Condensed Matter Seminar
Physics Department
University of Virginia
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Looking beyond the spin-orbit Mott phase:

Exploring the metal-insulator transition in 5d-Mott insulators

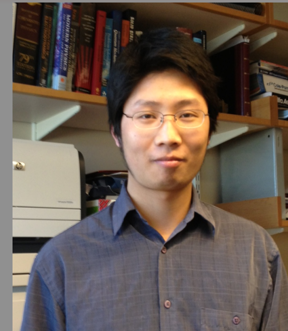
Stephen D. Wilson
Assistant Professor, Boston College Department of Physics

DMR-1056625
(NSF-CAREER)

Research Collaborators


Chetan Dhital


Tom Hogan

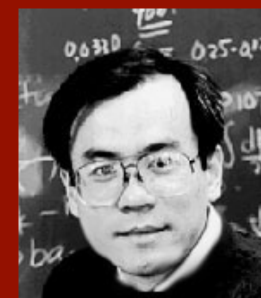

Xiang Chen


Mike Graf (μsR)


M. Naughton (torque)


V. Madhavan (STM)


C. Opeil (transport)


Z. Wang (theory)


Ken Burch (Optics)

<u>Oak Ridge</u>

Clarina de la Cruz–HFIR

Wei Tian –HFIR

<u>NIST NCNR</u>

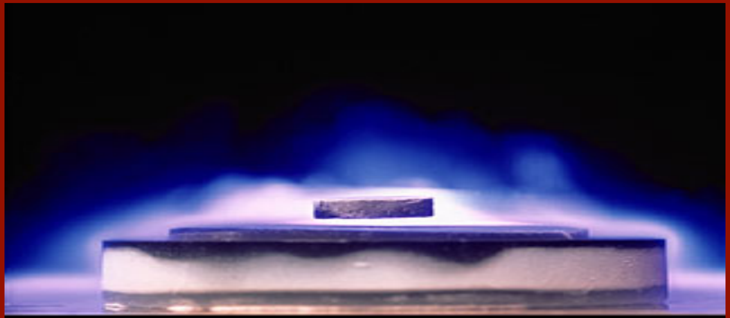
Jeff Lynn

<u>Chalk River Lab</u>

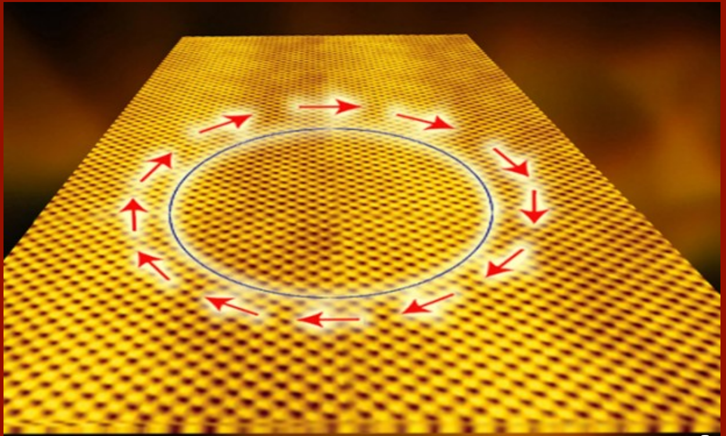
Zahra Yamani—CNRC

Talk Outline

- Introduction to Mott phase
  - Spin-orbit Mott phase
  - New phenomena of interest
- Introduction to R.P. series iridates
  - Platform for exploring S.O. Mott phase
- Exploring MIT in S.O. Mott phase
  - B-site (TM-site) doping
  - A-site (AE-site) doping
- Conclusions



Correlated electron states: U essential



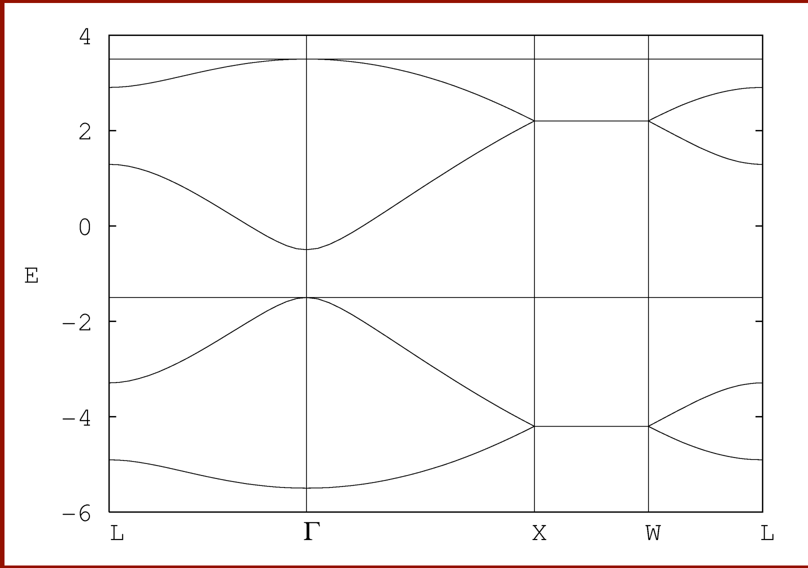
Dirac states: SOC essential

Introduction to spin-orbit Mott phase

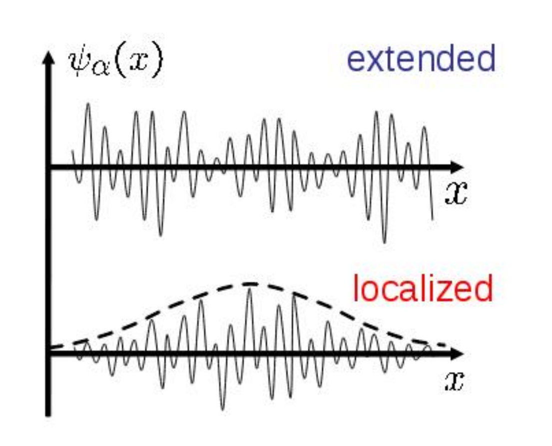
Types of insulating phases

Interactions with nuclei (ions):

Periodic: Band insulators



Disorder: Anderson localization

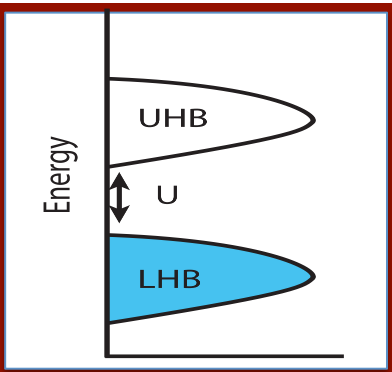


Strong/weak localization

P. W. Anderson

Interactions with other electrons:

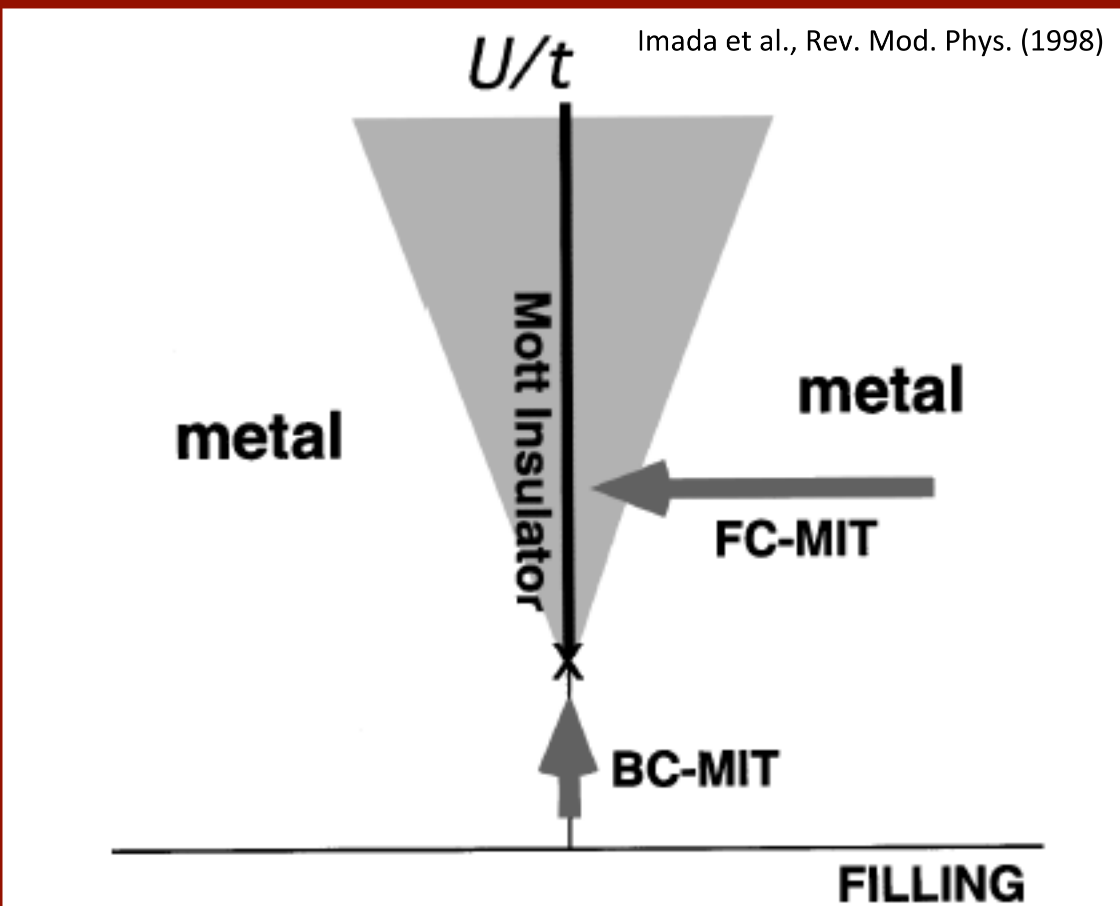
Nevill Mott

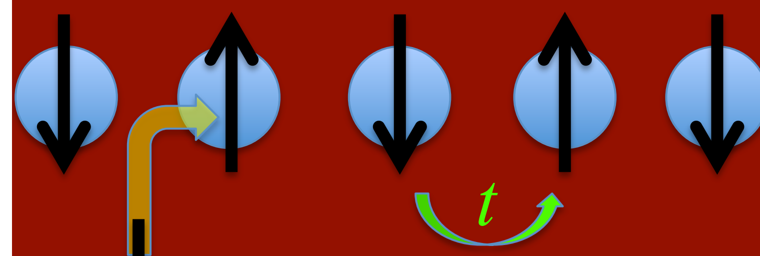


- Mott insulators:
  - Mott-Hubbard, Mott-Anderson ect…
- Wigner crystal

Generation of Mott phase:

- Consider special case of half-filled valence band:
  - Continuously increase on-site Coulomb repulsion, U





E' ≈ E + U

$$H = -\sum_{\langle ij \rangle, \sigma} t_{ij} \hat{c}^{+}_{i,\sigma} \hat{c}_{j,\sigma} + \sum_{i} U \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

Kinetic energy (hopping term)

Coulomb U (interaction term)

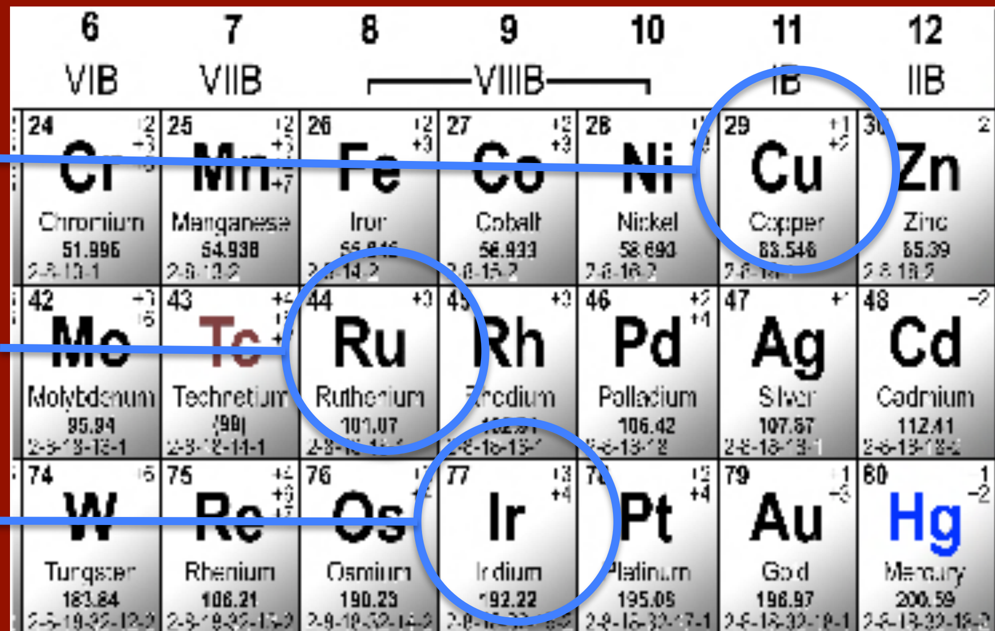
Transition metal oxides: $A_2(TM)O_4$

U

La_2CuO_4
U_{eff} ~ 5-7 eV

Ca_2RuO_4
U_{eff} ~ 2-4 eV

Sr_2IrO_4
U_{eff} ~ 1-2 eV



La_2CuO_4
λ < 0.10 eV

Ca_2RuO_4
λ ~ 0.15 eV

Sr_2IrO_4
λ ~ 0.5 eV

λ_{SO}

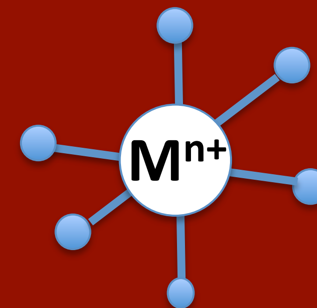
$\lambda_{SO} \sim Z^4$

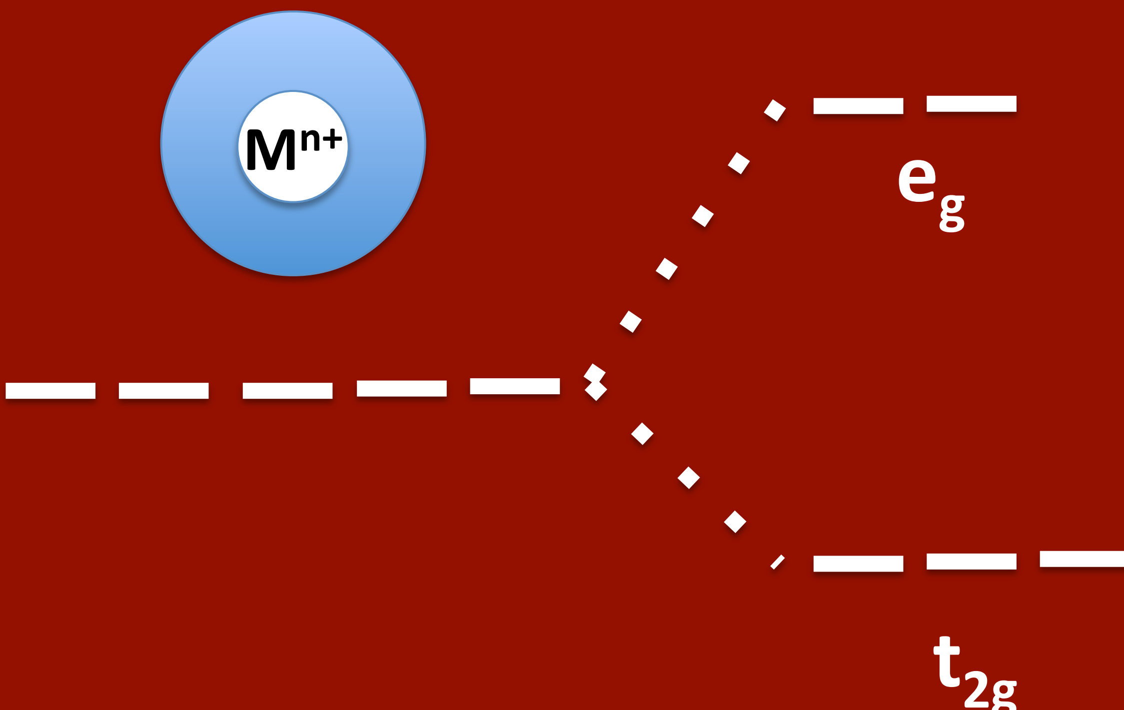
Examples:

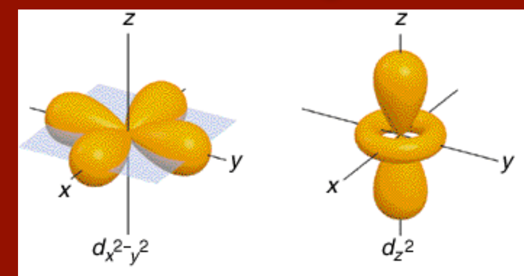
3d-electron	*4d*-electron	*5d*-electron
La_2CuO_4—AFI	Sr_2RuO_4 —PMM	Sr_2IrO_4 —AFI
La_2NiO_4 —AFI	Ca_2RuO_4—AFI	Ba_2IrO_4—AFI
La_2CoO_4—AFI	Sr_2RhO_4 —PMM	Ca_2IrO_4—AFI

Electronic configurations in perovskites

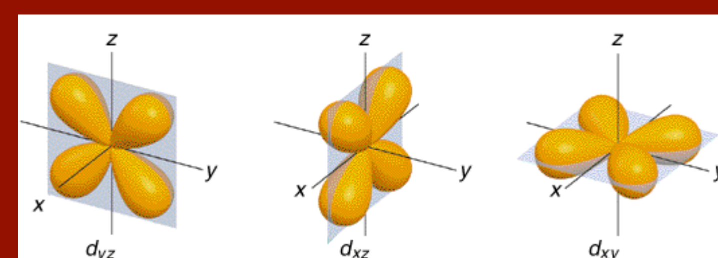
- Put *d*-valence electron in octahedral field







