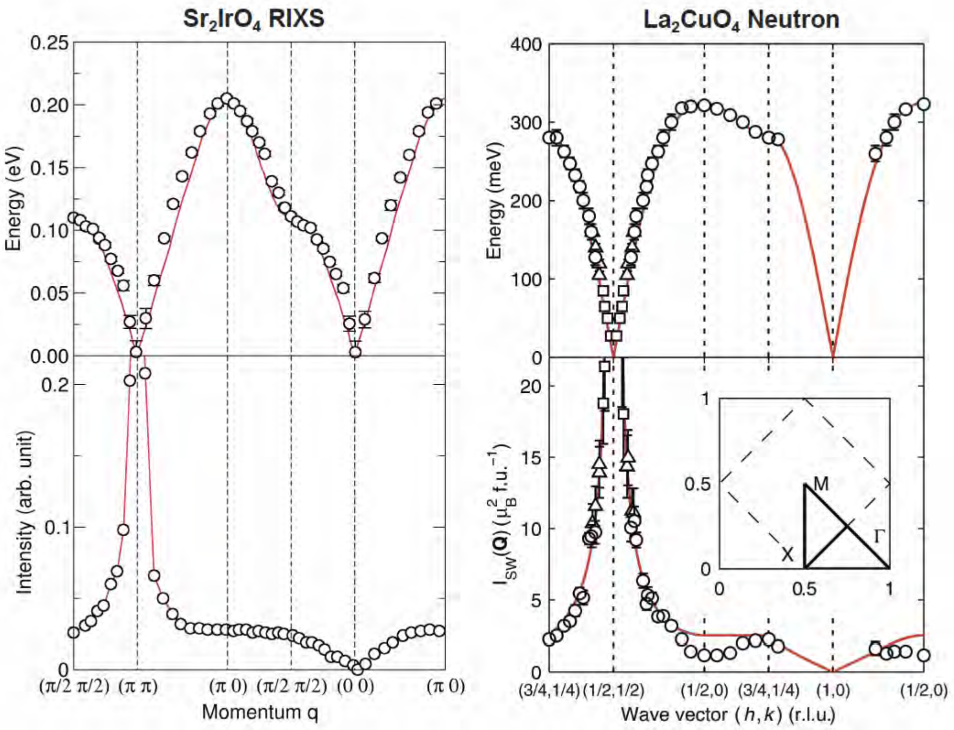
Sr_2IrO_4: Link to cuprates

- Comparing Sr_2IrO_4 and La_2CuO_4:
  - Analogous magnetic and crystal structure
  - Mott insulators
  - Spin-1/2 moments
  - Spin-excitations described by isotropic 2D Heisenberg

$$\mathcal{H} = \sum_{i,j} J_{ij}\vec{S}_i.\vec{S}_j + \Gamma S_i^z S_i^z + D(S_i^x.S_j^y - S_i^y S_j^x)$$

- But, SOC varies greatly: λ_{SO}[Ir]≈0.5eV and λ_{SO}[Cu]≈0.01eV
  - **Debate on anisotropy and spin-gap size: 0-20 meV**

Magnetic excitations



J. Kim et al., PRL 2012

Coldea et al., PRL 2000

Phys. Rev. B **92**, 020406 (2015)
Phys. Rev. B **96**, 075162 (2017)
Phys. Rev. B **93**, 024405 (2016)
Phys. Rev. B **89**, 180401 (2014)

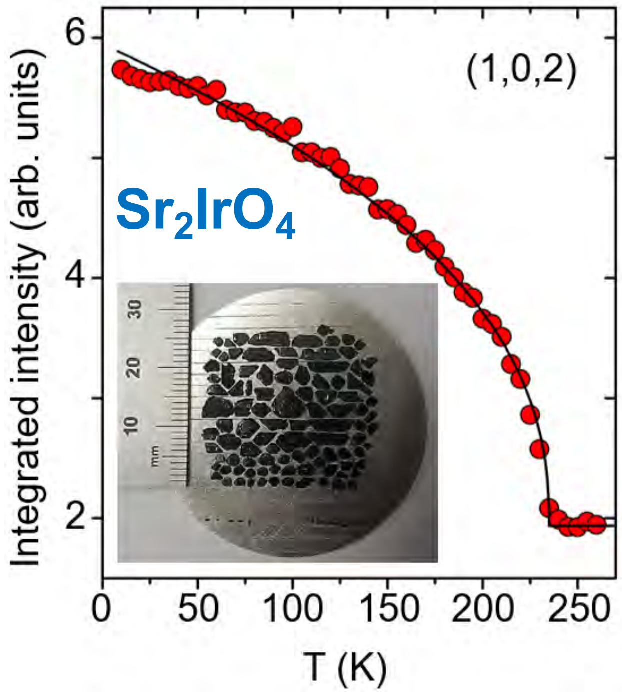
(1) Are the magnetic interactions 2D?

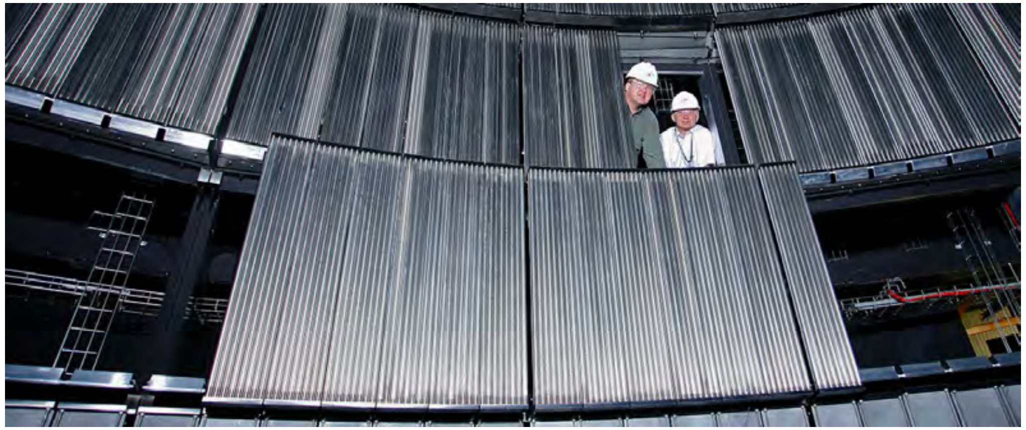
(2) Probe spin-gap to access role of anisotropy?

Inelastic neutron scattering (INS)

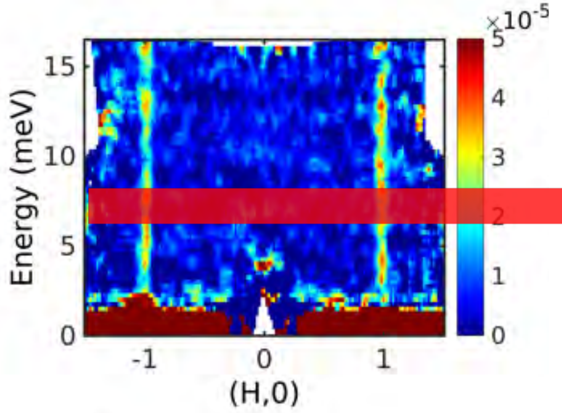
- Powerful insights into magnetic interactions
- Measures 4D (H,K,L,Energy) with sub-meV resolution
  - S(Q,ω) exactly defined

- <u>Challenges for iridates:</u>
  - Small crystal size → Coaligned ~100 crystal array for total mass 1.1 grams
  - Neutron absorption → Optimized sample and instrument geometry with large detectors
  - Small moment (~$0.25\mu_B$) → All of the above



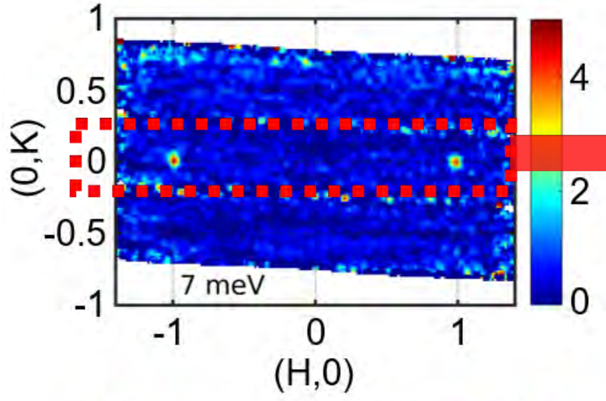


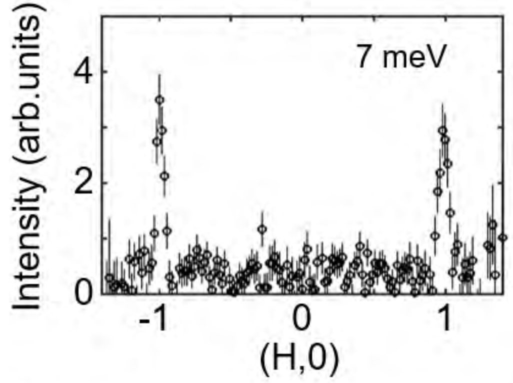
INS results for Sr_2IrO_4: Looking at (H,K,L,E) data at 10 K

