Program: Intertwinned17
KITP, UCSB
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Unconventional Metals in Doped J_{eff}=1/2 Mott Insulators

Unusual magnetism and competing electronic states in Ruddlesden-Popper series strontium iridates

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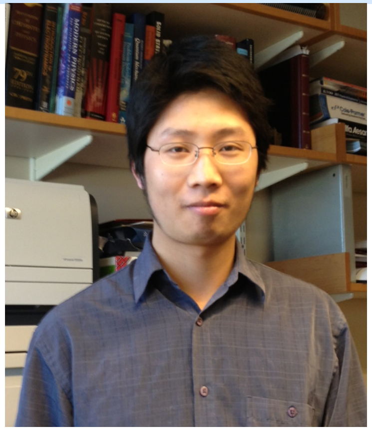
Santa Barbara

Collaborators

Tom Hogan



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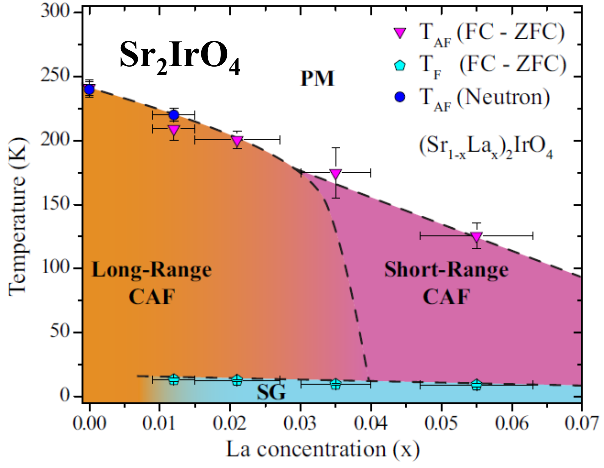
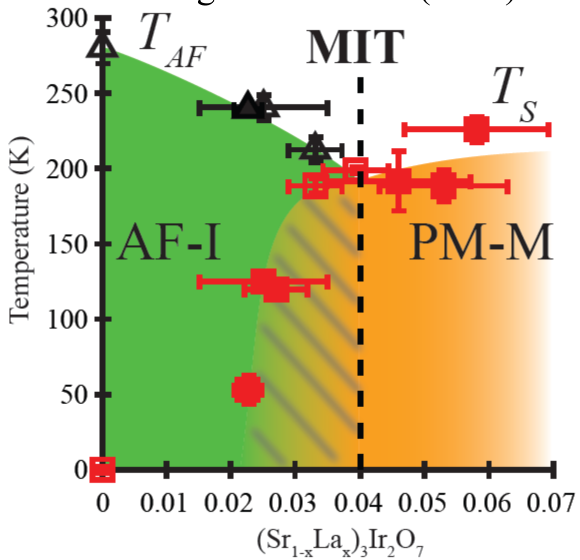
Talk Outline

- Ruddlesden-Popper iridates as J_{eff}=1/2 Mott insulators
  - Monolayer Sr_2IrO_4
  - Bilayer $Sr_3Ir_2O_7$
- Effect of electron doping R.P. iridates
  - Competing electronic states
  - $(Sr_{1-x}La_x)_2IrO_4$
  - $(Sr_{1-x}Lax)_3Ir_2O_7$
- Effect of hole doping R.P. iridates
  - Intermediate SDW state beyond the collapse of Mott state
  - $Sr_3(Ir_{1-x}Ru_x)_2O_7$
- Summary

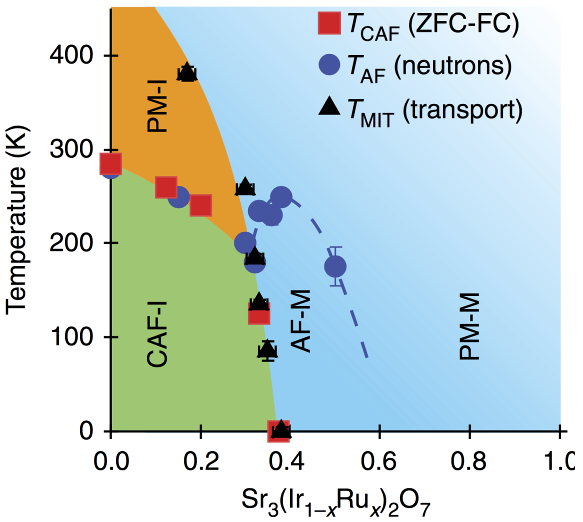
X. Chen et al. PRB (2015)



T. Hogan et al. PRL (2015)

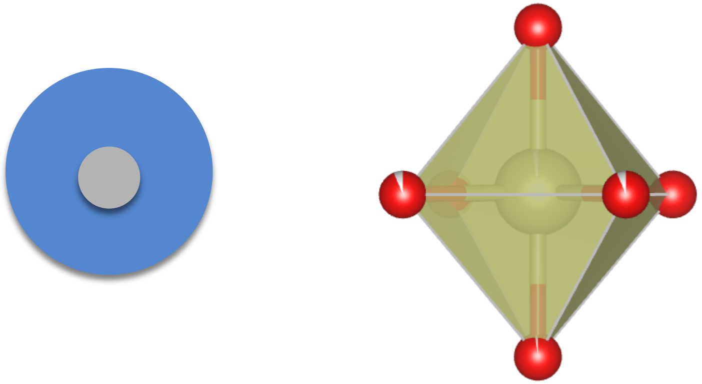


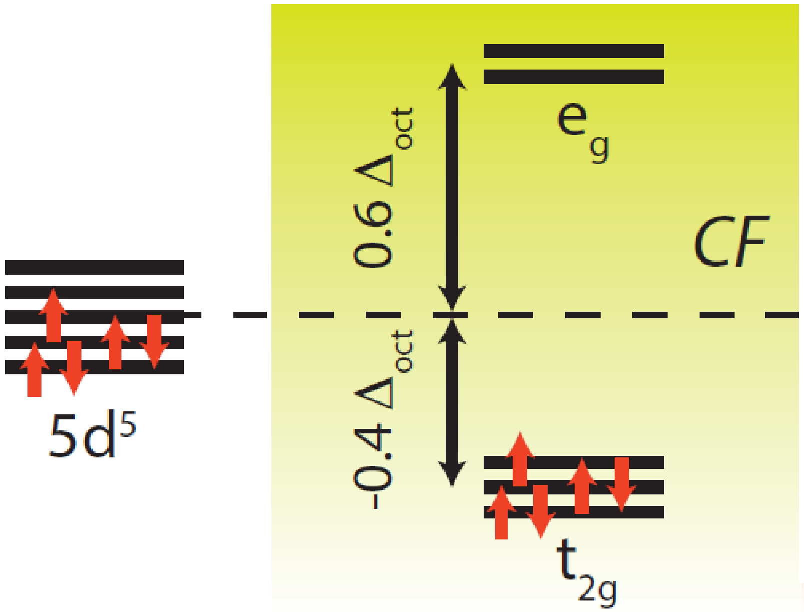
C. Dhital et al. Nat. Comm. (2014)



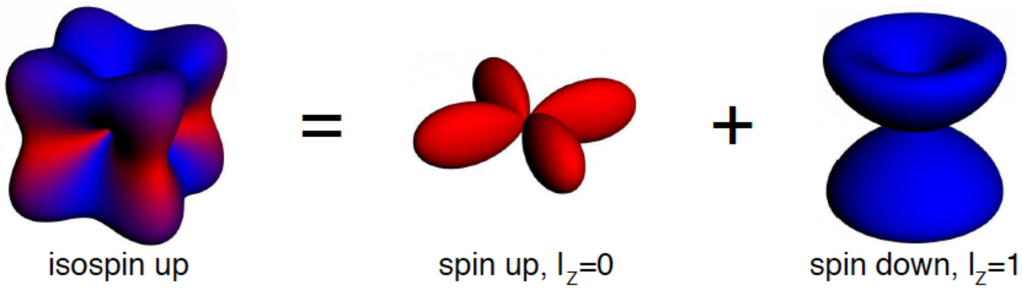
SOC in a cubic crystal field

Heavy transition metal cation with 5 valence electrons in a cubic crystal field



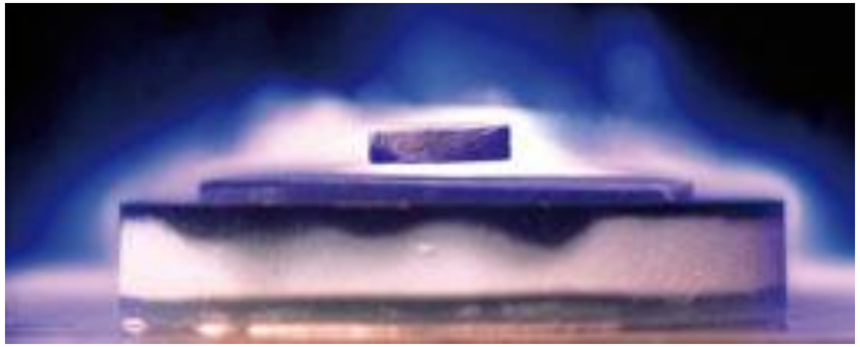


$$\left|J_{eff}=\frac{1}{2};m_J=\pm\frac{1}{2}\right\rangle=\frac{1}{\sqrt{3}}\left(\left|xy,\mp\sigma\right\rangle\mp\left|yz,\pm\sigma\right\rangle+i\left|zx,\pm\sigma\right\rangle\right)$$



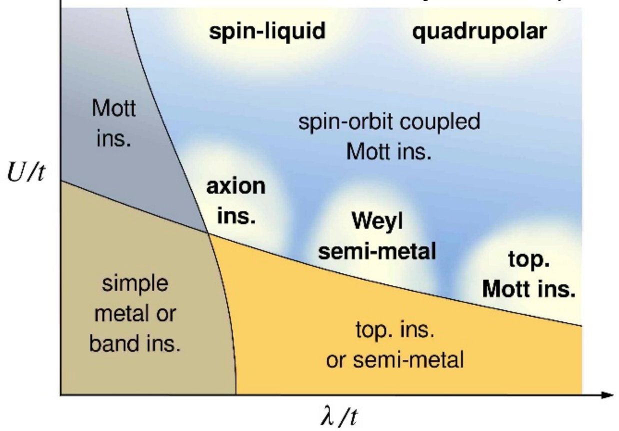
Phys. Rev. Lett. 102, 017205 (2009)

New phases in/near J_{eff}=1/2 Mott states

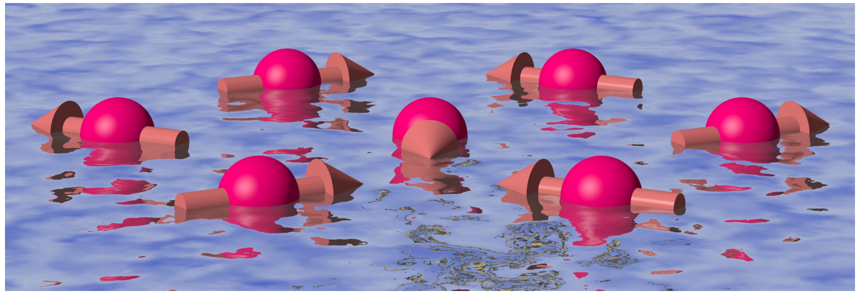


Layered lattices with corner sharing IrO_6 octahedra
R.P. iridates Sr_2IrO_4, $Sr_3Ir_2O_7$, …

Witzak-Krempa et al.,
Ann. Rev. Cond. Matt. Phys. 5, 57 (2014).



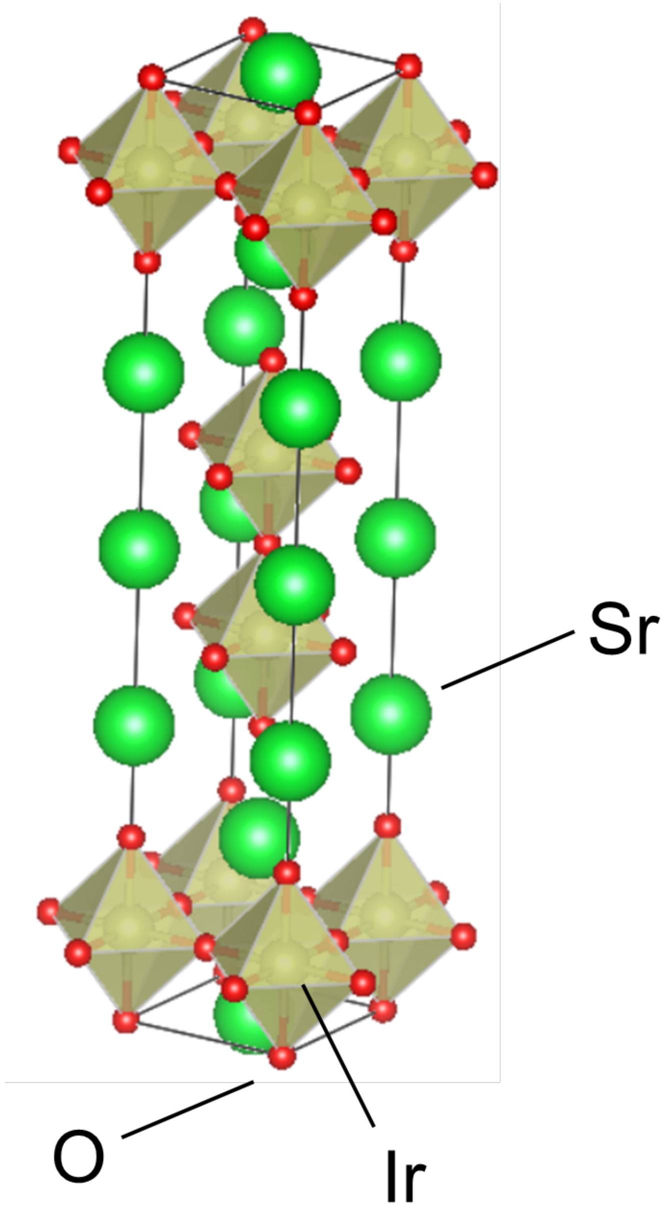
Pyrochlore lattice of IrO_6 octahedra
$RE_2Ir_2O_7$, $Y_2Ir_2O_7$, $Bi_2Ir_2O_7$ …

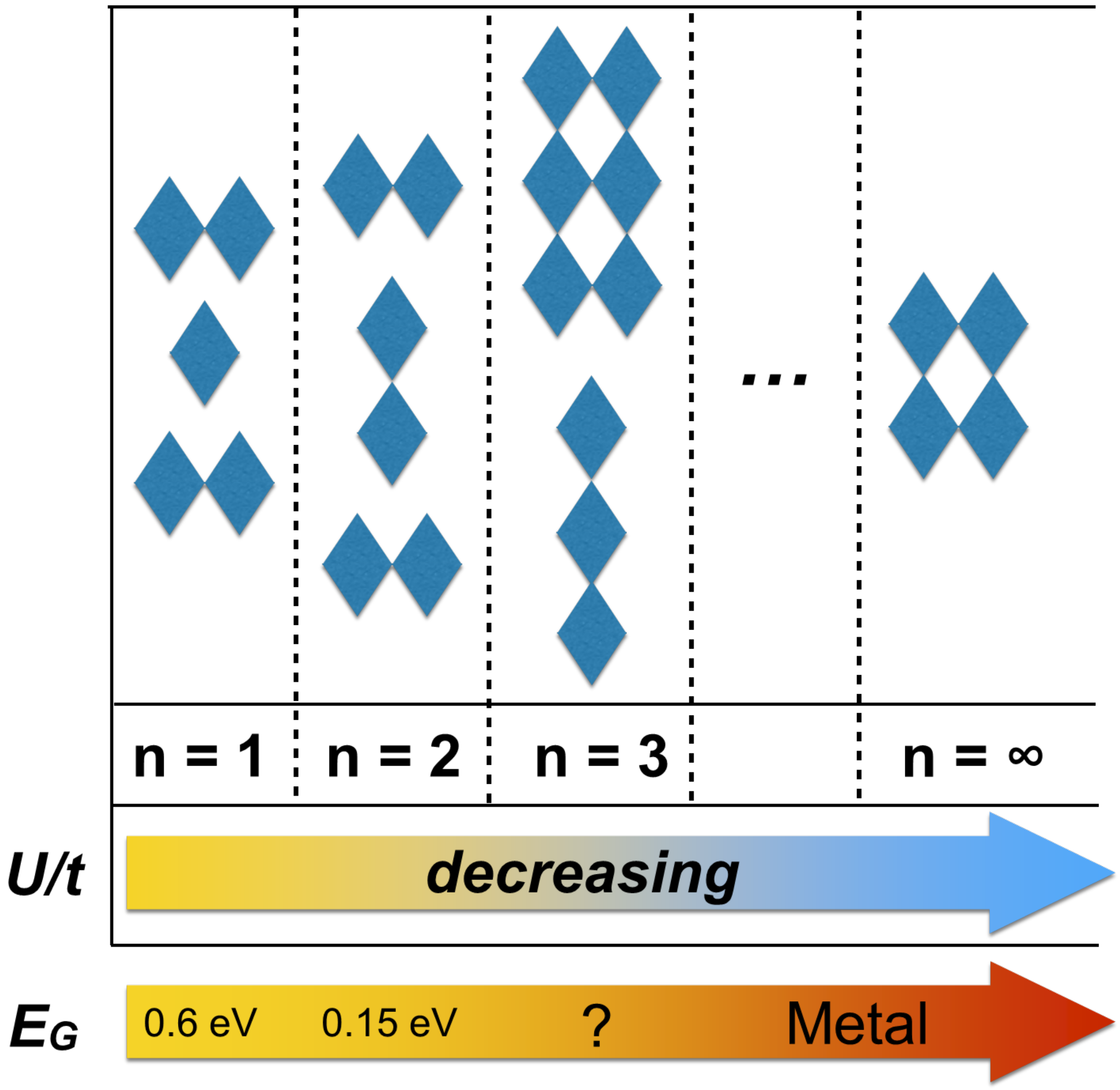


Frustrated lattices with edge-sharing IrO_6 octahedra
$\alpha\beta\gamma$-Na_2IrO_3, $Na_4Ir_3O_8$,…

Multilayer modulation of J_{eff}=1/2 wavefunction,
5d-3d electron interfaces, 111 iridate films

