LSMatter Workshop
June 20, 2015

The Influence of Bulk Electron Doping in Ruddelsden-Popper Strontium Iridates

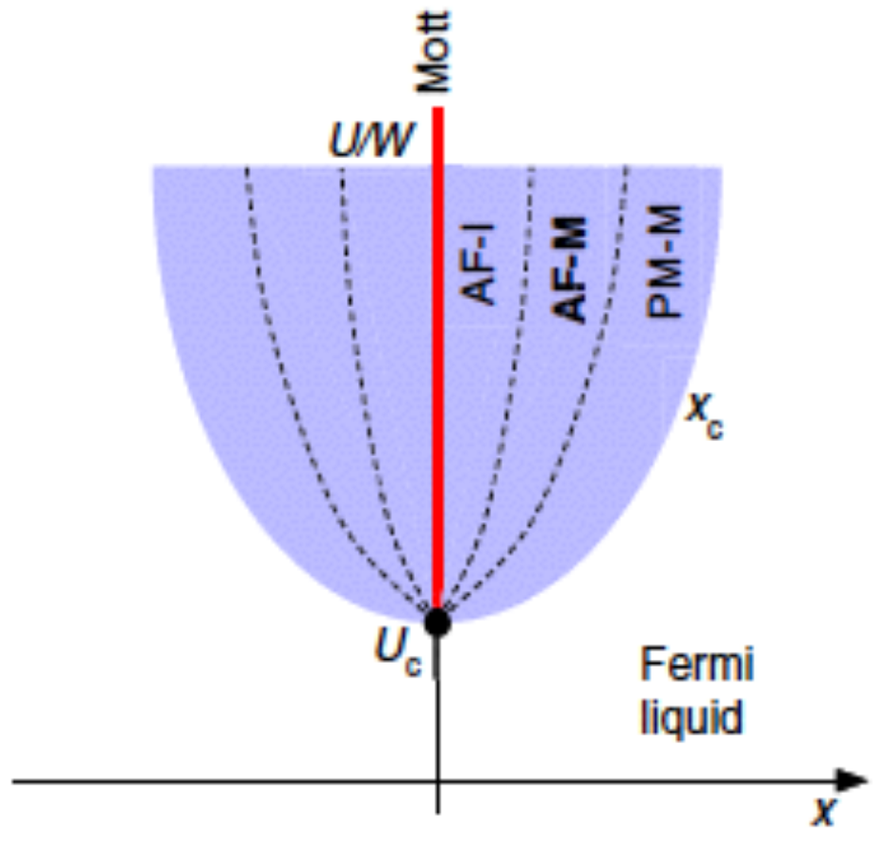
Carrier substitution at the weak limit of the Mott state

Stephen Wilson
Assistant Professor
Materials Department
UCSB

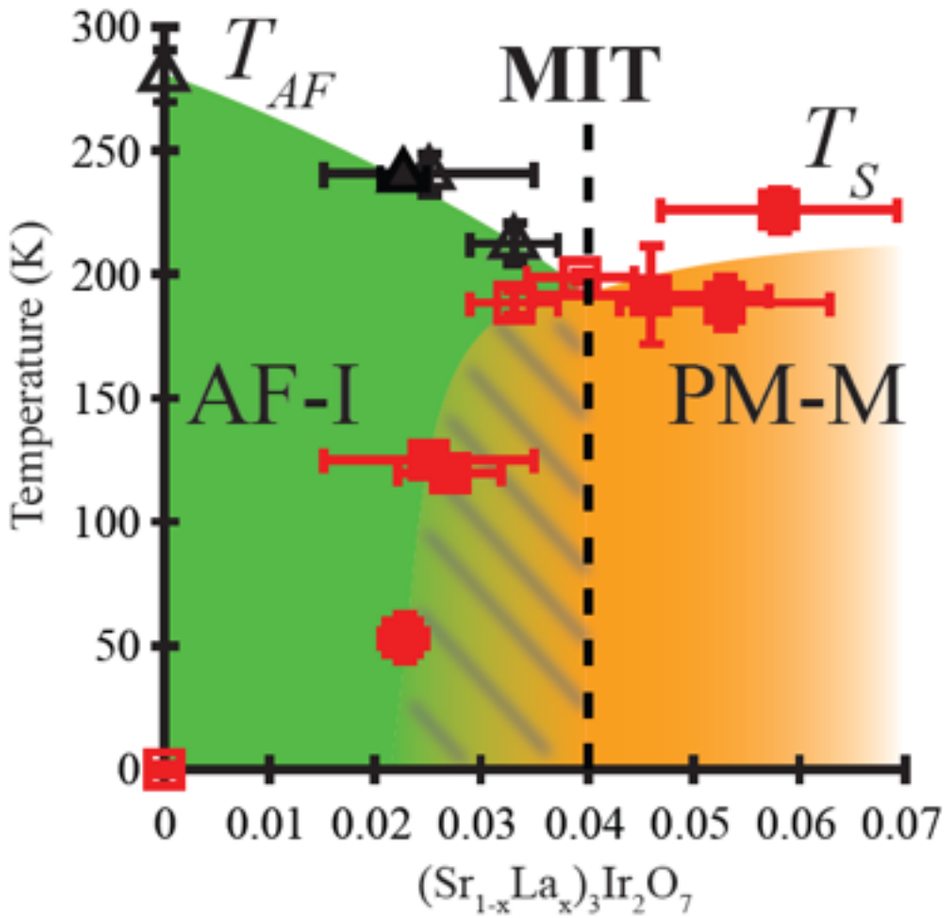
NSF CAREER
DMR-1056625

Outline

- Introduction and motivation
  - New states at intermediate coupling: electronic U, magnetic J, bandwidth t, spin-orbit λ
  - Spin orbit Mott state/R.P. Iridates
    - Platform for exploring MIT as weak limit is approached
- Electron-doping in $Sr_{2n+1}Ir_nO_{3n+1}$
  - How does MIT manifest?
  - Competing order/new states?
  - How does MIT evolve as U/t changes?
- Conclusions



C. Yee et al. PRX (2014)

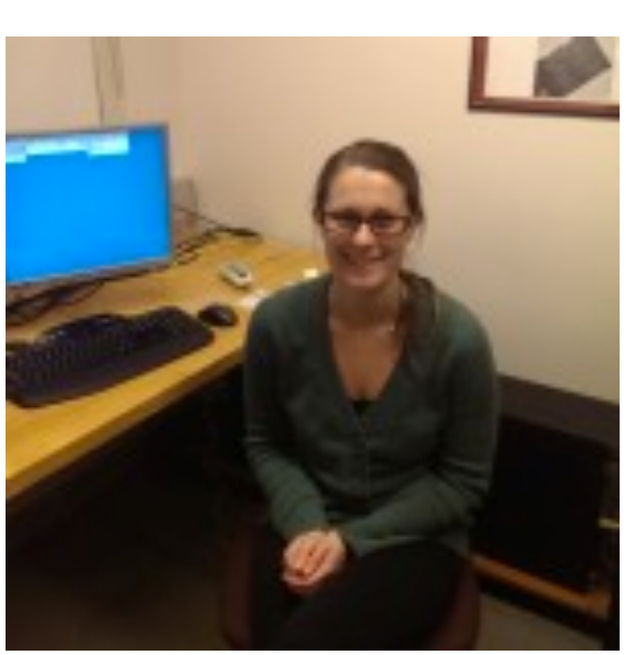


T. Hogan et al. PRL (in press)

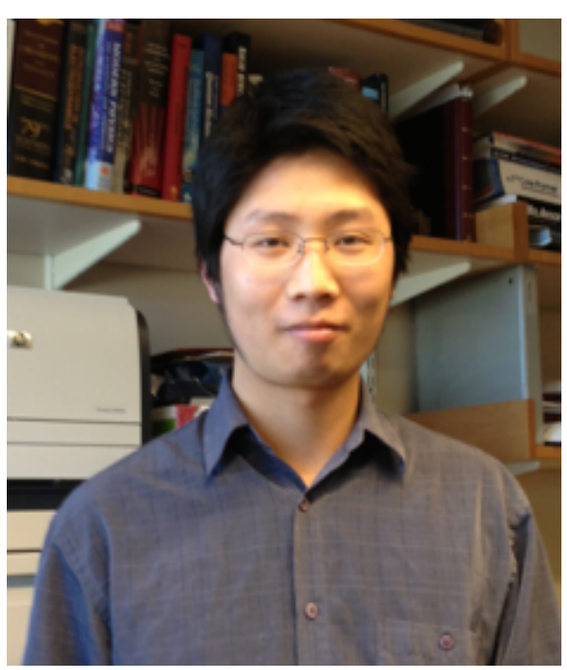
Students:

Tom Hogan

Ryan Need



Rebecca Dally



Xiang Chen

Collaborators:

UCSB

Boston College

NCNR NIST

Paul Scherrer Inst.