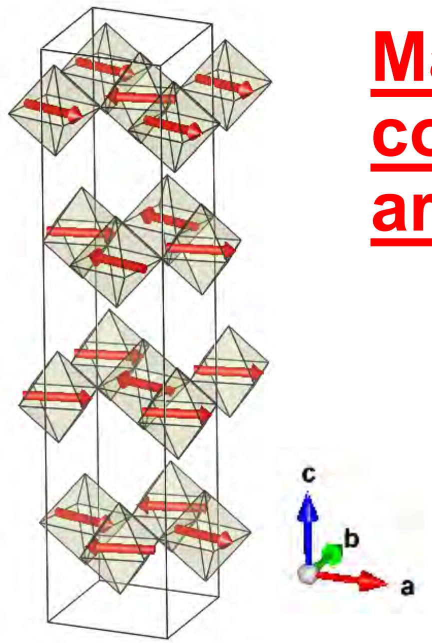


(H,K) momentum dependence

→ in-plane magnetic interactions



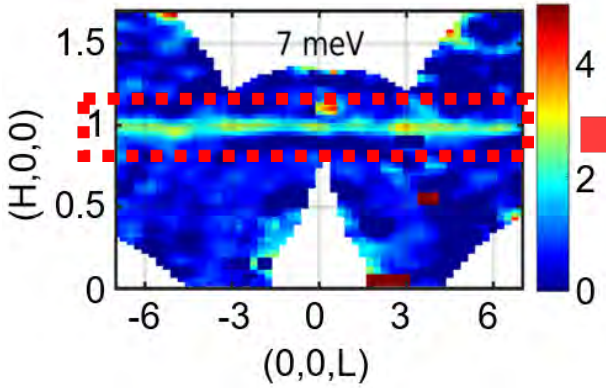


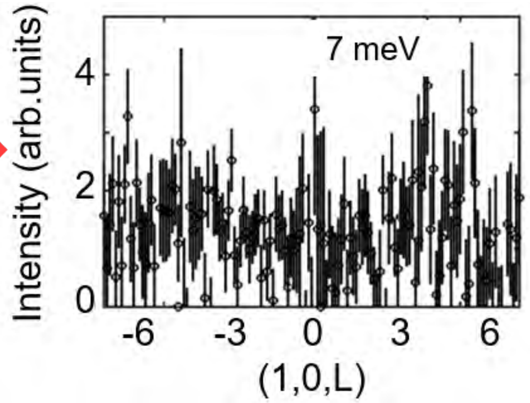


Magnetic correlations are strongly 2D

No momentum dependence along L

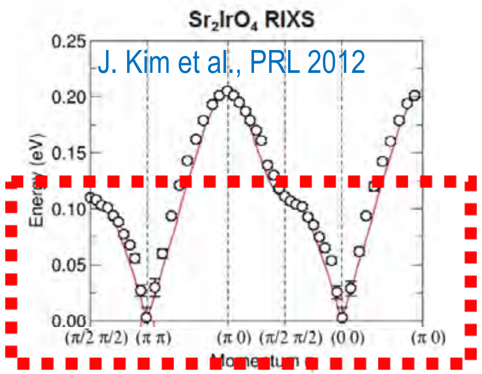
→ Negligible magnetic interactions between layers

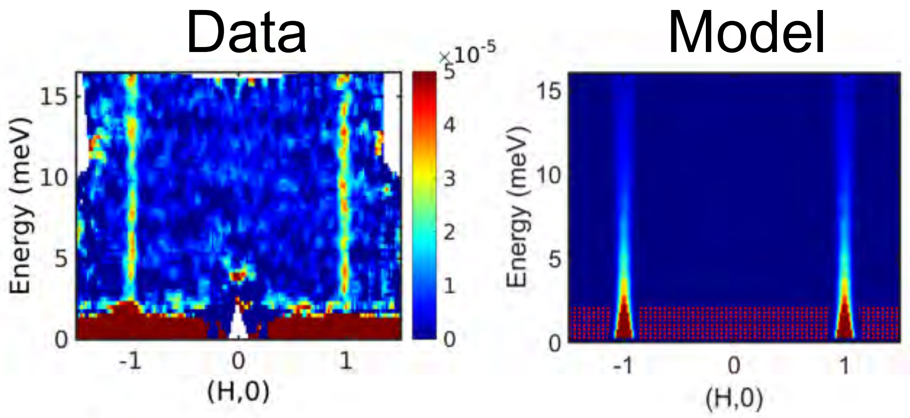


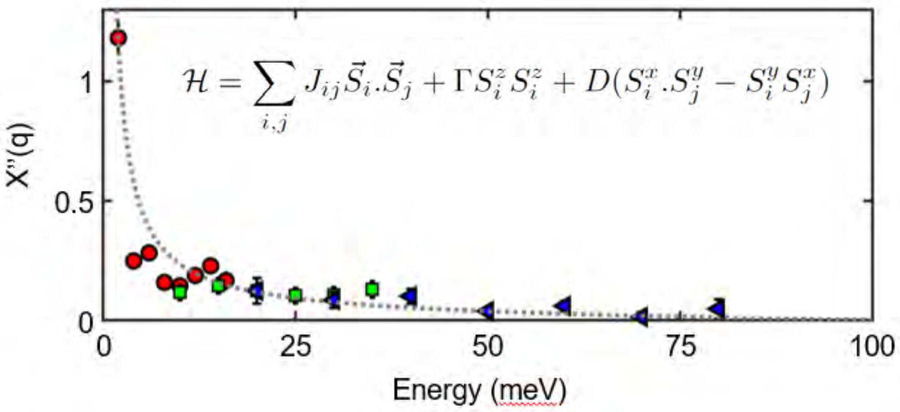


INS results for Sr_2IrO_4: Model dispersion

- RIXS studies have covered full in-plane magnetic excitations with ~30 meV resolution.
- Magnetic excitations measured up to 80 meV with neutrons.
- Modelled with isotropic 2D Heisenberg model.
- <u>Low energy in-plane spin-gap?</u>



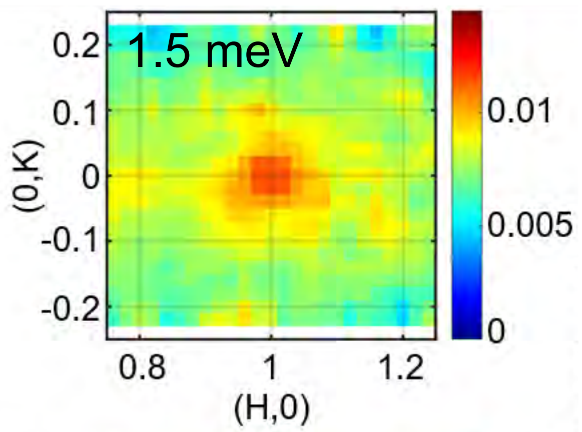


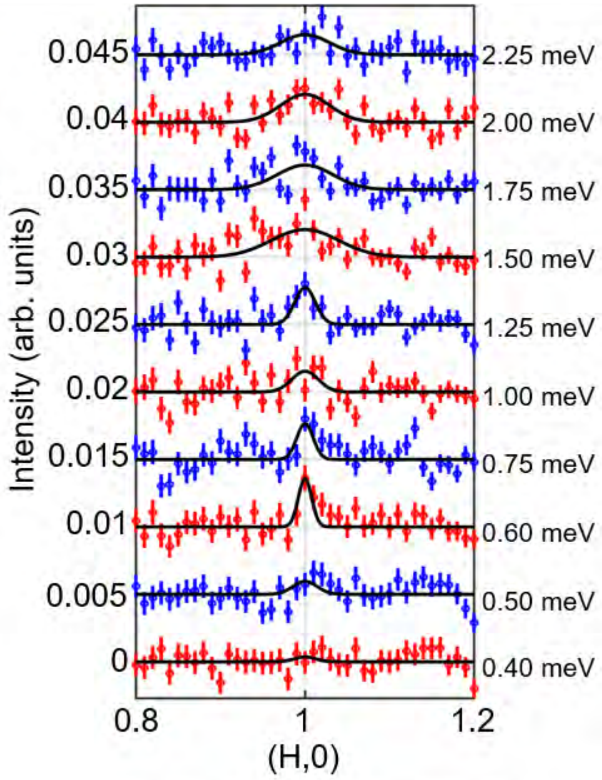


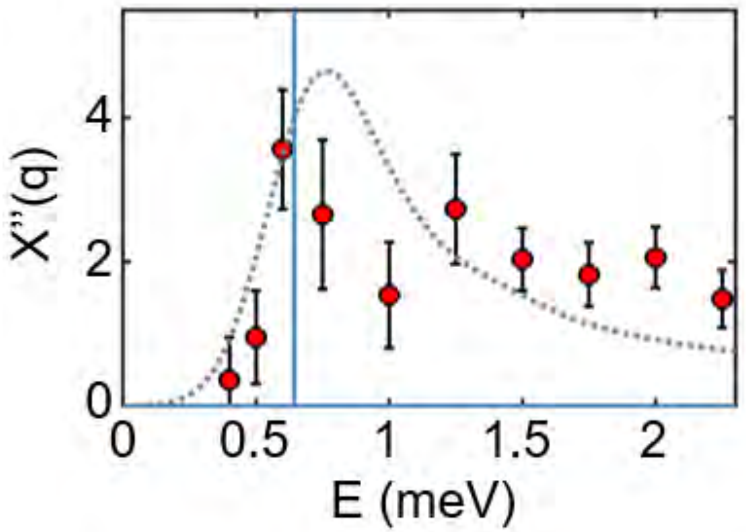
INS results for Sr_2IrO_4: Low energy neutron measurements

- Instrument resolution of 0.1 meV (CNCS)

Spin-gap of 0.6(1) meV







Finite, but small on the scale of excitations (~200 meV)