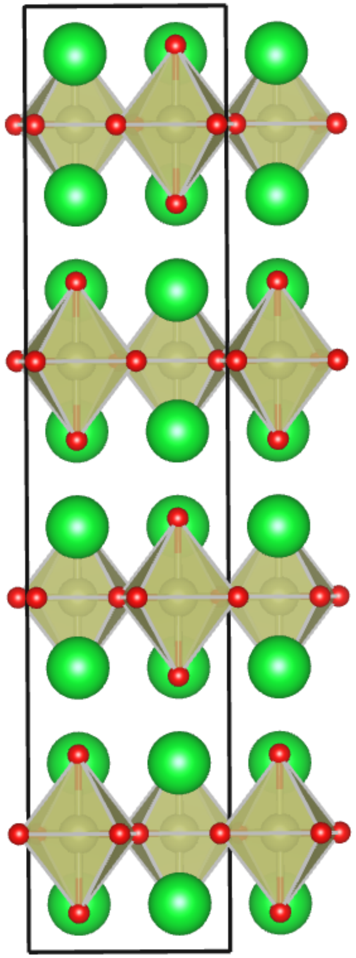
R.P. Iridates: $Sr_{n+1}Ir_nO_{3n+1}$

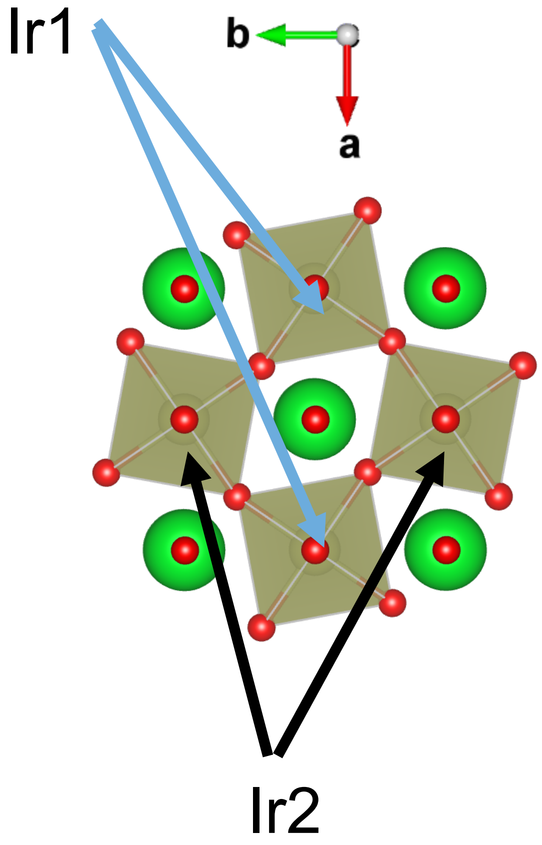




Okada et al. Nat. Mat. (2012) Dai et al. PRB (2014)

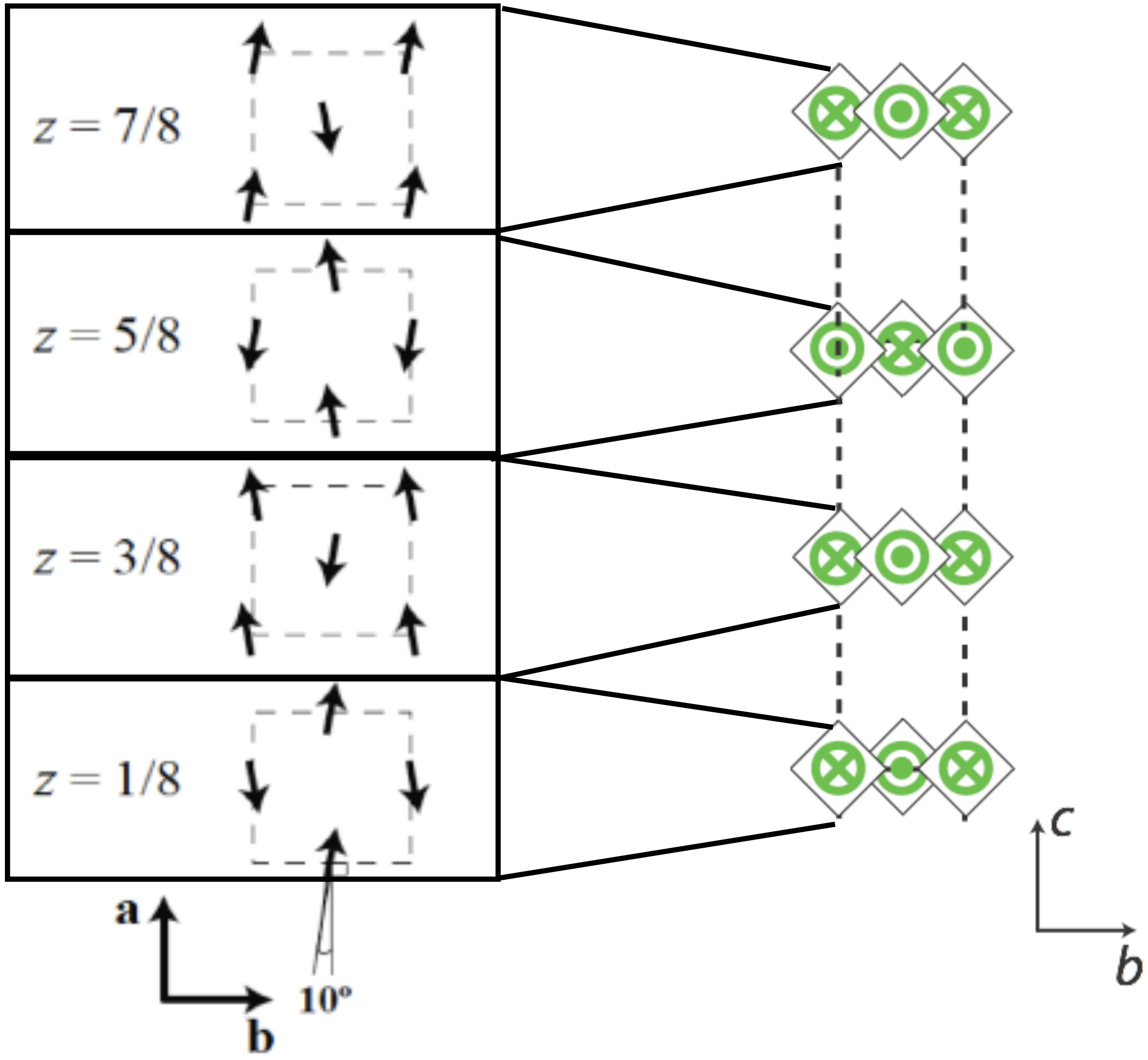
Electron-doping Sr_2IrO_4: La substitution





$I4_1/a$ most likely space group:
Torchinsky et al., PRL (2015)

Dhital et al., PRB (2013)



High-T_c predicted in *n*-type Sr_2IrO_4

PRL **106,** 136402 (2011) PHYSICAL REVIEW LETTERS week ending 1 APRIL 2011

Twisted Hubbard Model for Sr_2IrO_4: Magnetism and Possible High Temperature Superconductivity

Fa Wang and T. Senthil

$$H = -\sum_{\langle jk\rangle,\alpha} (t + i\epsilon_\alpha \epsilon_j \tilde{t}) d^\dagger_{j,\alpha} d_{k,\alpha} - \sum_{\langle\langle jk\rangle\rangle,\alpha} t' d^\dagger_{j,\alpha} d_{k,\alpha} - \sum_{\langle\langle\langle jk\rangle\rangle\rangle,\alpha} t'' d^\dagger_{j,\alpha} d_{k,\alpha} + U \sum_j d^\dagger_{j,\uparrow} d_{j,\uparrow} d^\dagger_{j,\downarrow} d_{j,\downarrow},$$

n-type iridate maps to *p*-type cuprate

PRL **110,** 027002 (2013) PHYSICAL REVIEW LETTERS week ending 11 JANUARY 2013

Monte Carlo Study of an Unconventional Superconducting Phase in Iridium Oxide $J_{\mathrm{eff}} = 1/2$ Mott Insulators Induced by Carrier Doping

Hiroshi Watanabe,* Tomonori Shirakawa, and Seiji Yunoki

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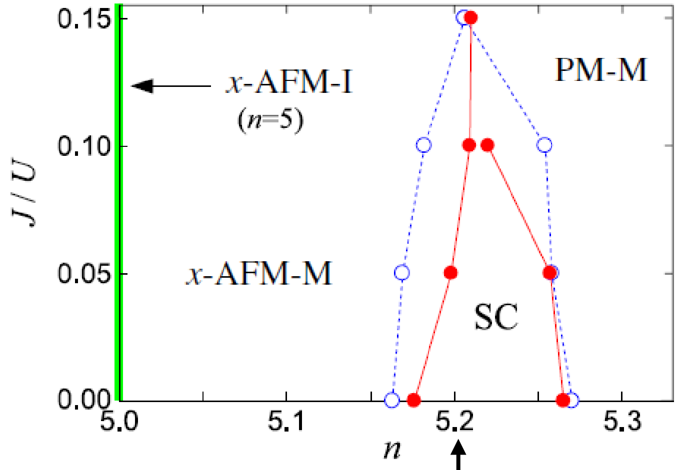
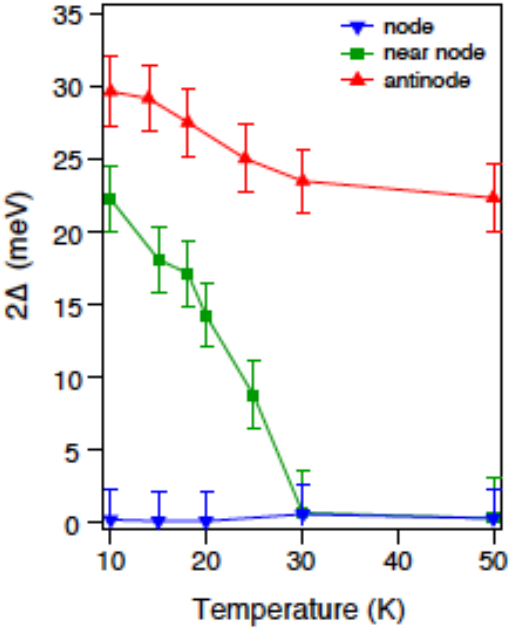
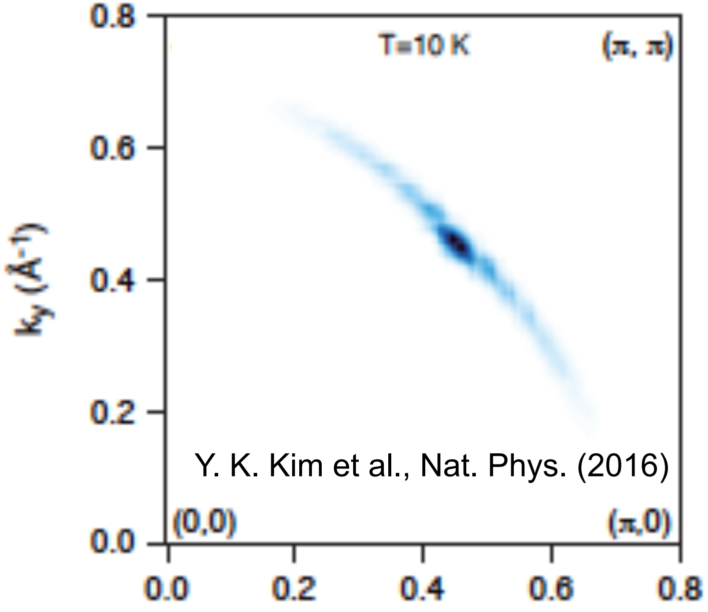
Other studies predicting SC, some also in *p*-type Sr_2IrO_4



SC predicted near 20% electrons/Ir cation

Unusual metals in surface doped Sr_2IrO_4



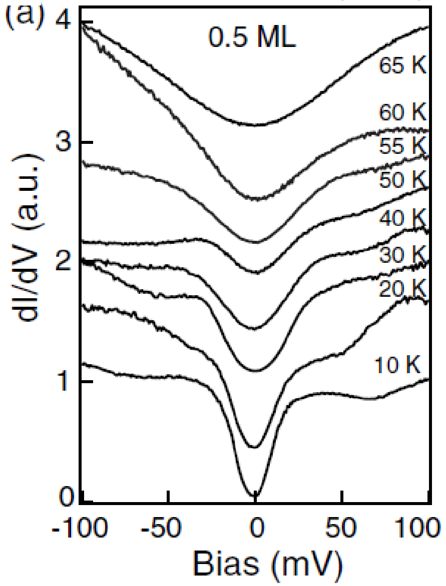


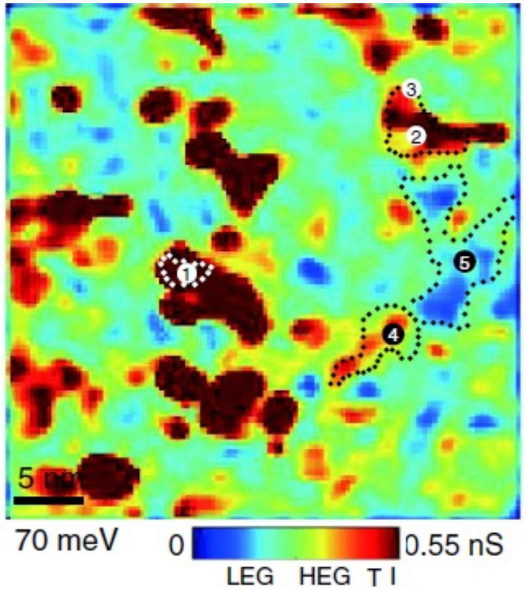
Y. K. Kim et al., Nat. Phys. (2016)

Nodal metal phase apparent around ~7% electrons/Ir

D-wave gap opens under surface K-dosing

Yan et al., PRX (2015)



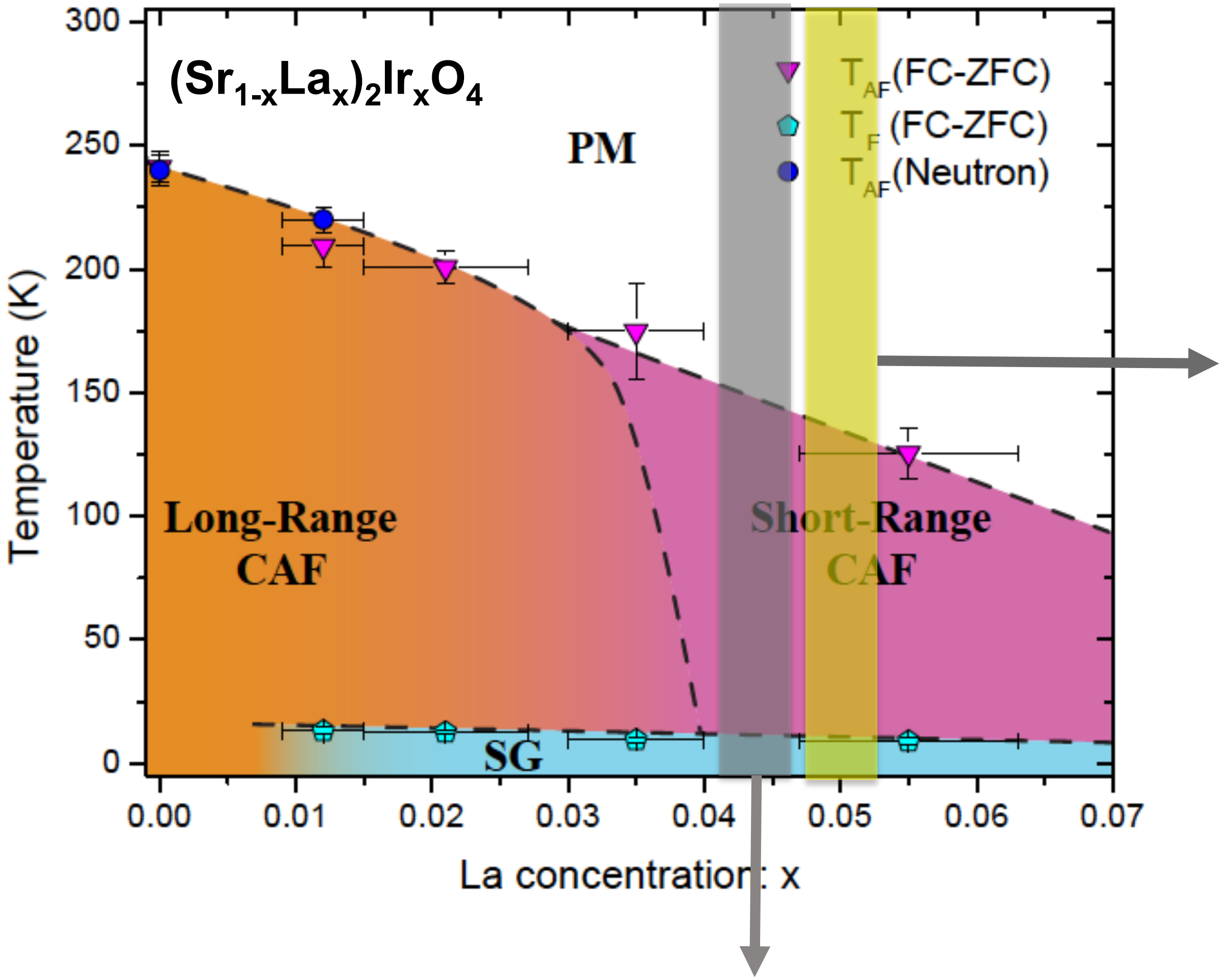


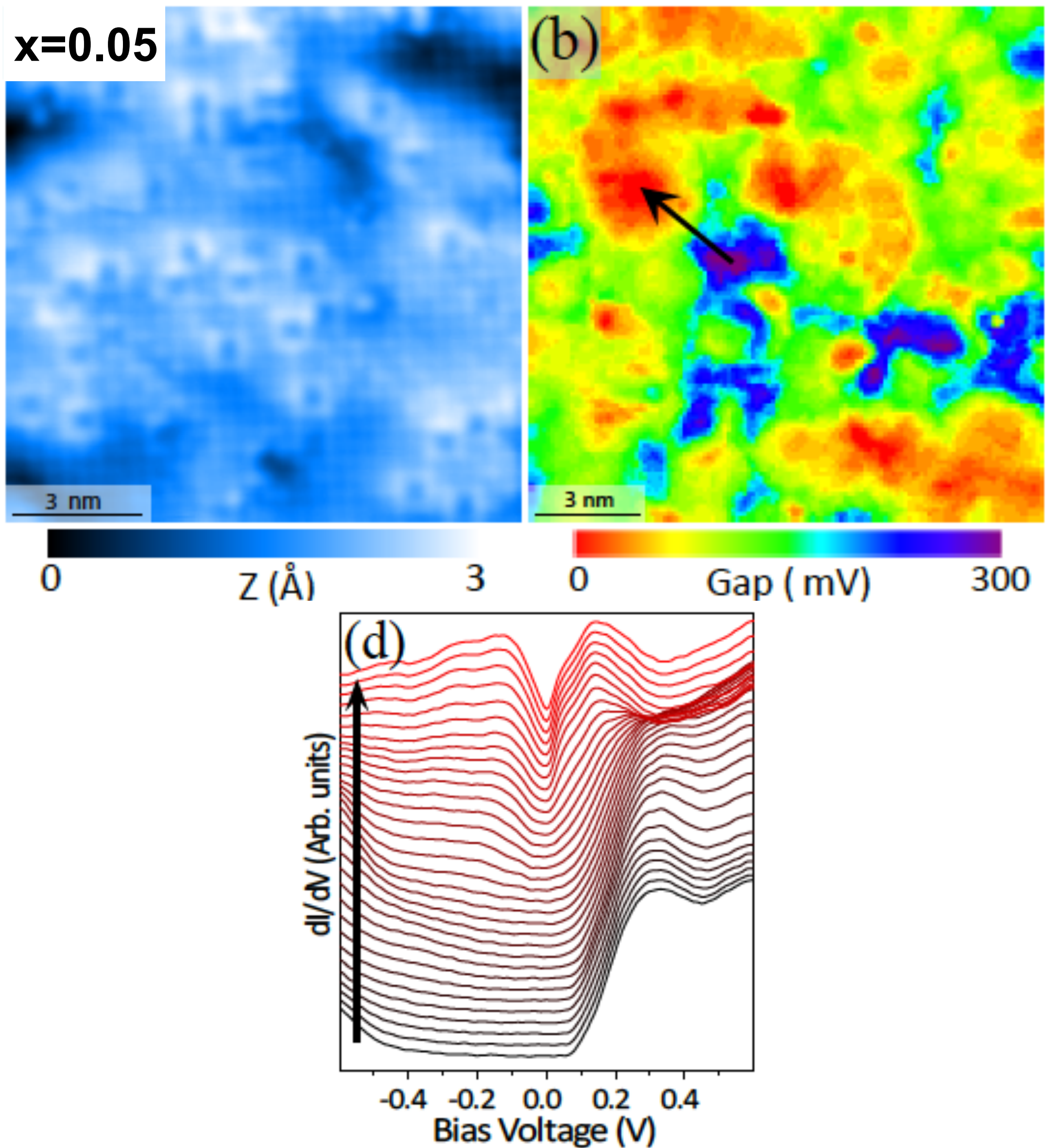
V-shaped gap opens around 50K at 0.5 ML

Surface electronically phase separated (coexisting insulating and metallic patches)