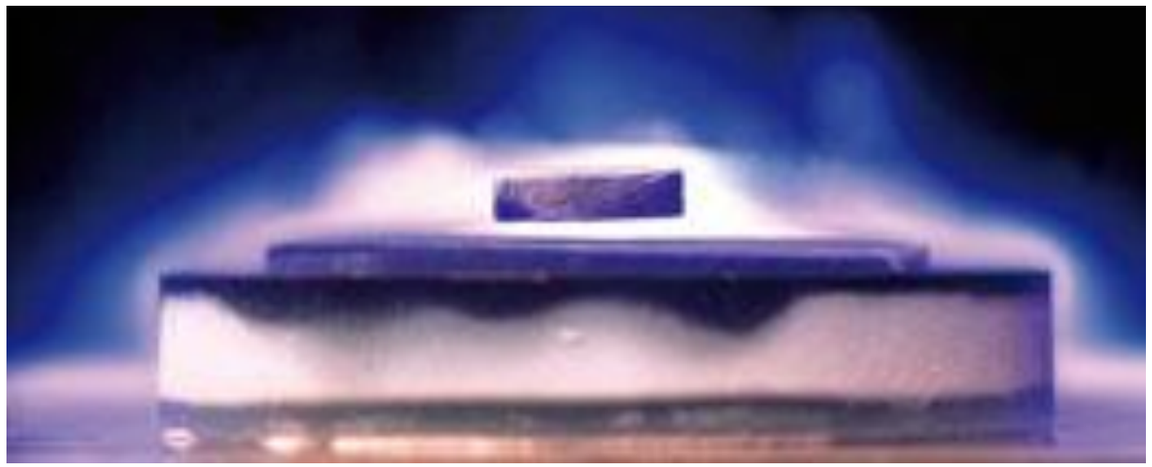
UIUC

Oak Ridge National Lab

Chalk River Laboratories

APS, ANL

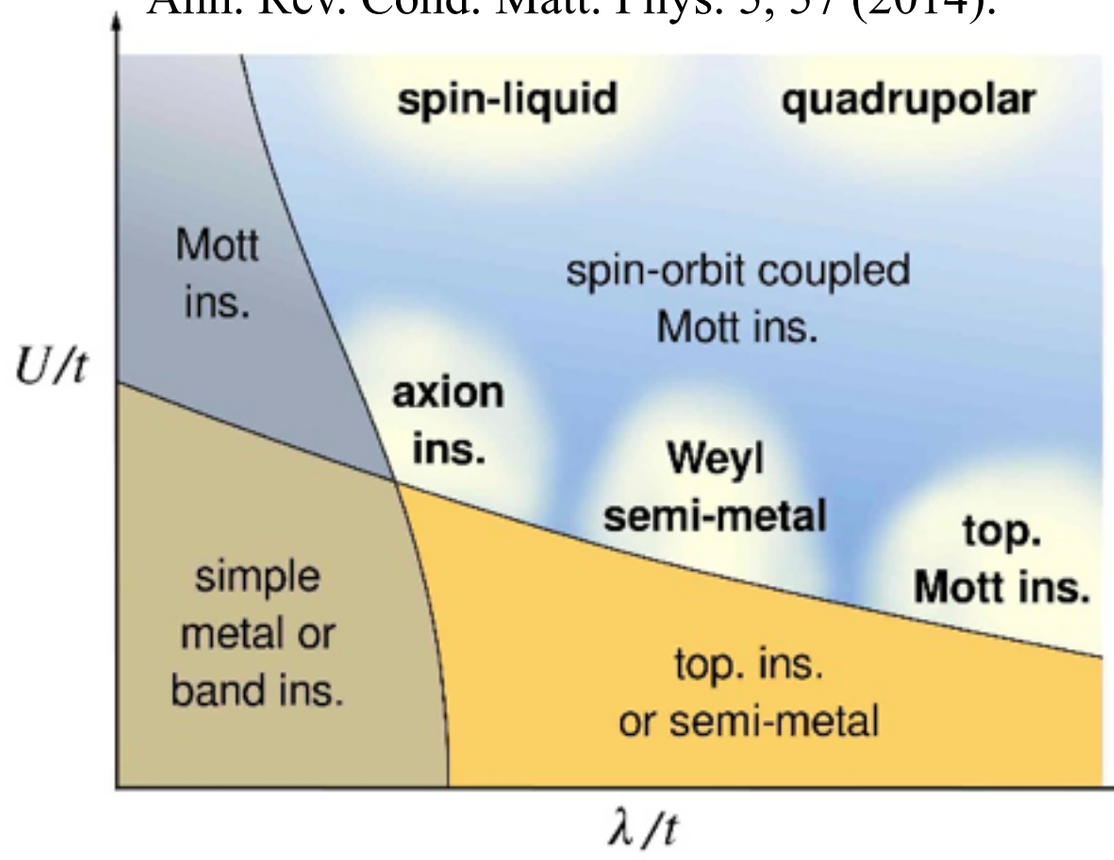
New Phases in materials with large U and large ƛ



Sr_2IrO_4,
R.P. perovskite iridates

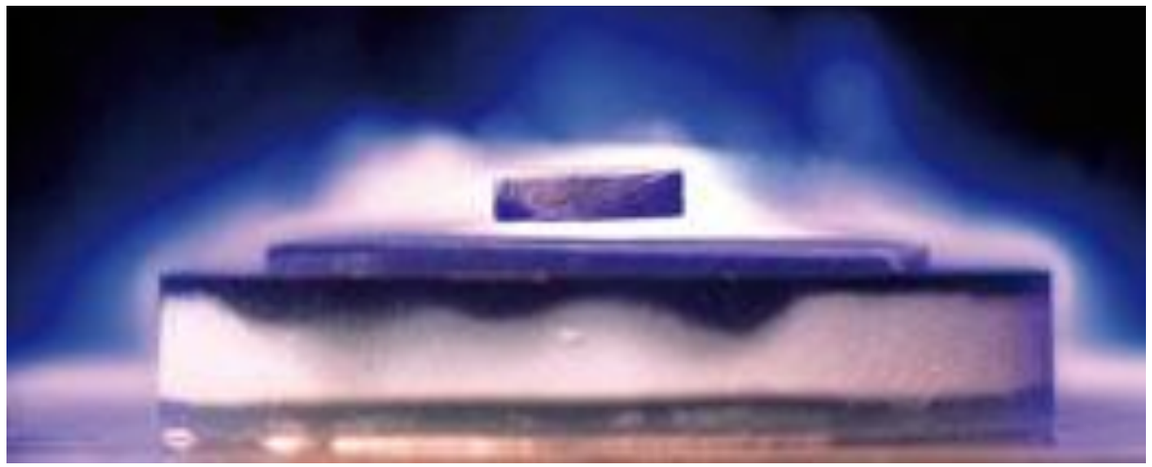
A_2IrO_3,
$Na_4Ir_3O_8$, …

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



$A_2Ir_2O_7$
(A=Y, Ln)

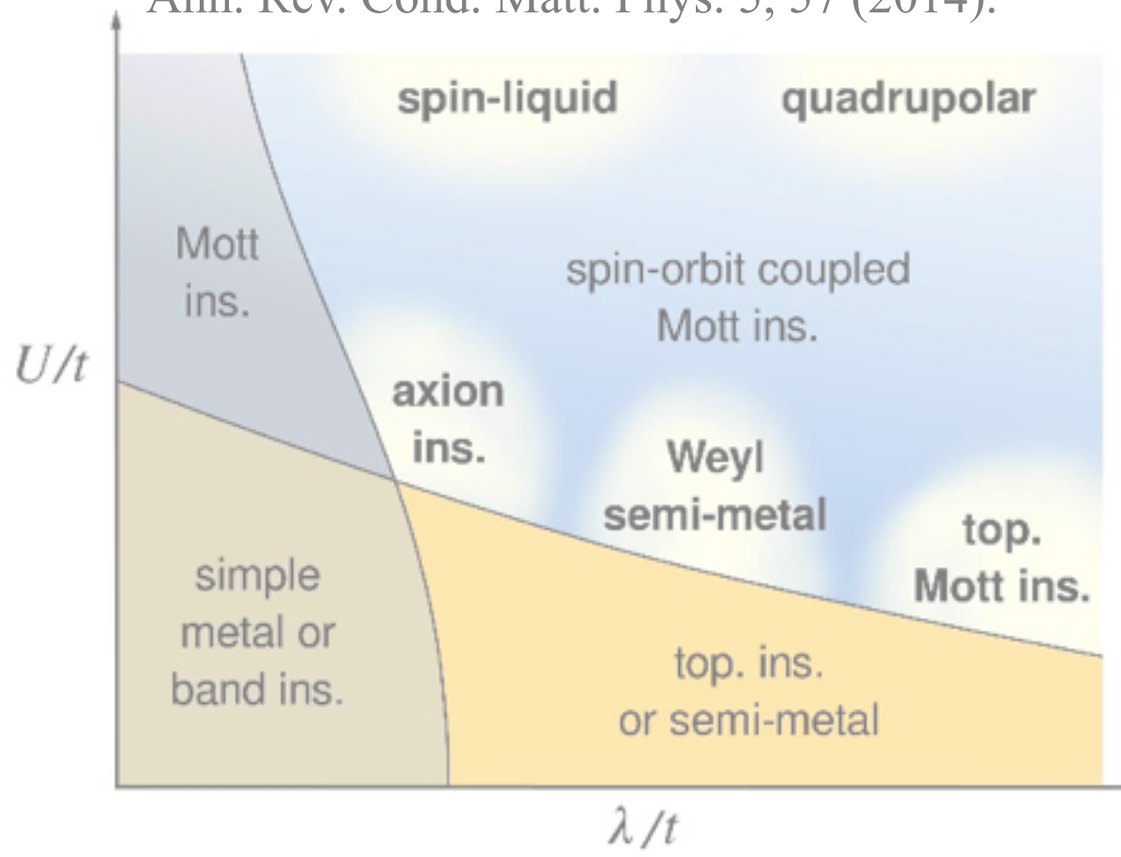
New Phases in materials with large U and large ƛ



Sr_2IrO_4,
R.P. perovskite iridates

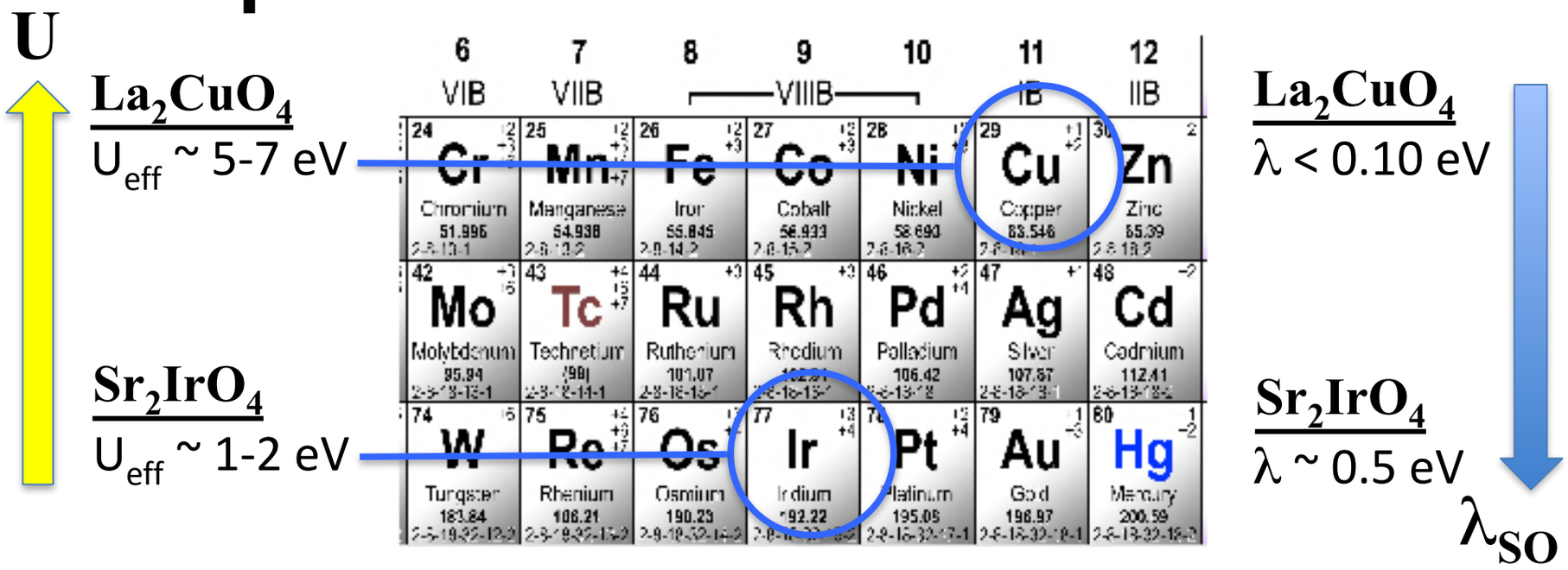
A
Na

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



A
(A=Y, Ln)