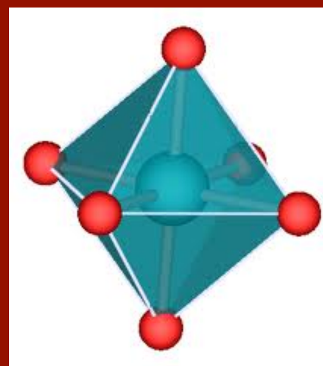
Oriented toward ligand ions



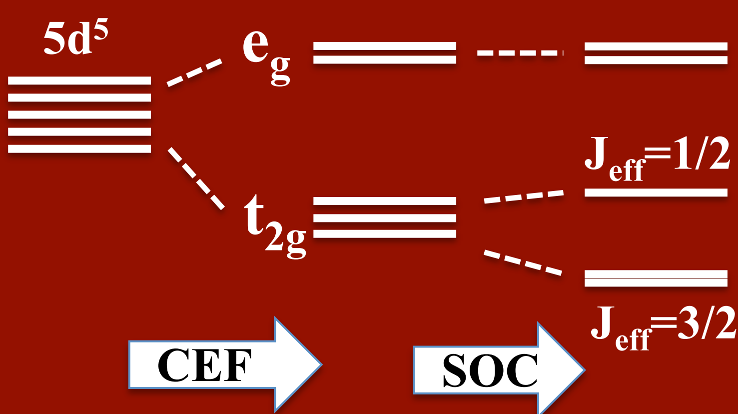
Oriented between ligand ions

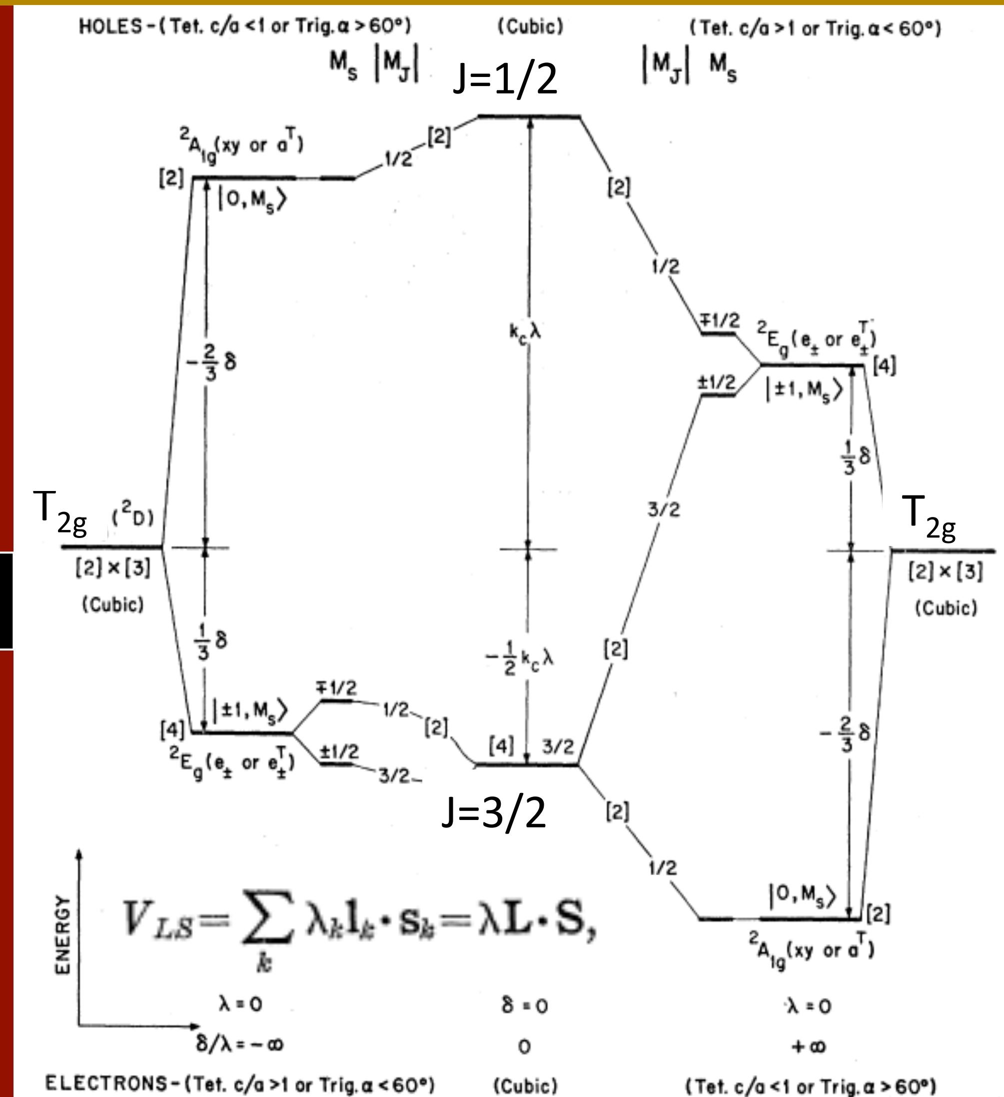
Spin-orbit coupling TM oxides

- Relativistic spin-orbit example:



$5d^5$ states in perovskite



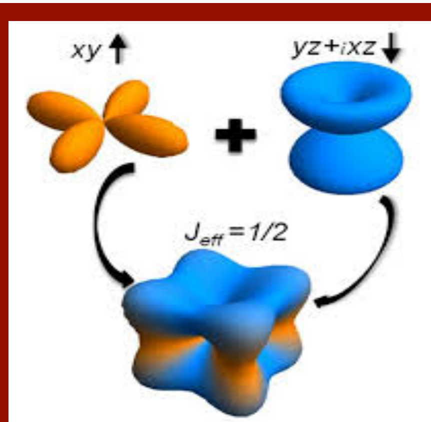


John Goodenough Phys. Rev. 1968

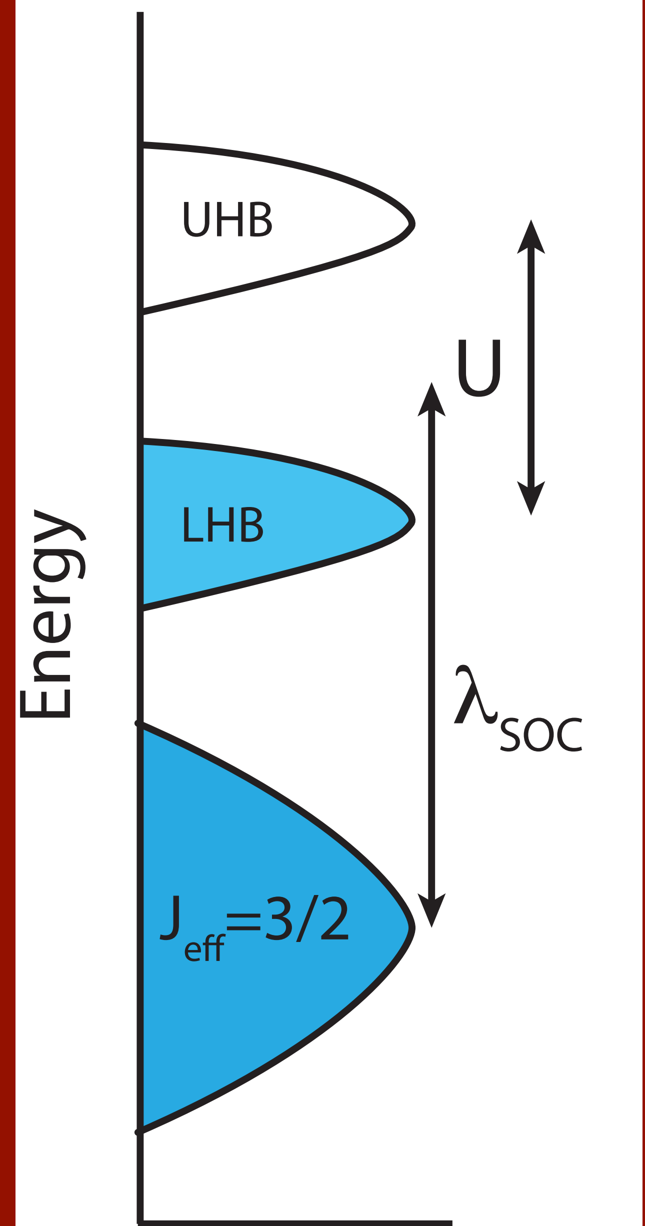
Spin-orbit driven Mott phase

- New form of Mott phase:
  - Both SOC and U are essential

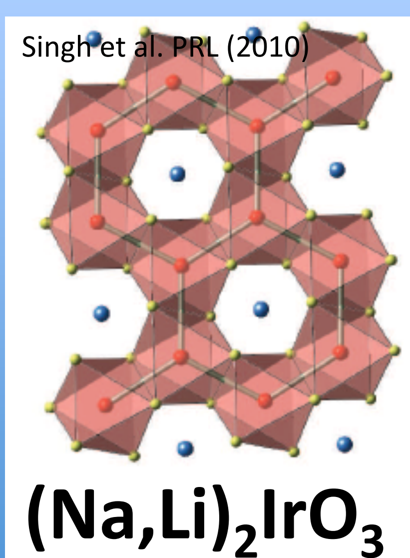
$$\left|J_{eff}=\frac{1}{2};m_J=\pm\frac{1}{2}\right\rangle=\frac{1}{\sqrt{3}}\left(\left|xy,\mp\sigma\right\rangle\mp\left|yz,\pm\sigma\right\rangle+i\left|zx,\pm\sigma\right\rangle\right)$$

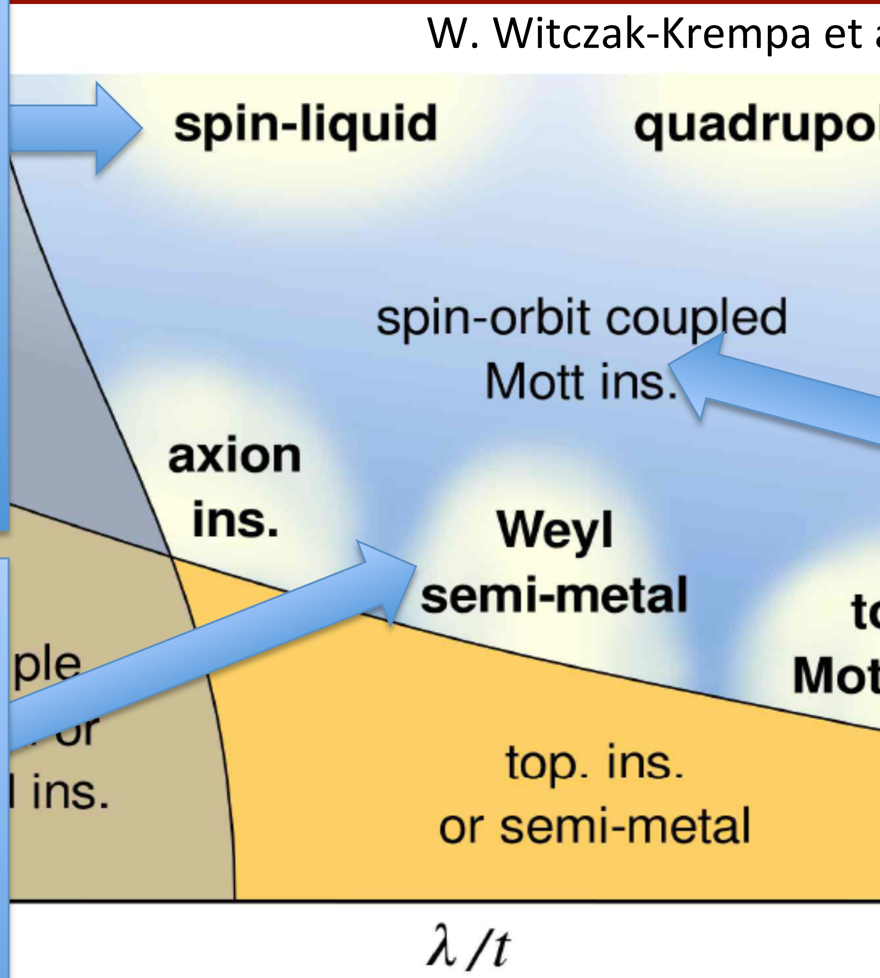


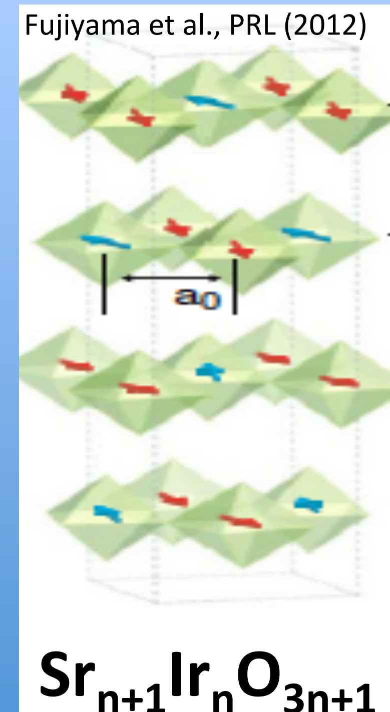
- Caveats:
  - Mixing between J_{eff}=3/2, J_{eff}=1/2
  - Hybridization with e_g orbitals
  - J_{eff}=1/2 G.S. still debated

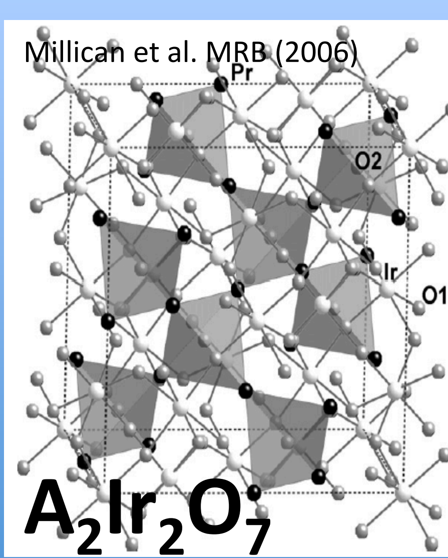


New phases in combined SOC + U







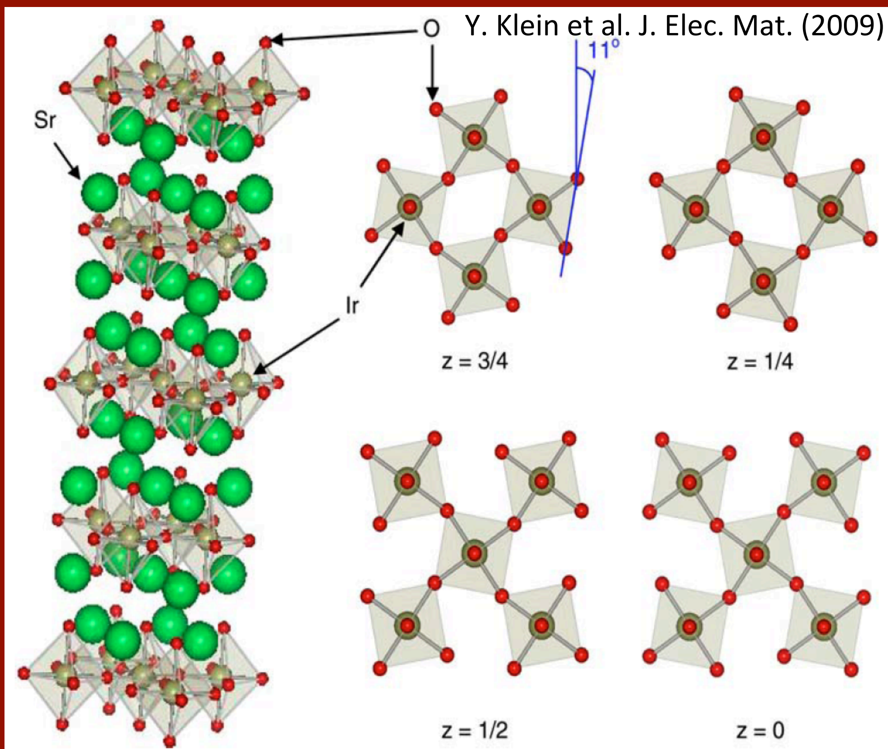


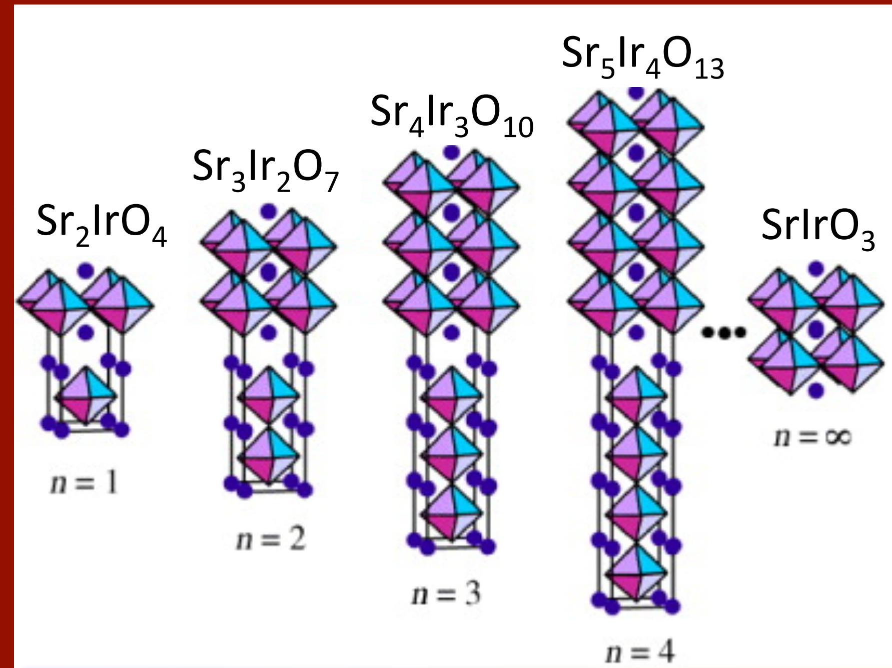
$$\ldots t_{ij,\alpha\beta}\, c^{\dagger}_{i\alpha} c_{j\beta} + \mathrm{h.c.} + \lambda \sum_i \boldsymbol{L}_i \cdot \boldsymbol{S}_i + U \sum_{i,\alpha} n_{i\alpha}(n_{i\alpha} - 1)$$