Phase diagram of bulk *n*-type Sr_2IrO_4

X. Chen et al., PRB (2016)

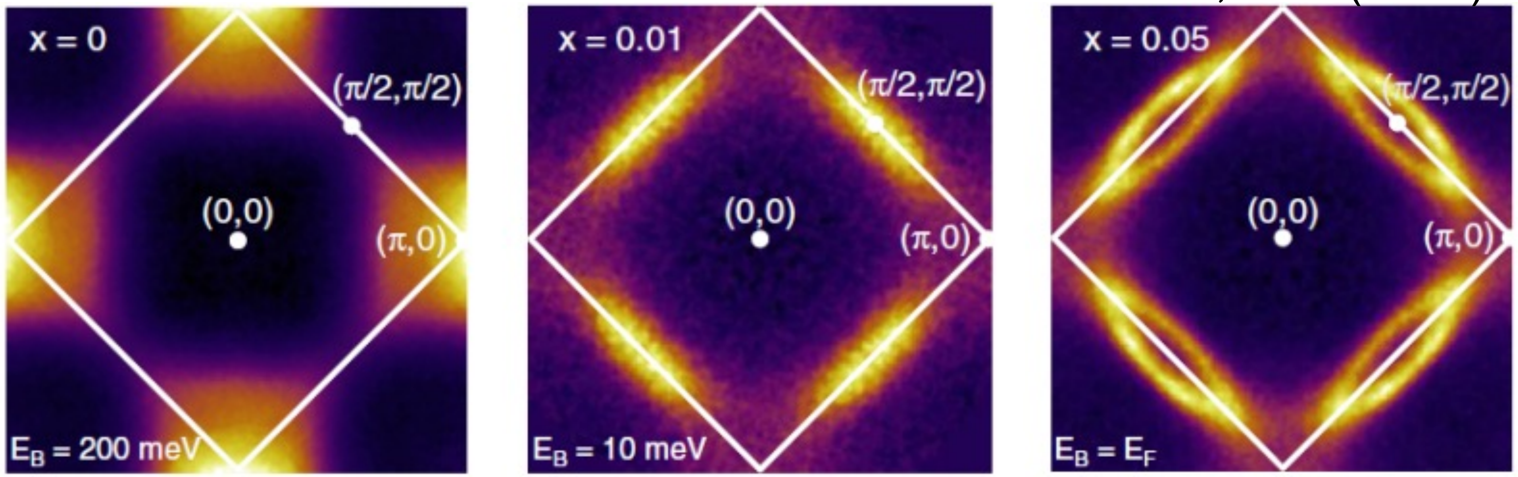




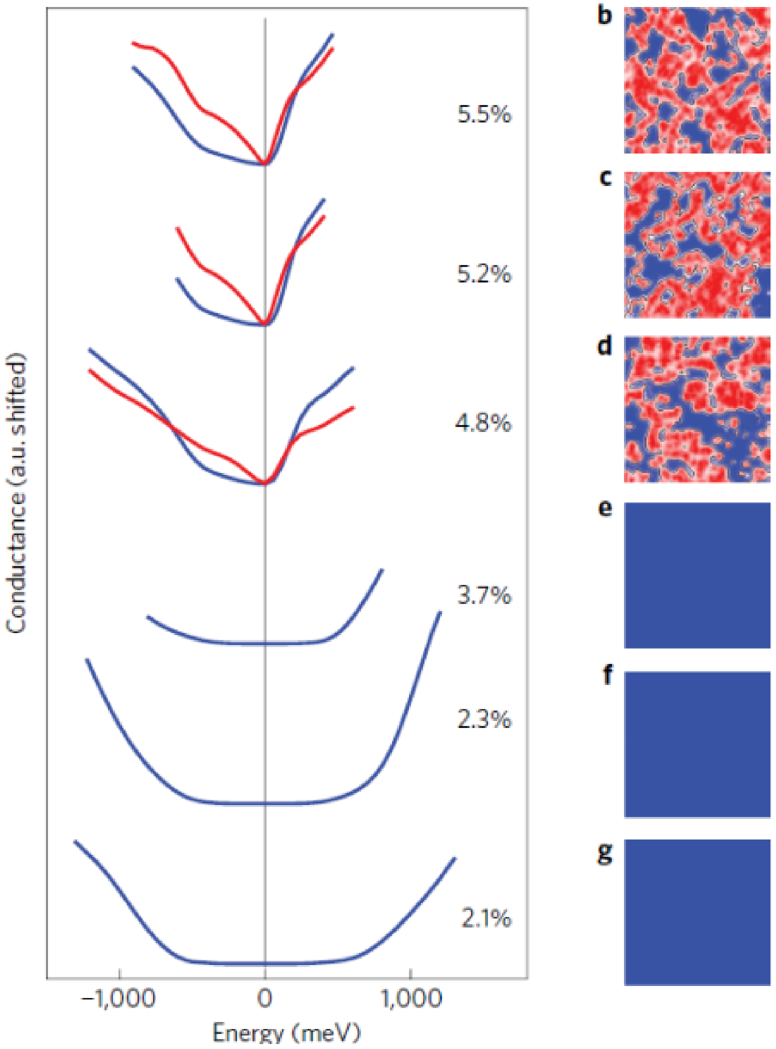
Comparable to 1 ML surface K doping

Quasiparticles in bulk $(Sr_{1-x}La_x)_2IrO_4$

de la Torre et al., PRL (2015)



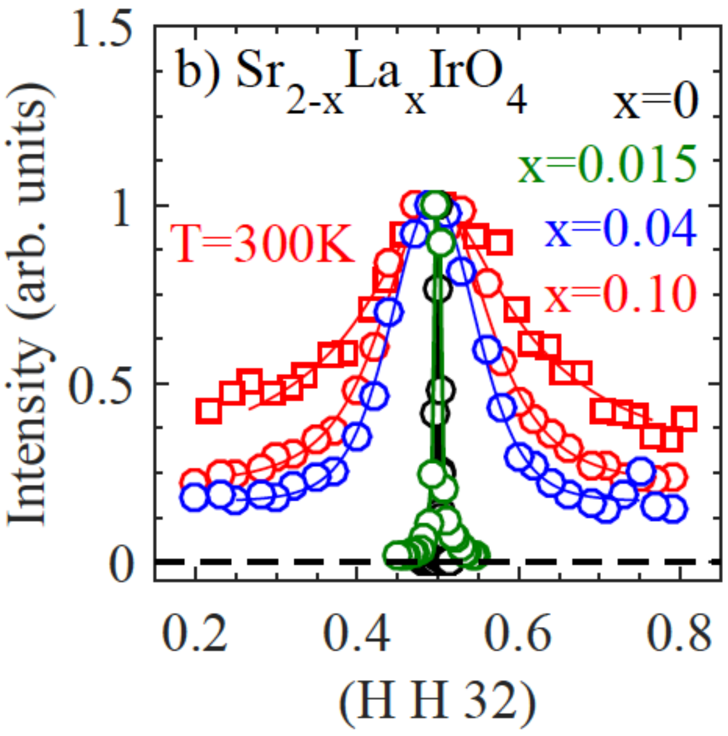
- Nodal FS observed at x=0.05
- Pseudogap-like state
- STM observes the formation of phase separated state above x=0.04 (8% electrons/Ir)
  - Metallic regions stabilize near this boundary

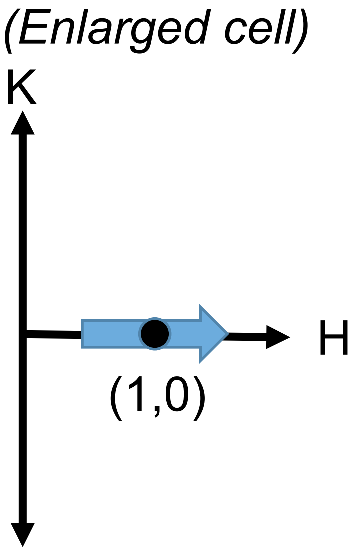


Battisti et al., Nat. Phys. (2016)

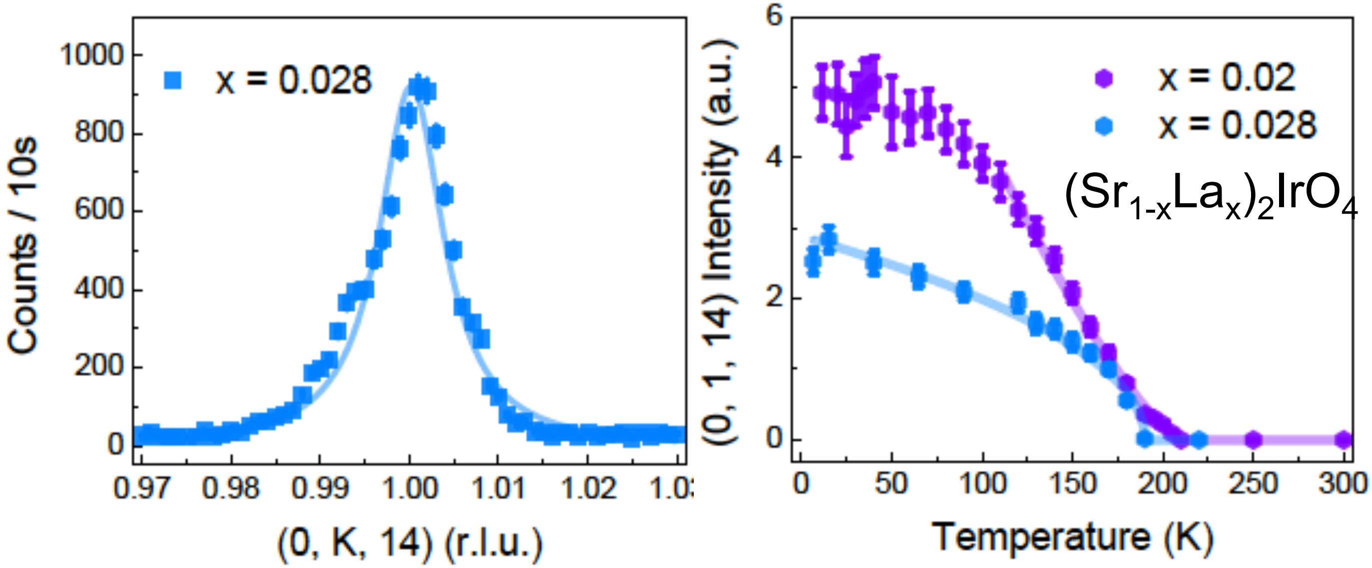
Remnant AF order in $(Sr_{1-x}La_x)_2IrO_4$

Gretarsson et al. PRL (2016)





X. Chen et al., unpublished

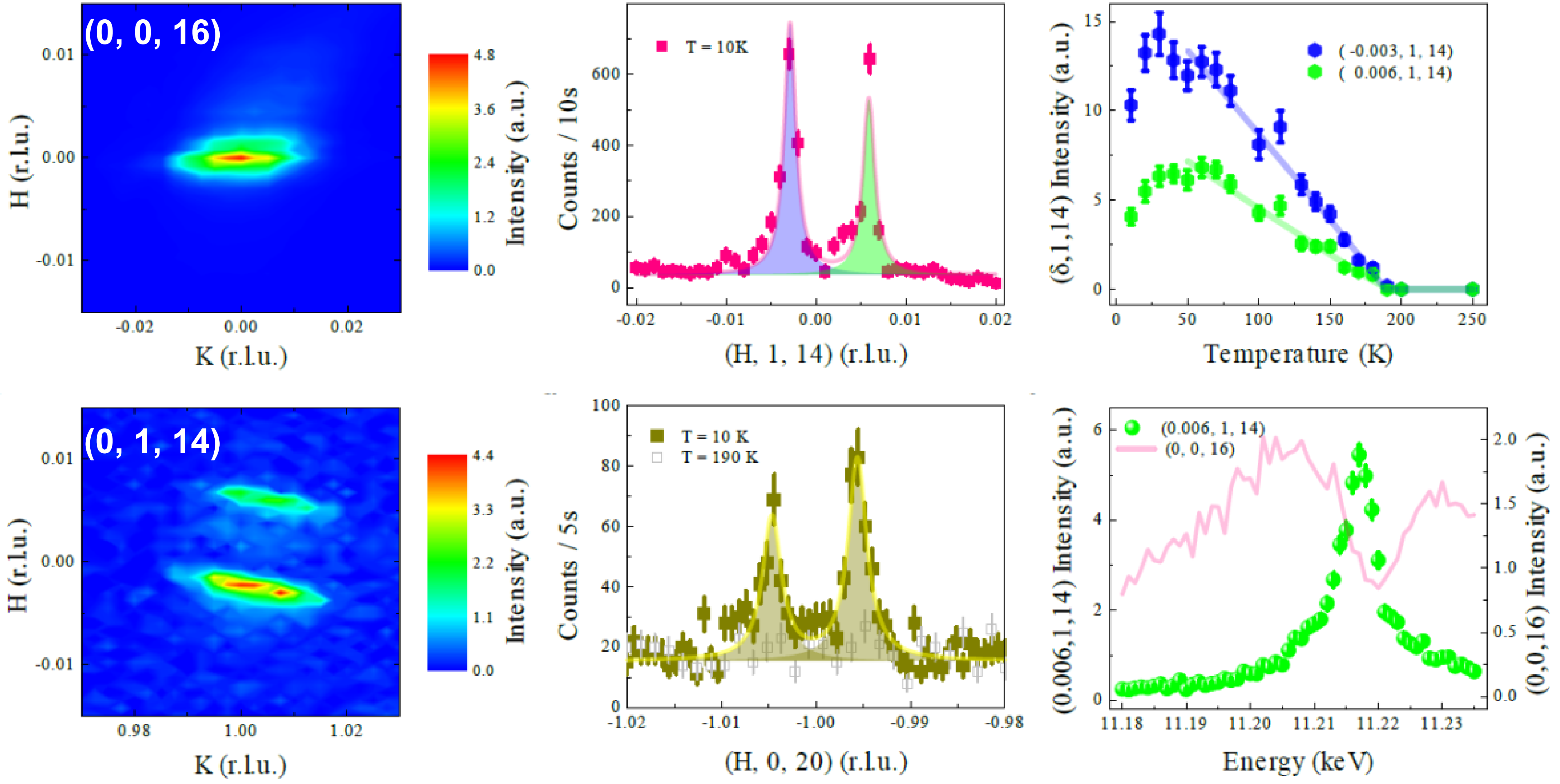


RIXS measurement shows transition to short-range order at 4% electrons/Ir and survival of short-range AF order at highest doping levels

REXS measurements show coexisting long-range AF order up to 5% electrons/Ir (coexisting diffuse and Bragg scatter)

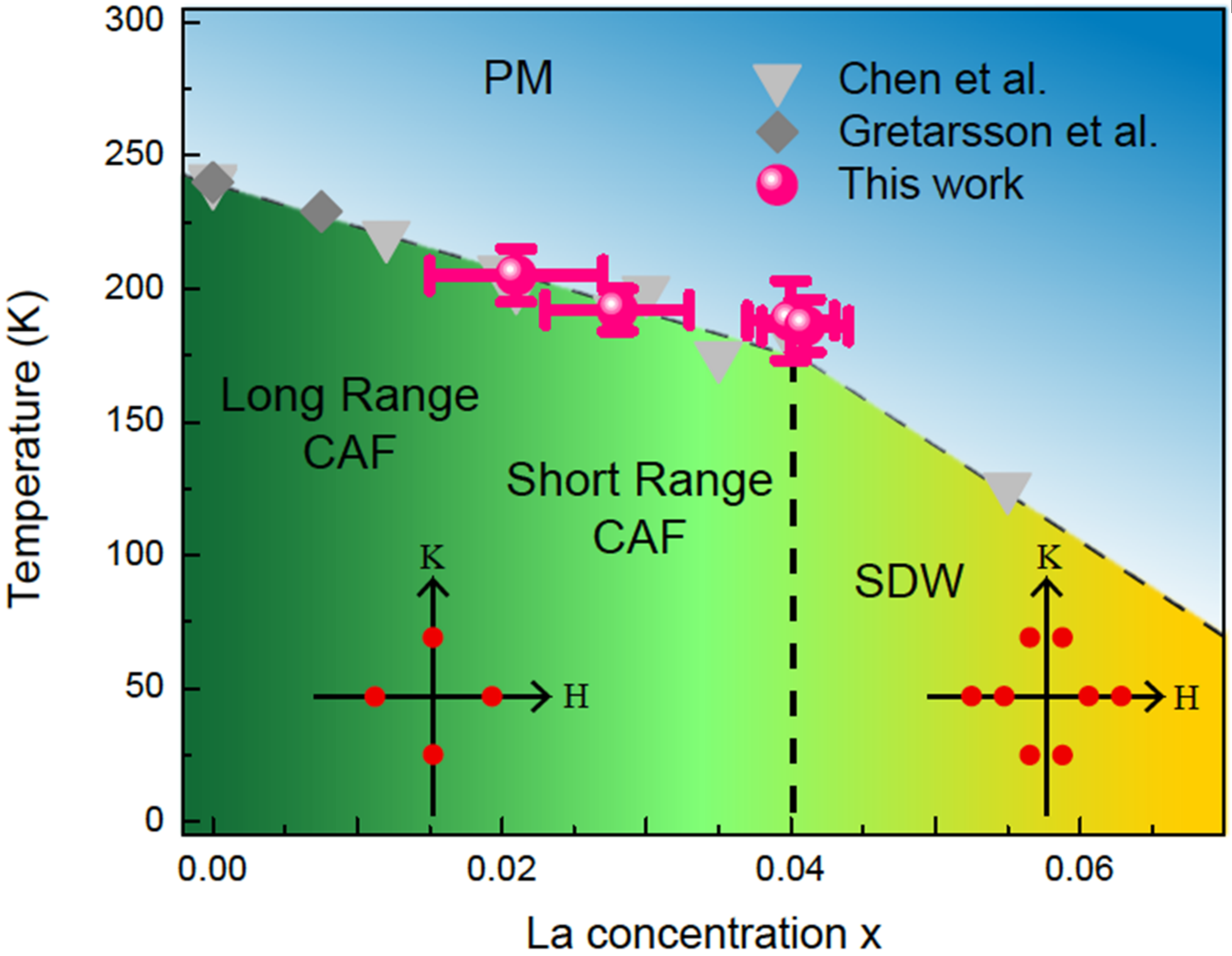
Is magnetism supported in metallic regions?