Spin Orbit Mott State



Spin Orbit Mott State

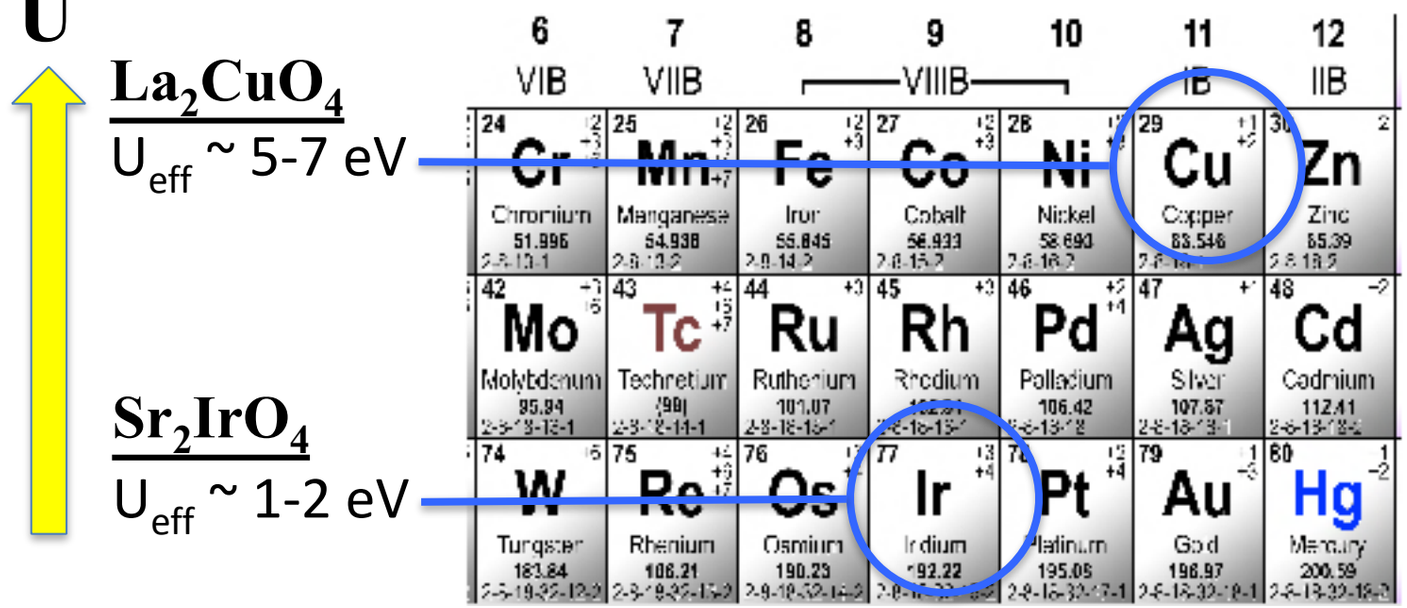
U

La_2CuO_4

U_{eff} ~ 5-7 eV

Sr_2IrO_4

U_{eff} ~ 1-2 eV



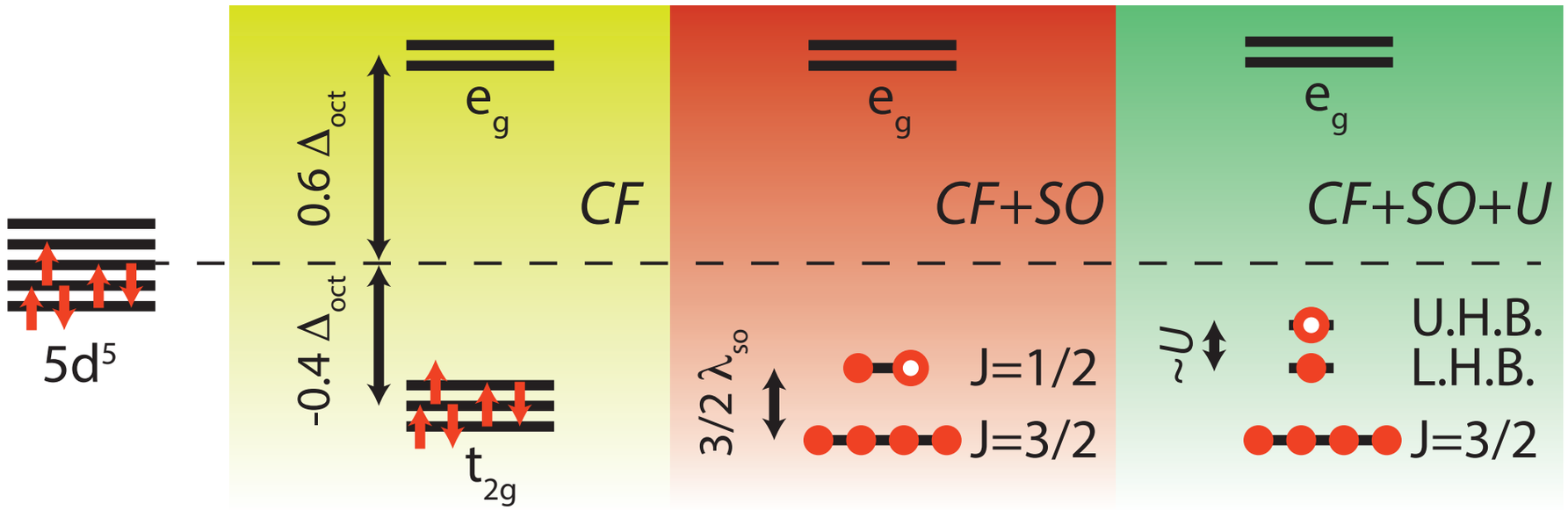
La_2CuO_4

$\lambda < 0.10$ eV

Sr_2IrO_4

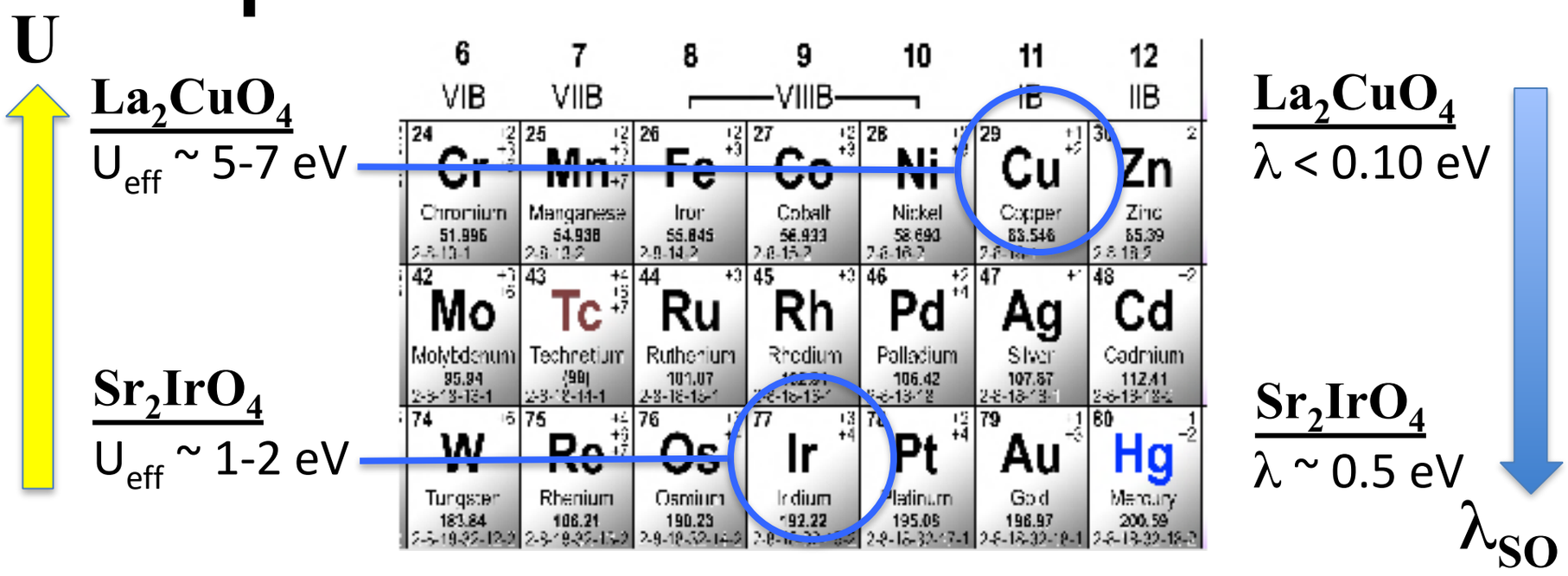
$\lambda \sim 0.5$ eV

λ_{SO}



A Mott state reliant on both strong ƛ and appreciable U

Spin Orbit Mott State



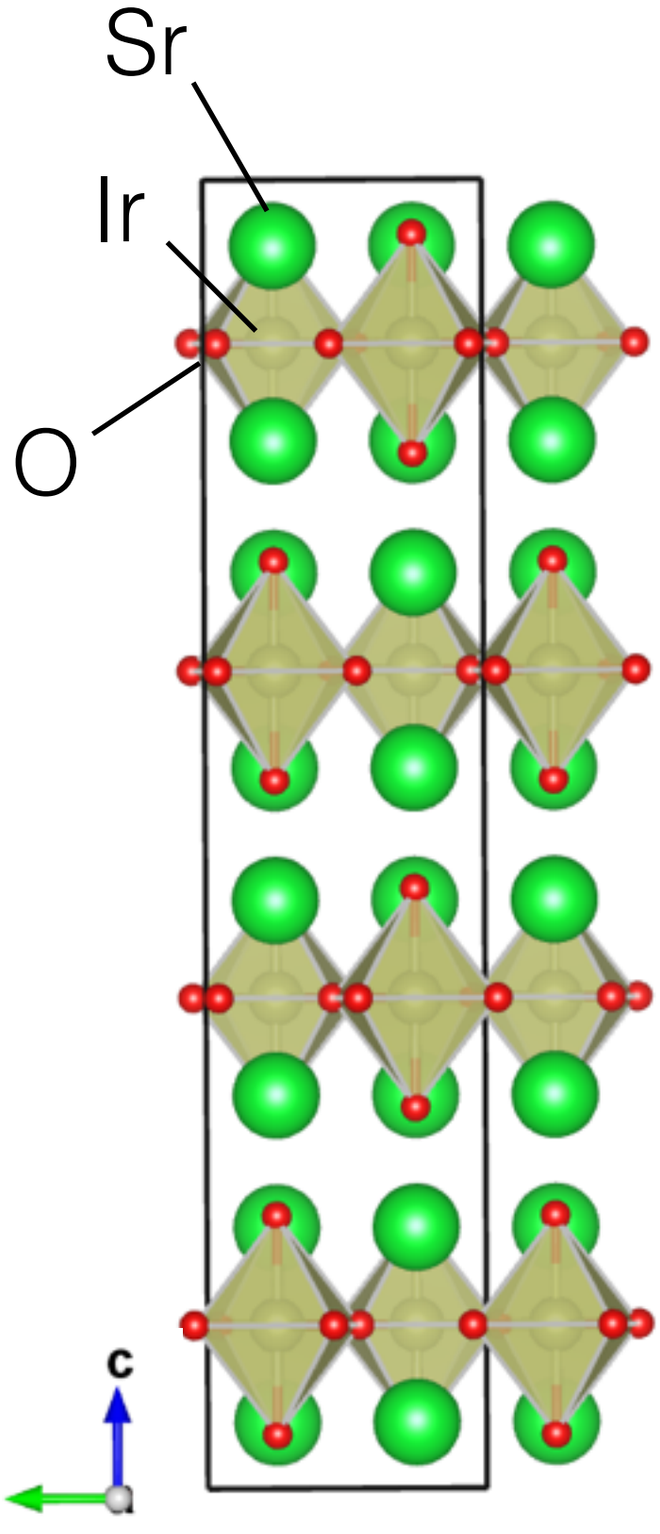
A Mott state reliant on both strong ƛ and appreciable U

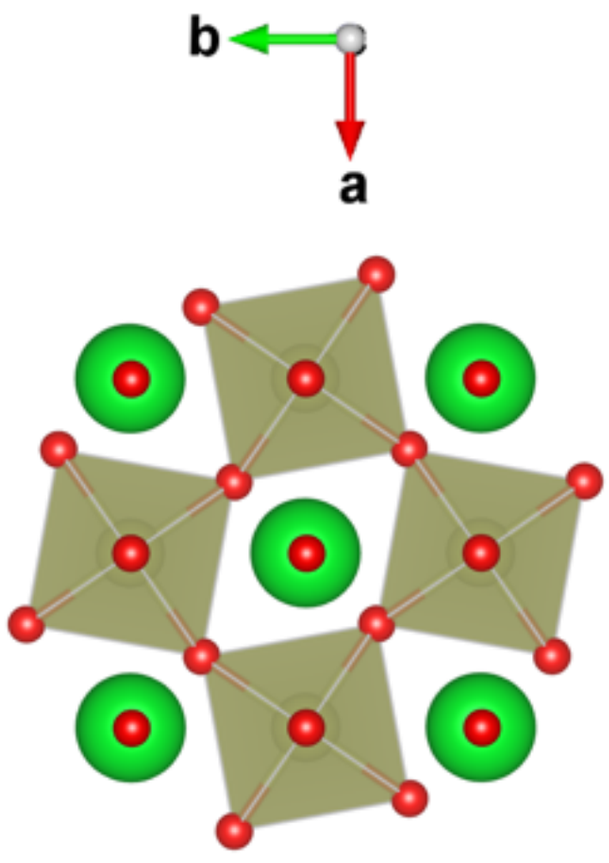
PRL 101, 076402 (2008) PHYSICAL REVIEW LETTERS week ending 15 AUGUST 2008