Spin-orbit Mott phase in $Sr_3Ir_2O_7$

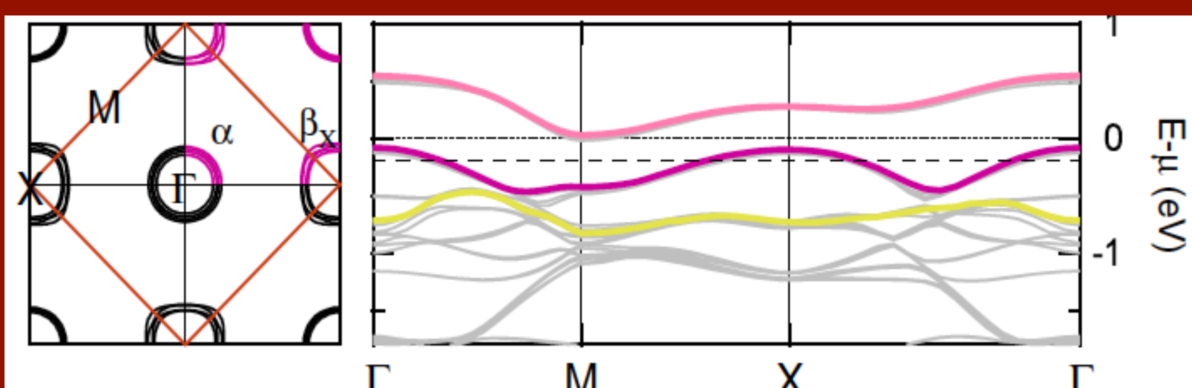
RP iridates: SOC Mott Phases

Seminal example: Sr-214





2D AF Insulator → **3D Paramagnetic Metal**



B. J. Kim et al., PRL (2008)

LDA + SOC + U

U ~ 2 eV

λ ~ 0.4 eV

E_G ~ 600 meV

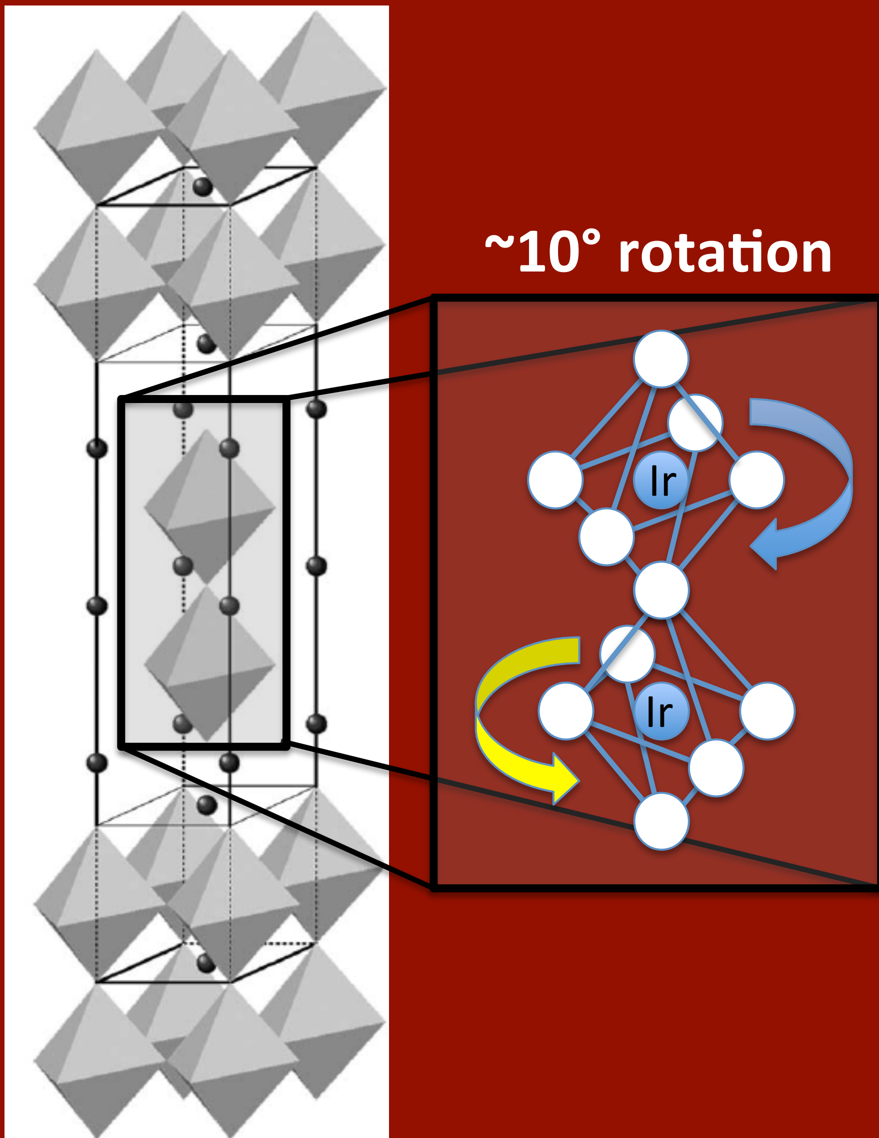
SOC Mott phase in $Sr_3Ir_2O_7$

LDA + SOC + U: U ~ 1.5 eV

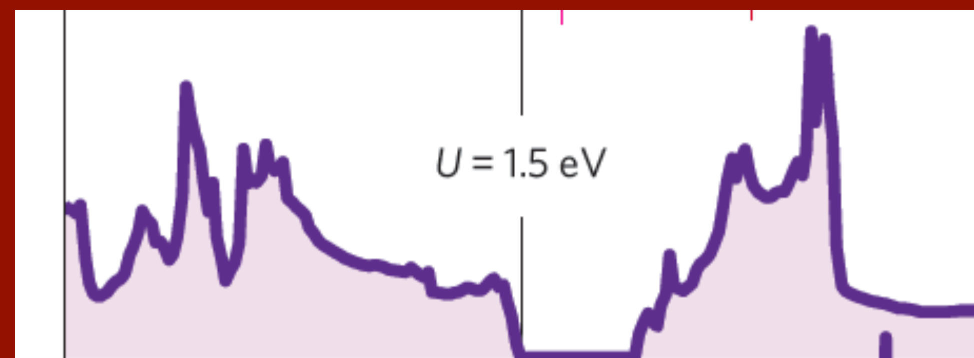
λ ~ 0.5 eV

E_G = 130 meV

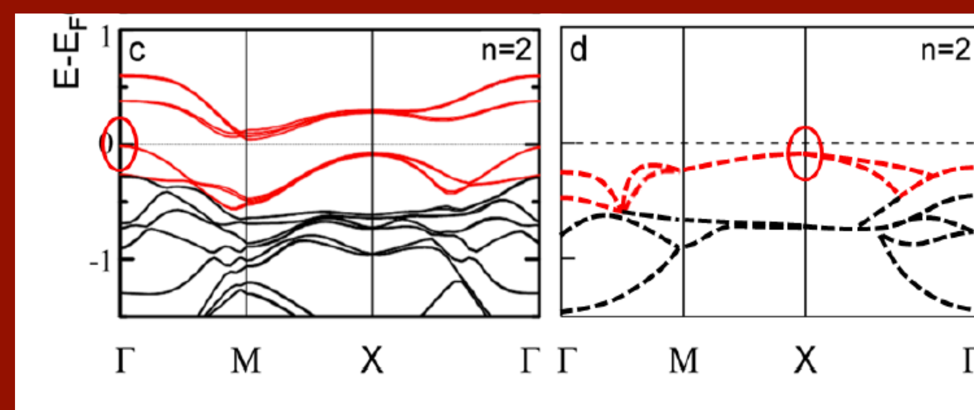
~10° rotation



Matsuhata et al., (2004)

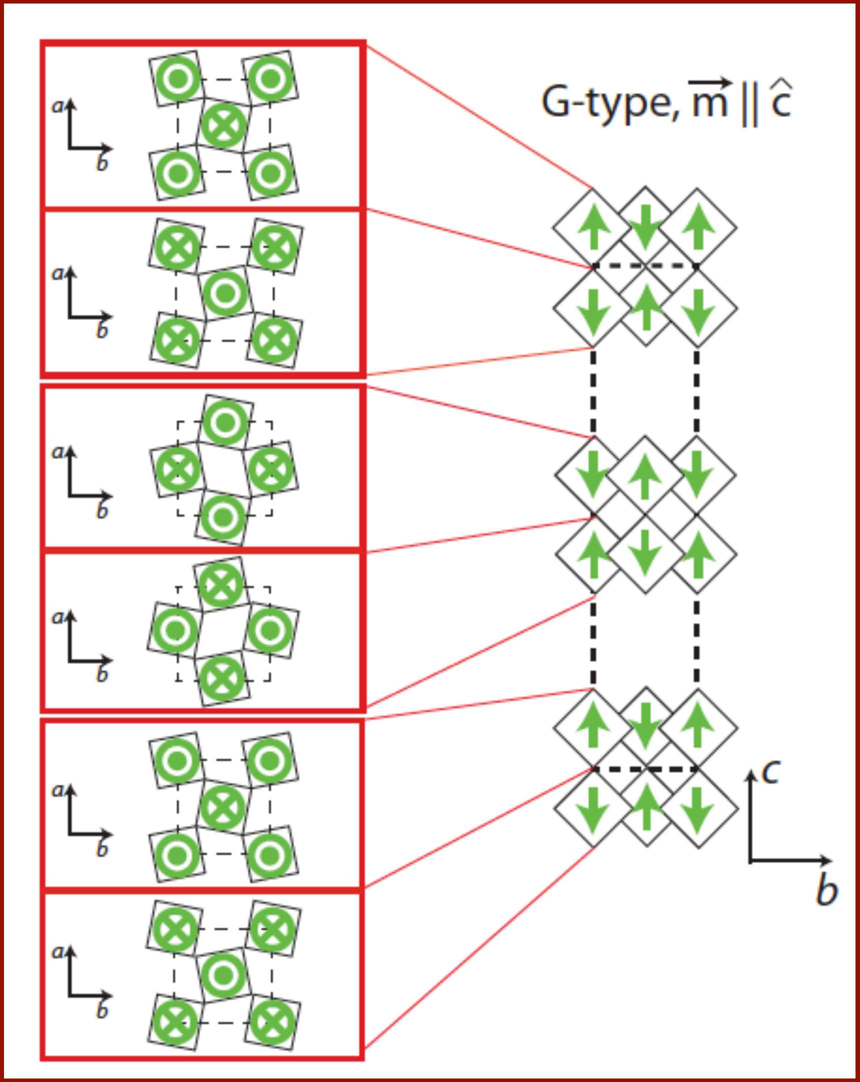


Y. Okada et al., Nat. Mat. (2013)

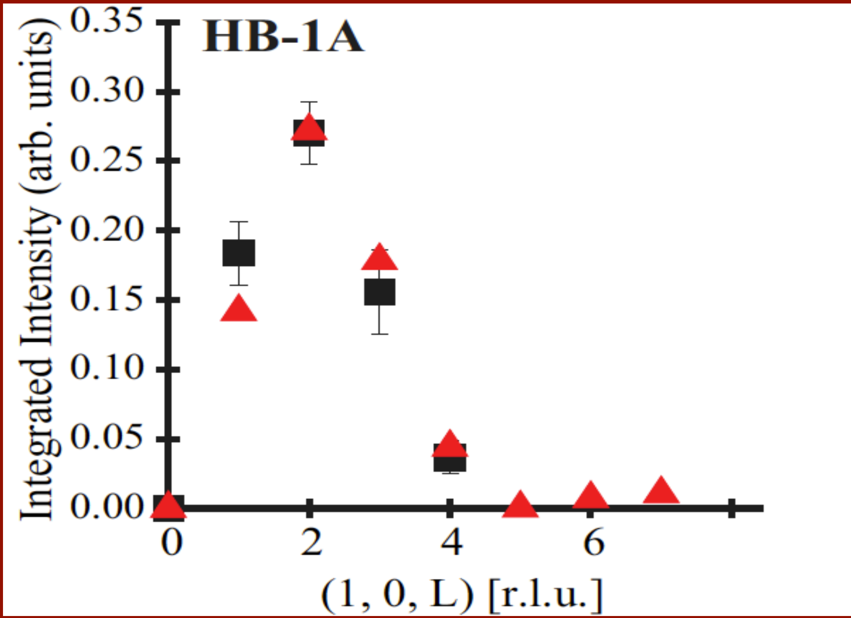


Q. Wang et al., PRB (2013)

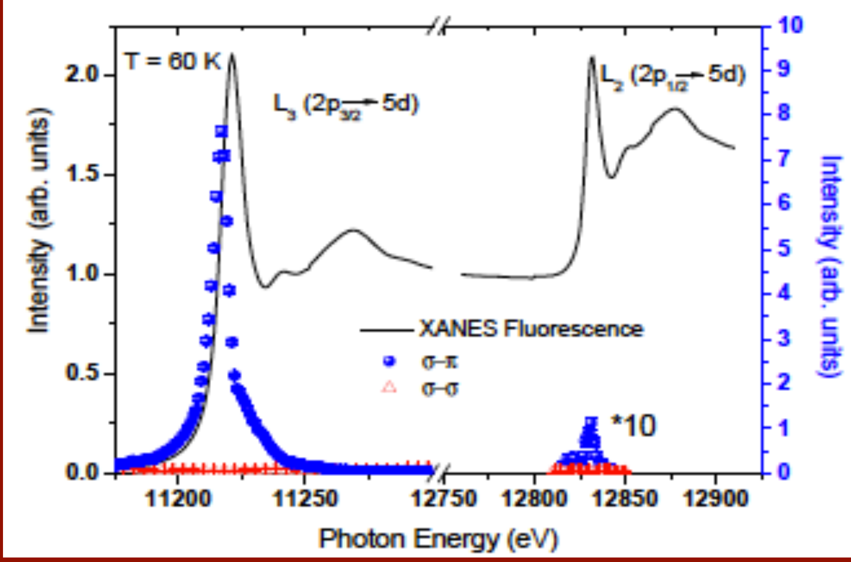
Spin Structure of $Sr_3Ir_2O_7$



- $m_{AF} = 0.36 +/- 0.06\ \mu_B$



Chetan Dhital et al. PRB (2012).



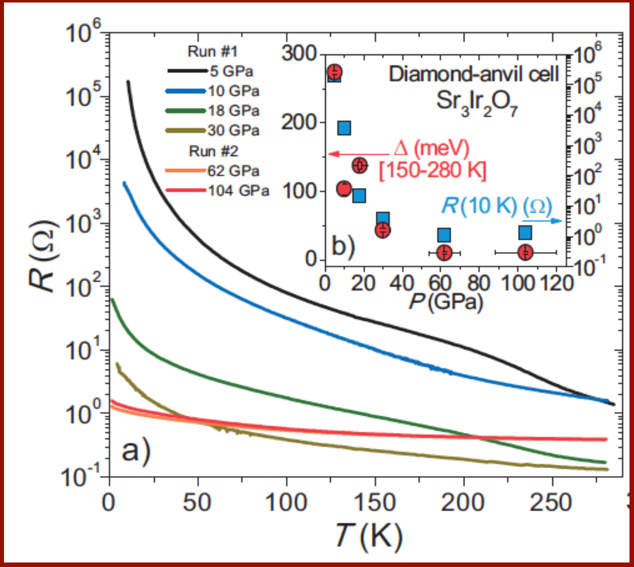
S. Bossegia et al. PRB (2012).

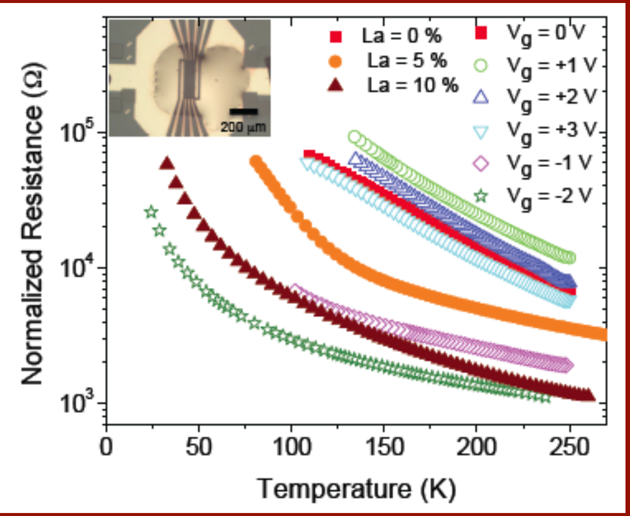
Why chemically dope?

- Goal: drive through MIT
  - Destabilize Mott phase
- Insulating state robust under B-field
- Pressure insufficient:
  - 104 GPa tested (Zocco et al.)
- Gating insufficient:
  - J. Ravichandran et al.
  - Discrepancy between film and bulk crystal

- Most viable way:
  - Substitute on either A- and B- sites
  - Oxygen tuning still under exploration

Zocco et al. arxiv (2013)

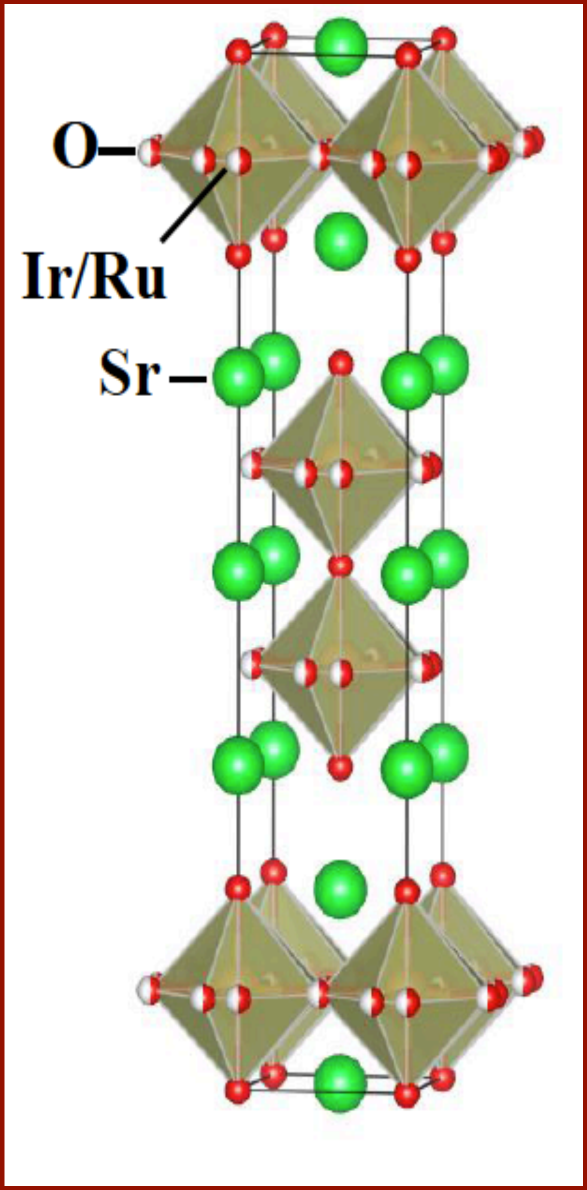




J. Ravichandran et al., arxiv

MIT in spin-orbit Mott phase: B-site substitution

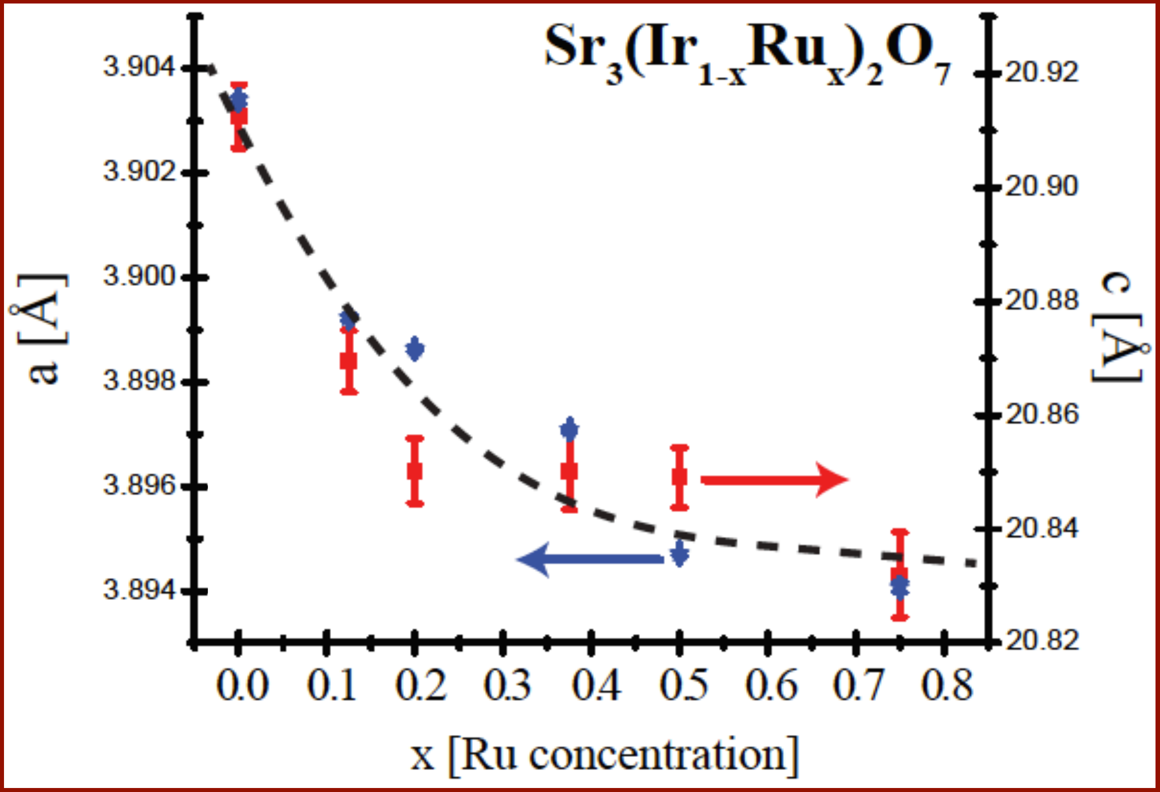
B-site doping (Ru onto Ir-sites)