$(Sr_{1-x}La_x)_2IrO_4$, x=0.04 (8% electrons/Ir)

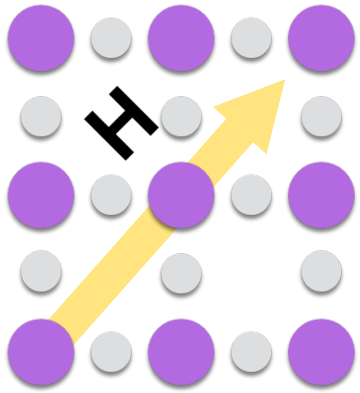


Formation of IC order via correlated metallic regions in $(Sr_{1-x}La_x)_2IrO_4$

X. Chen, unpublished

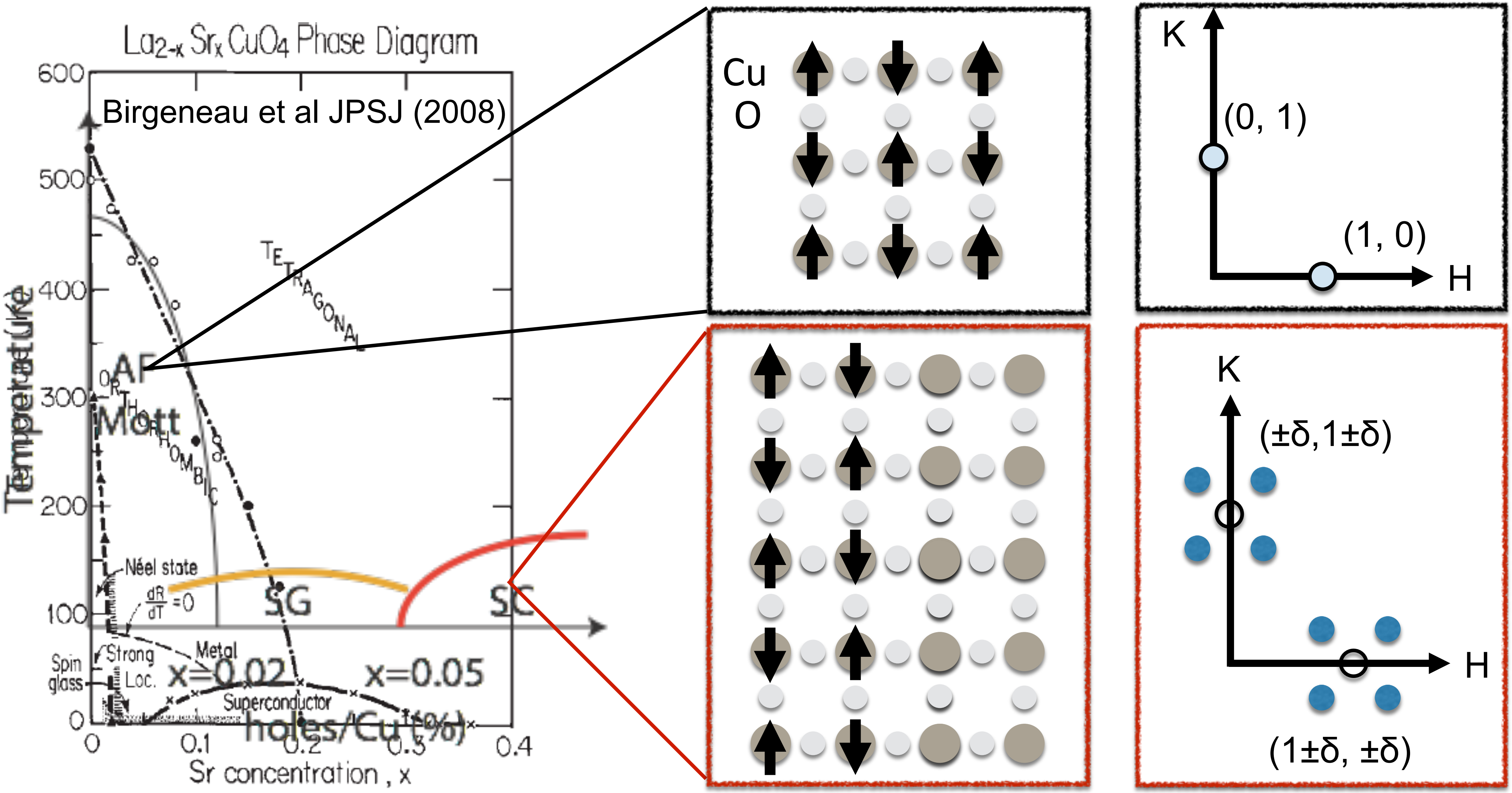


Incommensurate SDW state forms at FS threshold



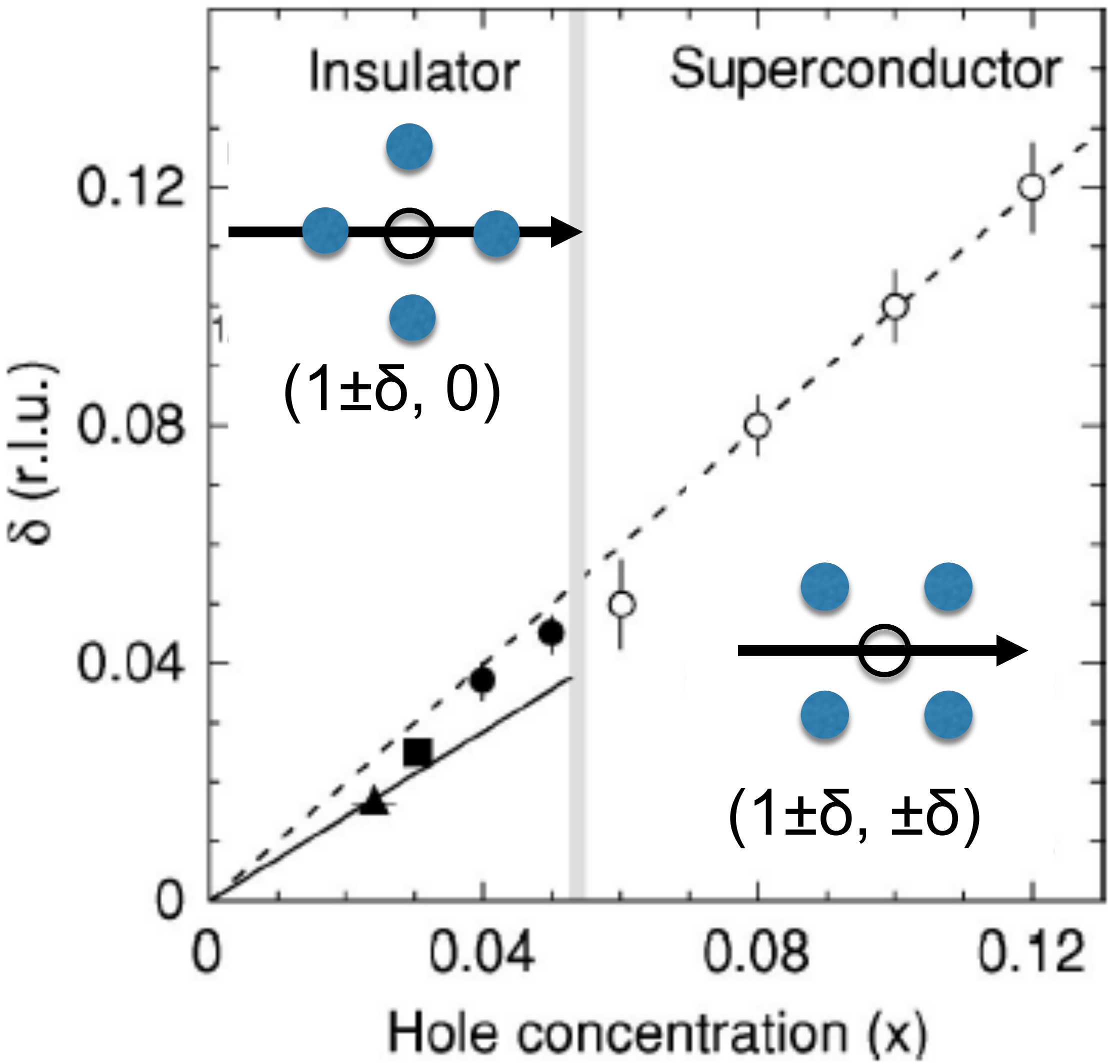
- Splitting along Ir-Ir bond diagonal
- Short-range along c (~8 unit cells)
- Necessarily lives phase separated with short-range AF correlations

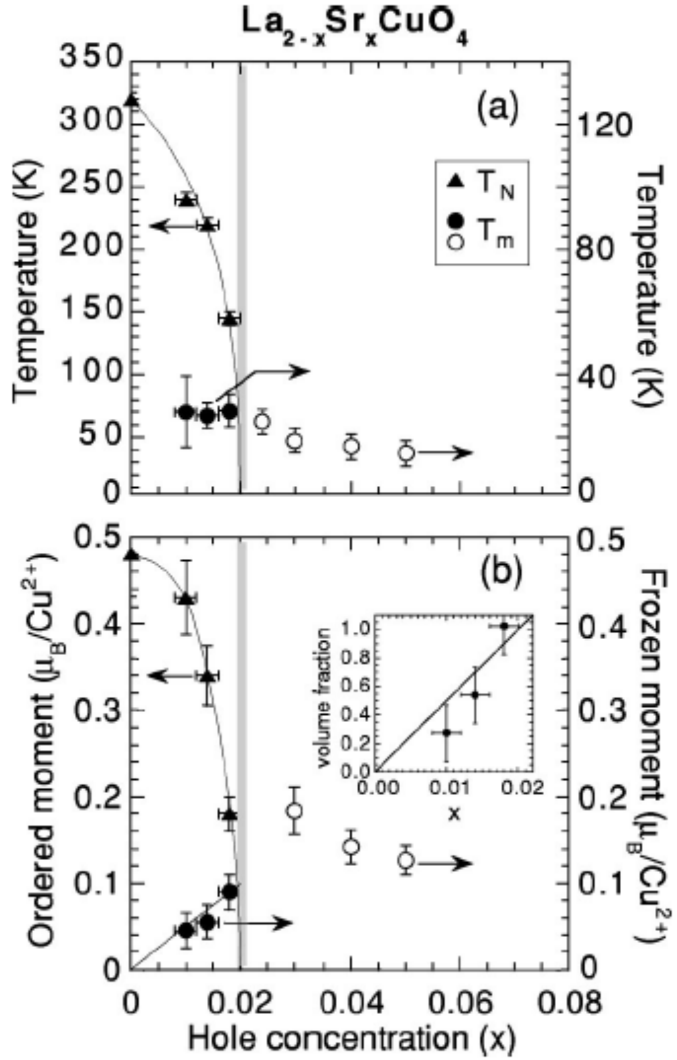
Comparison to monolayer cuprates



"Diagonal" spin density wave in cuprates

Matsuda et al., PRB (2001)



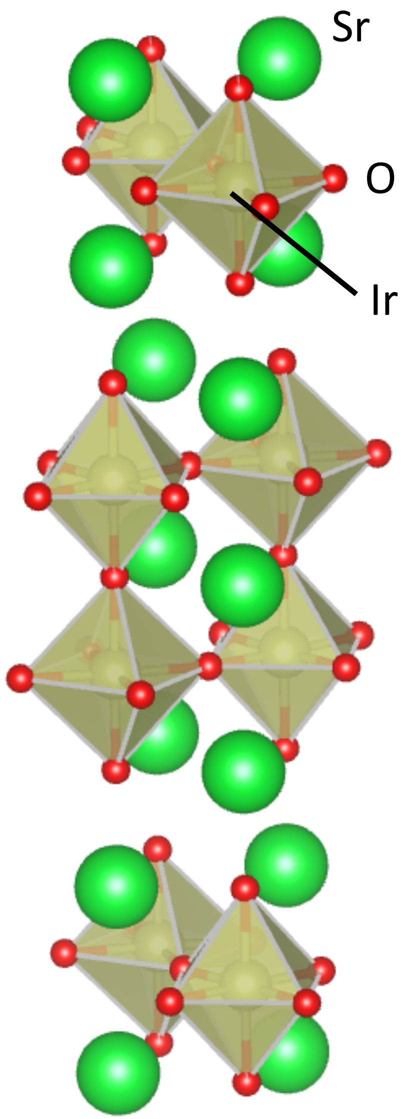


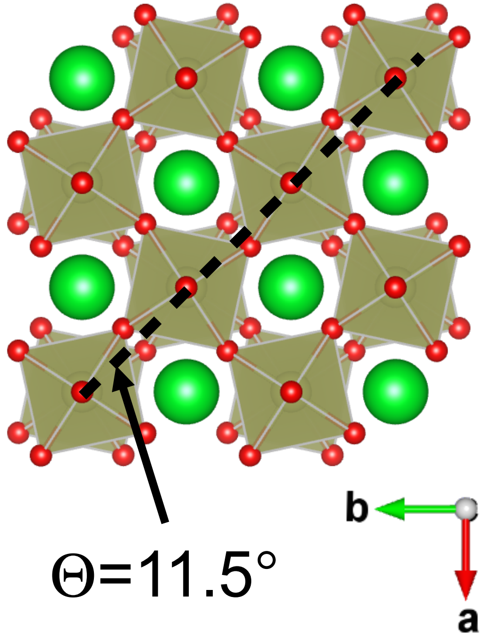
Matsuda et al., PRB (2002)

Summary for $(Sr_{1-x}La_x)_2IrO_4$

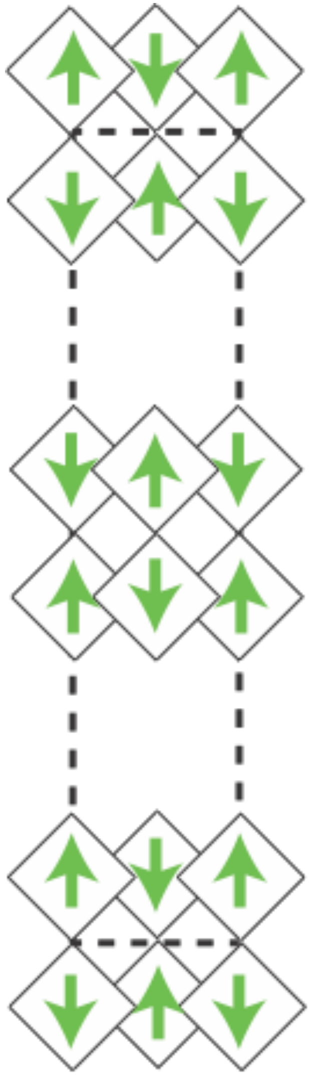
- Behaves like very lightly doped monolayer cuprate
- Electronic phase separation persists to current limits of electron-doping
  - Insulating regions coexist with metallic patches on nanoscale
  - Short-range AF order survives
- Metallic regions that form near 8% electrons/Ir are coincident with appearance of incommensurate spin state
  - Reminiscent of diagonal spin density wave state in monolayer cuprates
  - Suggestive of common magnetic instability
- Relation between incommensurate spin state and persistent AF correlations with pseudogap remains an open question