Novel $J_{eff} = 1/2$ Mott State Induced by Relativistic Spin-Orbit Coupling in Sr_2IrO_4

B. J. Kim,[1] Hosub Jin,[1] S. J. Moon,[2] J.-Y. Kim,[3] B.-G. Park,[4] C. S. Leem,[5] Jaejun Yu,[1] T. W. Noh,[2] C. Kim,[5] S.-J. Oh,[1] J.-H. Park,[3,4,*] V. Durairaj,[6] G. Cao,[6] and E. Rotenberg[7]

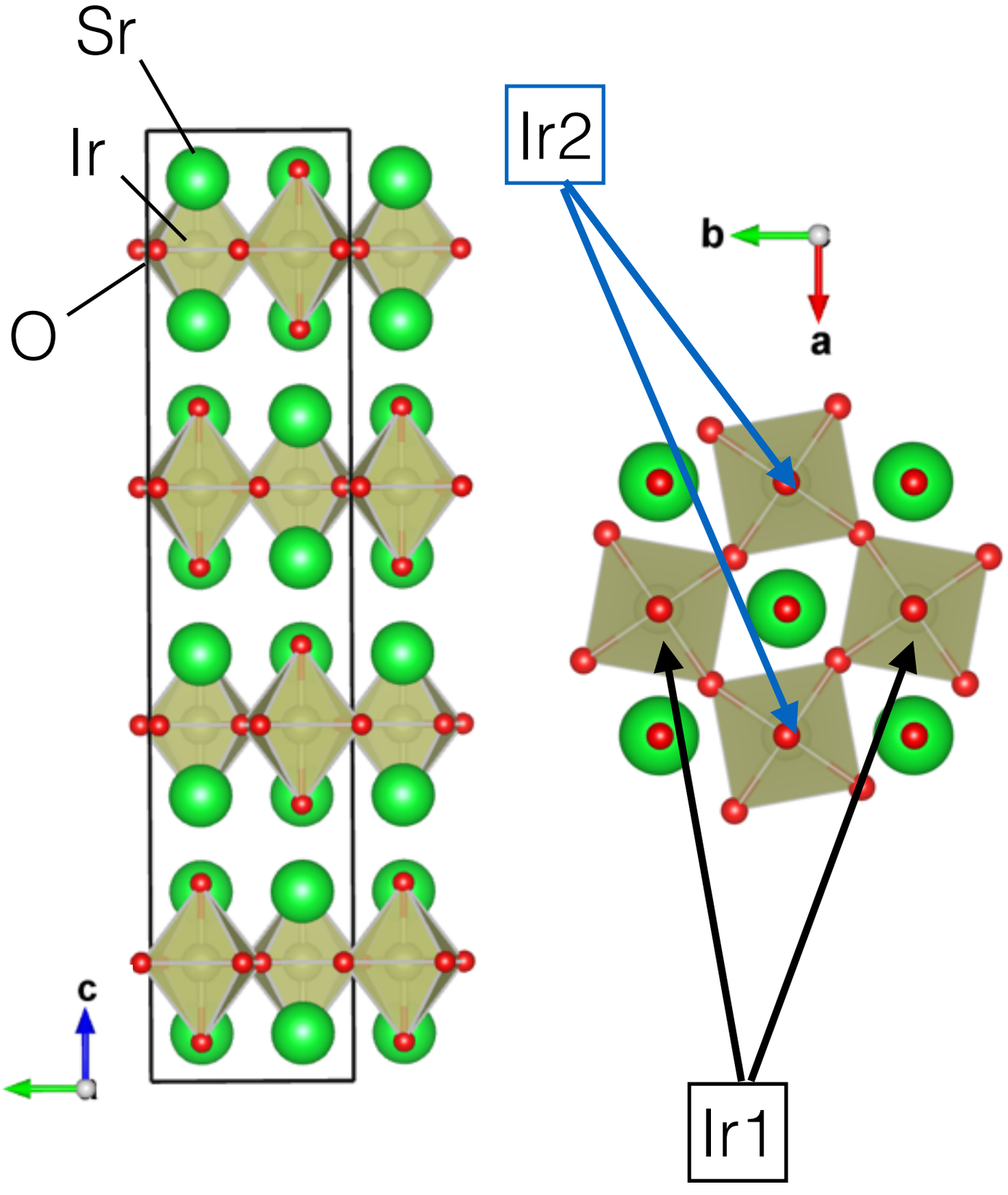
$n=1$ Sr_2IrO_4





- Puzzle of weak violations of $I4_1/acd$
  - Dhital et al., PRB (2013)
  - Ye et al., PRB (2014)
- $I4_1/a$ most likely space group (4/m point group)
  - Torchinsky et al., PRL (2015)