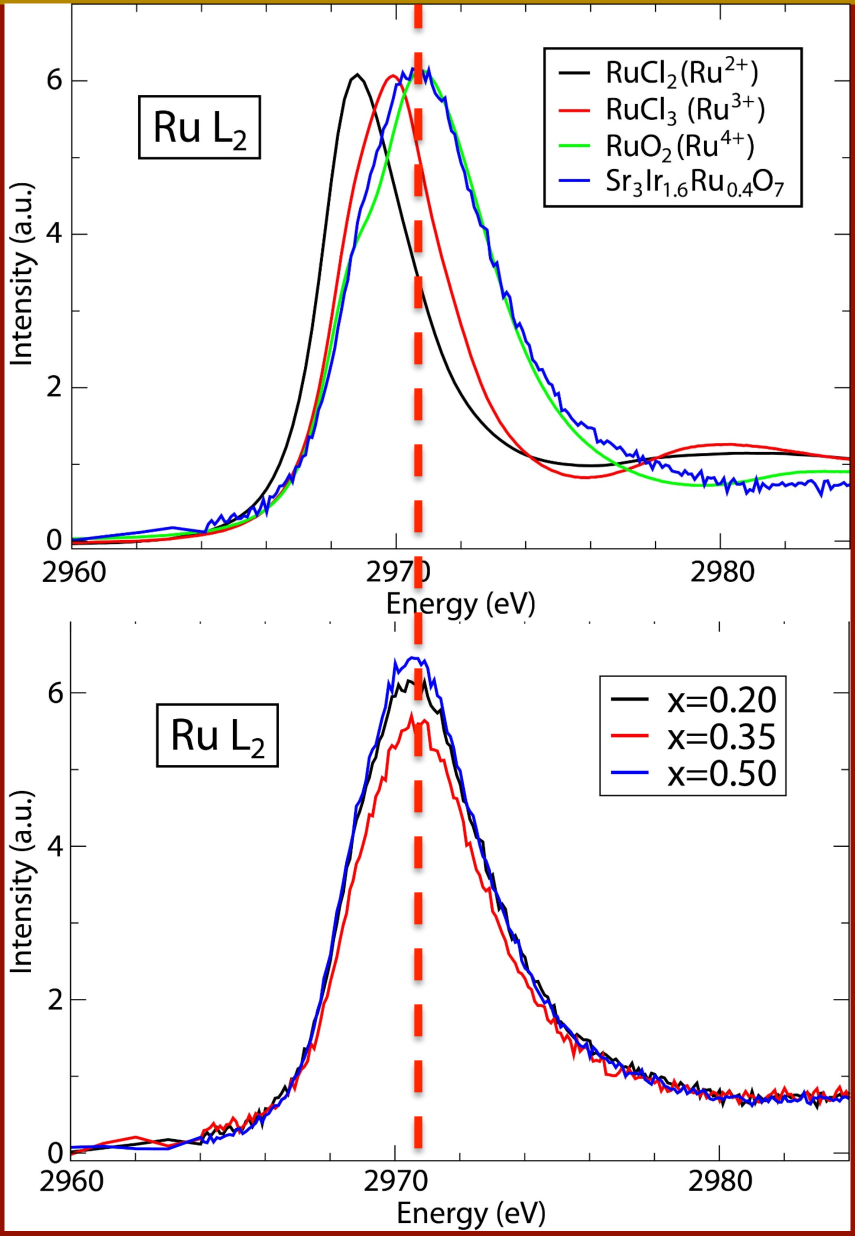
$Sr_3Ir_2O_7$ → $Sr_3Ru_2O_7$

$Sr_3Ir_2O_7$	B-site cation →	$Sr_3Ru_2O_7$
Ir^{4+} $5d^5$		Ru^{4+} $4d^4$

Chetan Dhital et al. Nature Comm. (2014).



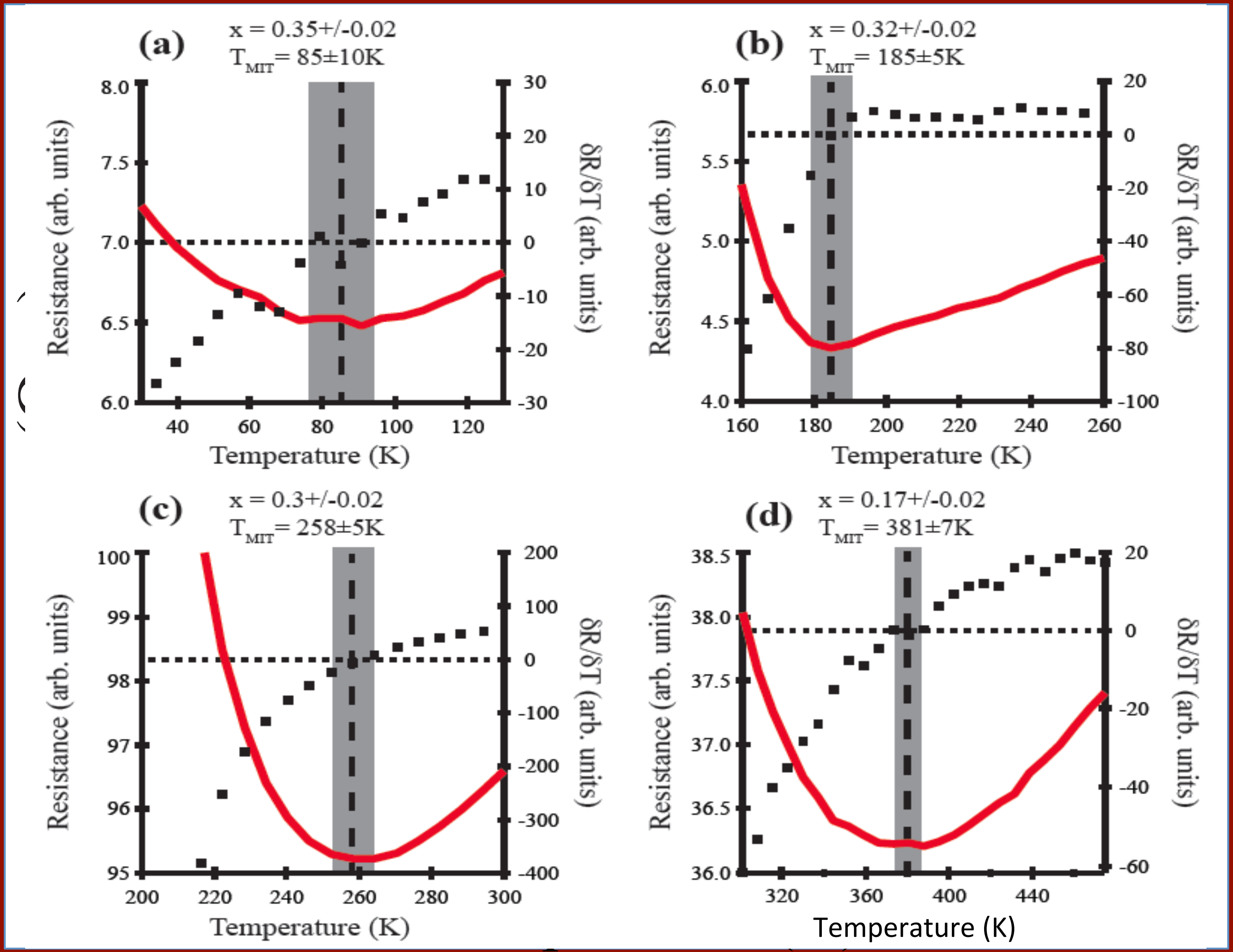
Ru-oxidation state in Sr-327



- XAS
  - Pat Clancy & Y. J. Kim (U. Toronto)
  - Ru oxidation state unchanged across series
- While-line spectra:
  - Consistent with Ru^{4+}
  - RuO_2 reference material

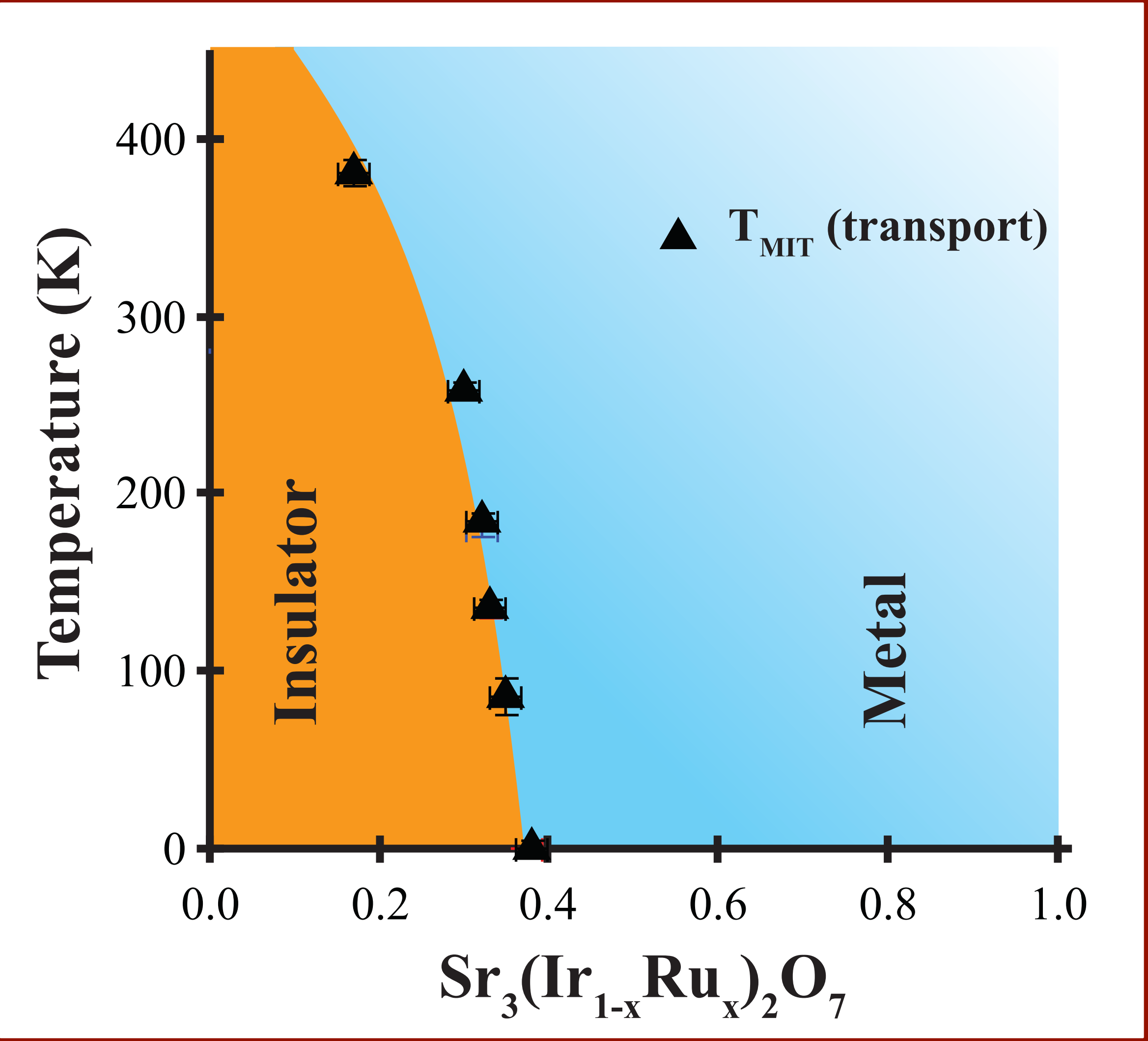
$Sr_3(Ir_{1-x}Ru_x)_2O_7$ Transport

Chetan Dhital et al. Nature Comm. (2014)

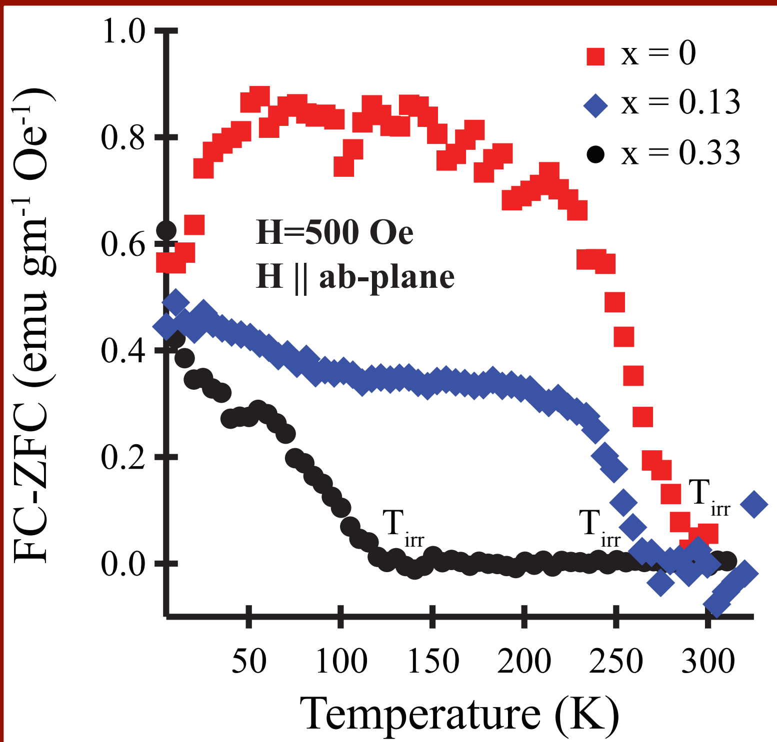


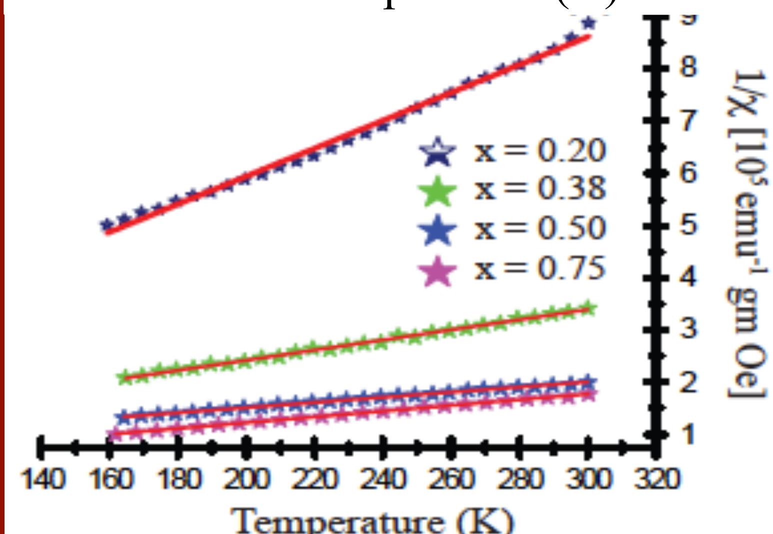
- MIT critical point at x=0.35
- Thermally driven MIT near critical point

T_{MIT} evolution with Ru doping



$Sr_3(Ir_{1-x}Ru_x)_2O_7$ Bulk Magnetization

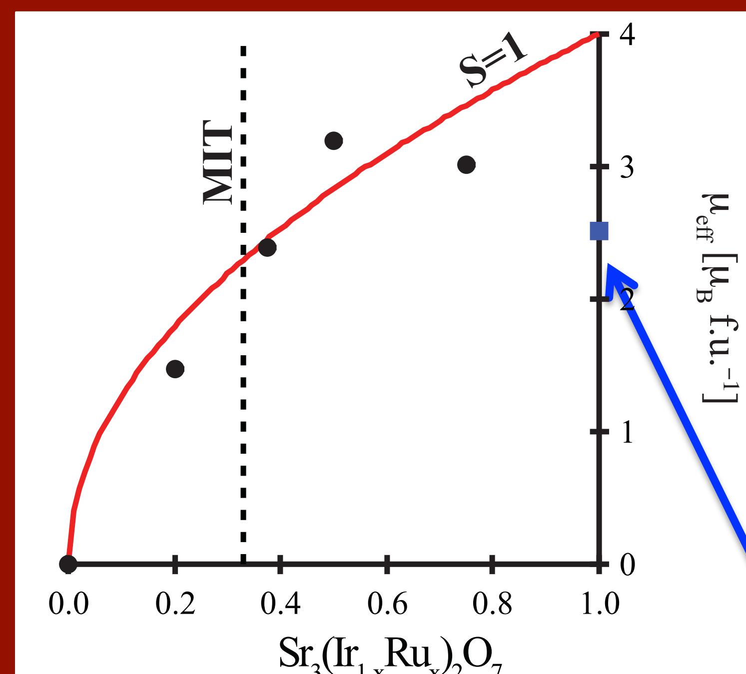




- T_{irr} decreases as x=0.35 approached
- Local moment behavior grows with increasing x
  - Curie-Weiss behavior beyond x=0.35