Electron-doping $Sr_3Ir_2O_7$: La substitution



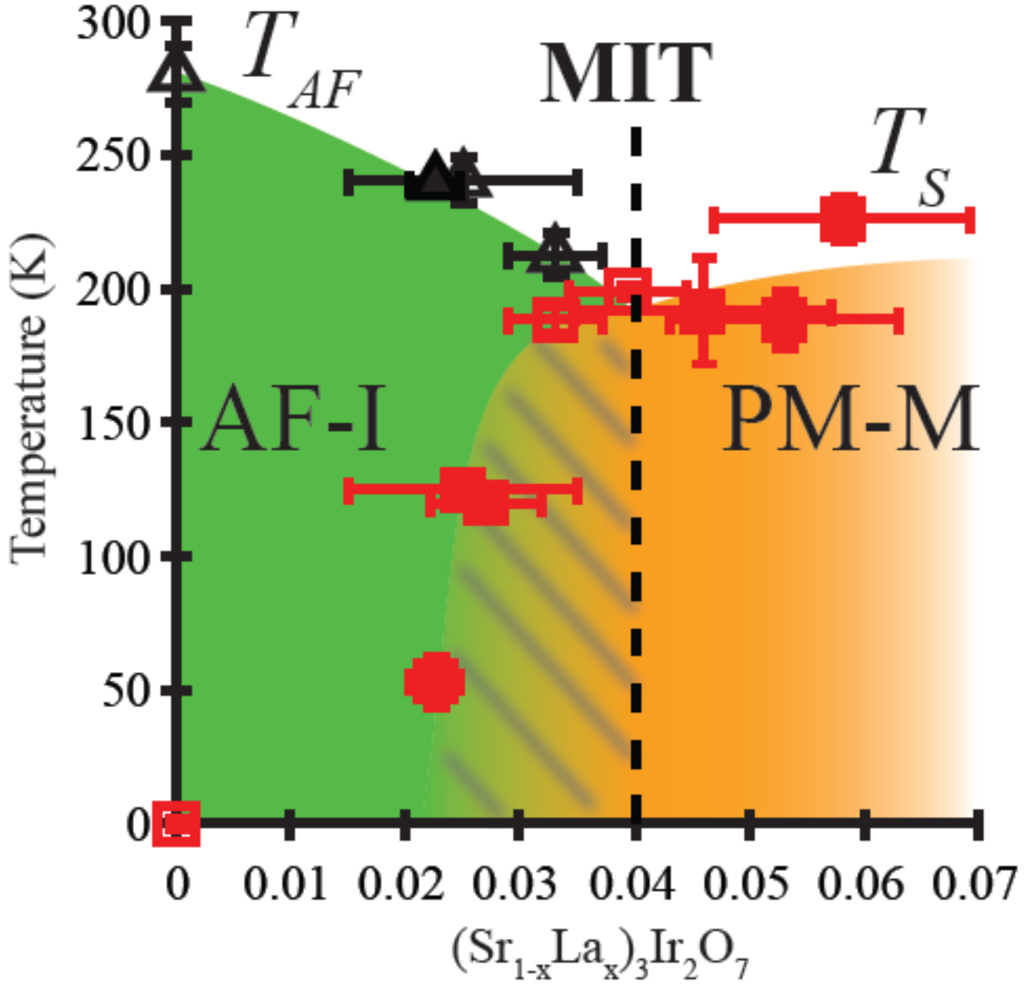


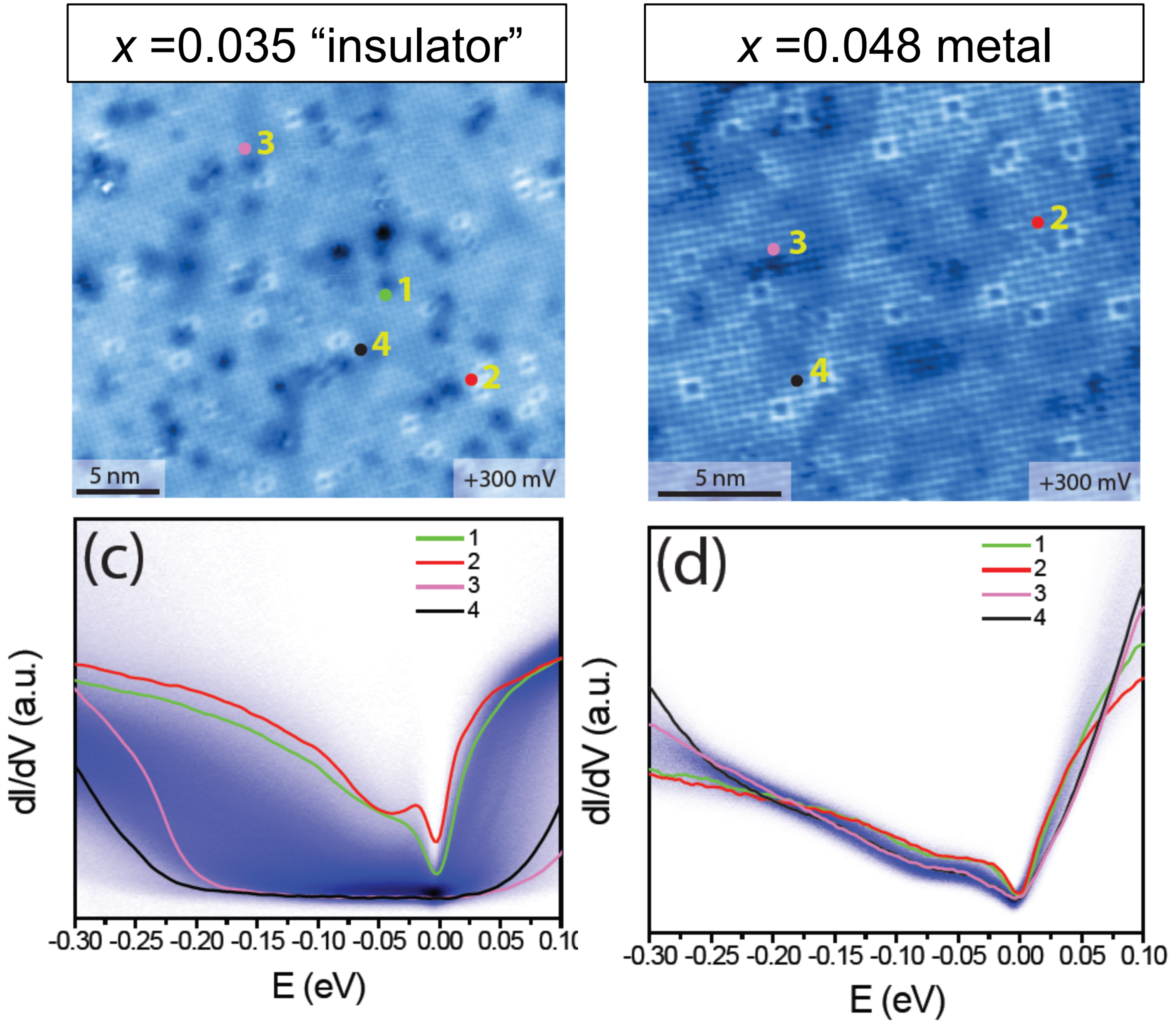
C2/c most likely space group:
Hogan et al., PRB (2016)



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

T. Hogan et al. PRL (2015)

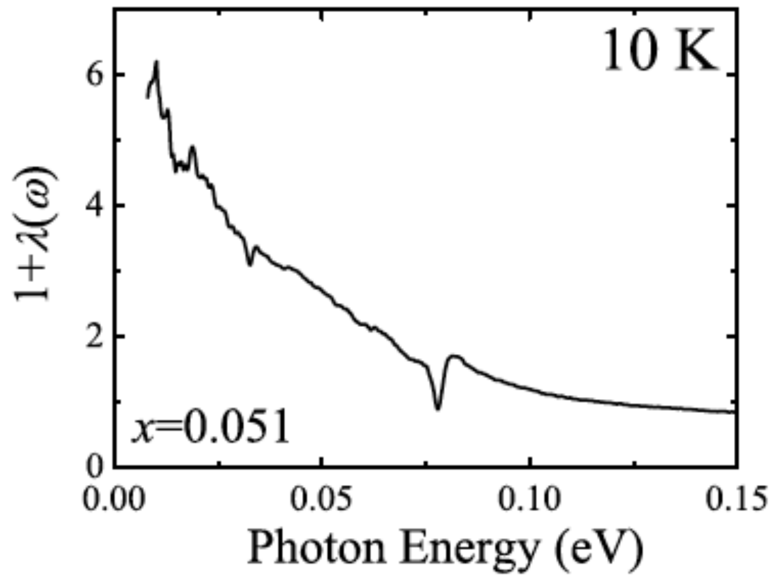




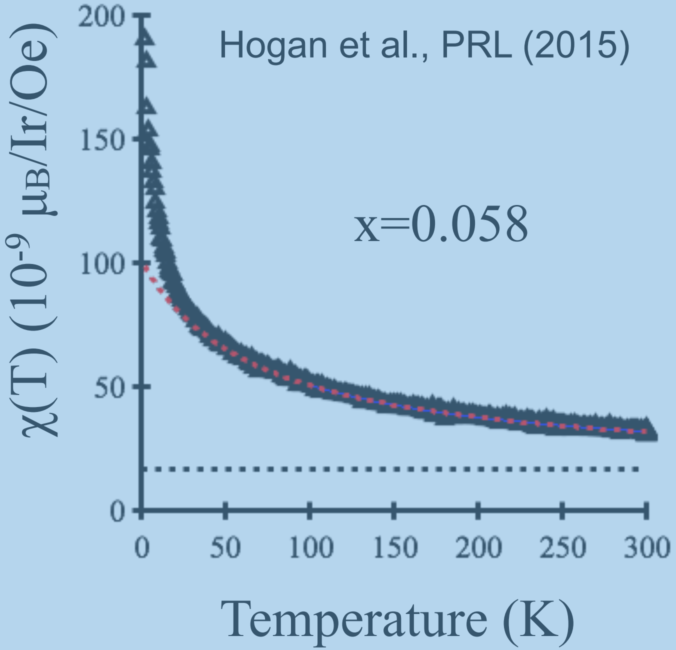
Correlation effects in metallic $(Sr_{1-x}La_x)_3Ir_2O_7$

G. Ahn et al. Sci. Rep. (2016)\

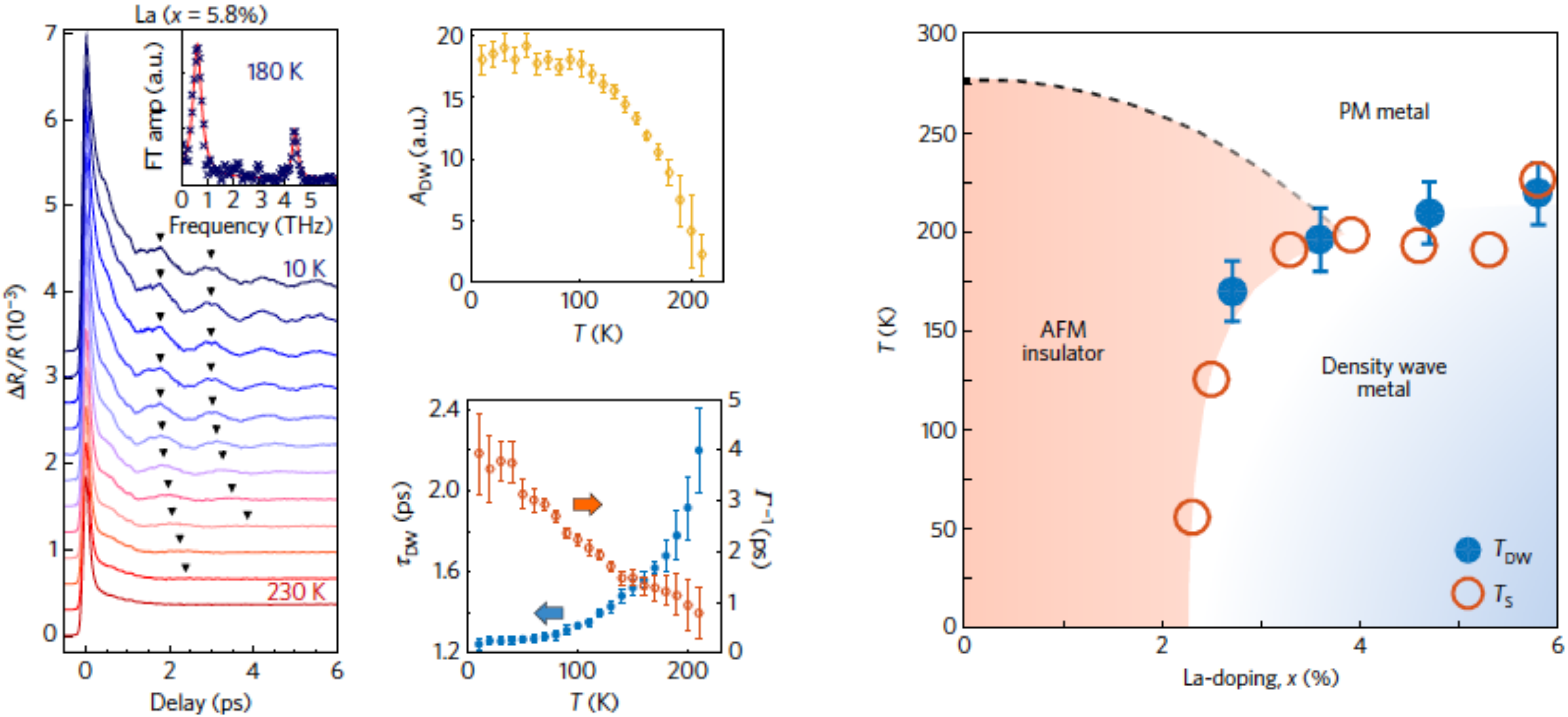
- Mass enhancement in frequency dependent conductivity
  - Extended Drude model: $$1 + \lambda(\omega) \equiv \frac{m^*(\omega)}{m_b}$$
  - Mass enhancement of ~6



- Curie-Weiss local moment behavior
  - m_{eff}=0.5 μ_B/Ir
  - Θ_{CW} = -70 K
  - Absent in parent system…

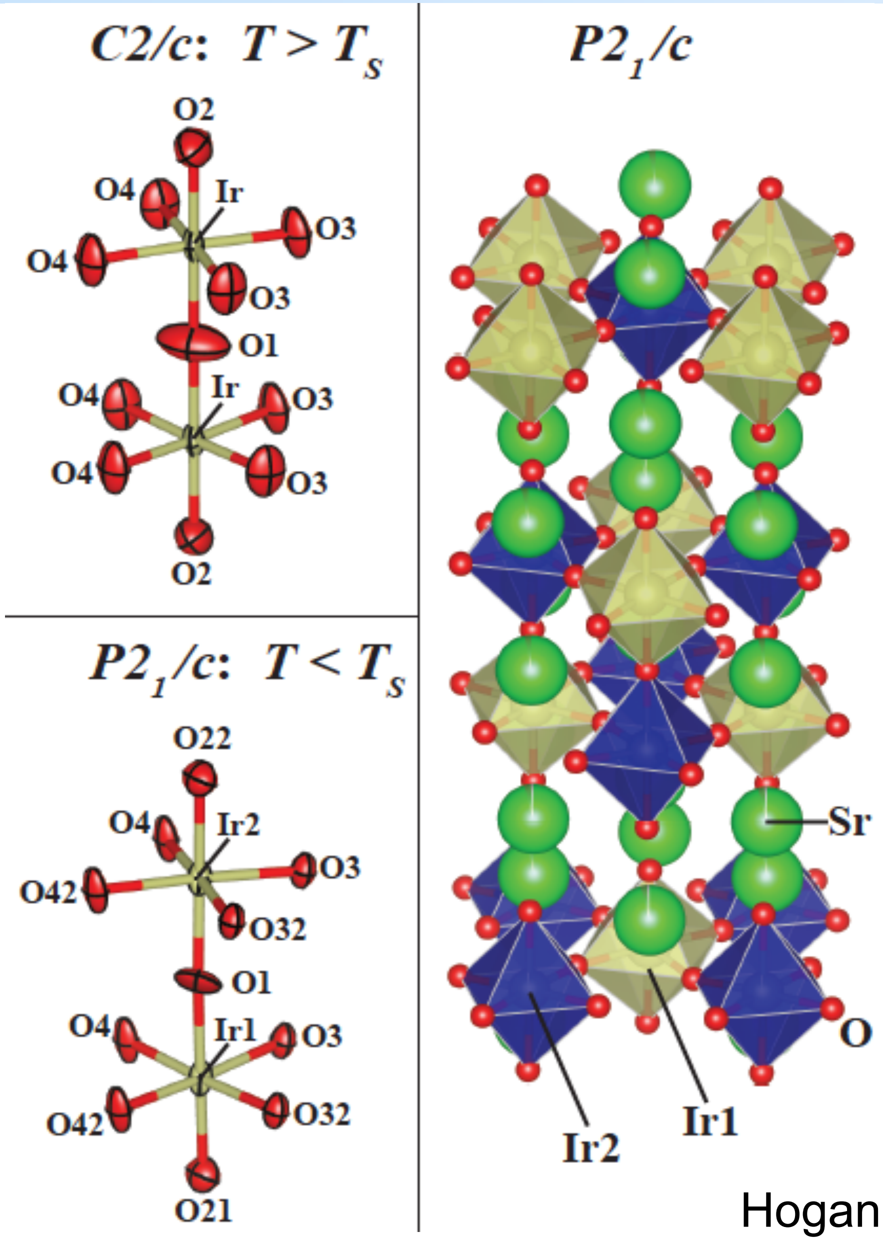


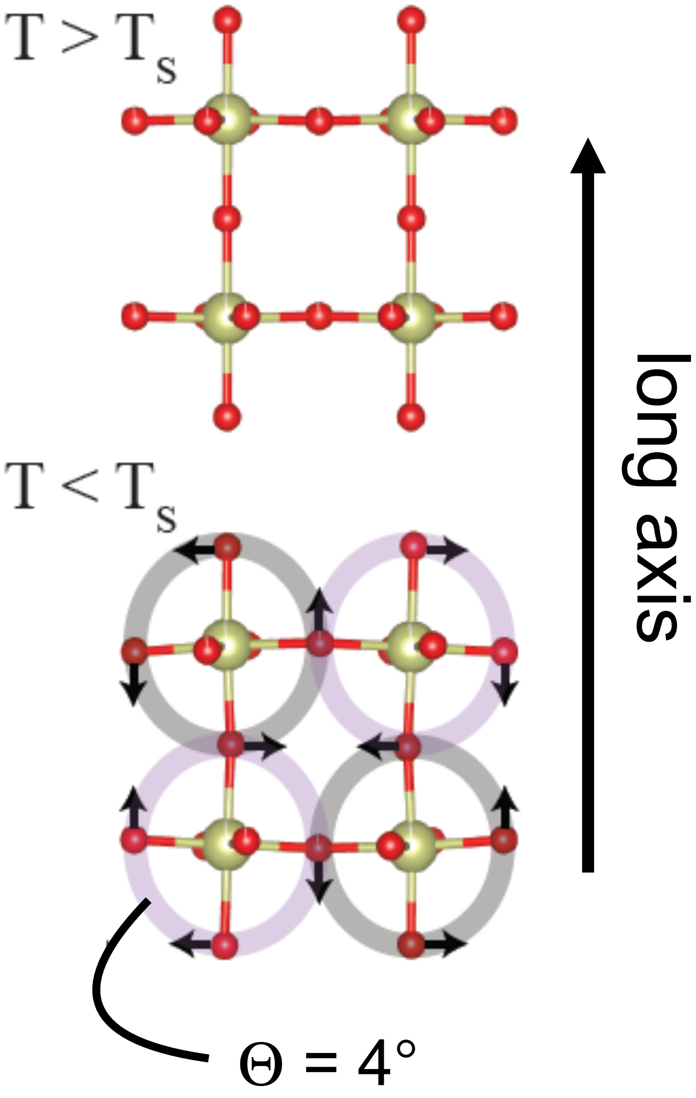
Charge density wave-like instability



H. Chu et al., Nature Materials

Structural distortion, T_S, coincident with T_{DW}





Hogan et al., PRB (2017)

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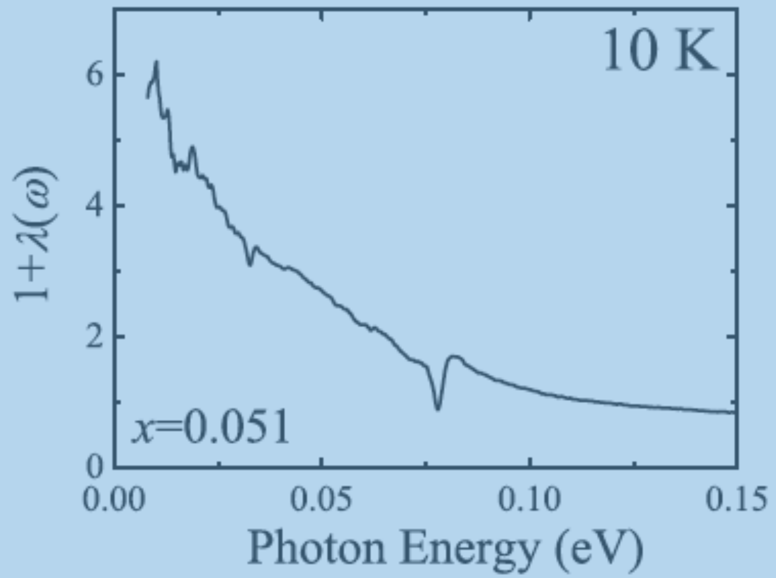