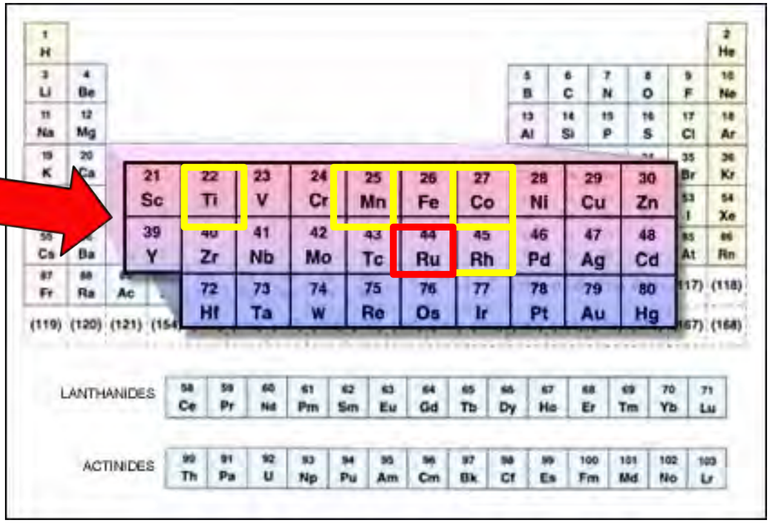
- Coupling of spins to lattice distortion potential route of spin-gap
- **2D isotropic Heisenberg model a good approximation in Sr_2IrO_4 and La_2CuO_4**

Doping to tune J_{eff}=1/2 state, magnetic structure and interactions?

Previous doping on Ir site

Rh doping creates Ir^{5+}/Ir^{4+}

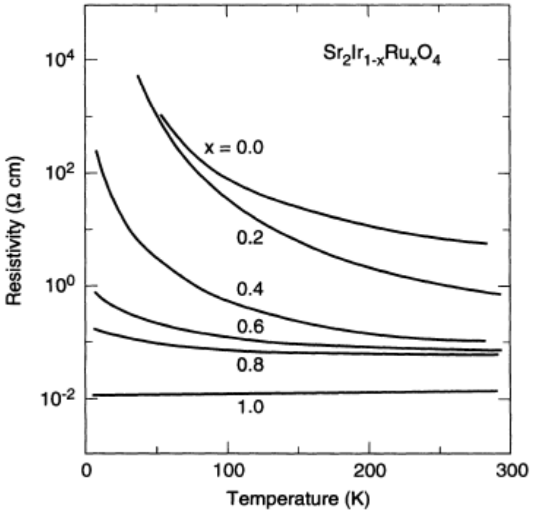
Try Ru doping

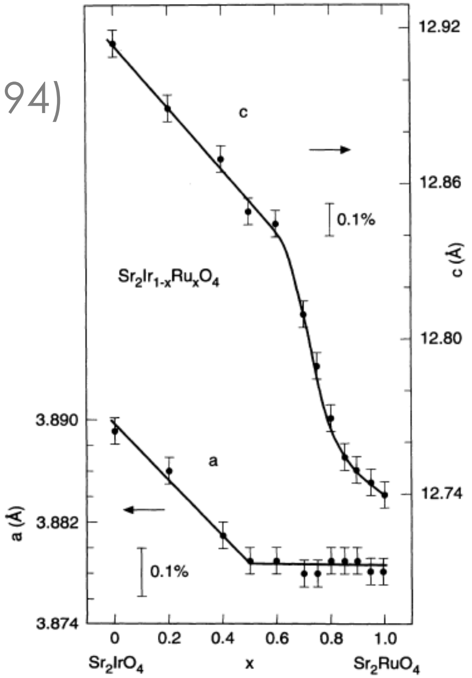


$Sr_2Ir_{1-x}Ru_xO_4$

- Sr_2IrO_4: Magnetic Mott-like insulator.
  - Ir^{4+} (d^5) with J_{eff}=1/2.
- Sr_2RuO_4: Metallic with unordered local moment.
  - Ru^{4+} (d^4) with S=1.

Cava et al., PRB **49**, 17 (1994)

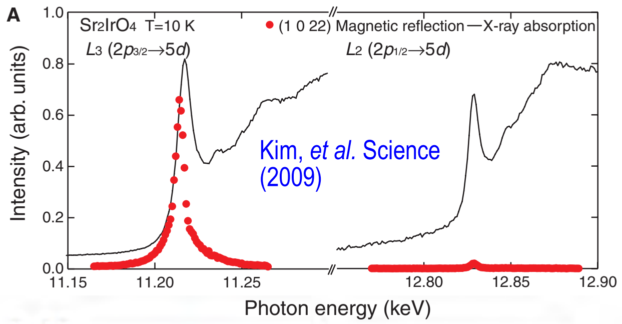




Resonant X-ray and XMCD/XAS:
$Sr_2Ir_{1-x}Ru_xO_4$ (x=0-0.4)

- J_{eff}=1/2 for all x.
- Ir^{4+} ($5d^5$) for all x.

Sr_2IrO_4



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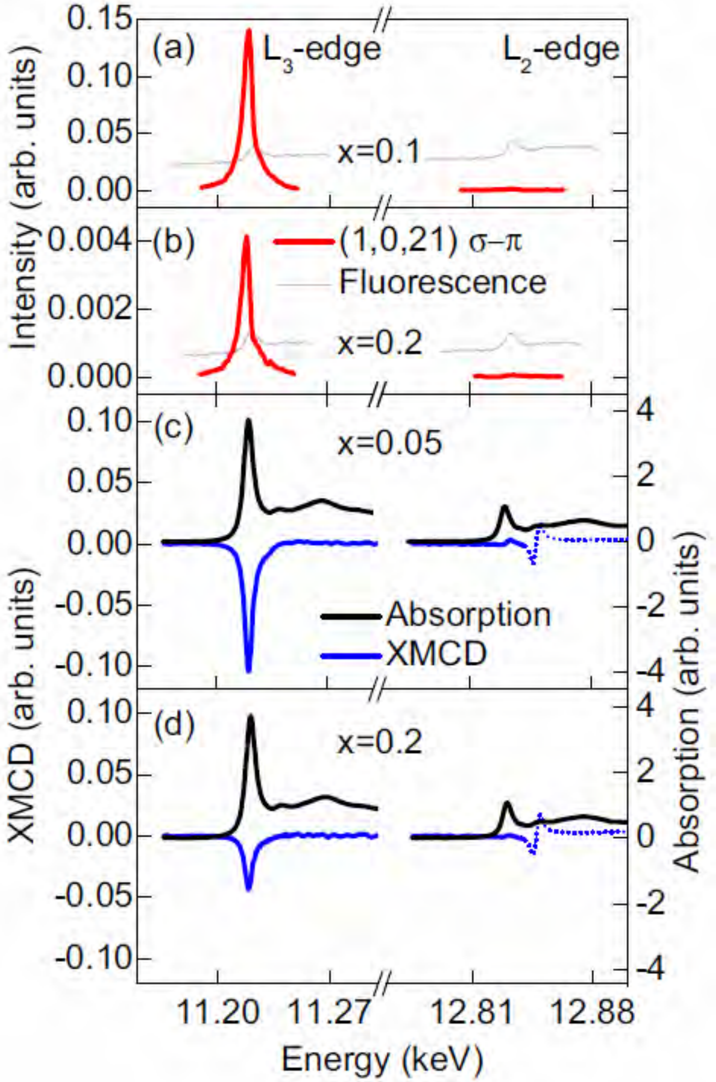
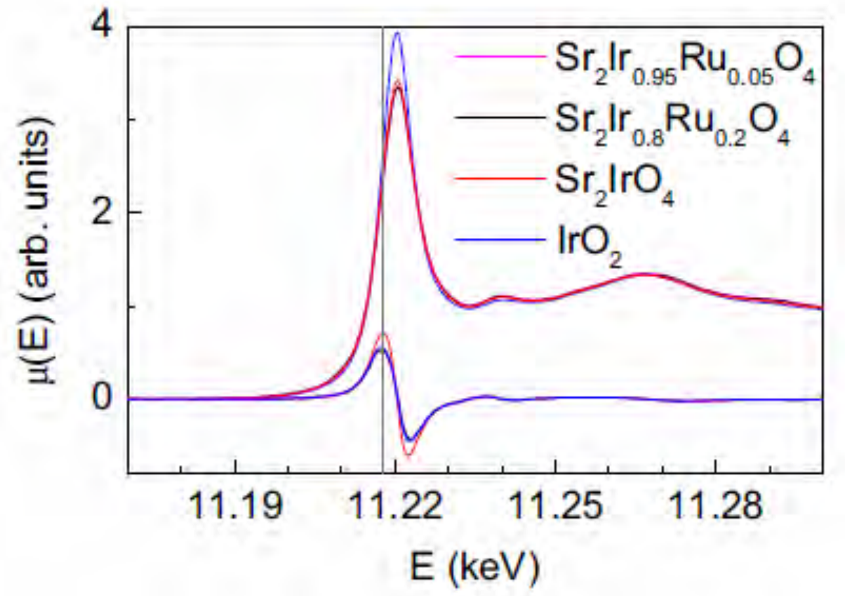
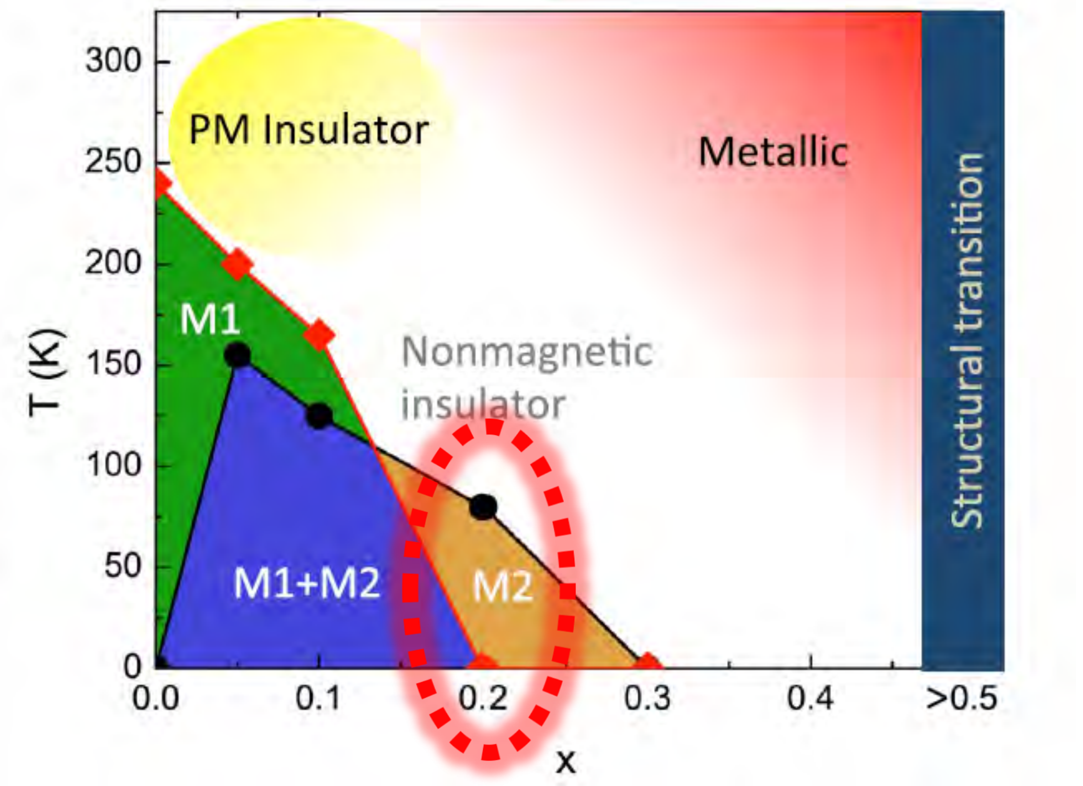
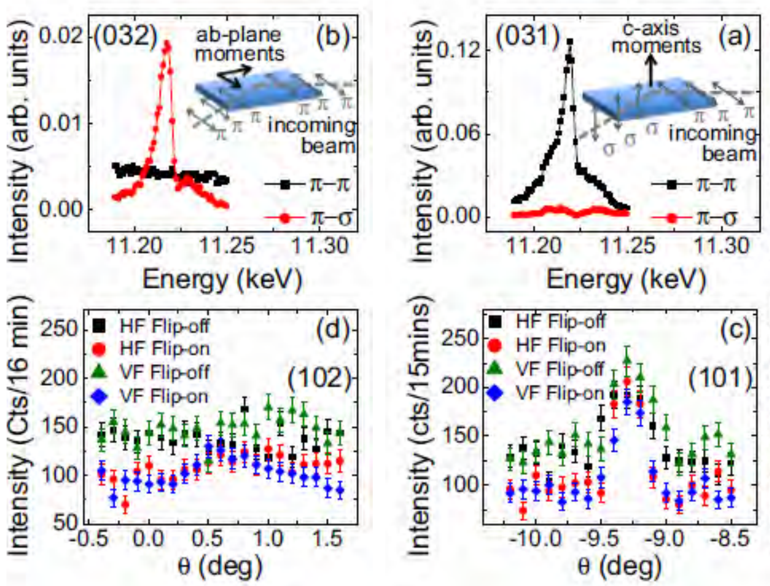