$n=1$ Sr_2IrO_4



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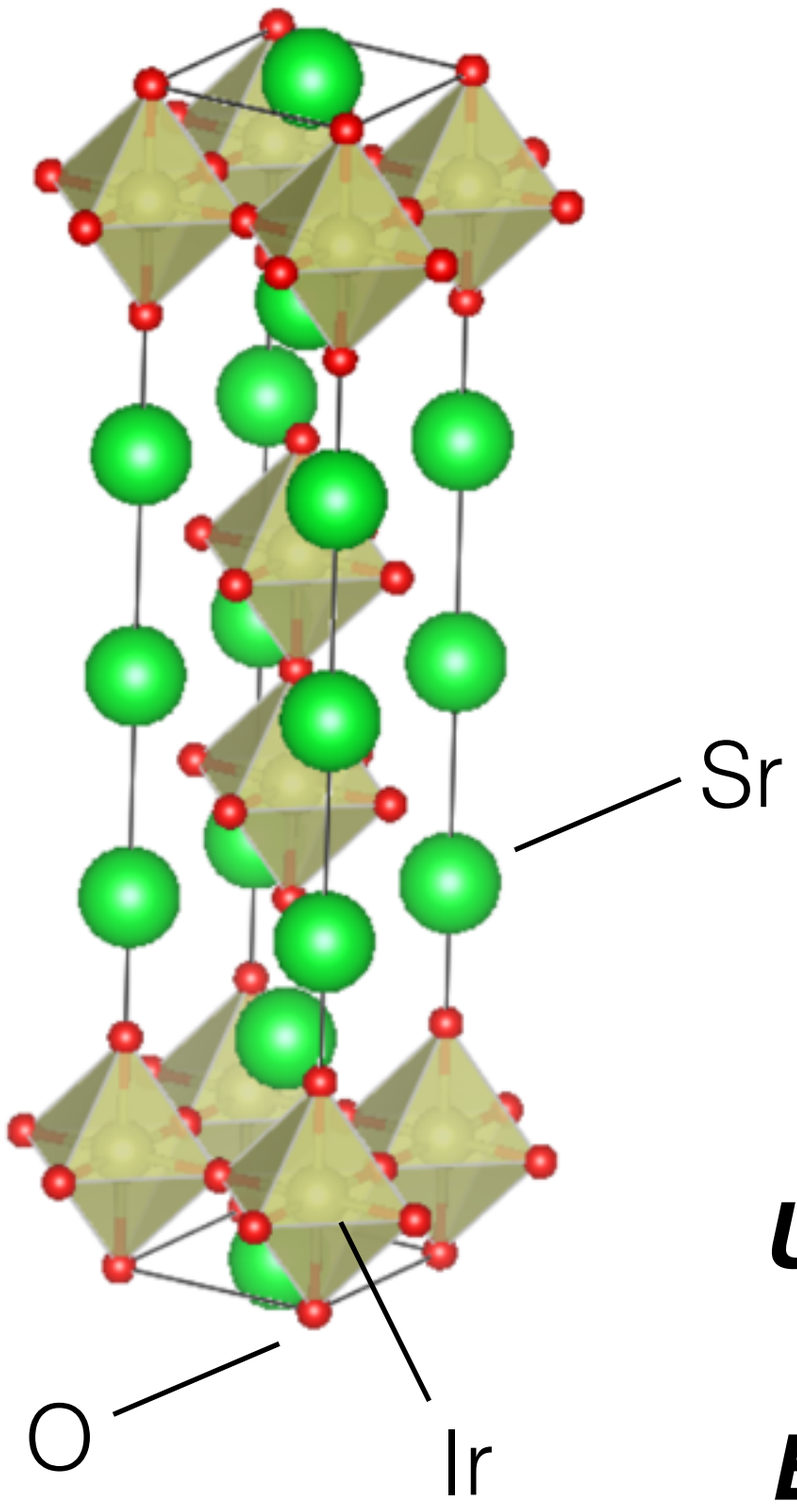
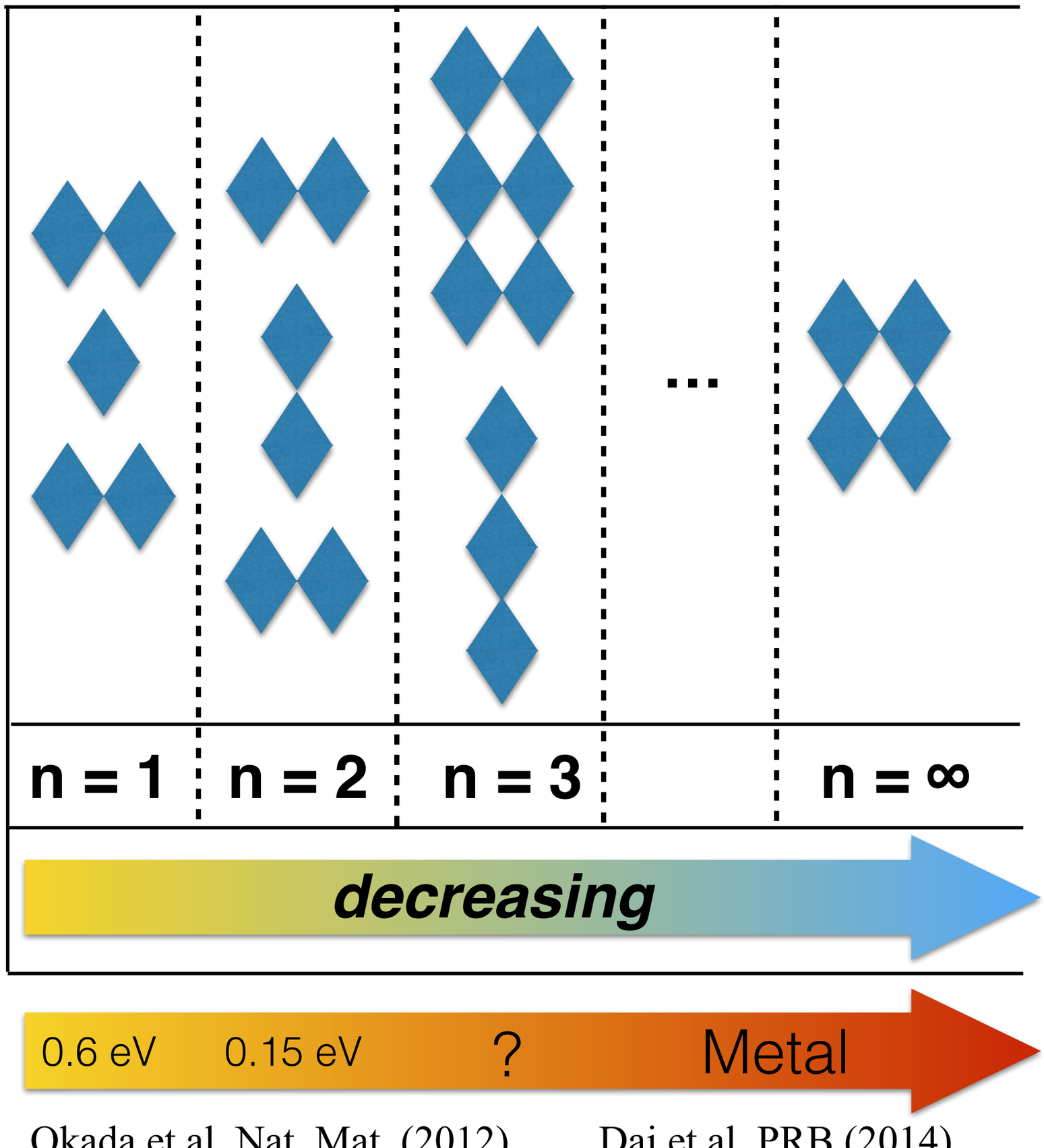
R.P. Iridates: $Sr_{2n+1}Ir_nO_{3n+1}$





Okada et al. Nat. Mat. (2012)

Dai et al. PRB (2014)