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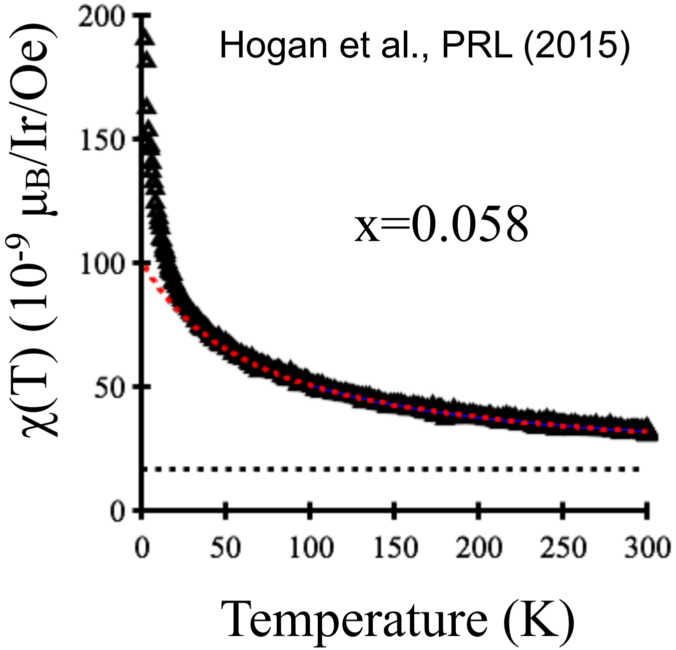
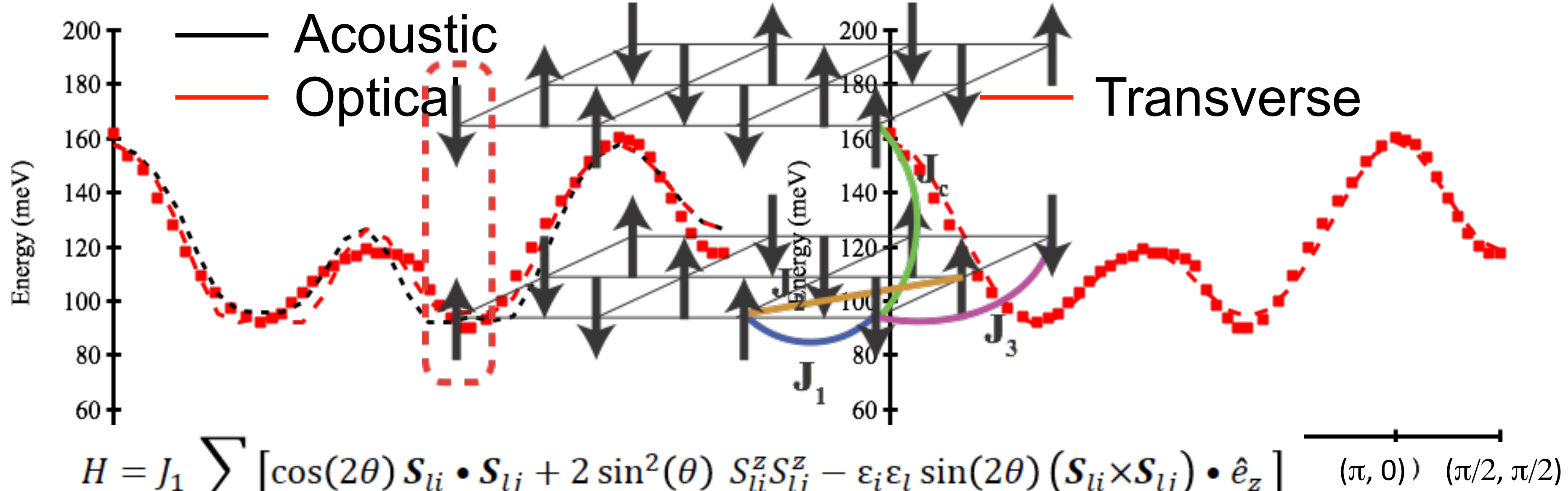
  - Absent in parent system…



Spin dynamics in parent $Sr_3Ir_2O_7$



$$H = J_1 \sum_{\langle i,j\rangle,l} \left[\cos(2\theta)\, \boldsymbol{S}_{li} \cdot \boldsymbol{S}_{lj} + 2\sin^2(\theta)\, S^z_{li} S^z_{lj} - \varepsilon_i \varepsilon_l \sin(2\theta) \left(\boldsymbol{S}_{li} \times \boldsymbol{S}_{lj} \right) \cdot \hat{e}_z \right]$$

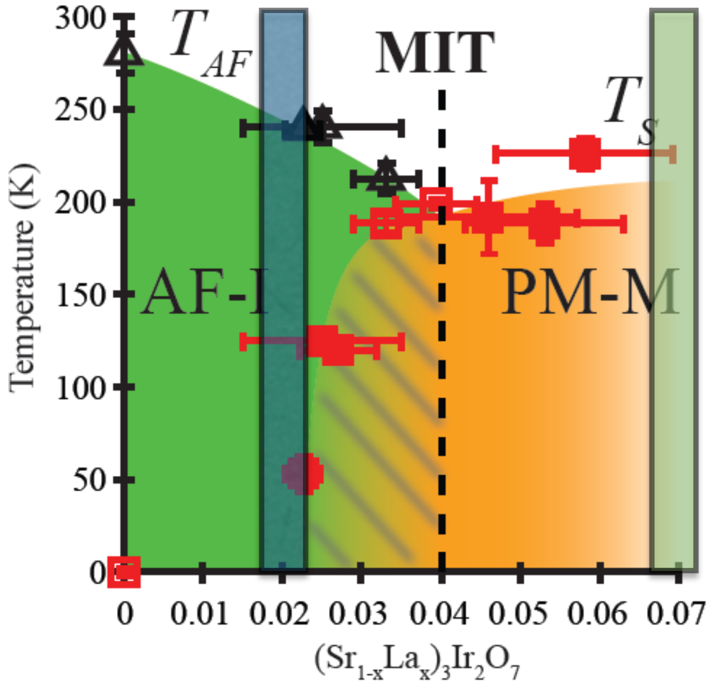
$$+ J_c \sum_i \boldsymbol{S}_{1i} \cdot \boldsymbol{S}_{2j} + J_2 \sum_{\langle\langle i,j\rangle\rangle,l} \boldsymbol{S}_{li} \cdot \boldsymbol{S}_{lj} + J_3 \sum_{\langle\langle\langle i,j\rangle\rangle\rangle,l} \boldsymbol{S}_{li} \cdot \boldsymbol{S}_{lj}$$

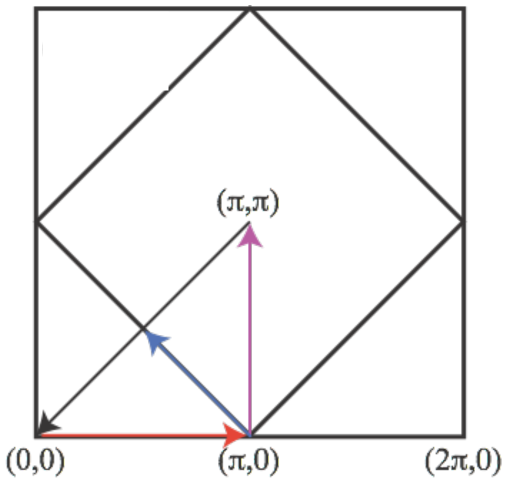
Kim et al., PRL (2012)

Sala et al., PRB (2015)

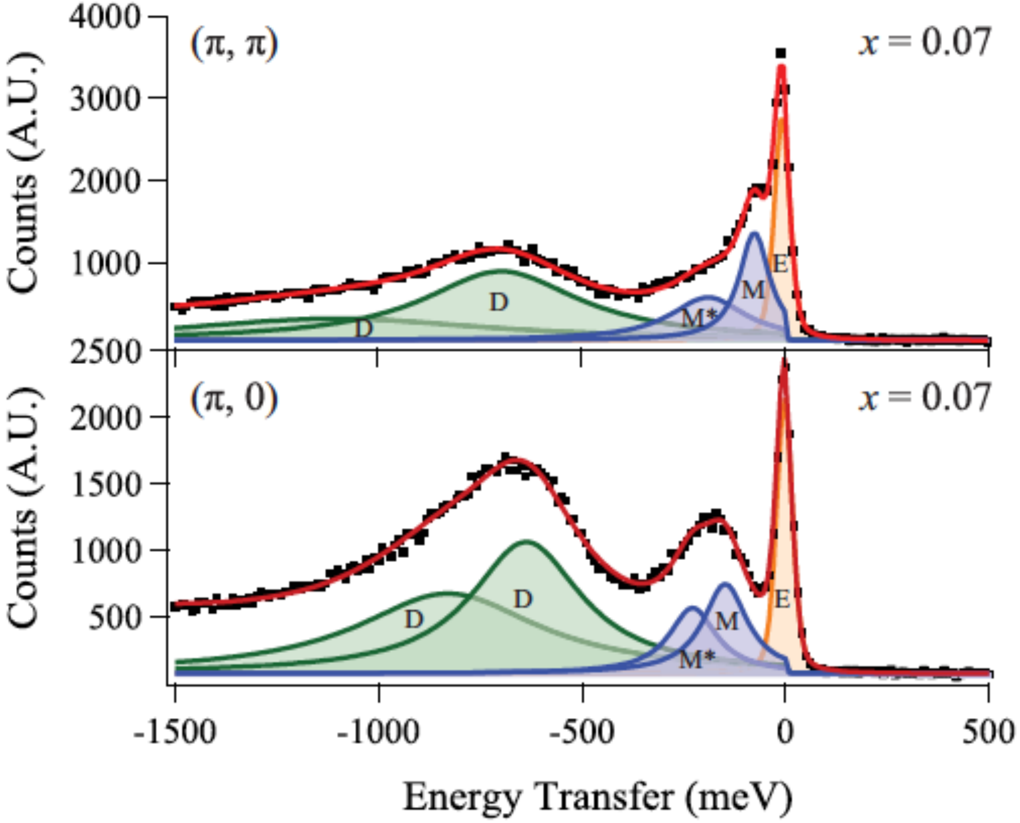
How do excitations evolve as static AF order collapses?

RIXS measurements of $(Sr_{1-x}La_x)_3Ir_2O_7$



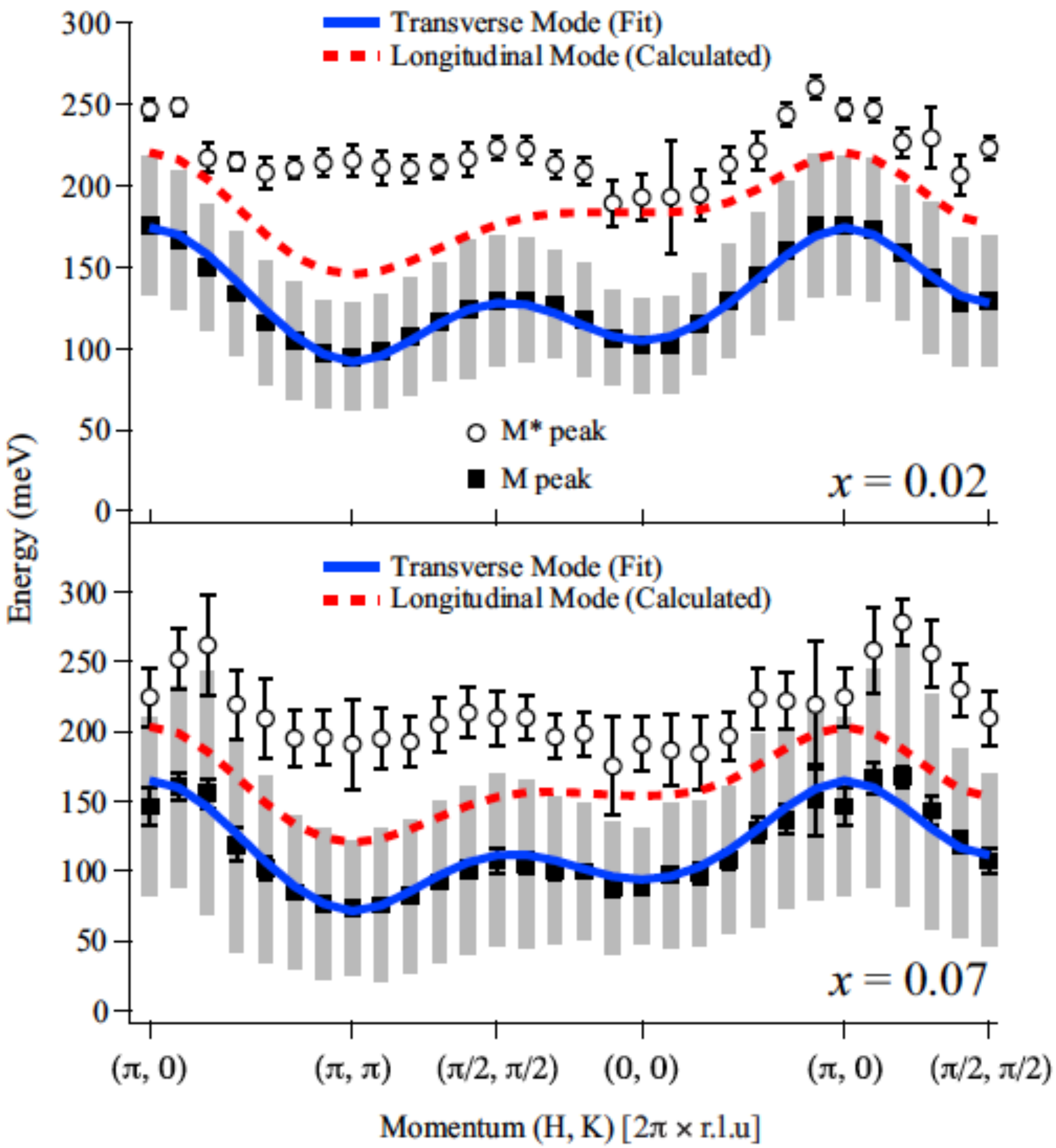


Hogan et al., PRB (2016)



Gapped magnons survive long after collapse of long-range AF order

Magnon dispersion in $(Sr_{1-x}La_x)_3Ir_2O_7$



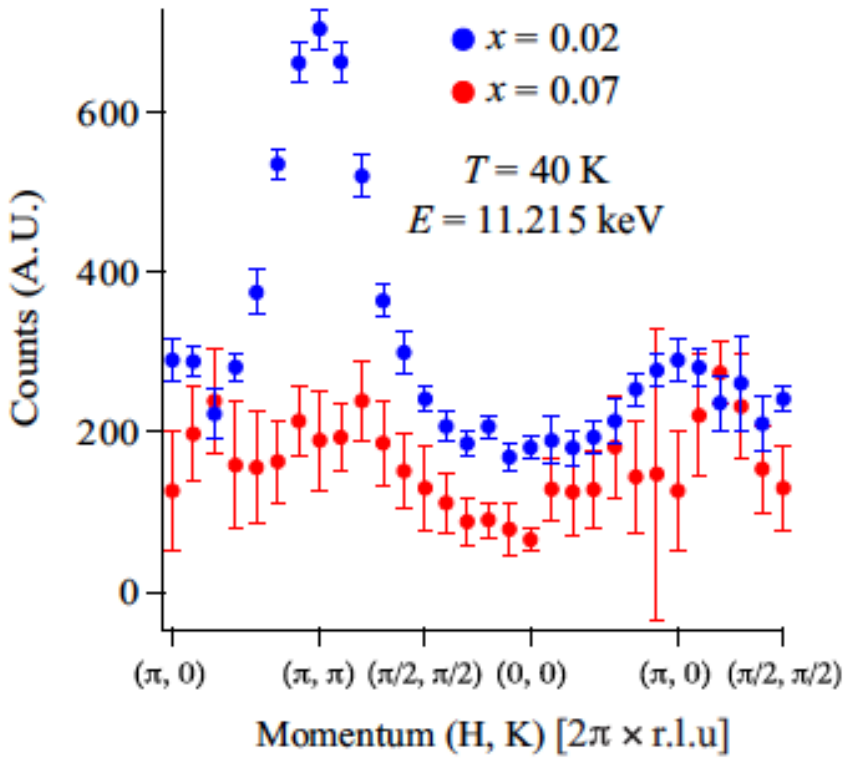
	$x = 0$ *Moretti Sala, et al.*	$x = 0.022$	$x = 0.071$	Units
J_1	26	37.7 ± 0.4	29.1 ± 0.7	meV
J_2	-15	-14.0 ± 0.3	-17.0 ± 0.6	meV
J_3	6	4.8 ± 0.3	5.2 ± 0.6	meV
J_c	90	87.6 ± 1.1	80.1 ± 2.3	meV
θ	37	41.2 ± 0.7	37.2 ± 1.7	degrees

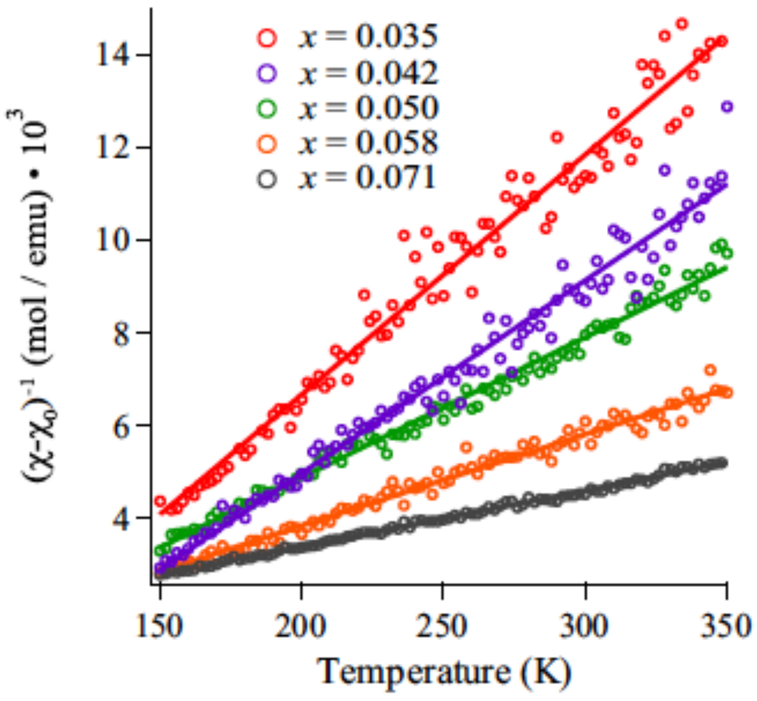
Magnon spectrum only weakly affected upon entering metallic state and loss of static order

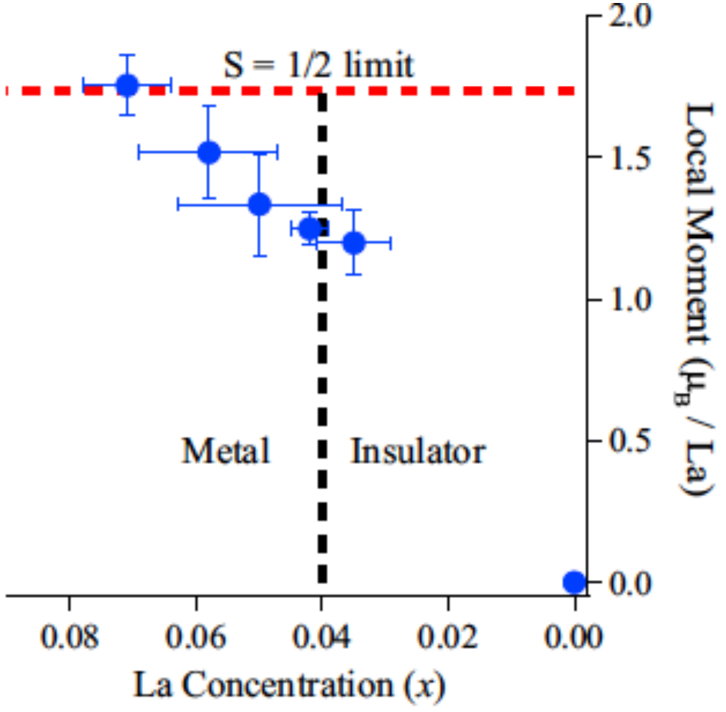
Carrier-induced damping of magnons has largest effect

Hogan et al., PRB (2016)

Hints of disordered dimer spin state





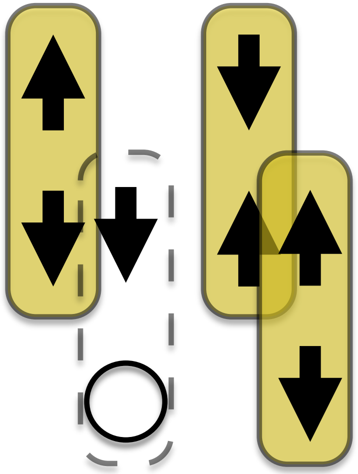


ZC spectral weight suppressed across MIT (vanishing AF order)