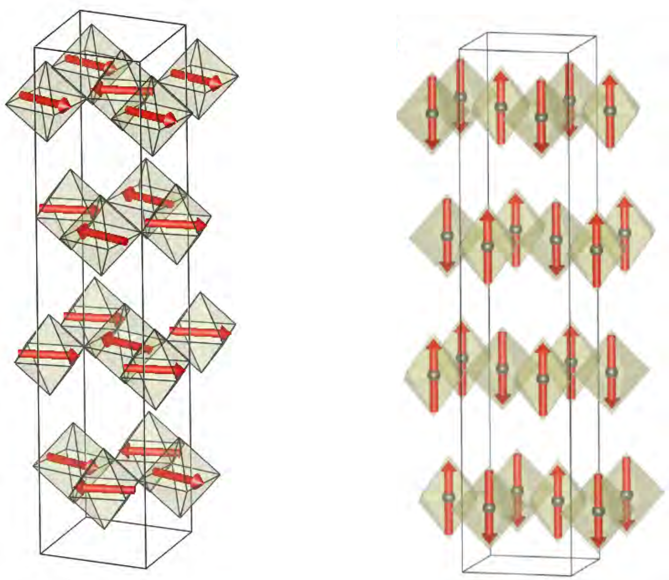


$Sr_2Ir_{1-x}Ru_xO_4$ Magnetic and MIT phase diagram

- Polarized neutron and resonant x-ray diffraction.
- Evolution from *ab*-plane to *c*-axis aligned moments.

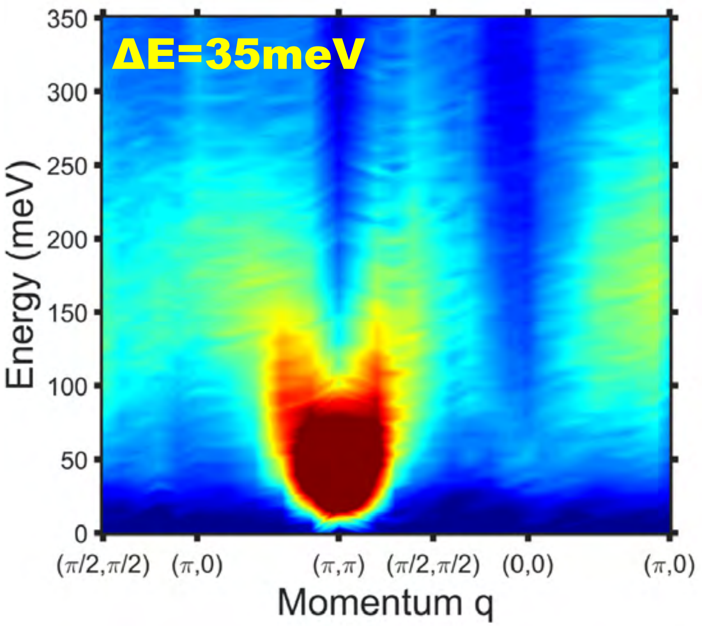


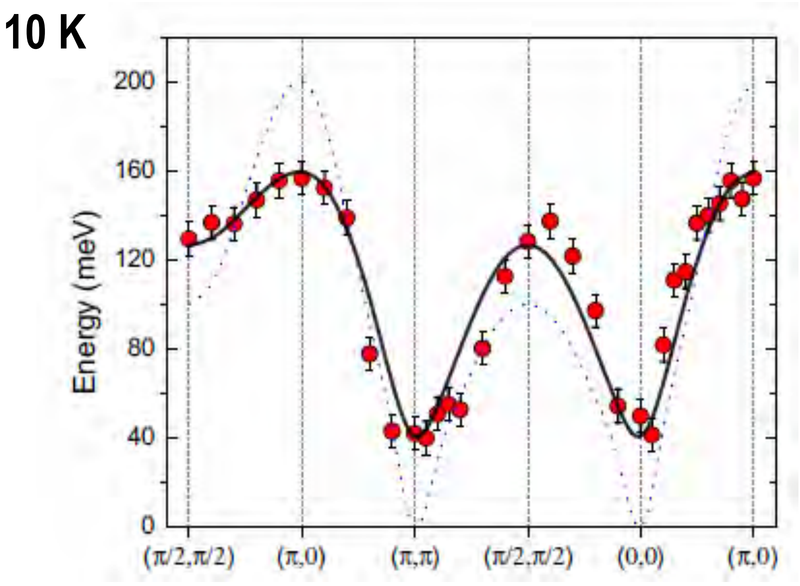




- Magnetism at large x → Percolation

RIXS: Spin-wave excitations in $Sr_2Ir_{0.8}Ru_{0.2}O_4$ at 10 K





Sr_2IrO_4 (dotted blue line):

J=60meV, J'=-20meV, J''=15meV

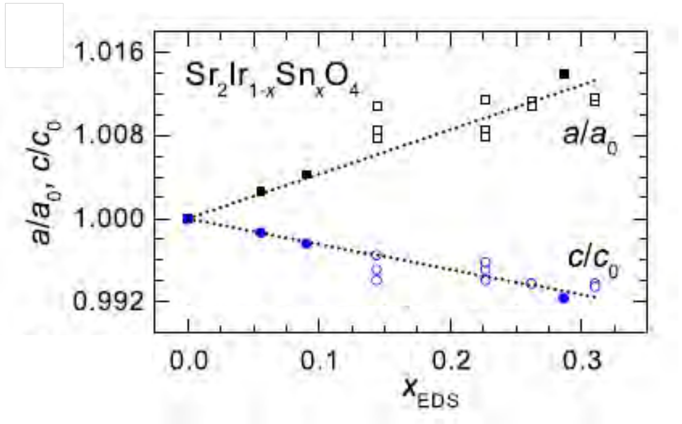
$Sr_2Ir_{0.8}Ru_{0.2}O_4$ (solid line):