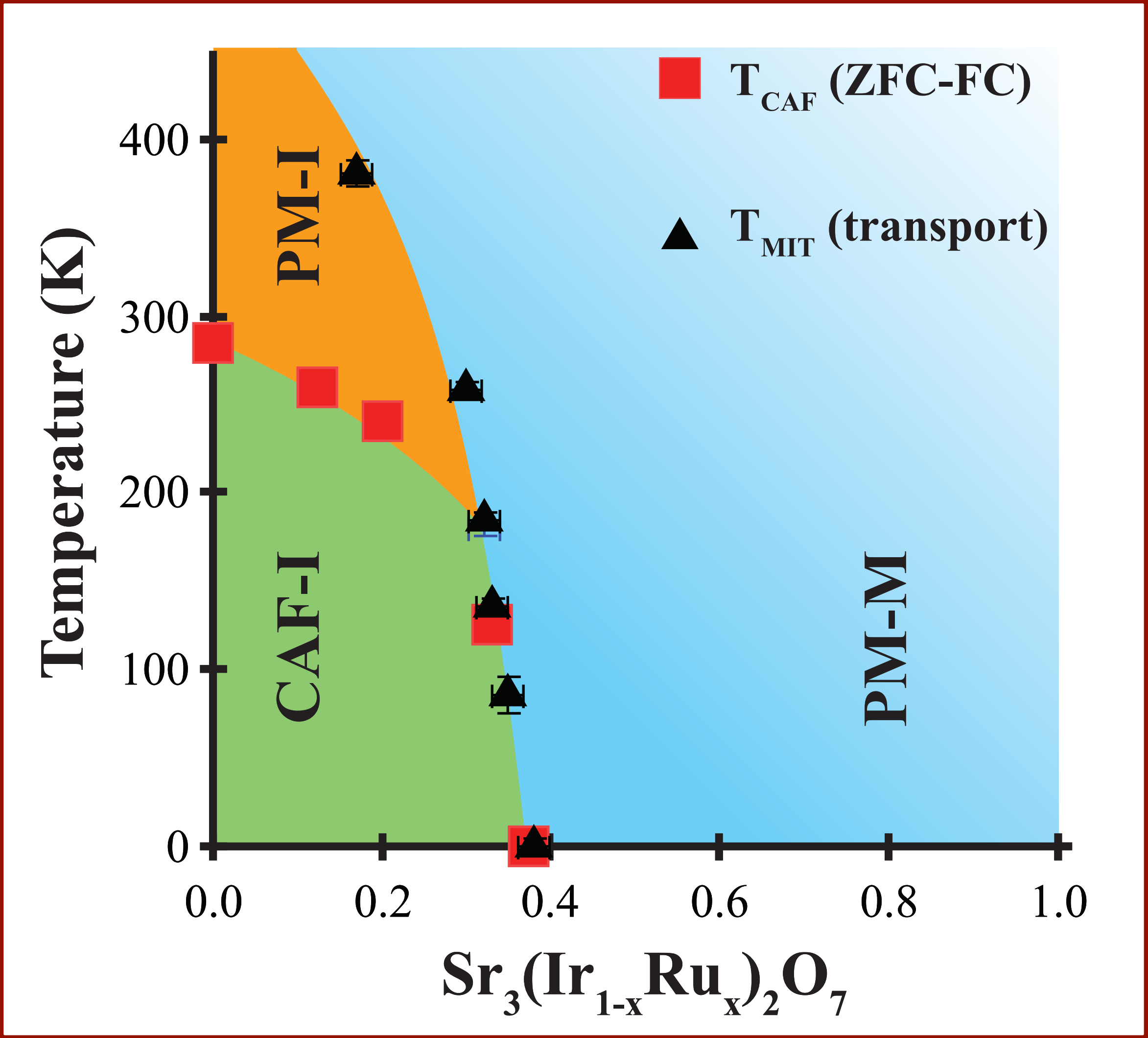
$$\chi(T) = C / (T + \Theta)$$

$$C = \frac{N_A}{3k_B} \mu_{eff}^2$$



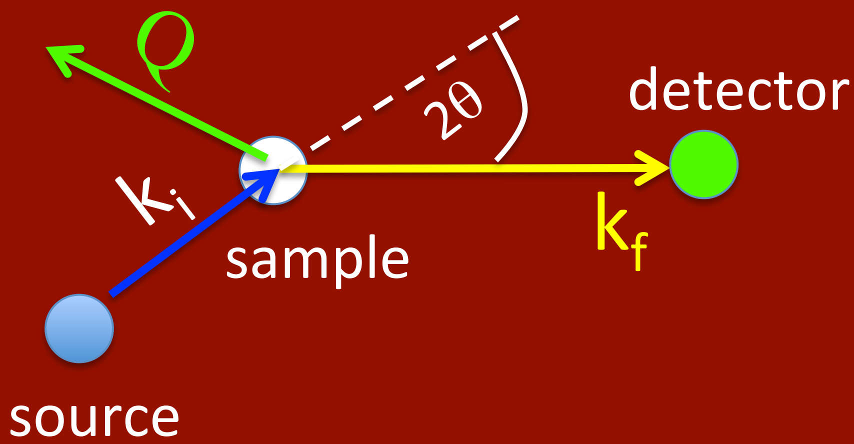
Ikeda et al.

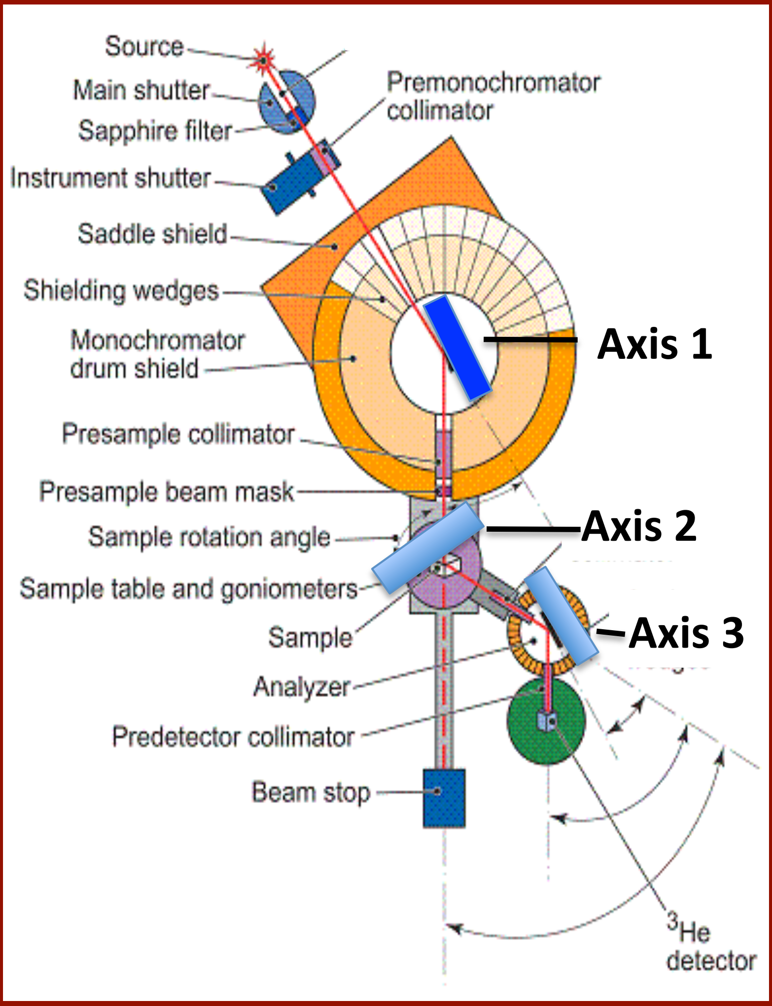
Phase diagram $Sr_3(Ir_{1-x}Ru_x)_2O_7$



Neutron scattering—TAS

- For diffraction: $|k_i|=|k_f|$; $Q=2k\sin\Theta$





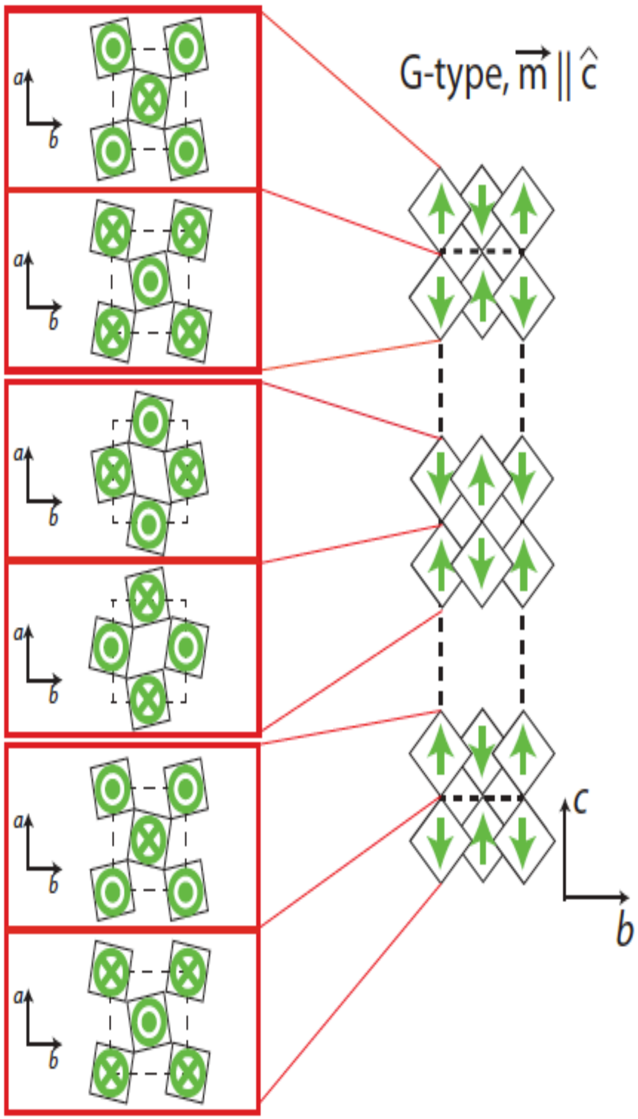
$$\left(\frac{\partial\sigma}{\partial\Omega}\right)_{elastic} = N\left(\frac{\gamma r_0}{2\mu_B}\right)^2 \frac{(2\pi)^3}{v_0}\sum_{\tau} e^{-2W}\partial(Q-\tau)\left|\hat{\tau}\times M(Q)\times\hat{\tau}\right|^2$$

Intensity

F.T. of $M(r)^2$

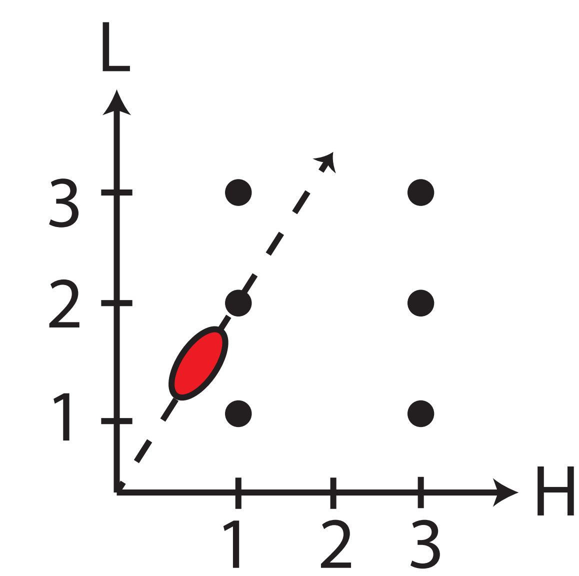
Normalize to known nuclear Bragg peaks to get N and m_{AF} in abs. units.

AF Bragg reflections

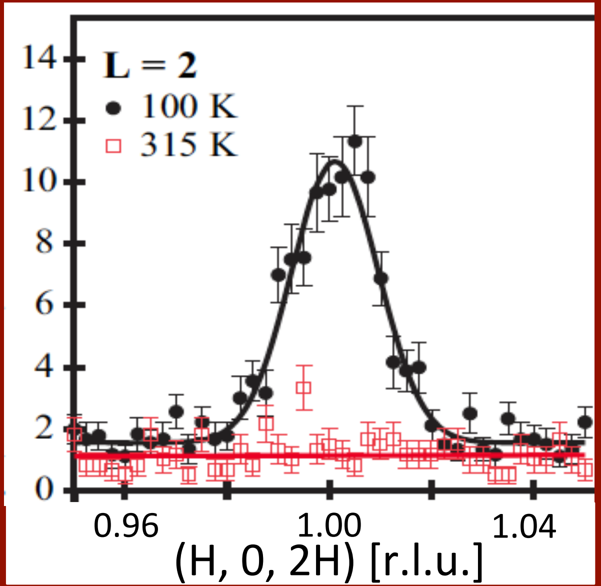


- Expect peaks at:
  - (odd, 0, L=odd)
  - (0, odd, L=even)
- Twin magnetic domains:
  - (odd, 0, L=integer) appear

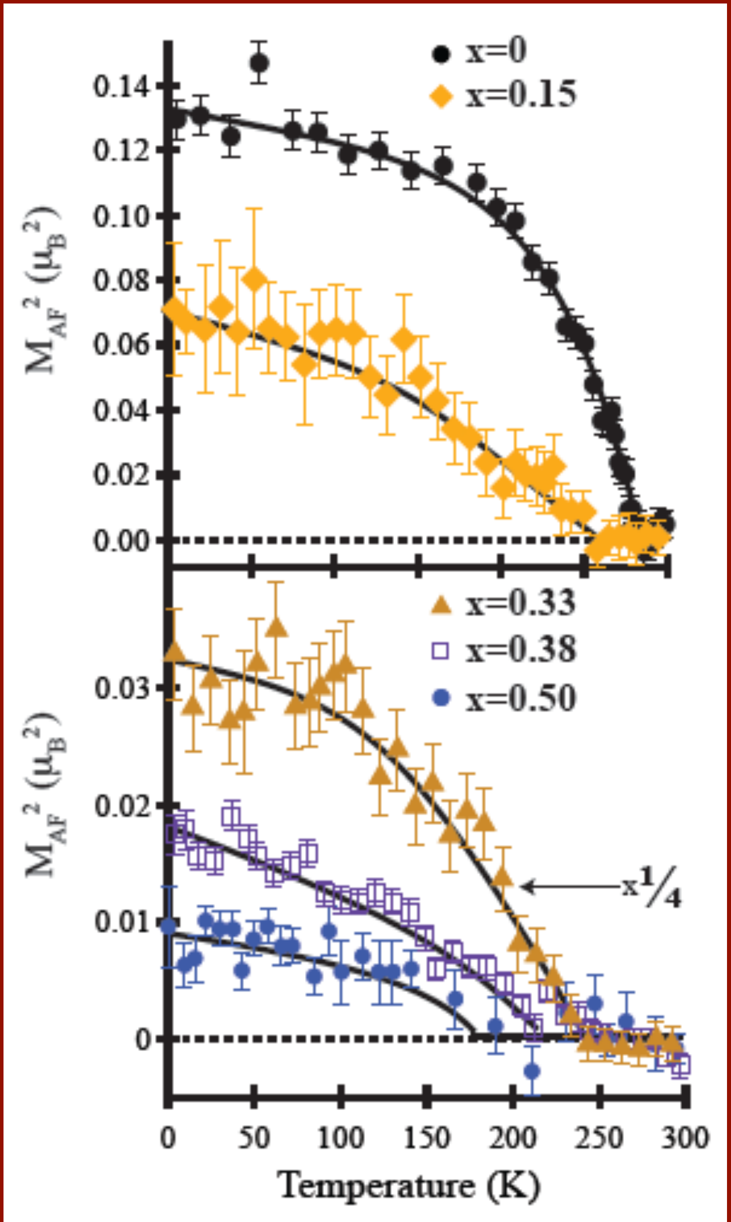
Map of reflections in (H, 0, L)