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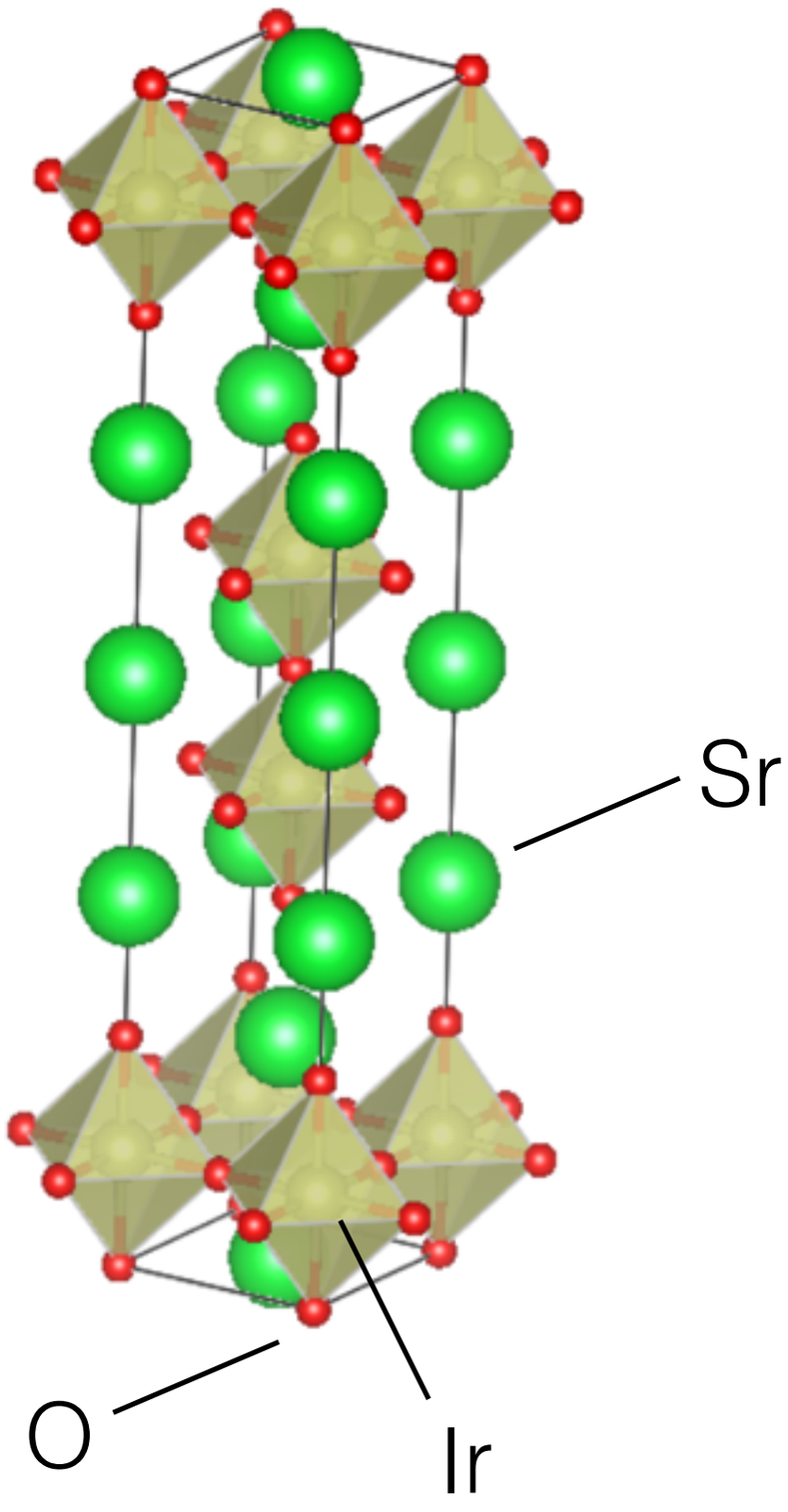
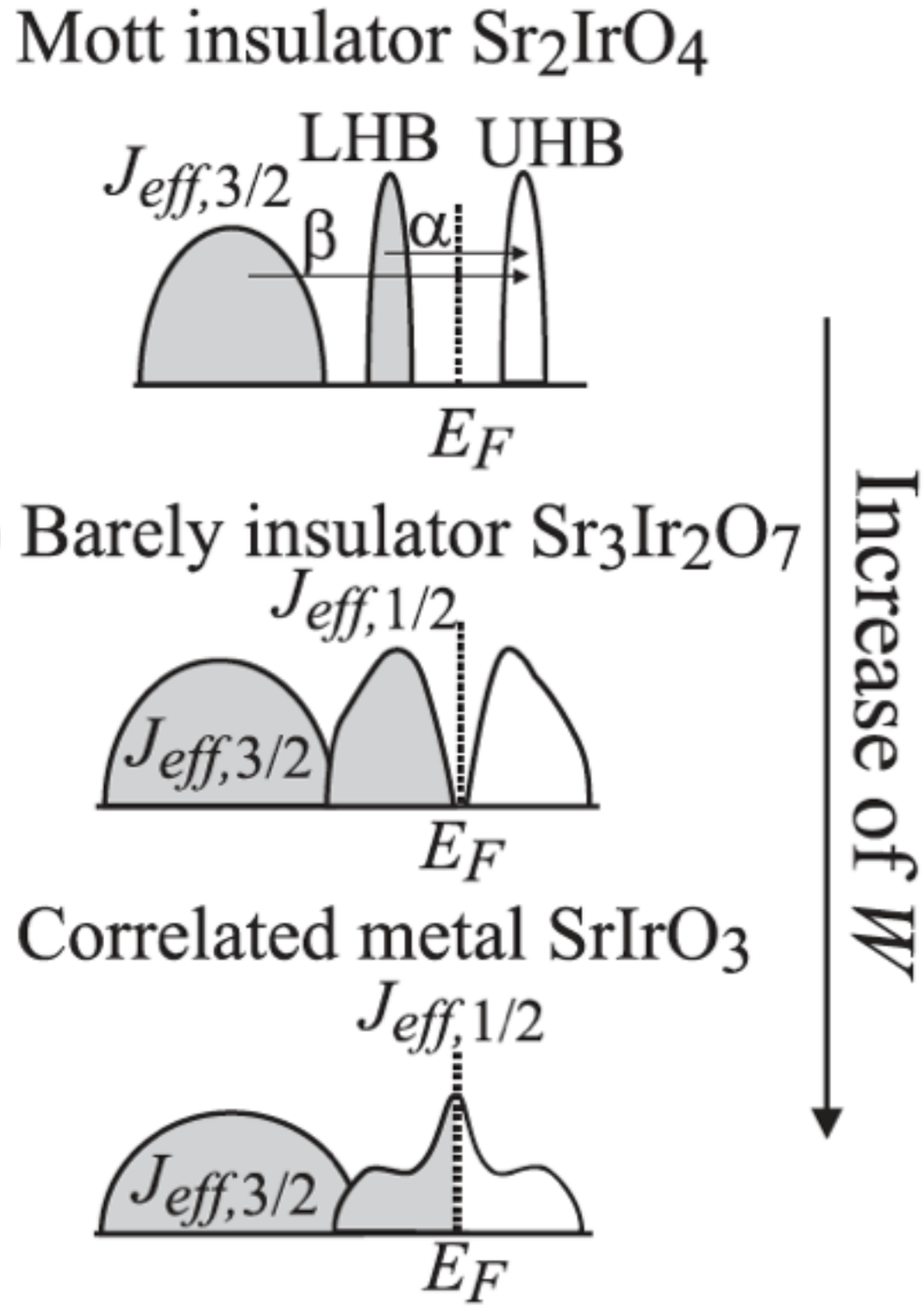
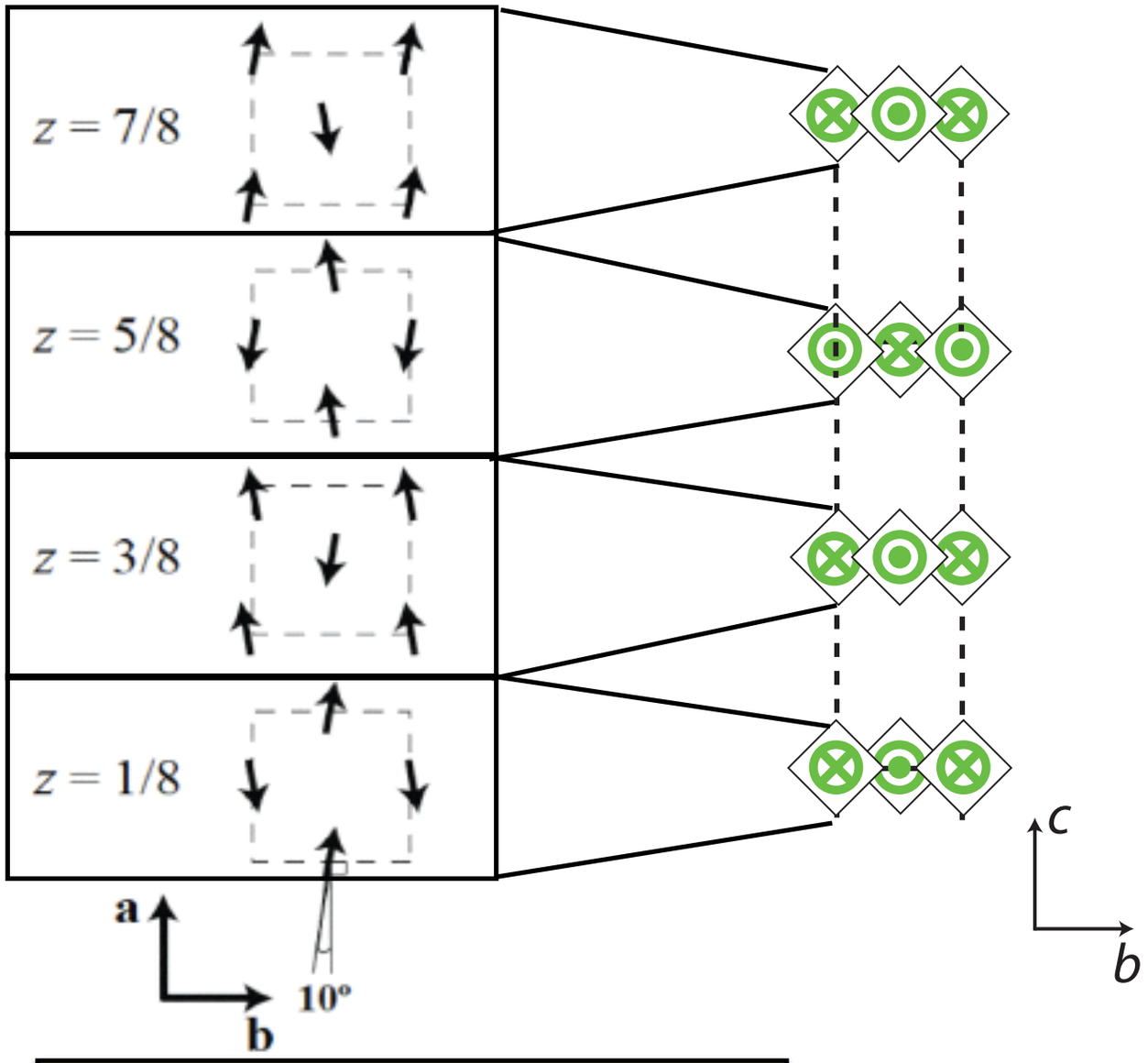




Moon et al. PRL (2008).

Magnetism in RP. Iridates

Dhital et al., PRB (2013)



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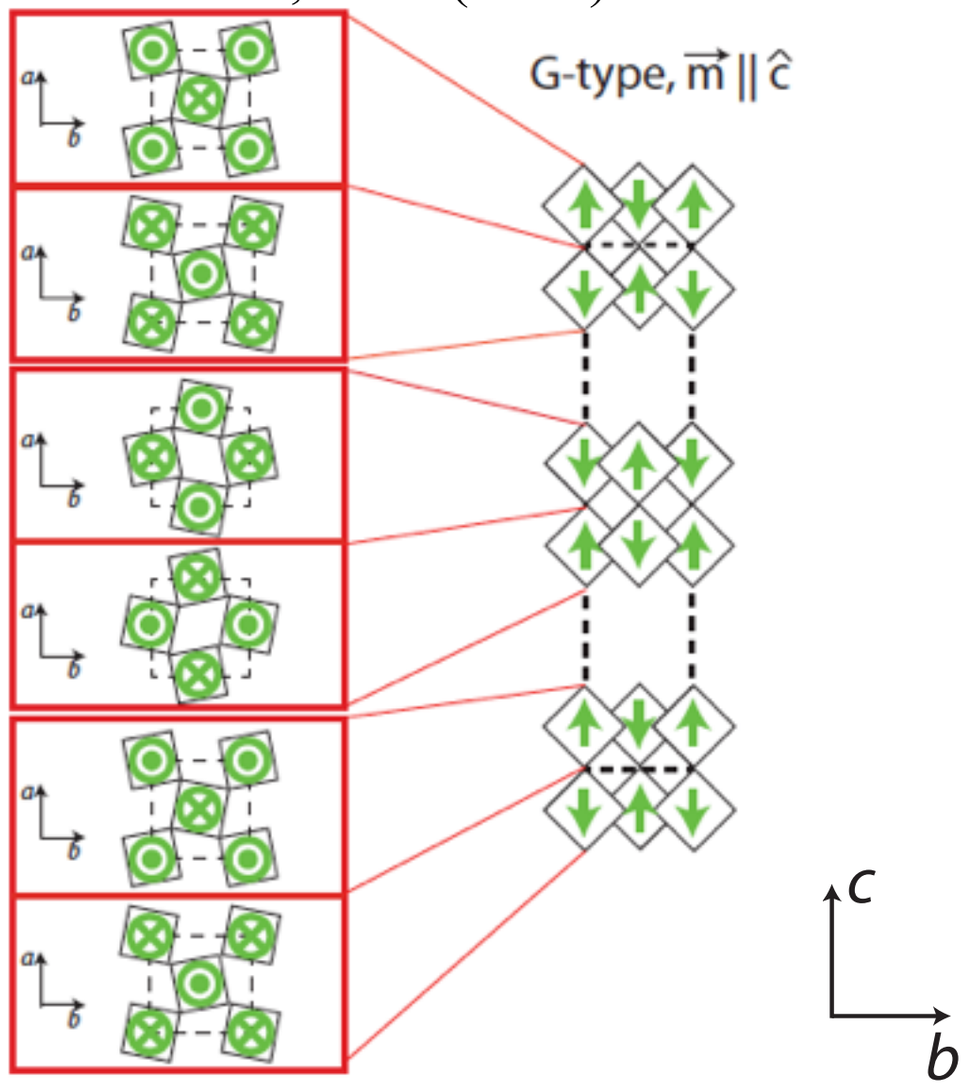