J=42meV, J'=-16meV, J''=0meV

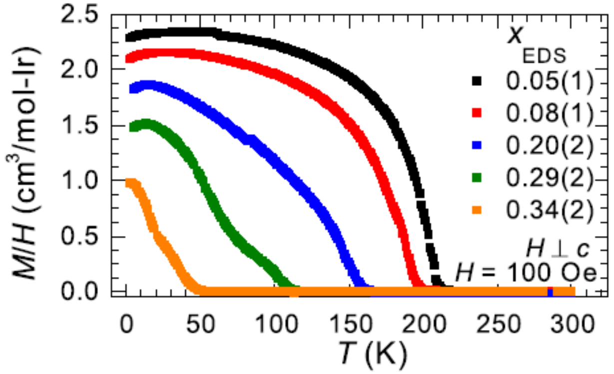
- **J''=0meV**: 1 in 5 Ir atoms disrupted by Ru substitution
  → interactions beyond J and J' suppressed.
- **Large spin gap of 40meV.**

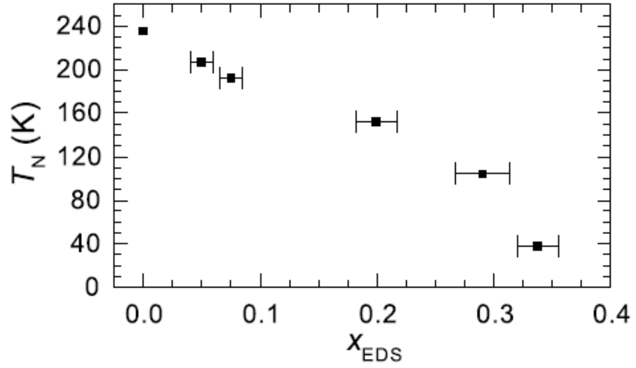
$Sr_2Ir_{0.8}Ru_{0.2}O_4$: measured anisotropy distinct from Sr_2IrO_4.

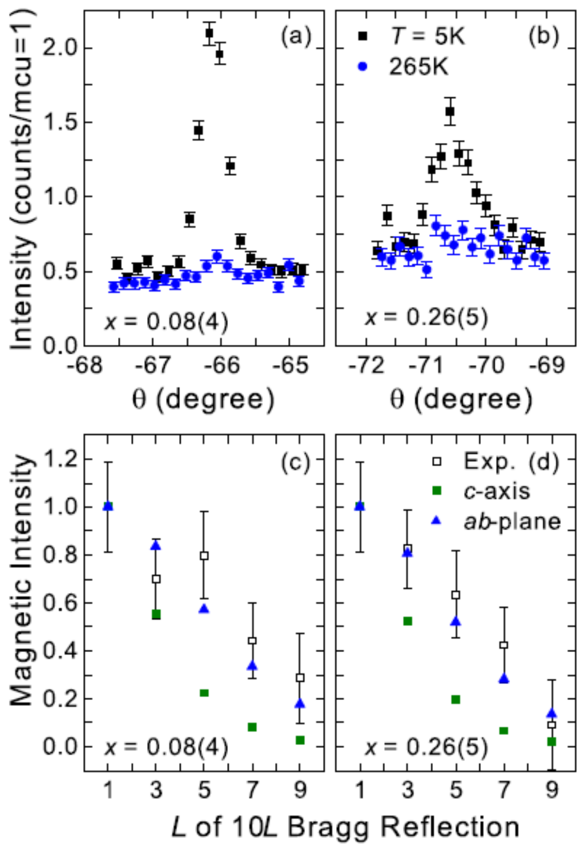
Non-magnetic Sn substitution on the Ir site: $Sr_2Ir_{1-x}Sn_xO_4$

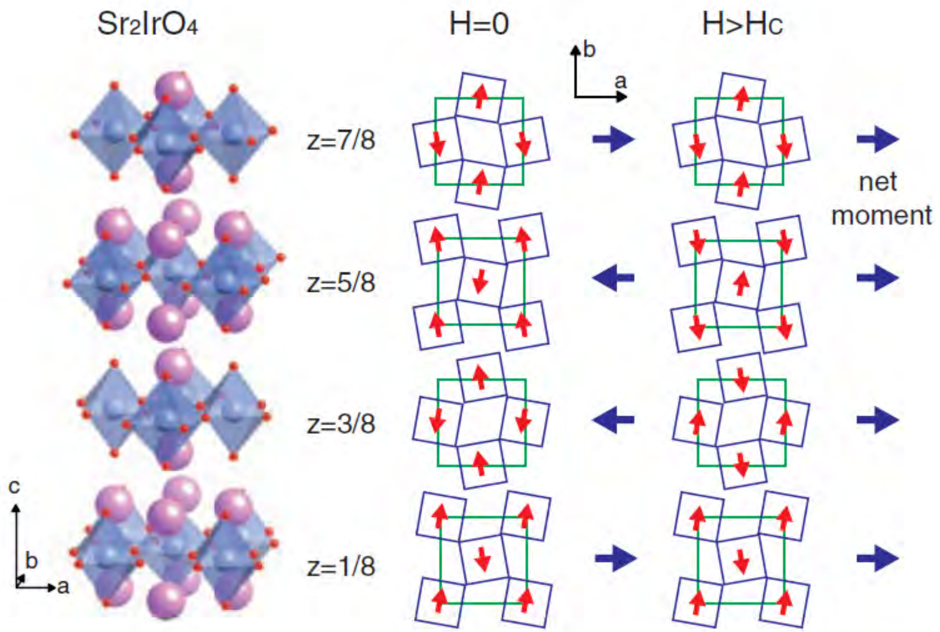


- Neutron scattering on x=0.05 and 0.2
  - magnetic structure altered from x=0
  - Distinct from Ru doping
  - Same structure as Sr_2IrO_4 in applied field









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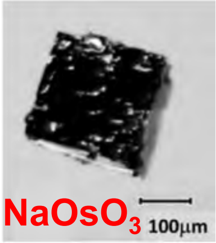

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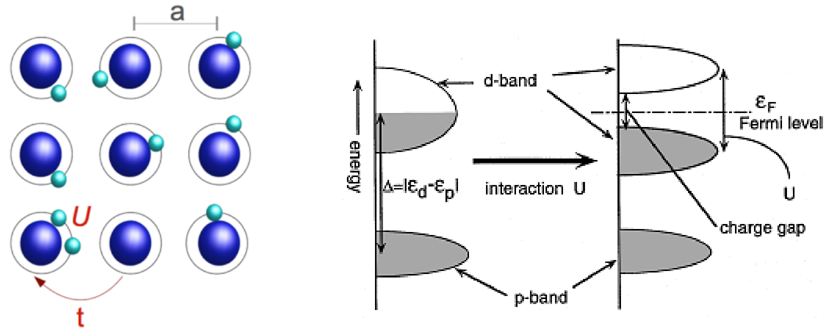

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$5d^5$ Iridates (SOC driven Mott MIT - 2009)

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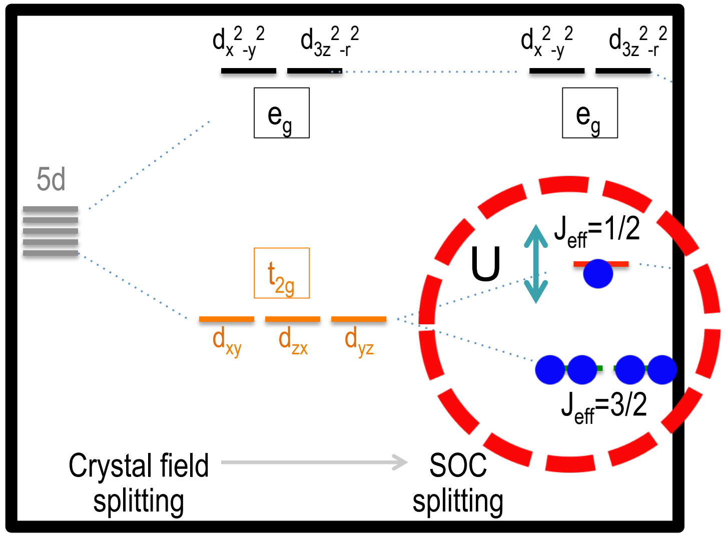

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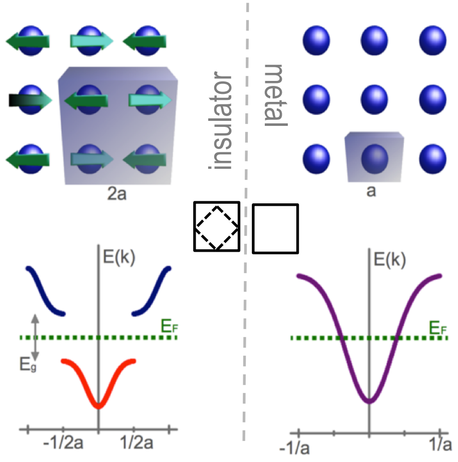


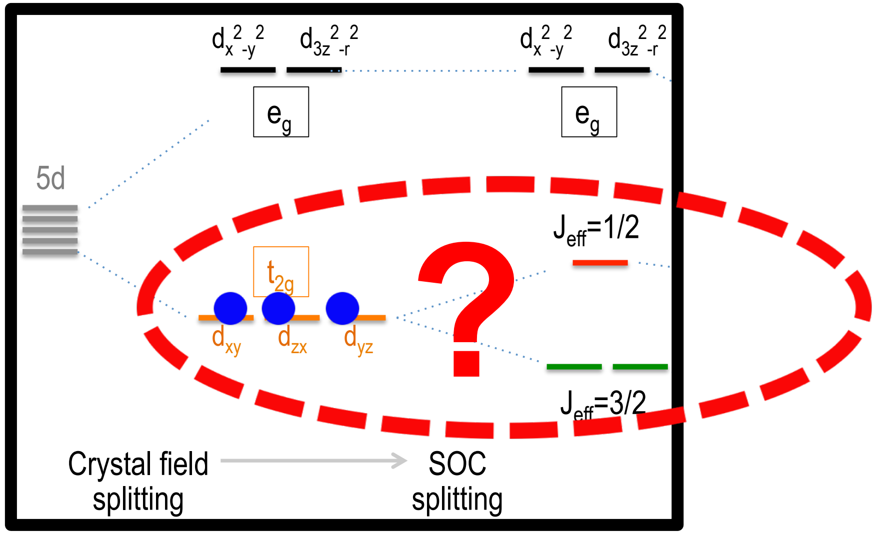
Slater: Ordered moments create mean field confining electrons.