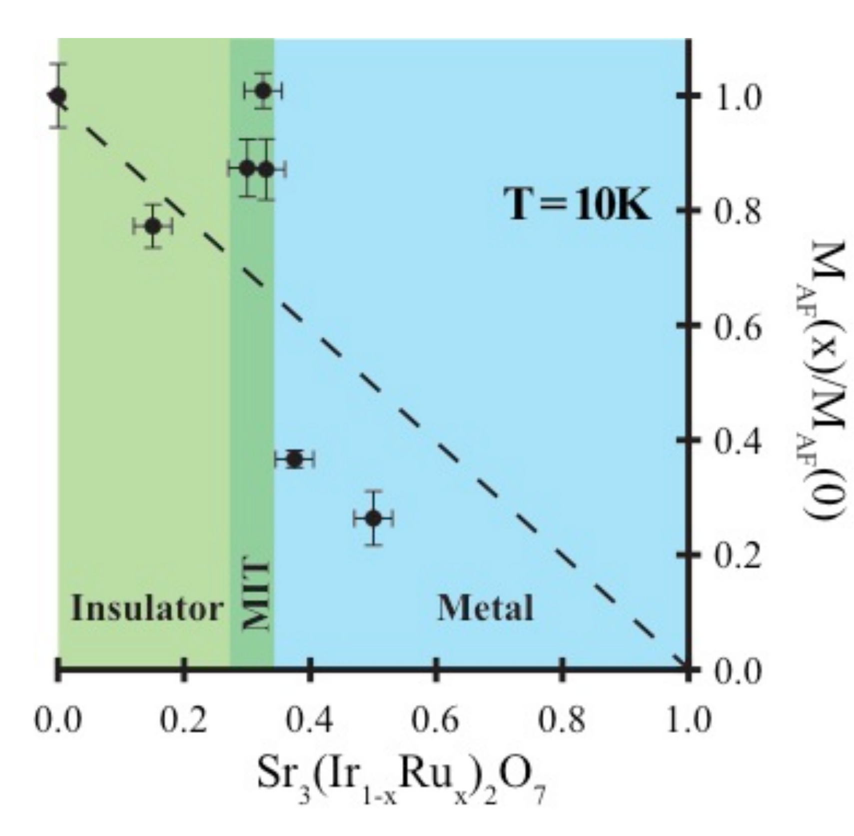
Measured (1,0,2) Sr-327



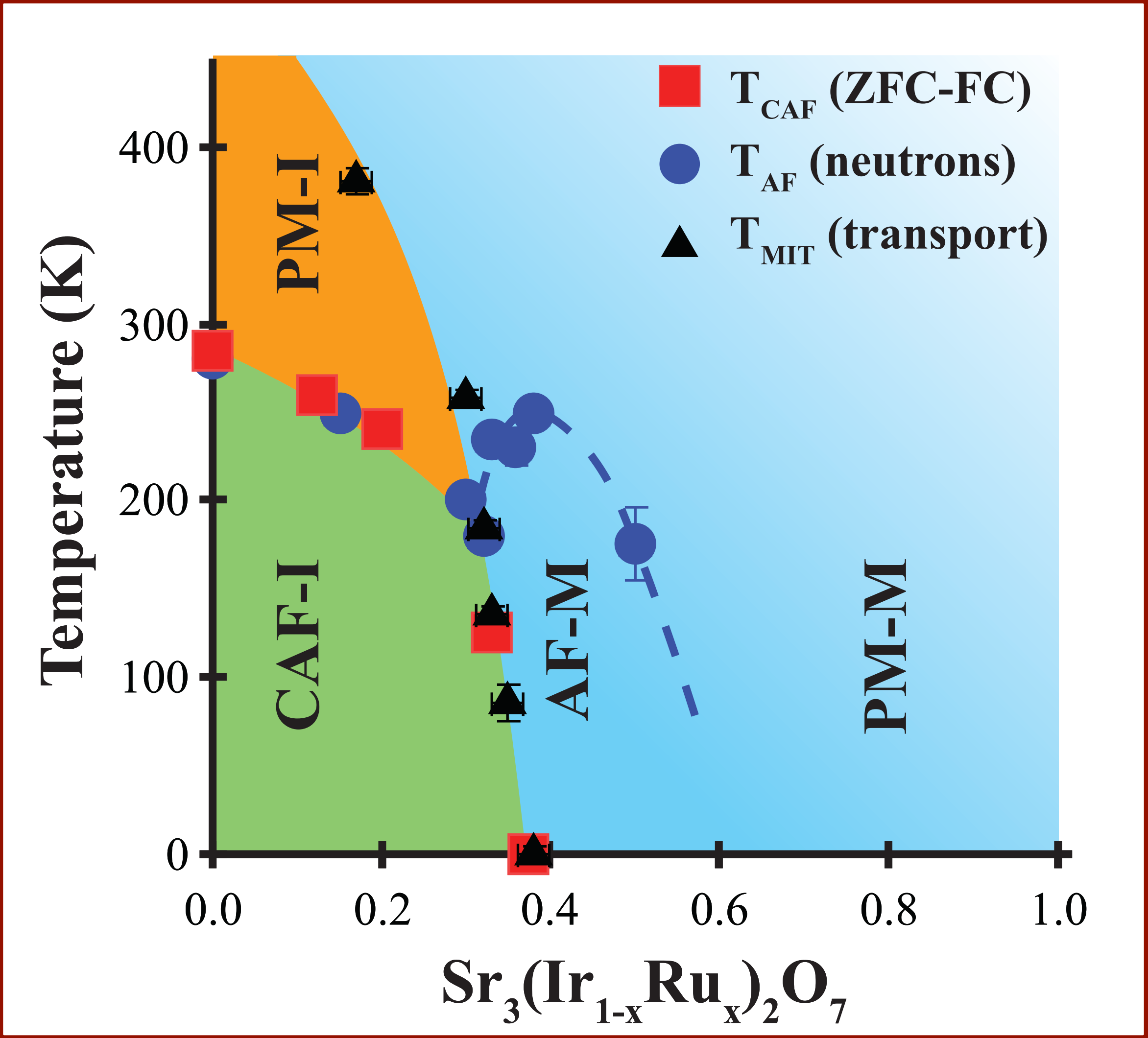
Neutron diffraction studies

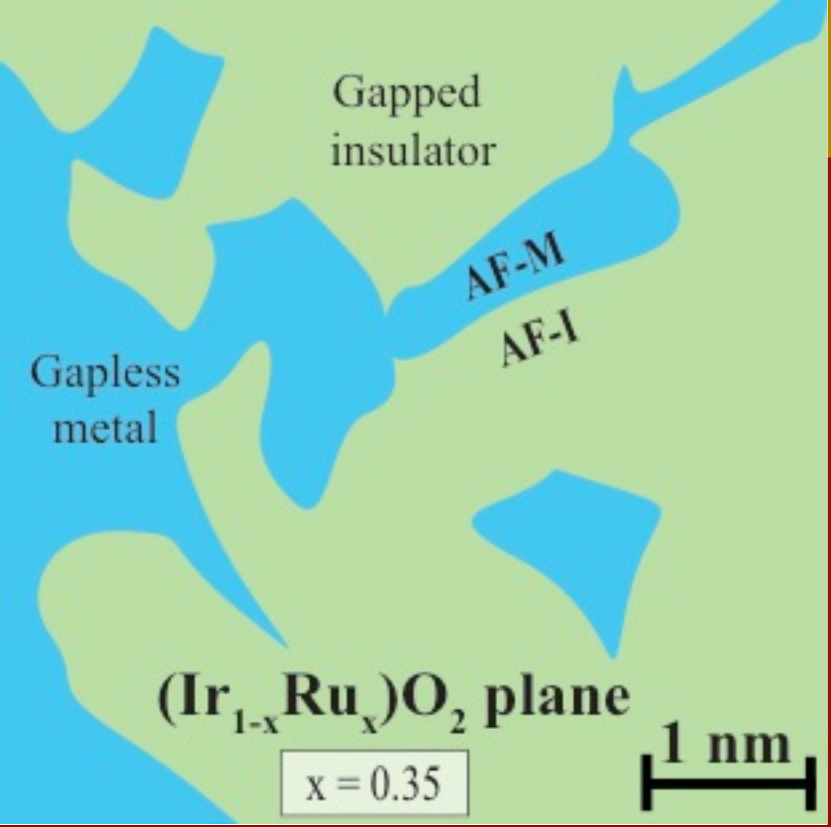


- Insulating regime: T_{AF} tracks T_{irr}
- AF order survives across MIT:
  - Same Q positions
- Enhanced moment near critical point



Phase diagram $Sr_3(Ir_{1-x}Ru_x)_2O_7$

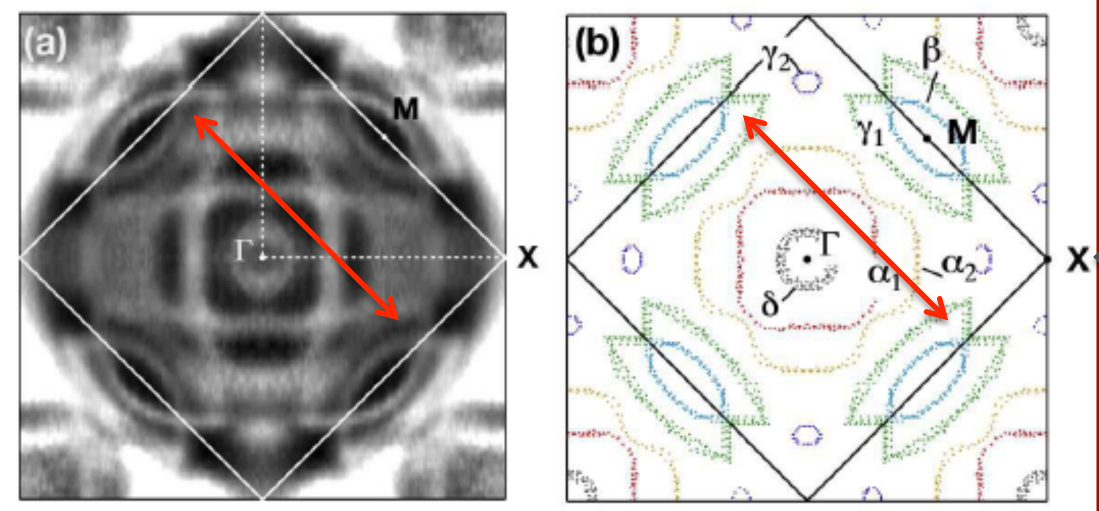




Phase separation?

- Phase separation of holes within AF insulating bckg.
  - S=1 local moment
  - Near-percolative behavior
- Proximity induced AF order in hole-rich regions
  - Large susceptibility needed…
  - Enhanced moment at maximum heterogeneous area

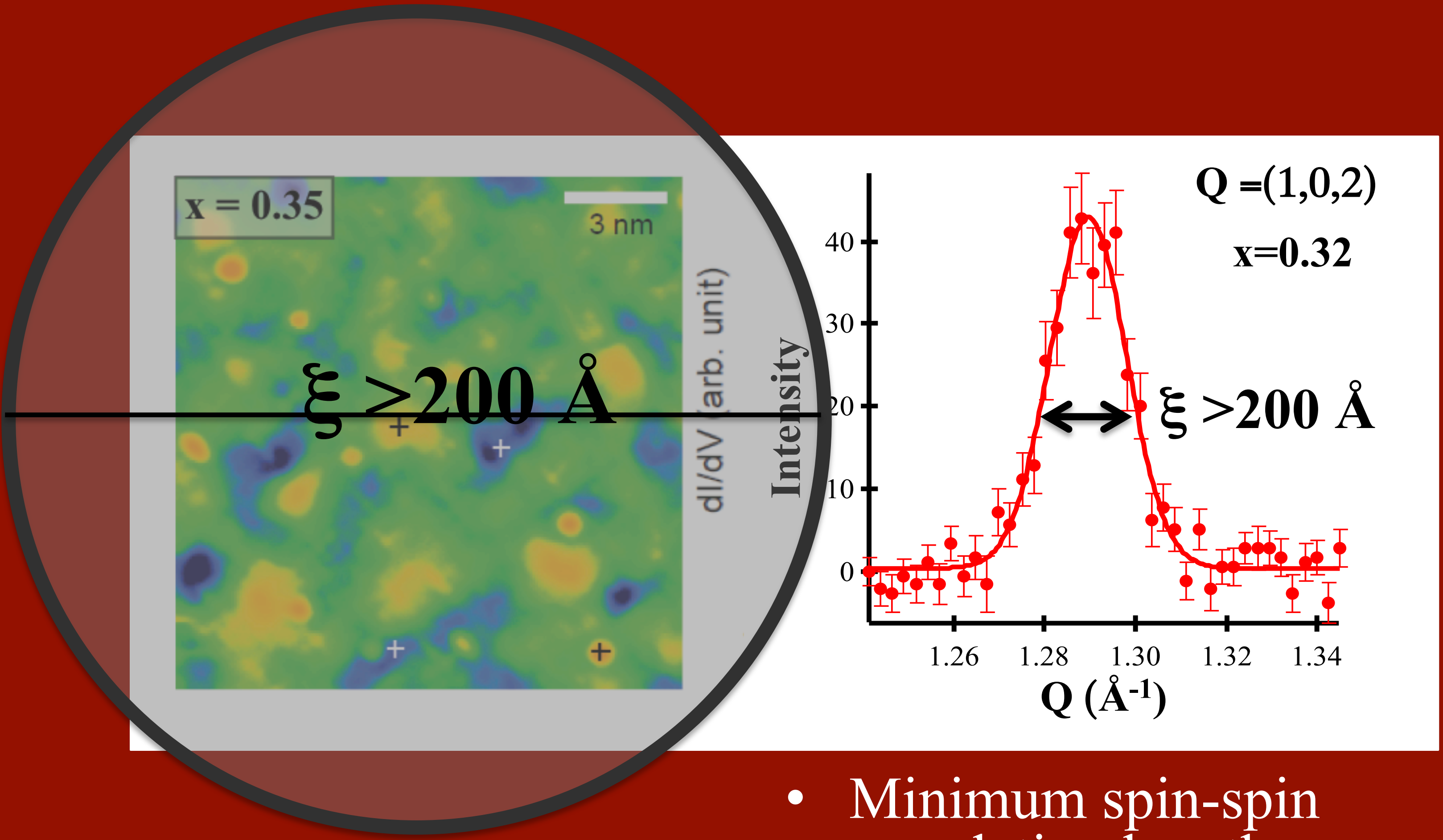
$Sr_3Ru_2O_7$



Divergent DOS nested at necessary Q in $Sr_3Ru_2O_7$

A. Tamai et al., PRL (2010).

Nanoscale phase separation resolved via STS



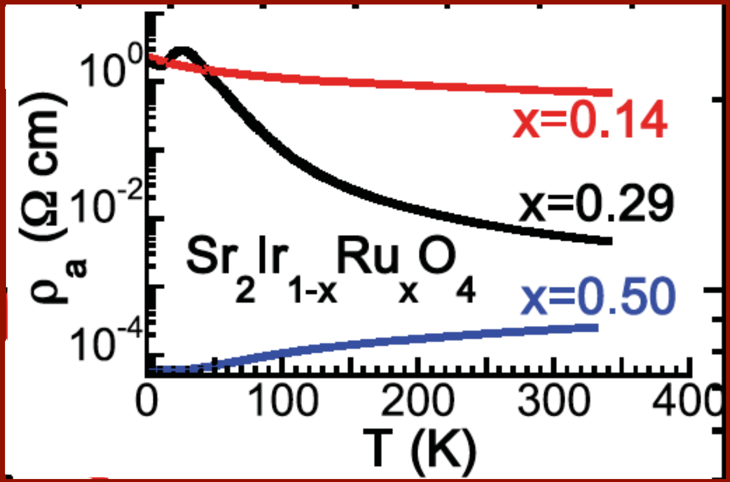
- Minimum spin-spin correlation length ξ_{min}>> electronic phase separated puddles

Generic percolation with B-site doping?

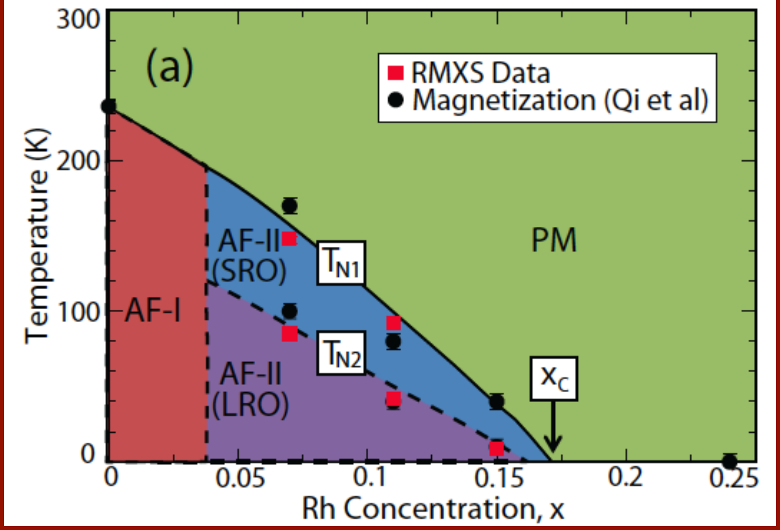
- $Sr_2Ir_{1-x}Ru_xO_4$
  - MIT between $0.29<x_c<0.50$
  - No studies of AF to date
- $Sr_2Ir_{1-x}Rh_xO_4$
  - $x_c=0.17$
  - Rh^{3+} replaces Ir^{4+} (induced Ir^{5+})
  - Percolation threshold 34%
  - Holes remain localized

MIT not thermodynamic PT with B-site doping (percolation/ interface physics)

T. F. Qi et al. Phys. Rev B (2012).

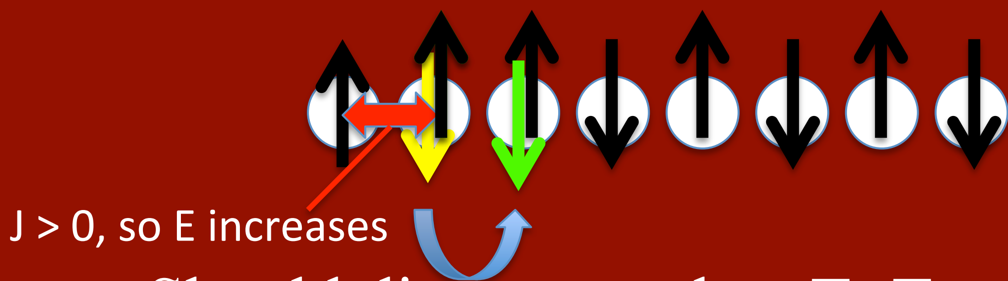


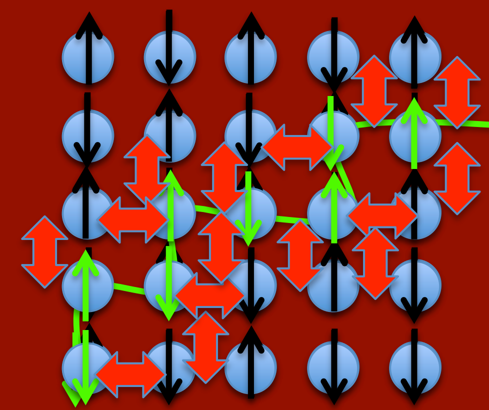
J. P. Clancy et al. arxiv (2013).



Mechanisms for carrier localization

- Doped carriers into magnetic semiconductor:
  – Nagaev: d-d interactions into self-localized states



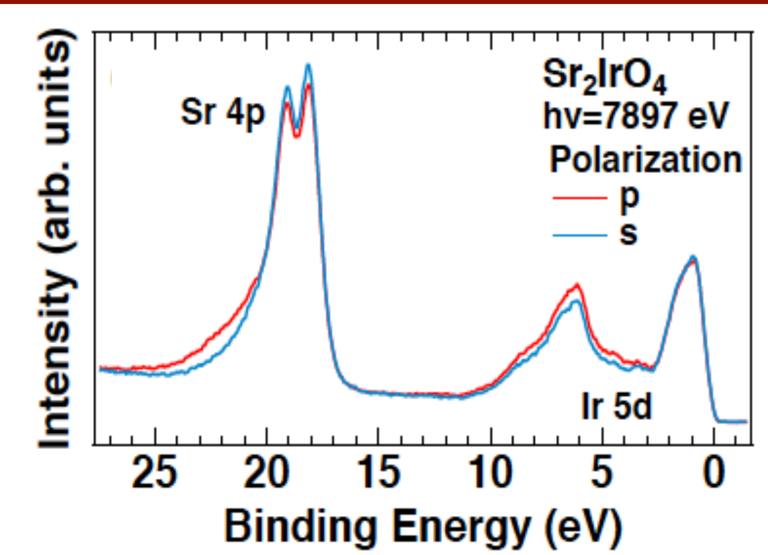


  – Should disappear when $T>T_N$
- Disorder mechanism: Anderson localization
  – Large negative magnetoresistance, VRH transport
- Emery et al. t-J model of phase separation
  – Correlation driven, U essential
- Single-ion mechanism
  – Local variation in λ_{SOC}

MIT in spin-orbit Mott phase: A-site substitution

A-site doping (La onto Sr-sites)

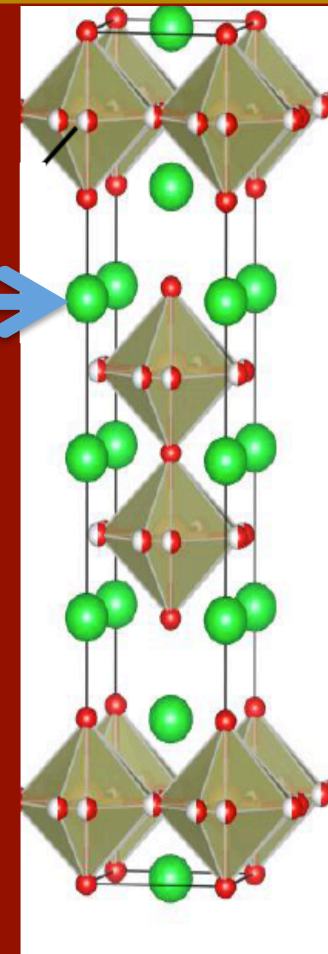
Suga et al., arxiv (2013)