Remnant weight has weak momentum dependence

Local moments induced by La-substitution

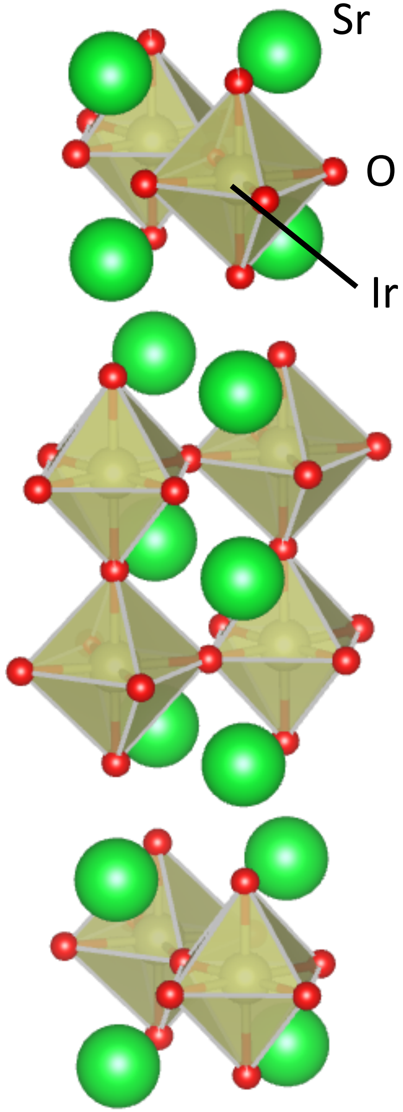
Induced moment approaches *J*=1/2 per electron doped

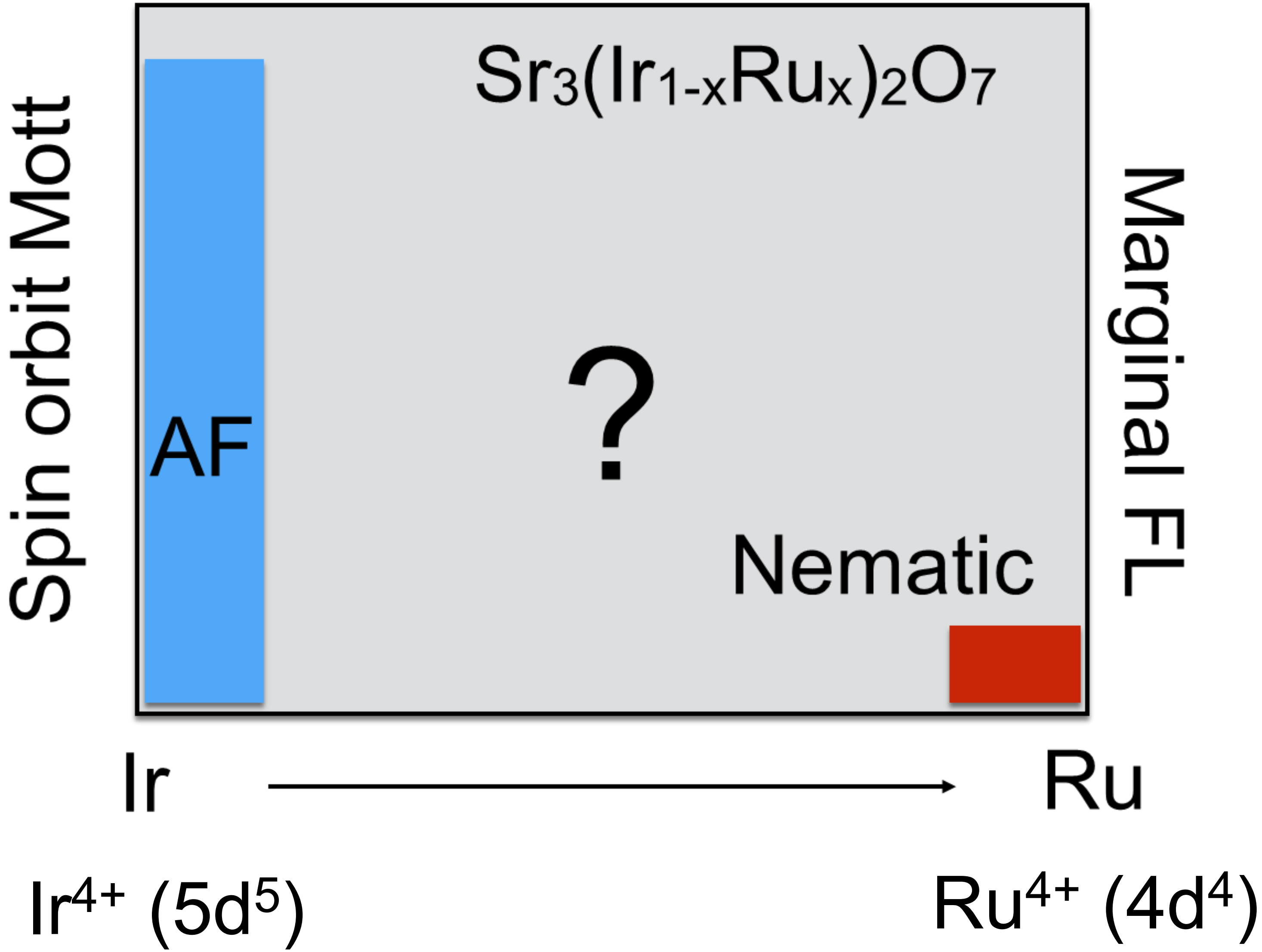


Summary for $(Sr_{1-x}La_x)_3Ir_2O_7$

- Electron doping $Sr_3Ir_2O_7$ causes first order collapse of spin-orbit Mott state
  - Competing charge density wave (-like) instability appears
  - Lattice distortion accompanies this density wave order
- Magnetism's evolution into nearby correlated metallic state is unusual
  - Static AF order vanishes at MIT
  - Robust spin dynamics persist up to the limit of doping
  - Gap value, bandwidth, and modeled exchange parameters only weakly affected
  - Evolution of spectral weight and magnon dispersions best modeled via bond operator derived model
- Local moments appear with doped electrons and scale as S=1/2 impurities
  - Suggest potential disordered dimer state…

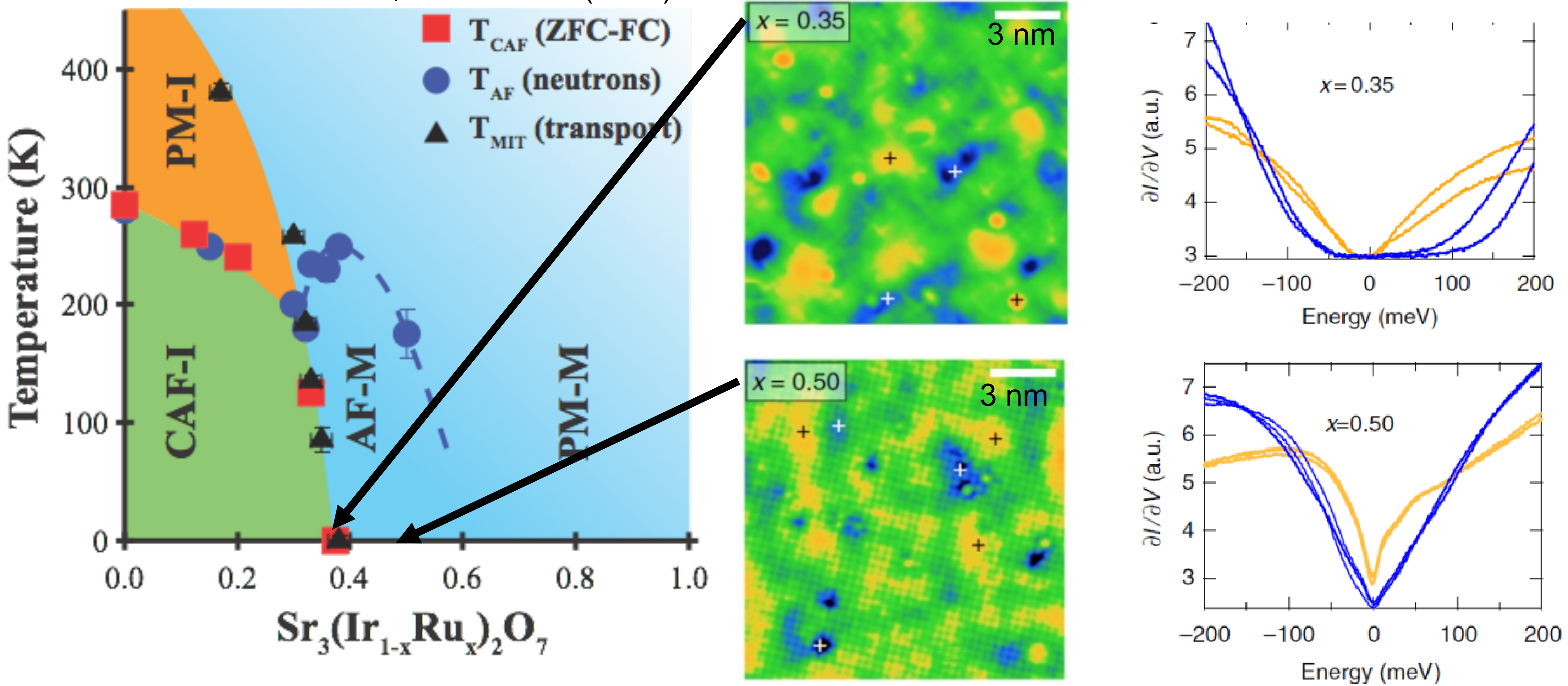
$Sr_3Ir_2O_7$: B-site substitution





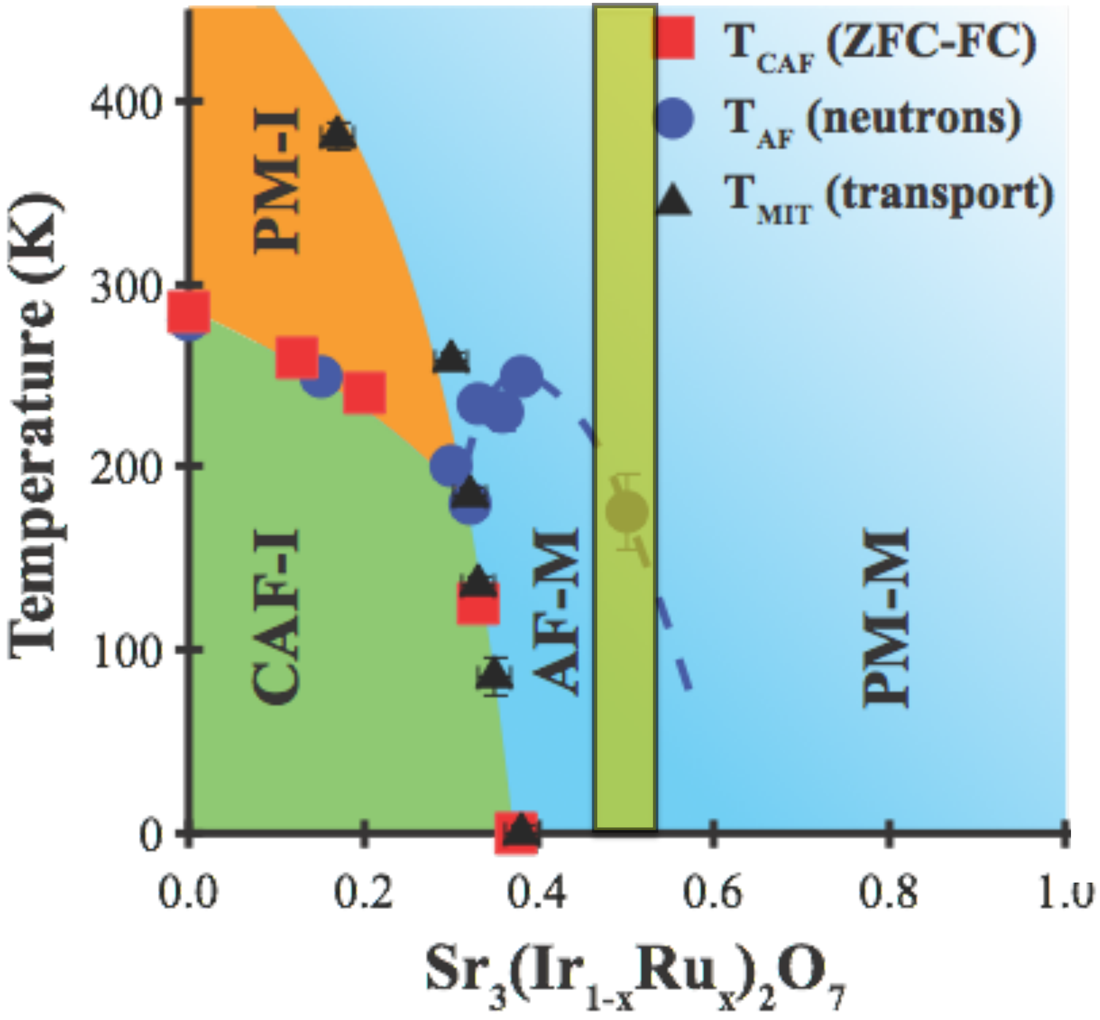
MIT in $Sr_3(Ir_{1-x}Ru_x)_2O_7$

Dhital et al., Nat. Comm. (2013)

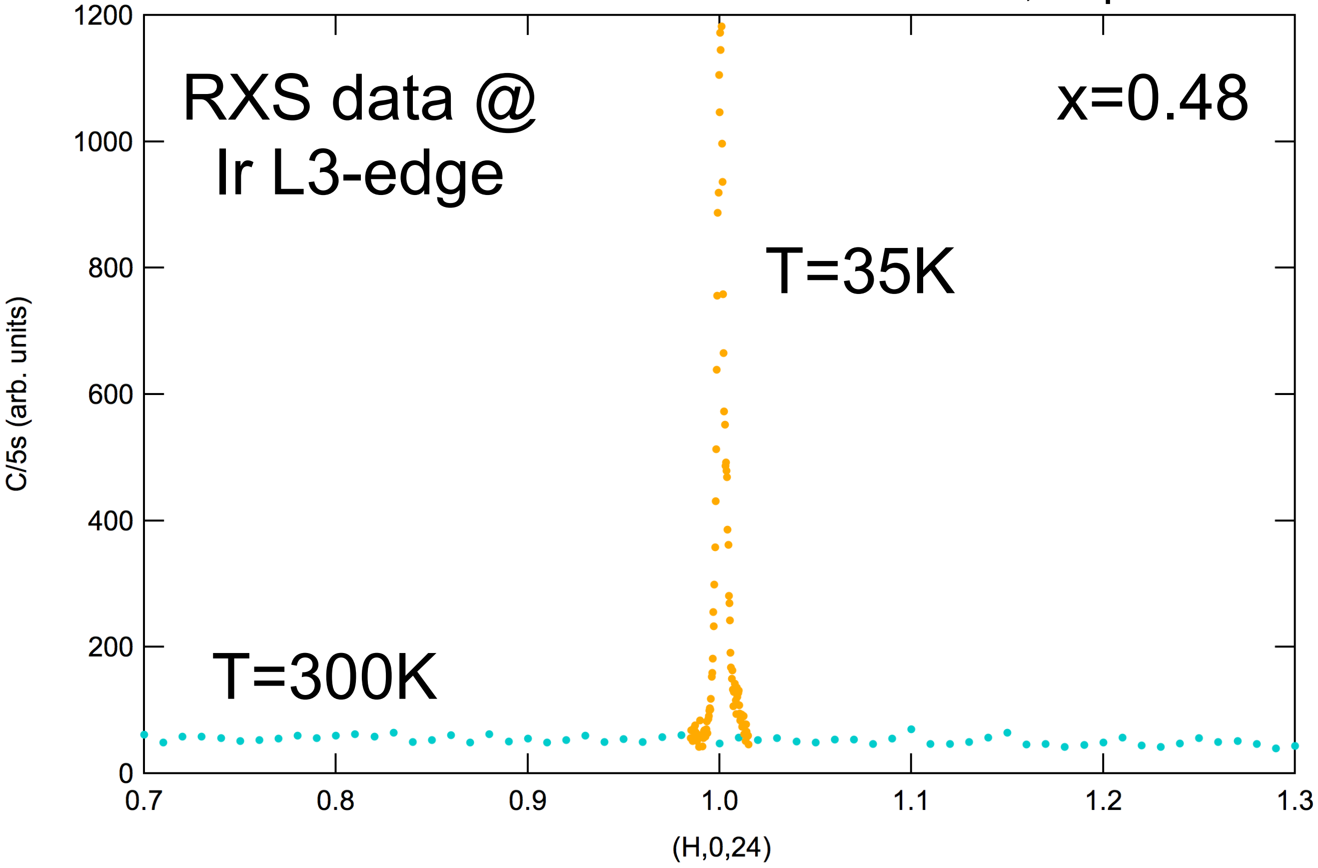


Intermediate SDW state in $Sr_3(Ir_{1-x}Ru_x)_2O_7$

Dhital et al., Nat. Comm. (2013)