Lifshitz: Small *U* and large hybridization drive renormalization of the Fermi surface

L-S coupling

$5d^3$ Osmates (Magnetic MIT: Slater/Lifshitz - 2009)

$NaOsO_3$ (2009)



Osmium 5d^3 compound: $NaOsO_3$

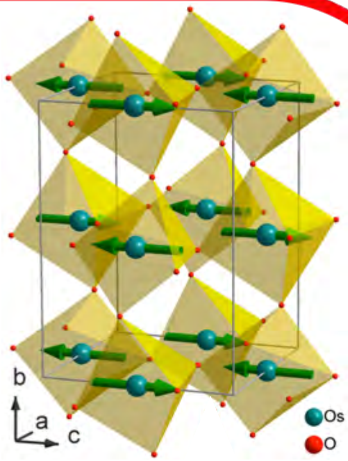
Metal-insulator transition coupled to magnetic order → Slater/Lifshitz/other?

$NaOsO_3$ (Perovskite)

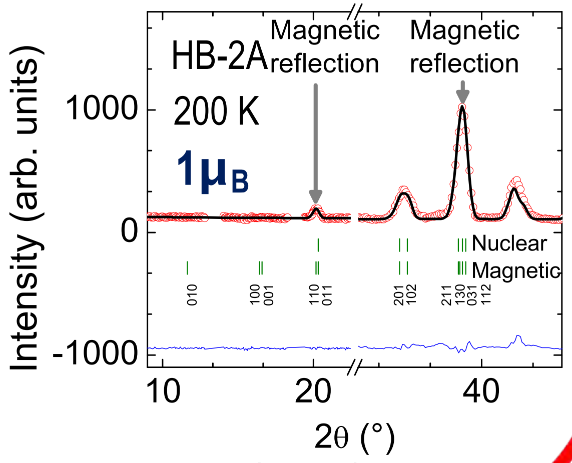
G-type AFM

T_N=410 K= T_{MIT}

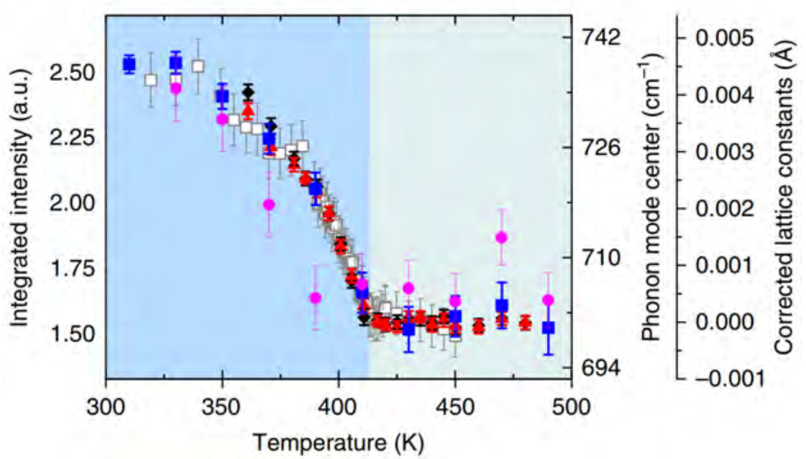
Reduced 1.0(1) μ_B



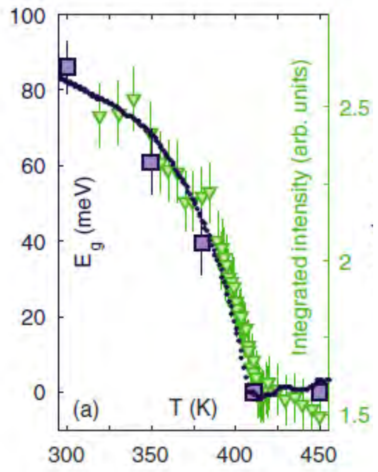
Neutron and resonant x-ray scattering



Calder *et al.*, PRL **108**, 257209 (2012)



Spin-phonon coupling at MIT



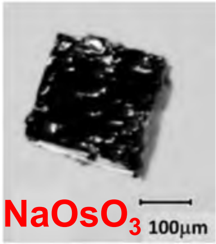
Spin-gap coupling at MIT.

- Inelastic neutron scattering showed largest phonon shift at spin-phonon transition.

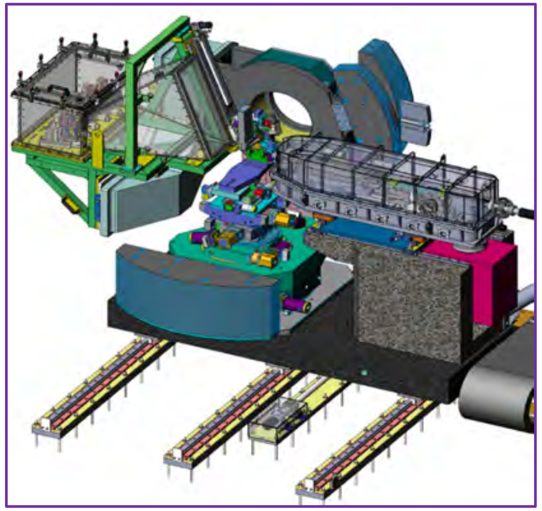
Calder *et al.*, Nature Communications **6** 8916 (2015).

Osmium L_3-edge RIXS development at APS and ESRF

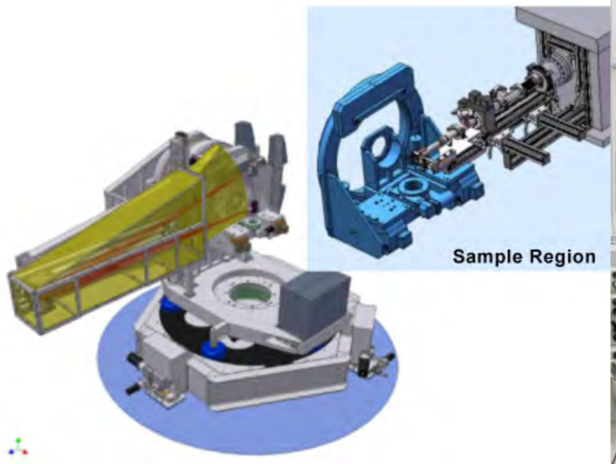
- Understand magnetic interactions through MIT
  - $NaOsO_3$ crystals too small for inelastic neutron scattering.
- Developed Os edge at APS and ESRF with instrument teams
- Best resolution ΔE=50 meV
  - Comparable to Ir L-edge RIXS

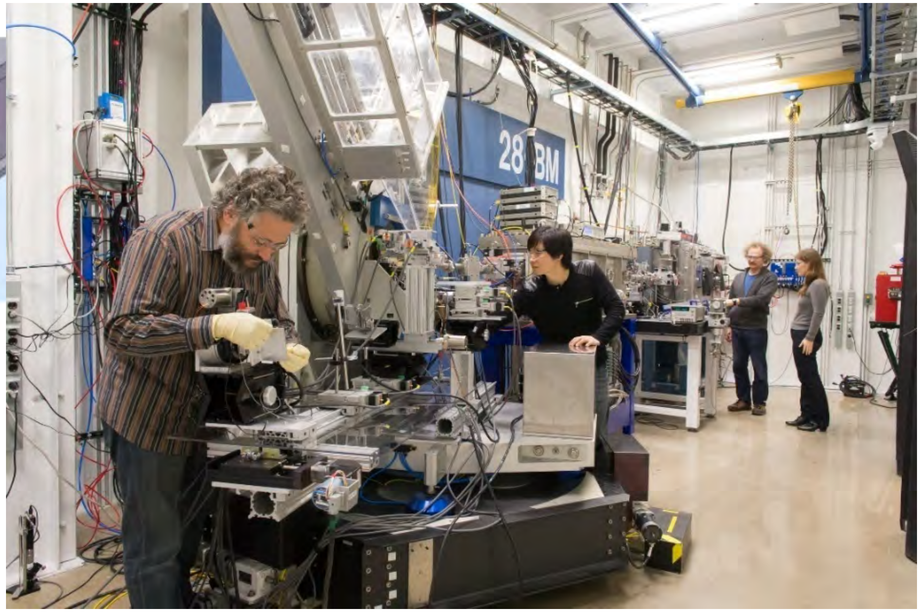


ESRF (ID-20)