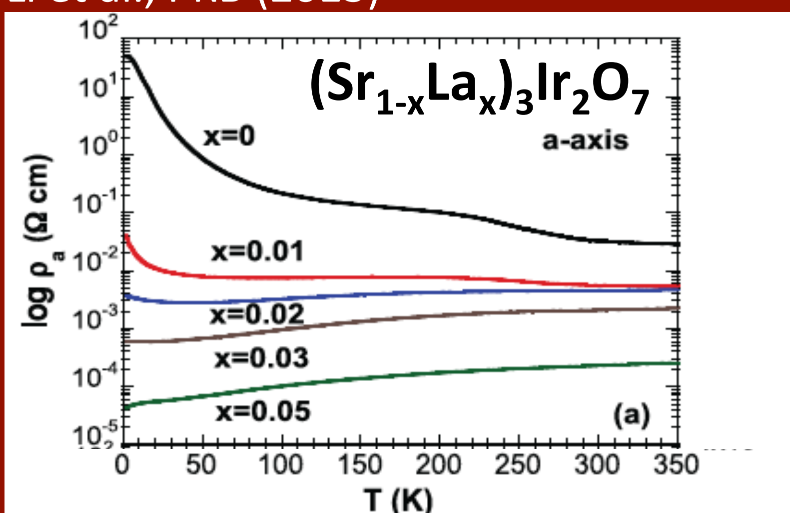
$(Sr_{1-x}La_x)_3Ir_2O_7$

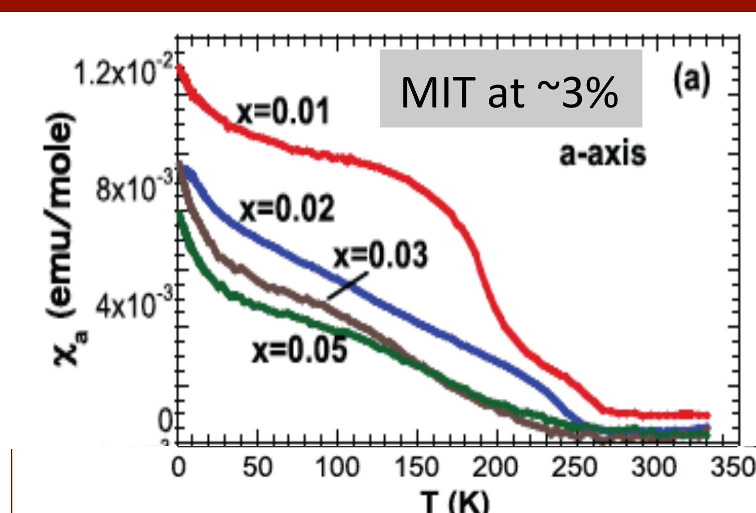
Sr^{2+} → La^{3+}

~10% size reduction

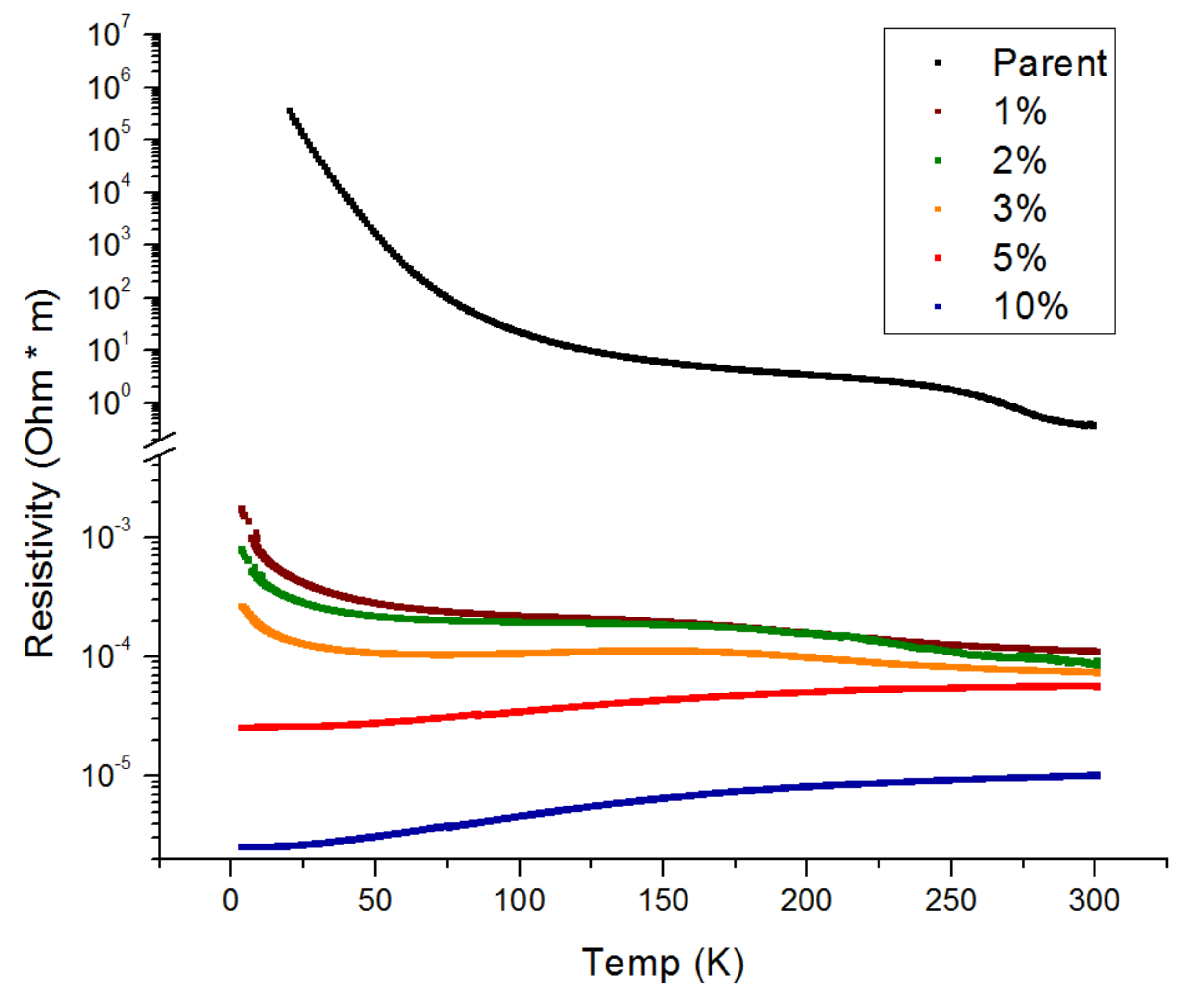


Li et al., PRB (2013)





Transport in La-doped Sr-327

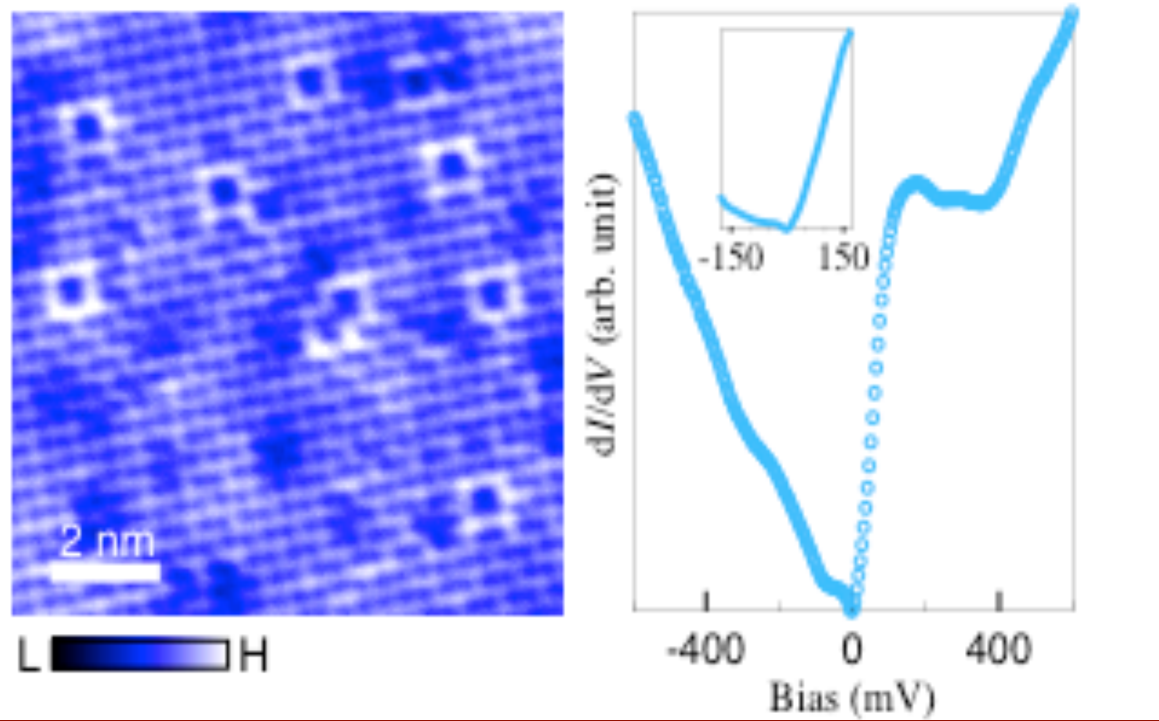


$(Sr_{1-x}La_x)_3Ir_2O_7$

- MIT at 4% La-doping
- Resolution ~1%

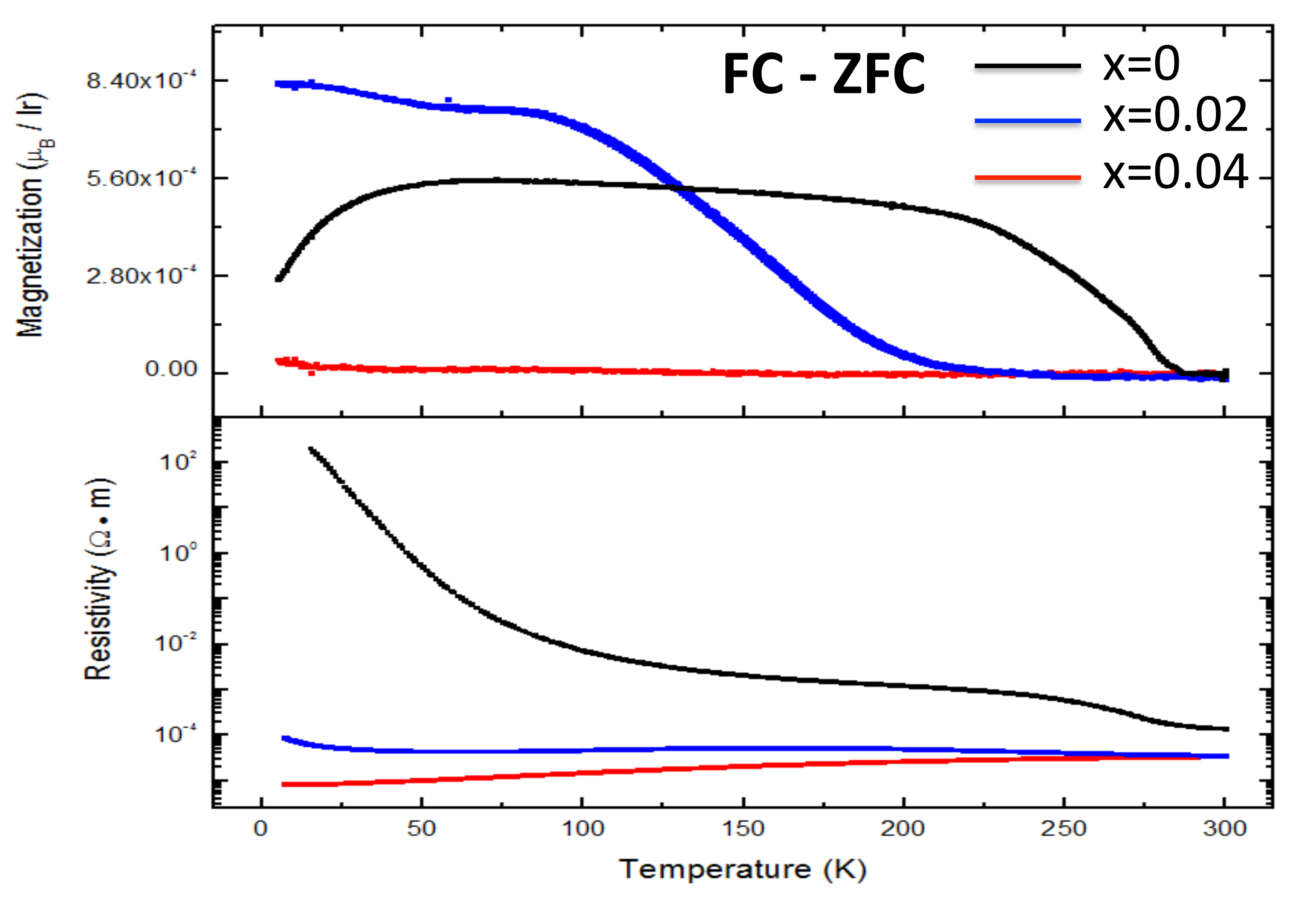
Globally metallic phase

V. Madhavan et al., unpublished



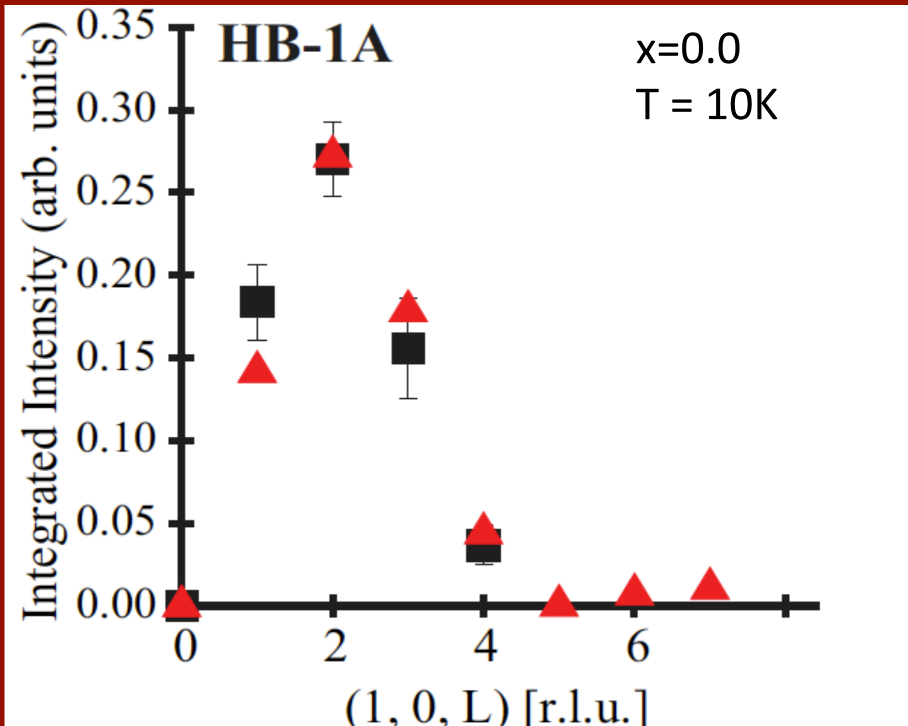
- Target 4% La-doped sample
  - Metallic transport
  - No gapped regions