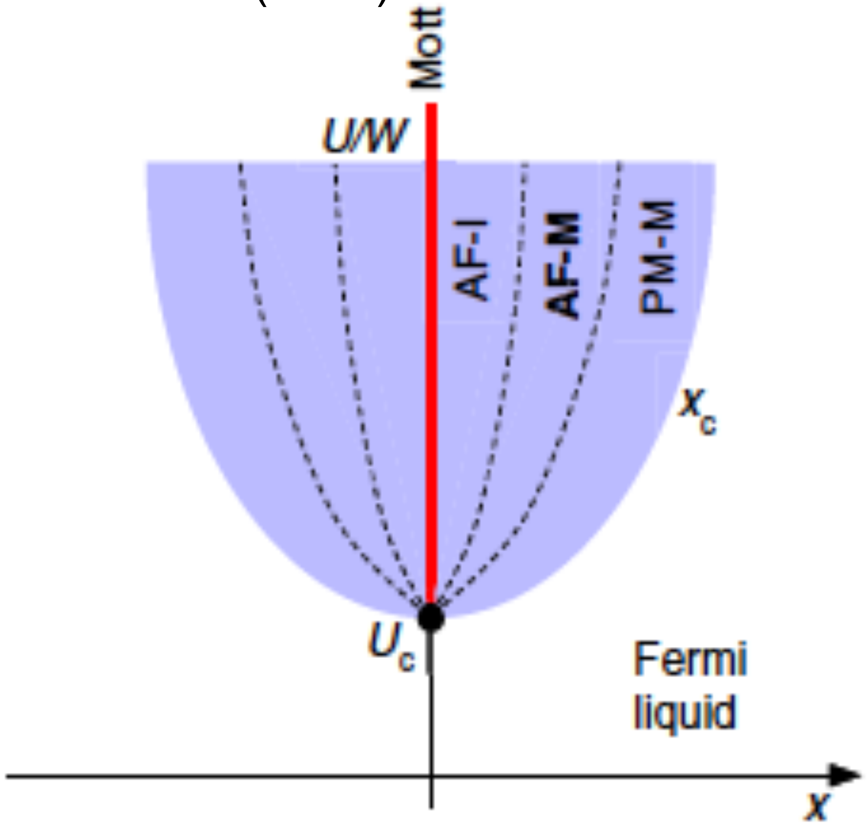
J. Schmehr, unpublished



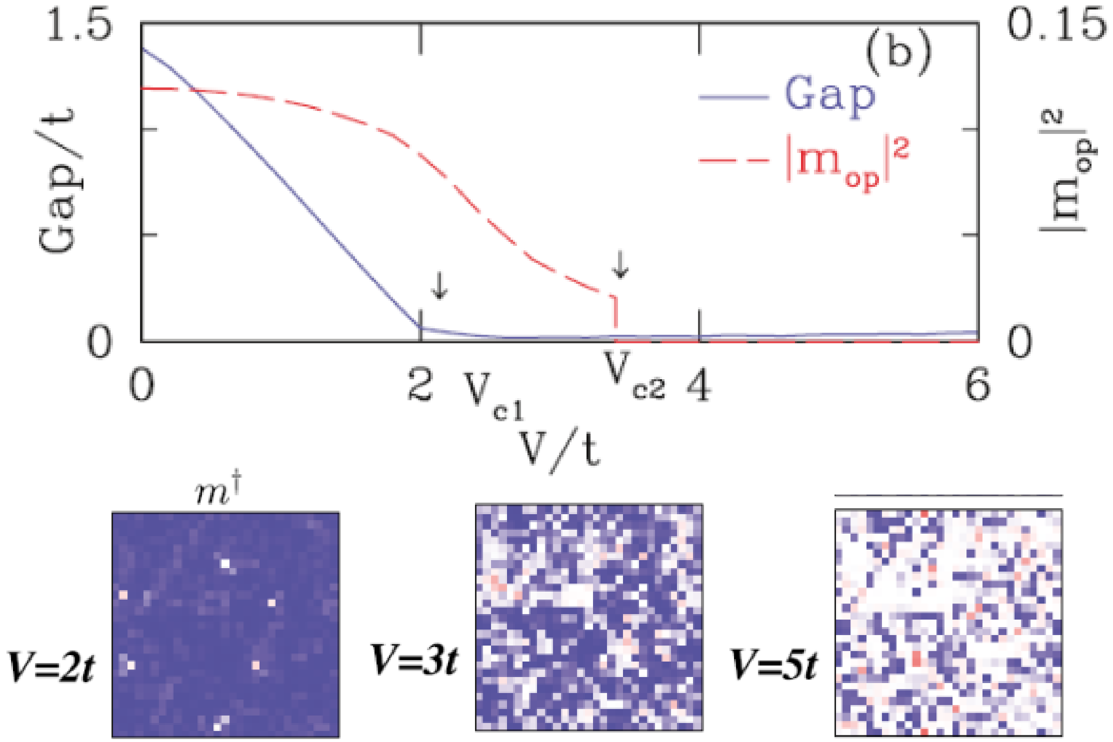
Long-range AF order survives
far beyond collapse of insulating state

Intermediate AF states beyond the Mott state

C. Yee et al. PRX (2014)



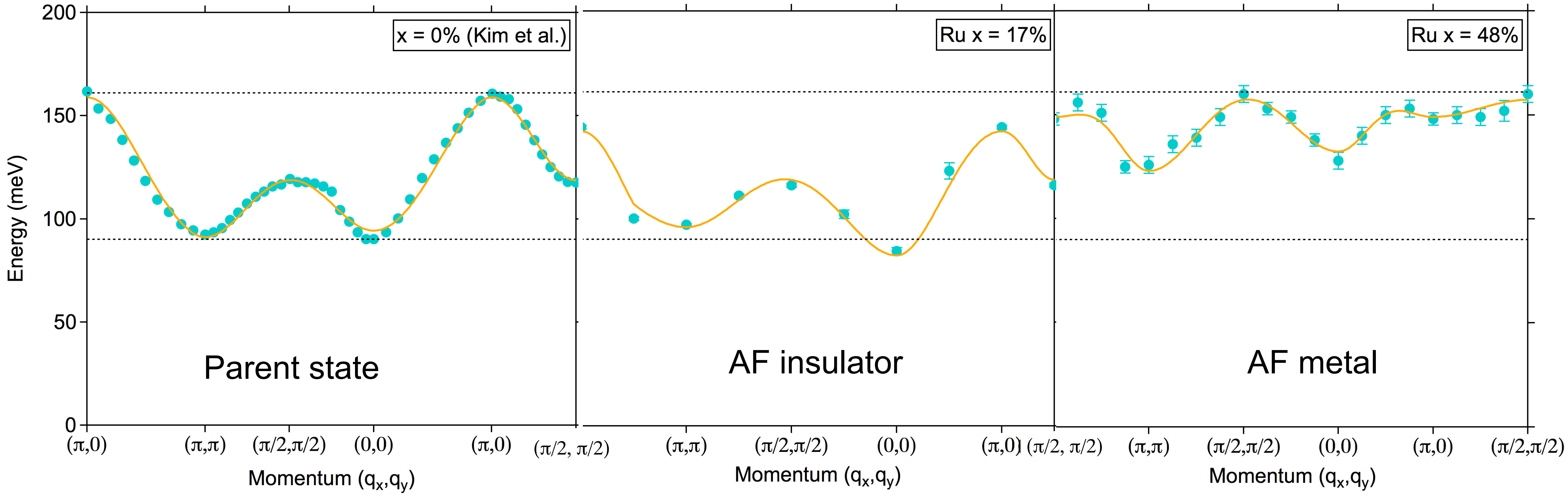
Heidarian et al. PRL (2004)



Thermodynamic arguments for first order transition into AF-M

Disorder driven collapse of Mott gap with extended survival of AF-M

Spin dynamics in intermediate SDW state

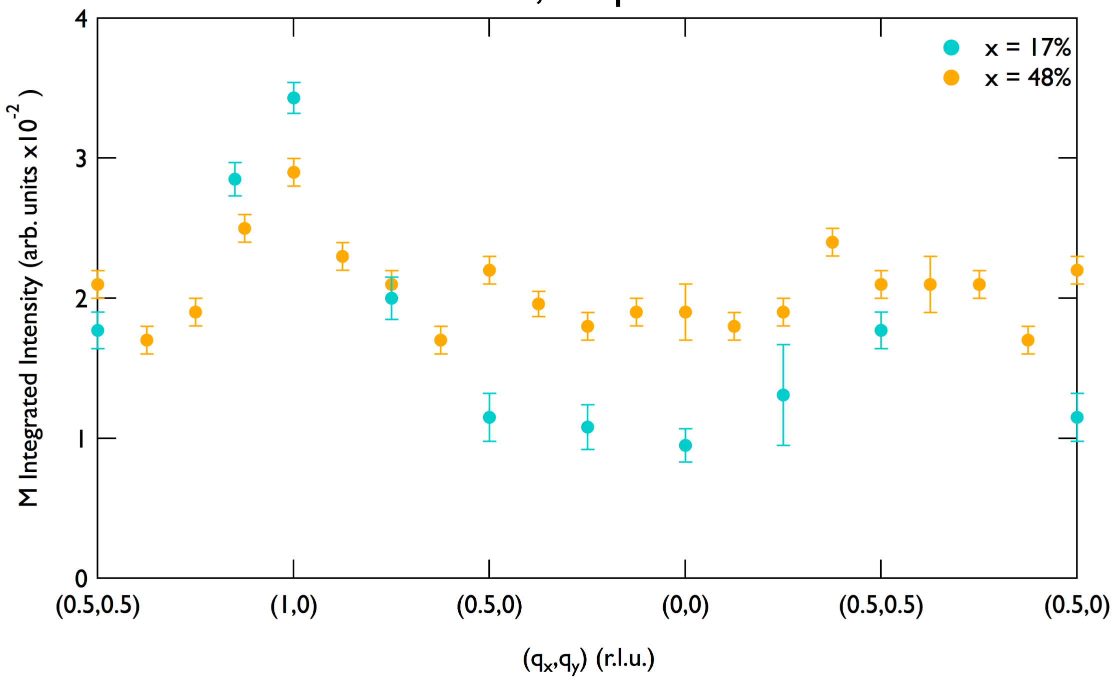


x	gap (meV)	bandwidth (meV)
0	90	72
17%	84	60
48%	125	35

Increasing gap and narrowing bandwidth with increased Ru content

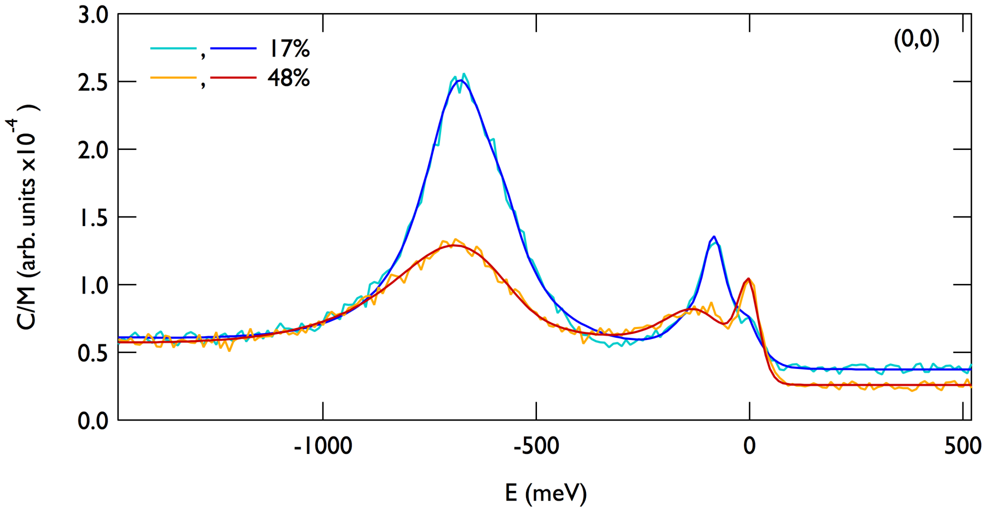
S(q) and J values in AF metallic phase

J. Schmehr, unpublished

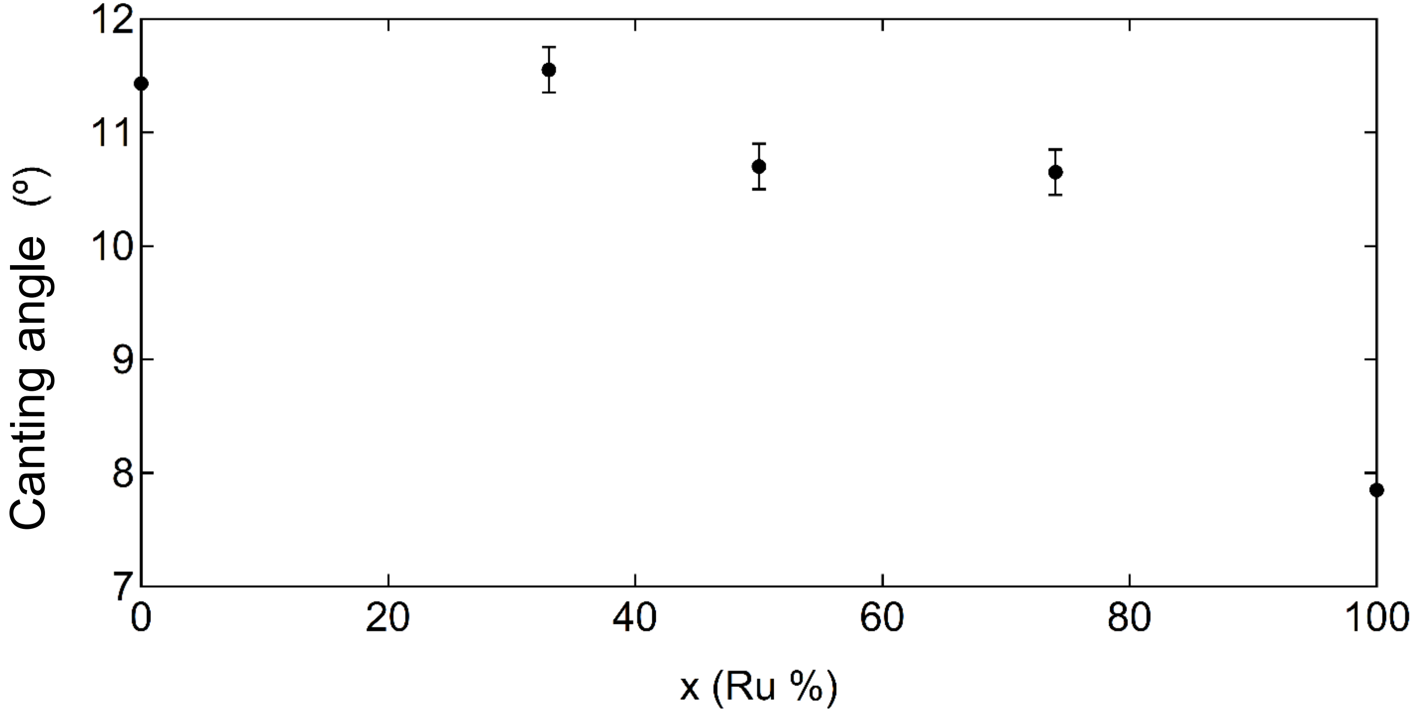


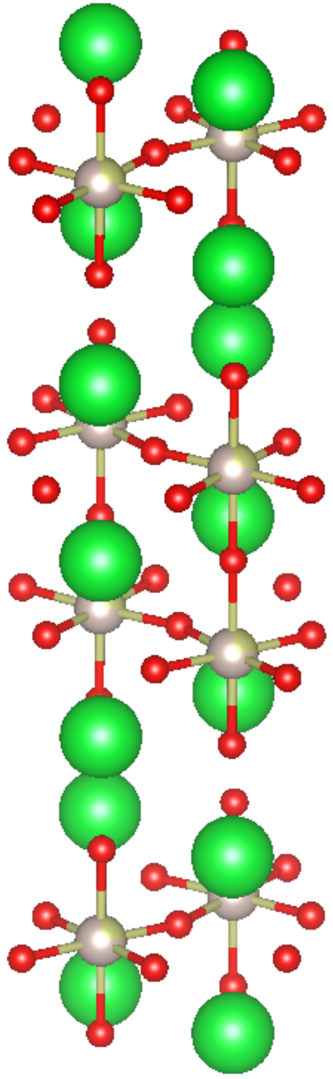
Parent Mott state	**AF metallic state**
J_1 = 26 meV	J_1 = 59 meV
J_2 = -15 meV	J_2 = 4 meV
J_3 = 6 meV	J_3 = 0 meV
J_c = 90 meV	J_c = 97 meV
Θ = 37 meV	Θ = 43 meV

- Difficult to parameterize
  - Evolution of Θ unknown
  - Anisotropy coupled to J_C
- Extended in-plane exchange damped out
- Interplane coupling largely unaffected
- Magnons heavily damped



Structure largely unperturbed in $Sr_3(Ir_{1-x}Ru_x)_2O_7$





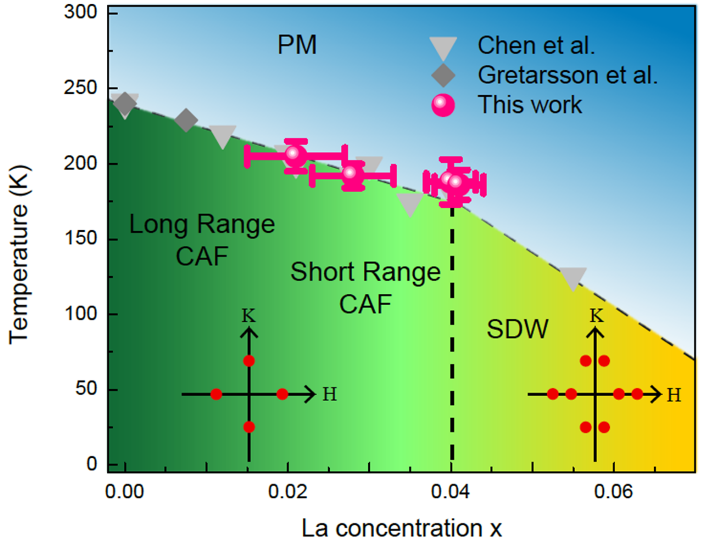
Heavy alloying with Ru is only a weak perturbation to the lattice structure

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

- Ru-alloying into is naively a strong perturbation to J_{eff}=1/2 state
  - Modified SOC
  - Added holes
  - S=1 impurities
- J_{eff}=1/2 Mott insulating phase survives locally until ~50% replacement of Ir with Ru
  - AF survives beyond this and remains stable within an inhomogenous, globally metallic, state
- Spin dynamics are unusual in new AF metallic state
  - Spin gap *increases*, bandwidth narrows
  - Disorder screens longer range J terms
  - Coupling between layers robust (in dimer type fitting)
- Work ongoing in exploring this state….

Unconventional metals in doped R.P. iridates

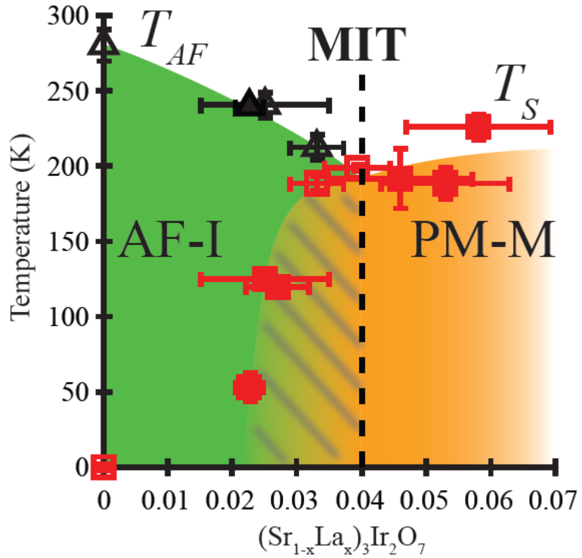
X. Chen et al., unpublished



Electron-doped Sr_2IrO_4 at 12% electrons/Ir acts like hole-doped La_2CuO_4 at 2% holes/Cu

Phase separated spin density wave state with "diagonal" character

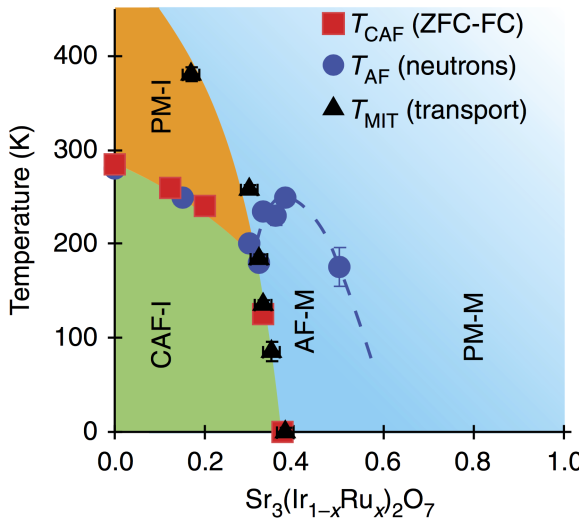
T. Hogan et al. PRL (2015)



Electron-doped $Sr_3Ir_2O_7$ reveals competing CDW-like instability

Persistent spin dynamics beyond collapse of static AF order. Phenomena suggestive of potential disordered dimer phase.

C. Dhital et al. Nat. Comm. (2014)



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