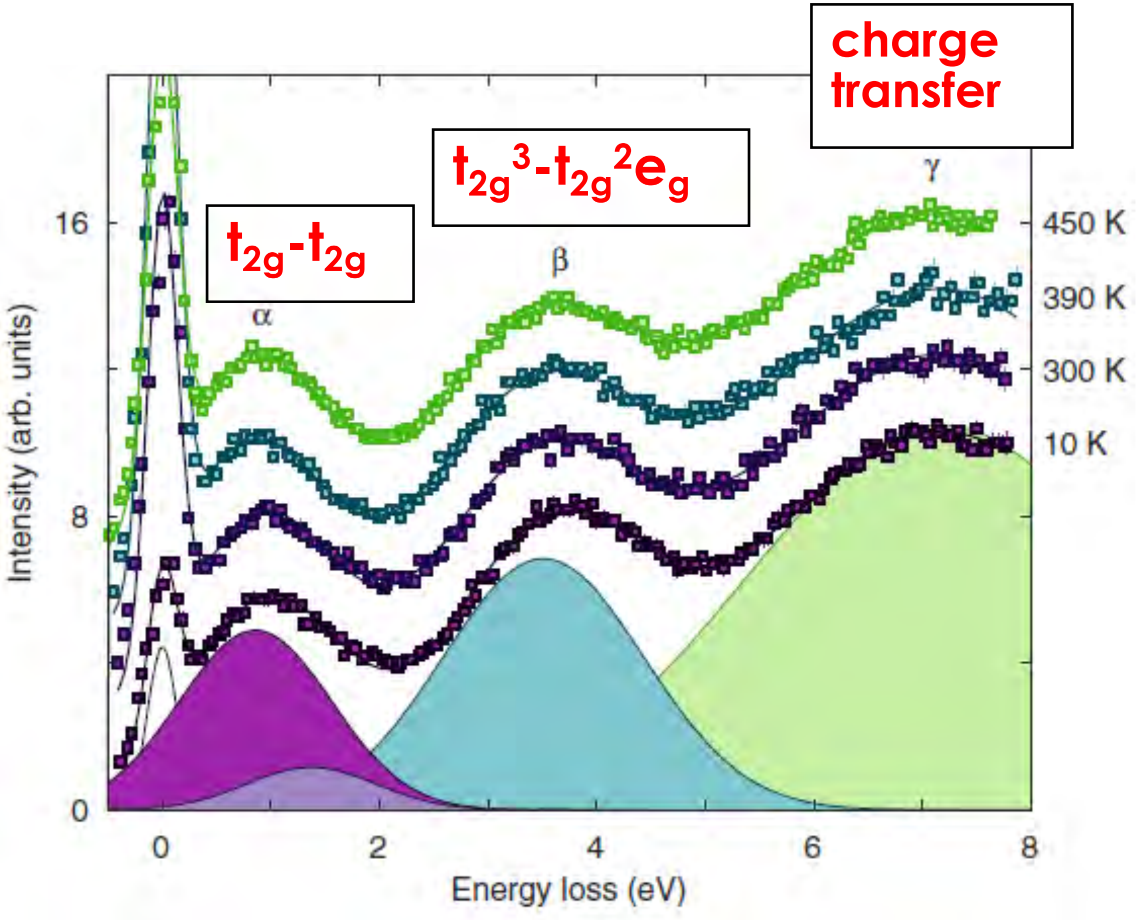
APS (MERIX, sector-27)



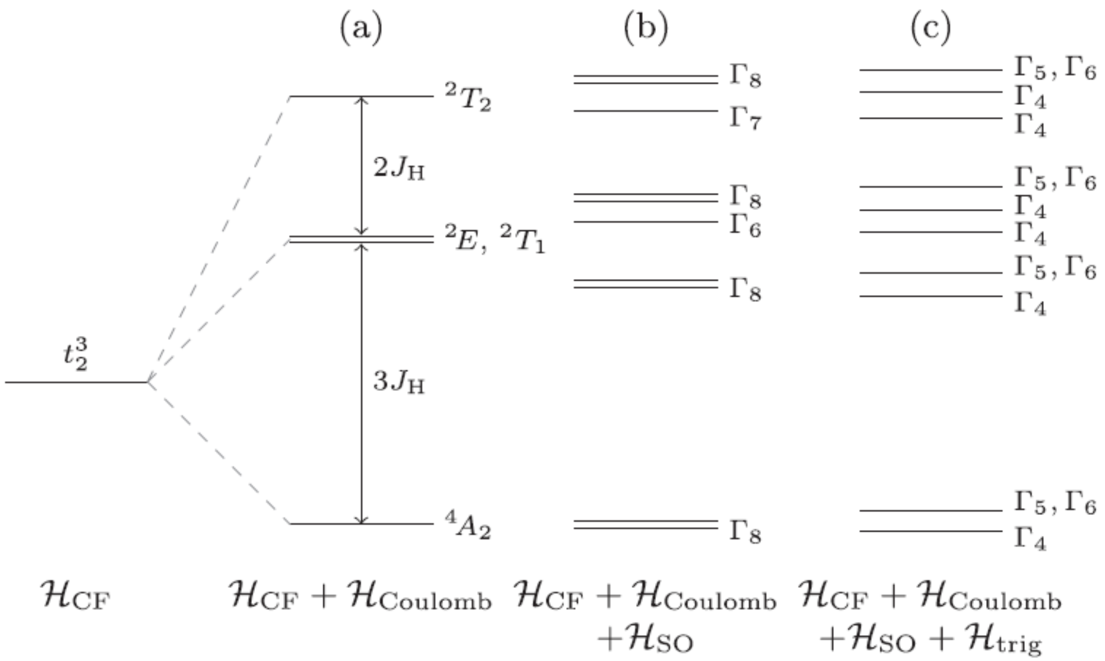


d-d excitations unchanged through MIT

- Features broader than ΔE=300 meV resolution



- Fit to **L.S** model (a)
- J_H=0.25(1)eV



$NaOsO_3$ Magnetic excitations in insulating regime (300 K)

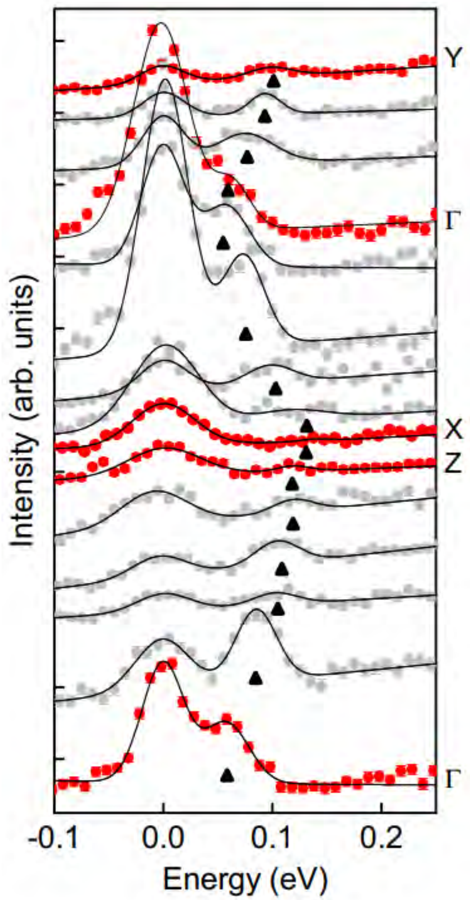
Well-defined spin waves

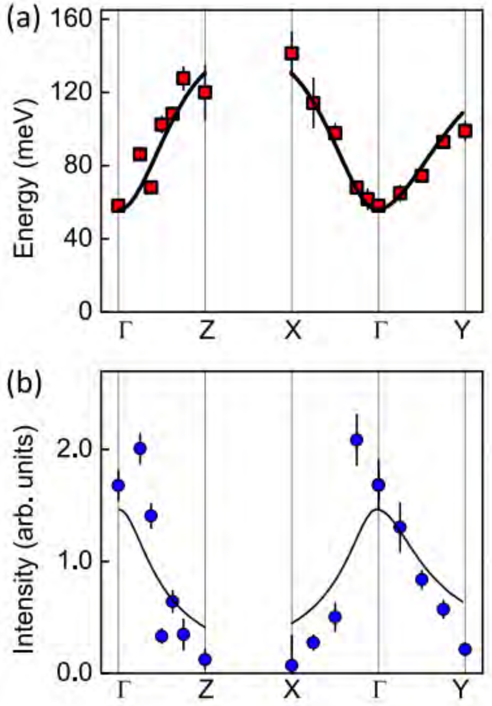
- Large spin-gap of ~50 meV
- Minimal model Hamiltonian within linear-spin wave theory.

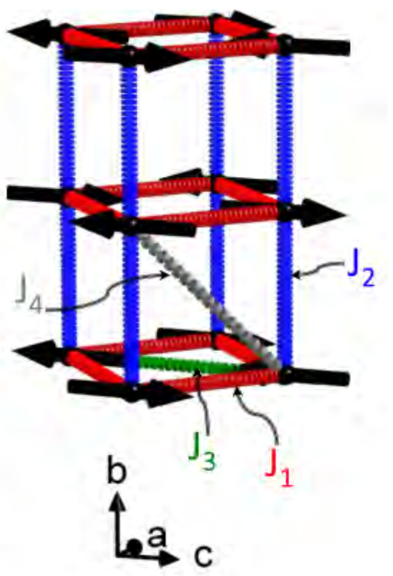
$$\mathcal{H} = J_1 \sum_{\mathrm{nn}} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\mathrm{nnn}} \mathbf{S}_i \cdot \mathbf{S}_j + \Delta$$

  - $J_1=J_2=14$ meV
  - Anisotropic term due to SOC.