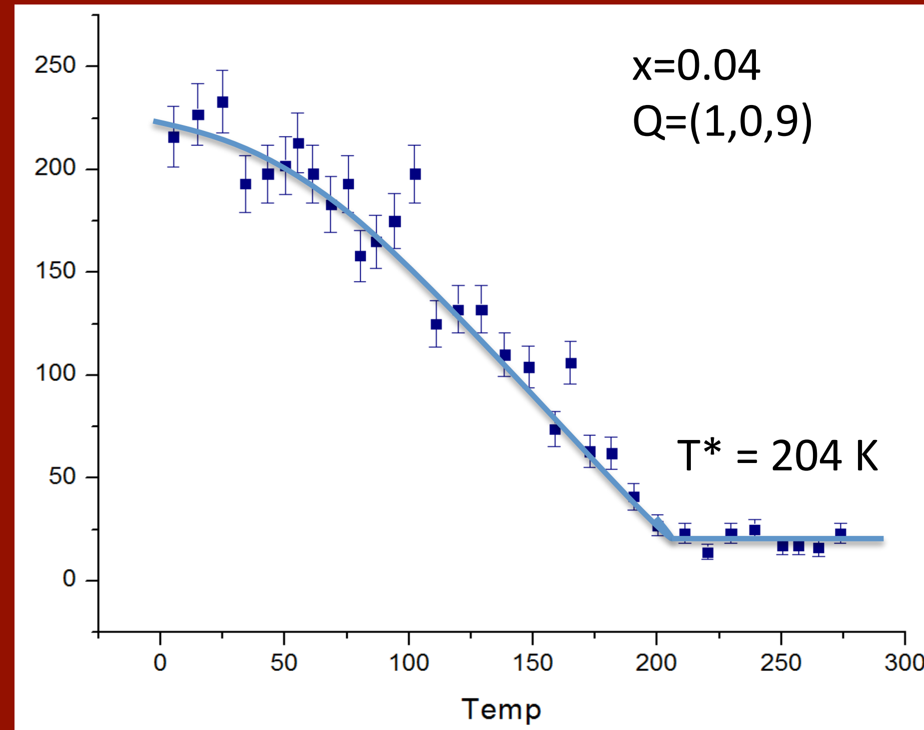
Magnetization in La-doped Sr-327



- Net magnetization suppressed near MIT

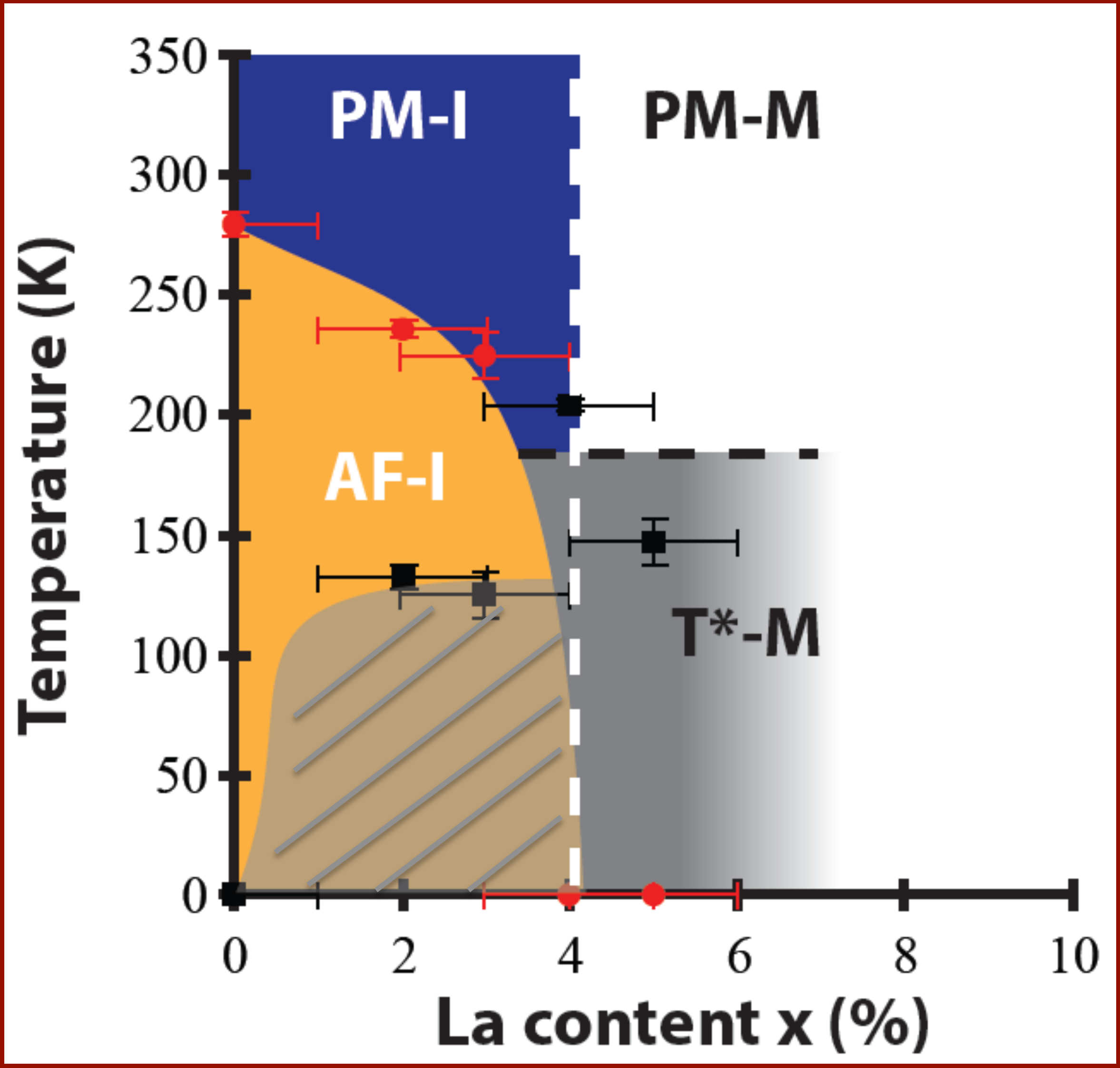
AF order and T* in La-doped Sr-327



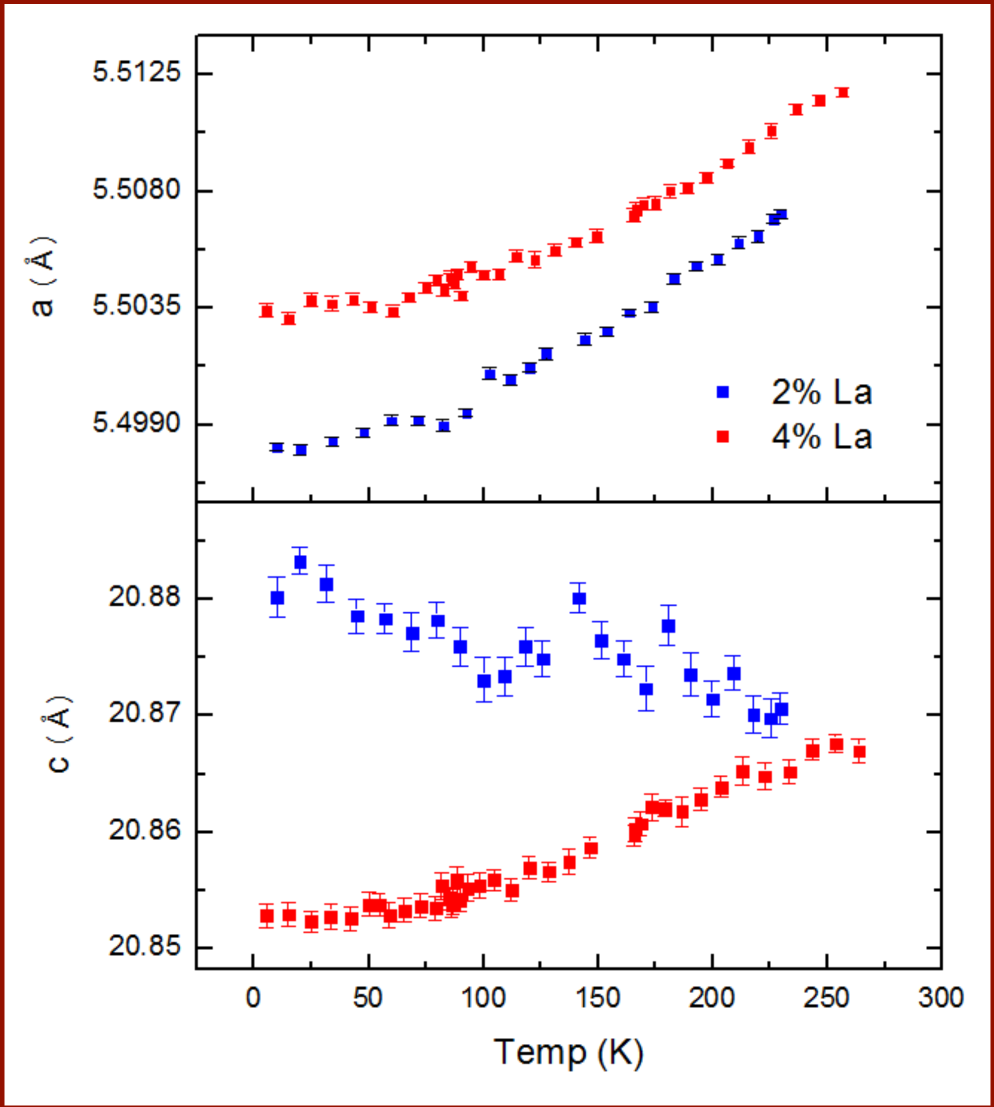


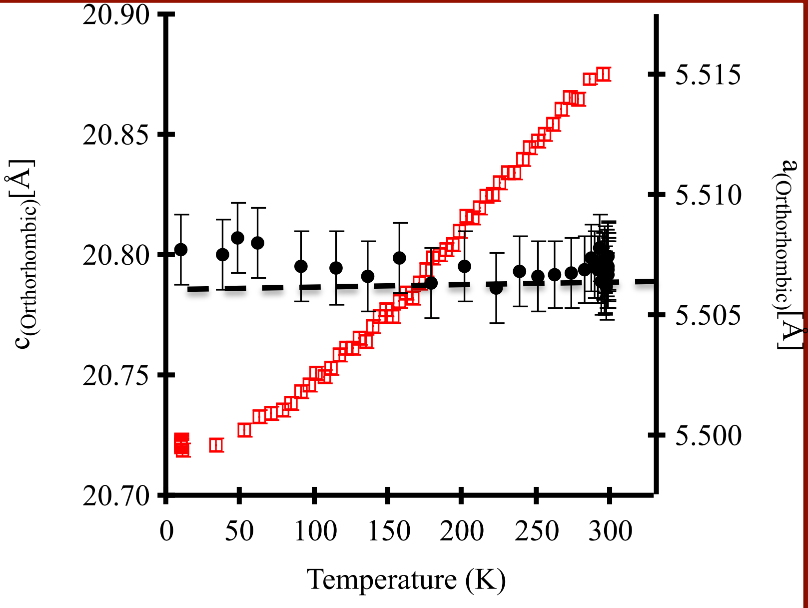
- Parent AF order strongly suppressed
  - Vanishes by 4% La-doping
- T* appears at (odd, 0, odd)-type positions
  - Survives across MIT

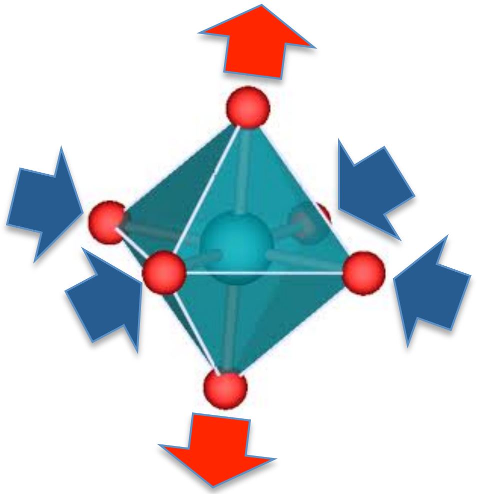
Tentative phase diagram $(Sr_{1-x}La_x)_3Ir_2O_7$



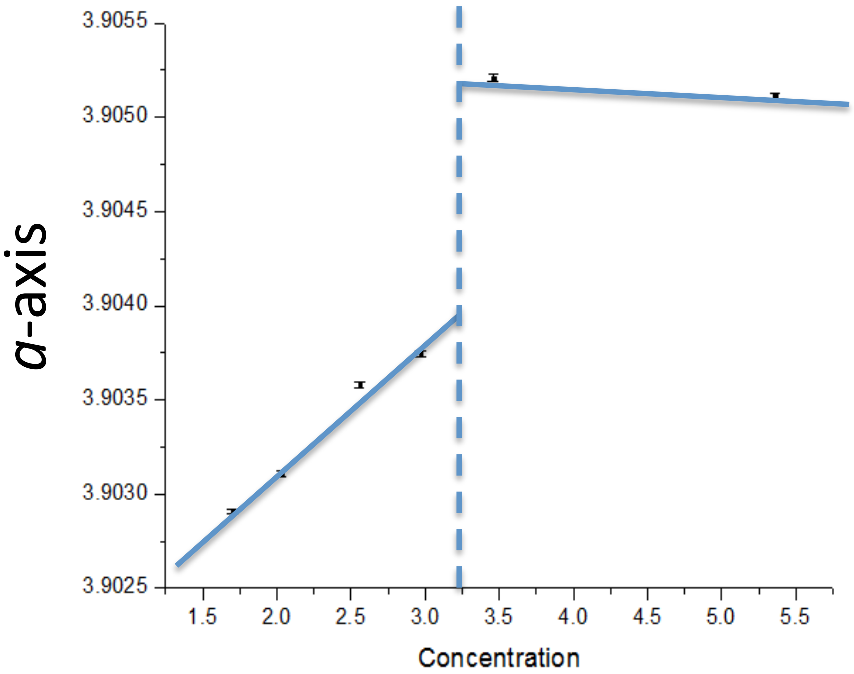
Anisotropic Expansion

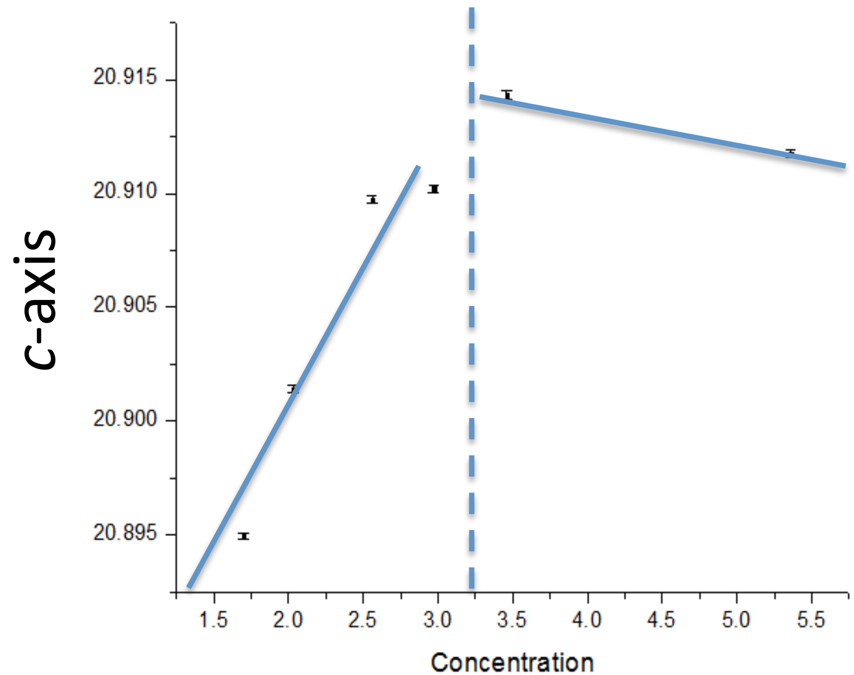






Lattice parameters vs La conc.

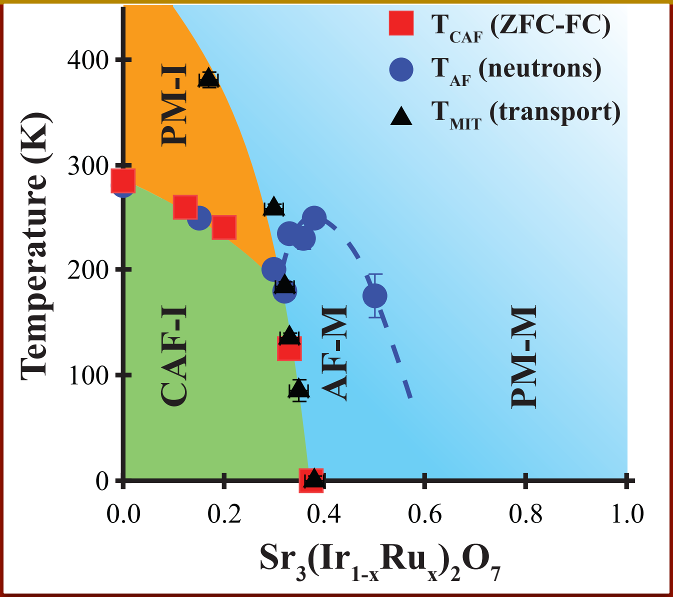


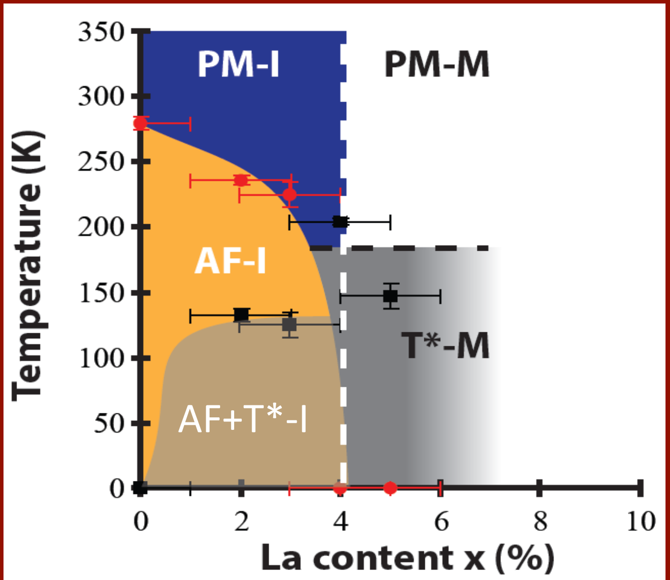


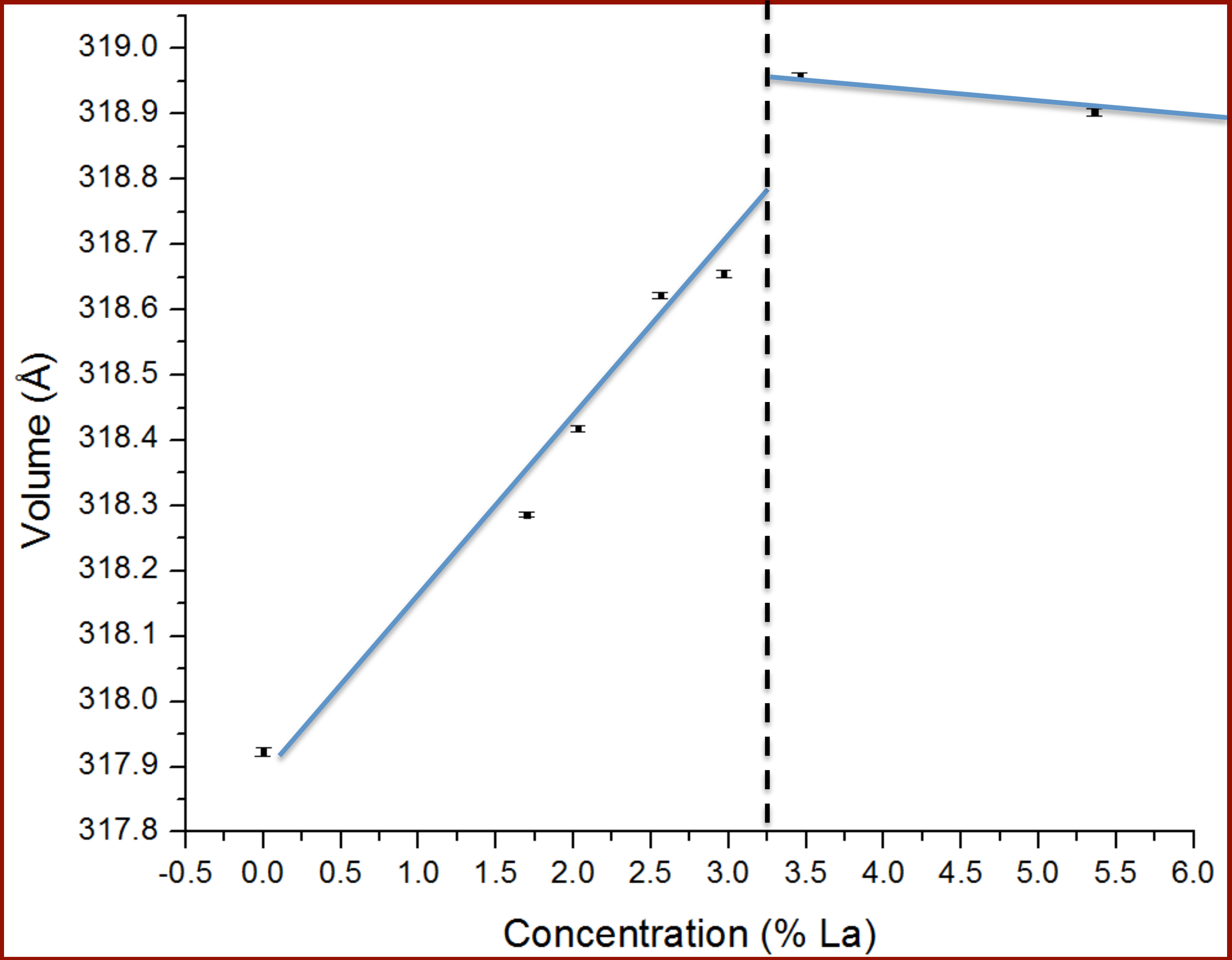
- Discontinuity in unit cell volume at x_c

Conclusions

- Explored mechanisms for doping in Sr-327
  - B-site: percolative mechanism
  - A-site: bandwidth tuning/carrier doping
- B-site doping: Nanoscale interface effects
  - SOC Mott phase/correlated metal
  - Enhanced moment at $x_{percolation}$
- A-site doping: First order transition out of Mott phase
  - T* simultaneous to MIT







Volume (Å)
319.0
318.9
318.8
318.7
318.6
318.5
318.4
318.3
318.2
318.1
318.0
317.9
317.8
-0.5
0.0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
5.0
5.5
6.0
Concentration (% La)