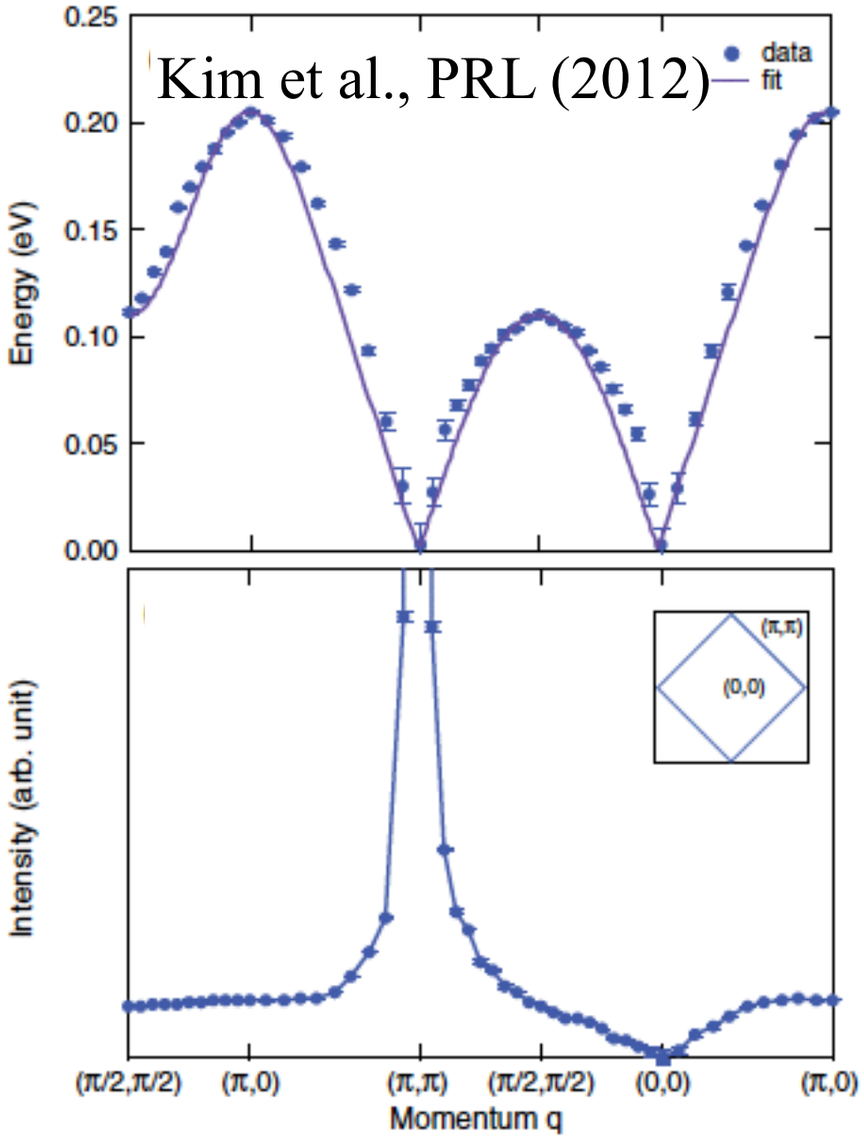
Sr_2IrO_4

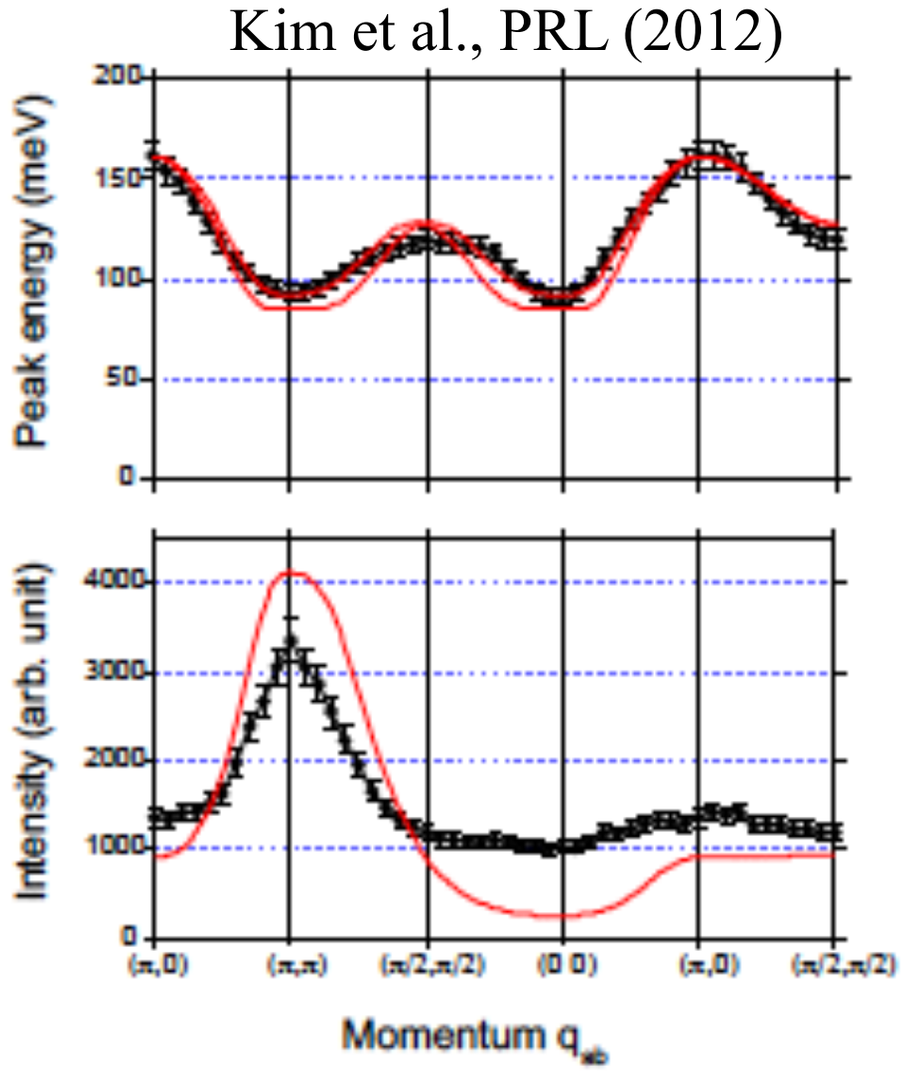
- $m_{AF} = 0.34\ \mu_B$
- $J_{NN} \sim 60$ meV
  $J_{NNN} \sim -20$ meV

$Sr_3Ir_2O_7$

- $m_{AF} = 0.36\ \mu_B$
- $J_{NN} \sim 93$ meV
  $J_{NNN} \sim 12$ meV
  $J_C \sim 25$ meV

Magnetism in RP. Iridates





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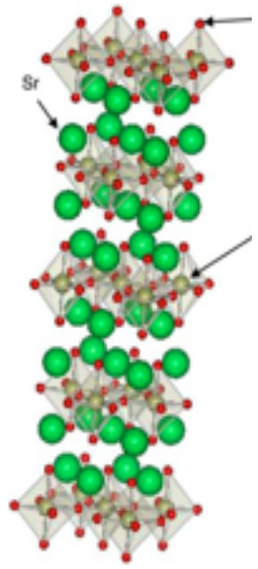
High-Tc in N-type Iridates



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

Fa Wang and T. Senthil

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

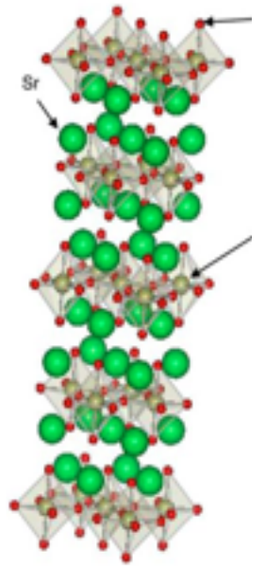
(Received 17 November 2010; published 30 March 2011)

$$H = -\sum_{\langle jk\rangle,\alpha}(t + \mathbb{i}\epsilon_\alpha\epsilon_j\bar{t})d^\dagger_{j,\alpha}d_{k,\alpha} - \sum_{\langle\langle jk\rangle\rangle,\alpha} t' d^\dagger_{j,\alpha}d_{k,\alpha}$$

$$- \sum_{\langle\langle\langle jk\rangle\rangle\rangle,\alpha} t'' d^\dagger_{j,\alpha}d_{k,\alpha} + U\sum_j d^\dagger_{j,\uparrow}d_{j,\uparrow}d^\dagger_{j,\downarrow}d_{j,\downarrow},$$

n-type iridate maps to p-type cuprate

High-Tc in N-type Iridates



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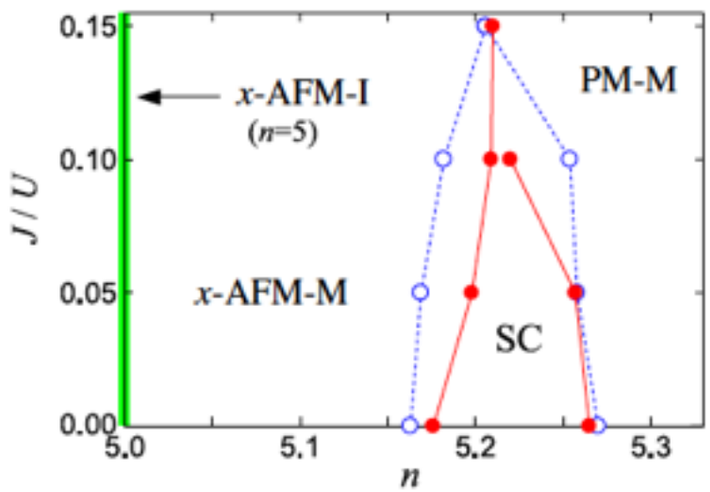
n-type iridate maps to p-type cuprate