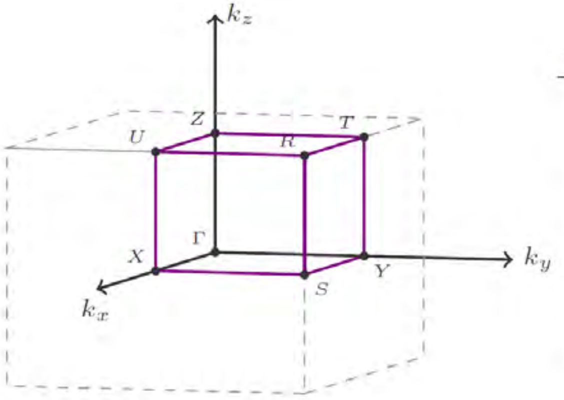
- Evidence of some damping.
  - Departure from local moment model.







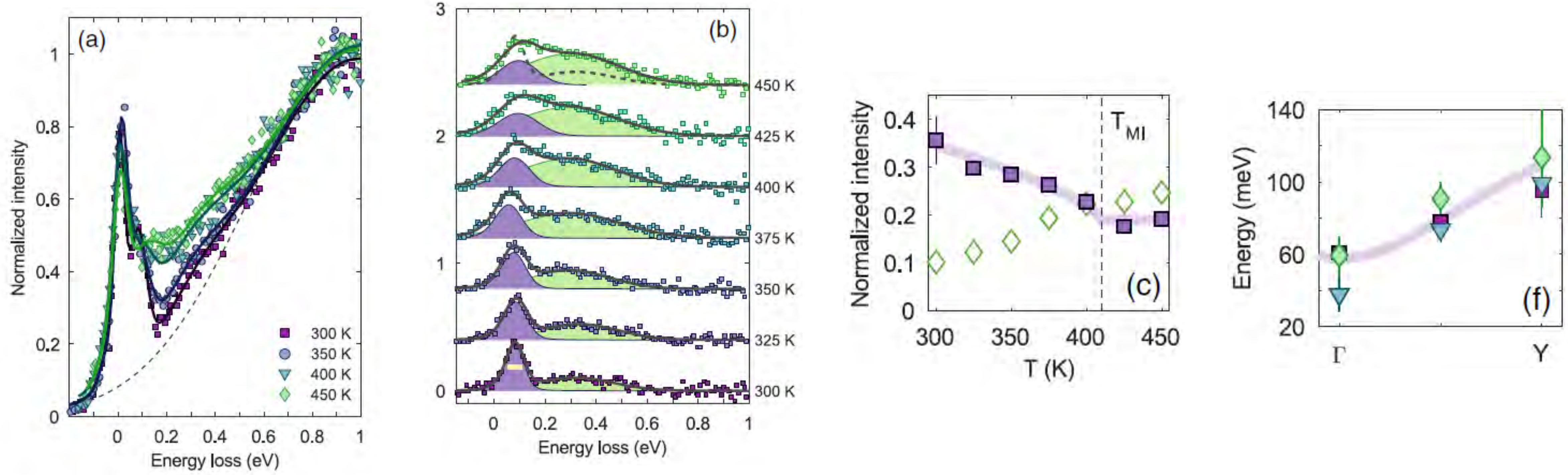
Resolution of ΔE=50 meV



Point	Position
Γ	(0, 0, 0)
X	($\frac{1}{2}$, 0, 0)
Y	(0, $\frac{1}{2}$, 0)
Z	(0, 0, $\frac{1}{2}$)
U	($\frac{1}{2}$, 0, $\frac{1}{2}$)
S	($\frac{1}{2}$, $\frac{1}{2}$, 0)
T	(0, $\frac{1}{2}$, $\frac{1}{2}$)
R	($\frac{1}{2}$, $\frac{1}{2}$, $\frac{1}{2}$)

Calder *et al.*, PRB: Rapid 95, 020413(R) (2017)

Temperature dependence of magnetic excitations



- Increasing T: Insulating (300 K) → MIT (410 K) → metallic (450 K)
  - Purple: Single magnon peak broadens and weakens.
  - Green: Concurrently a continuous increase in region 0.1-0.6 eV
- Resistivity in metallic regime consistent with Fermi liquid. Shi et al., PRB 80 161104 (2009)
- Simple picture considering Landau damping of spin waves does not fit data

Modelling low energy excitations in metallic regime

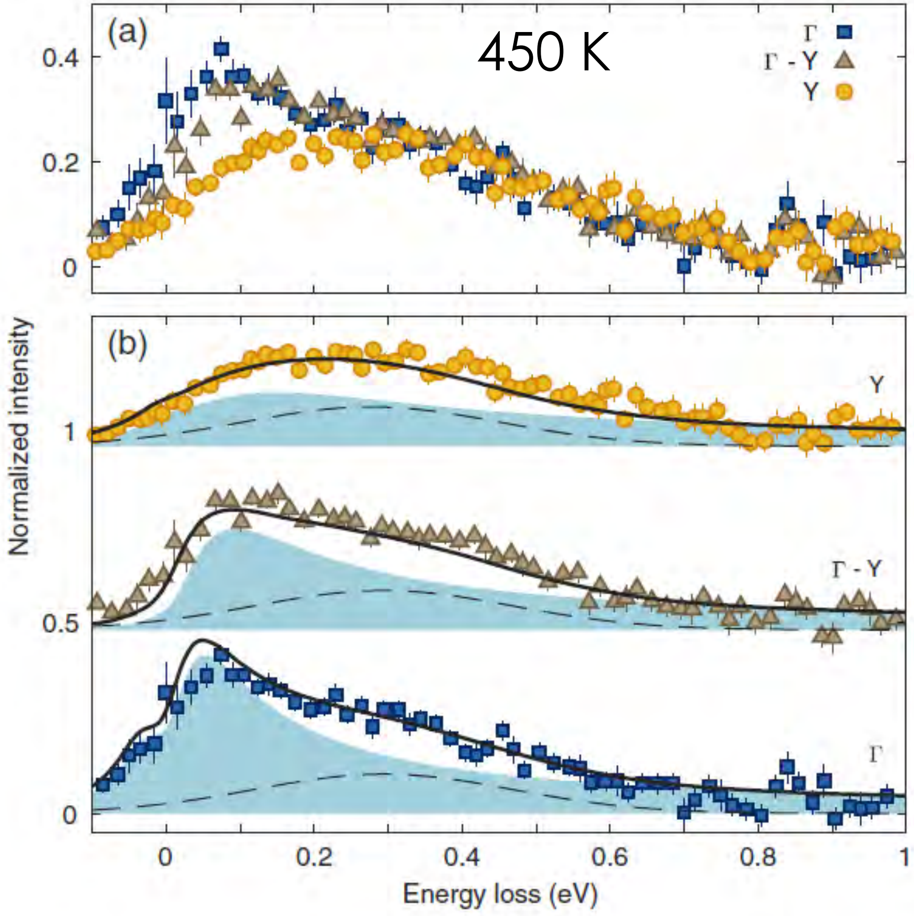
- Consider the self-consistent renormalization theory appropriate for a weakly antiferromagnetic Fermi liquid (WAFL)
  - Follow approach for unconventional superconductors

    Nature Phys 6 (2010), PRB 89 180503(R) (2014)

$$\chi''(\mathbf{Q}, E) \propto \frac{\chi_0 \Gamma E}{E^2 + \Gamma^2[1 + \xi^2(\mathbf{Q} - \mathbf{Q}_{\mathrm{AFM}})^2]^2}$$

- Where $\Gamma = a_0^2/\gamma\zeta^2$
  - with a_0^2 the Os-Os distance [3.8 Å]
  - ζ the spin-spin correlation length [$\zeta/a_0 \sim 1$]
  - γ the damping coefficient [0.02 meV^{-1}] (c.f. pnictides)

- **Excitations are consistent with paramagnetic spin fluctuations in a weakly AFM Fermi liquid**
  - **characteristic of a system close to itinerant limit**



Summary

- Neutron and resonant x-ray scattering combined offer powerful insights into 5*d* magnetism.

Sr_2IrO_4 ($5d^5$)

- Inelastic neutron scattering on single crystal iridates is feasible.
- Strongly 2D correlations.
- Near vanishing in-plane spin gap of 0.6 meV (excitations ~200meV).
- Doping on Ir site (Ru/Sn) induces strong change in magnetic exchange and anisotropy.

PRB:Rapid 92, 220407(R) (2016)
PRB 94, 165128 (2015)
PRB 98, 220402(R) (2018)
PRM 2, 094406 (2018)

$NaOsO_3$ ($5d^5$)

- 300K: close to Heisenberg local moment limit with anisotropy (SOC).
- T increases: continuous progression towards itinerant limit through MIT.
- 450 K: Consistent with weakly correlated paramagnetic spin fluctuations model (WAFL).
- Local moment to itinerant cross-over tuned with temperature in a single material.

PRL 120, 227203 (2018)
PRB 97, 184429 (2018)
PRB 95, 020413(R) (2017)