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

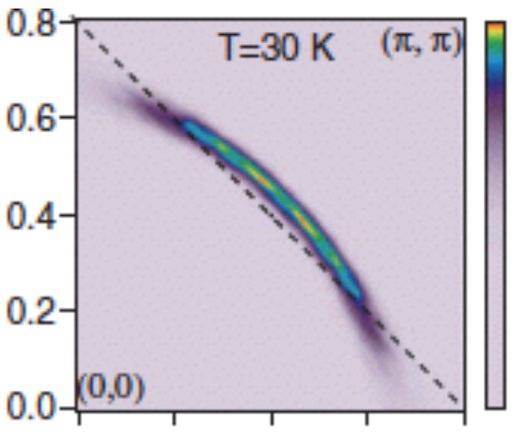
Hiroshi Watanabe,* Tomonori Shirakawa, and Seiji Yunoki

Computational Condensed Matter Physics Laboratory, RIKEN ASI, Wako, Saitama 351-0198, Japan, CREST, Japan Science and Technology Agency, Kawaguchi, Saitama 332-0012, Japan, and Computational Materials Science Research Team, RIKEN AICS, Kobe, Hyogo 650-0047, Japan

(Received 1 June 2012; published 9 January 2013)

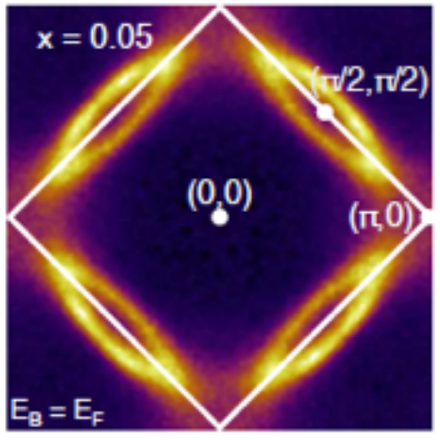


Exotic Metals in N-type RP Iridates



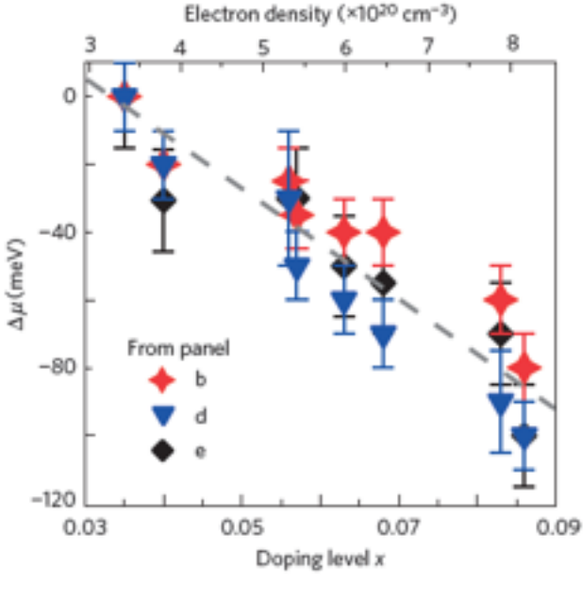
Fermi arcs in a doped pseudospin-1/2 Heisenberg antiferromagnet

Y. K. Kim,[1] O. Krupin,[1] J. D. Denlinger,[1] A. Bostwick,[1] E. Rotenberg,[1] Q. Zhao,[2] J. F. Mitchell,[2] J. W. Allen,[3] B. J. Kim[2,3,4]*



Collapse of the Mott gap and emergence of a nodal liquid in lightly doped Sr_2IrO_4

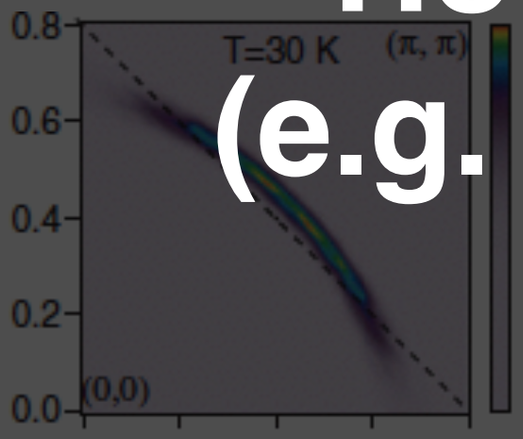
A. de la Torre,[1] S. McKeown Walker,[1] F. Y. Bruno,[1] S. Ricco,[1] Z. Wang,[2,1] I. Gutierrez Lezama,[1] G. Scheerer,[1] G. Giriat,[1] D. Jaccard,[1] C. Berthod,[1] T. K. Kim,[3] M. Hoesch,[3] E. C. Hunter,[4] R. S. Perry,[5] A. Tamai,[1] and F. Baumberger[1,2,6]

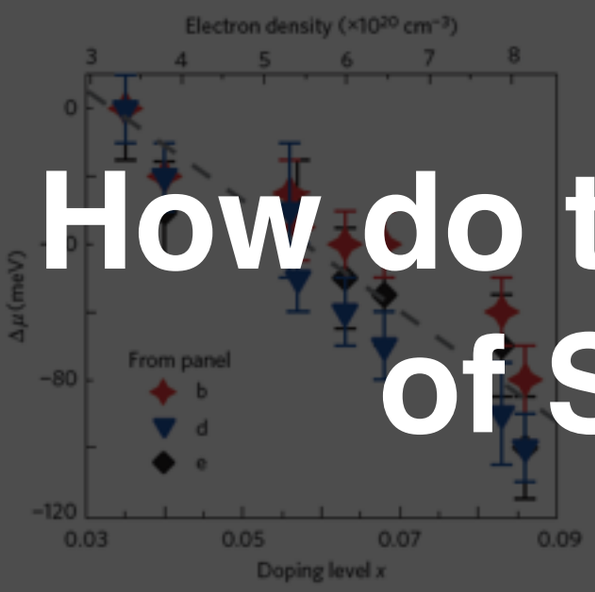


Spectroscopic evidence for negative electronic compressibility in a quasi-three-dimensional spin–orbit correlated metal

Junfeng He[1†], T. Hogan[1†], Thomas R. Mion[1], H. Hafiz[2], Y. He[3], J. D. Denlinger[4], S-K. Mo[4], C. Dhital[1], X. Chen[1], Qisen Lin[1], Y. Zhang[5], M. Hashimoto[3], H. Pan[1], D. H. Lu[3], M. Arita[6], K. Shimada[6], R. S. Markiewicz[2], Z. Wang[1], K. Kempa[1], M. J. Naughton[1], A. Bansil[2], S. D. Wilson[1,7] and Rui-Hua He[1*]

Exotic Metals in N-type RP Iridates

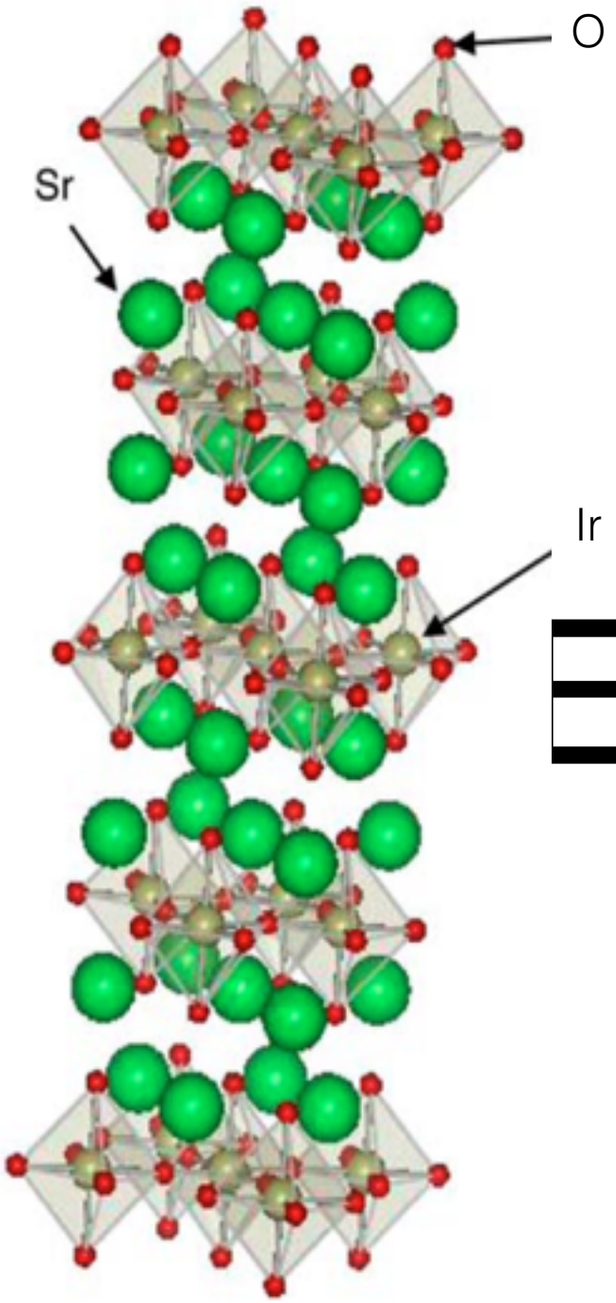




How do these states arise? (e.g. competing orders, broken symmetries)

How do the corresponding MITs manifest?

How do the MITs evolve as weak limit of SOM state approached?

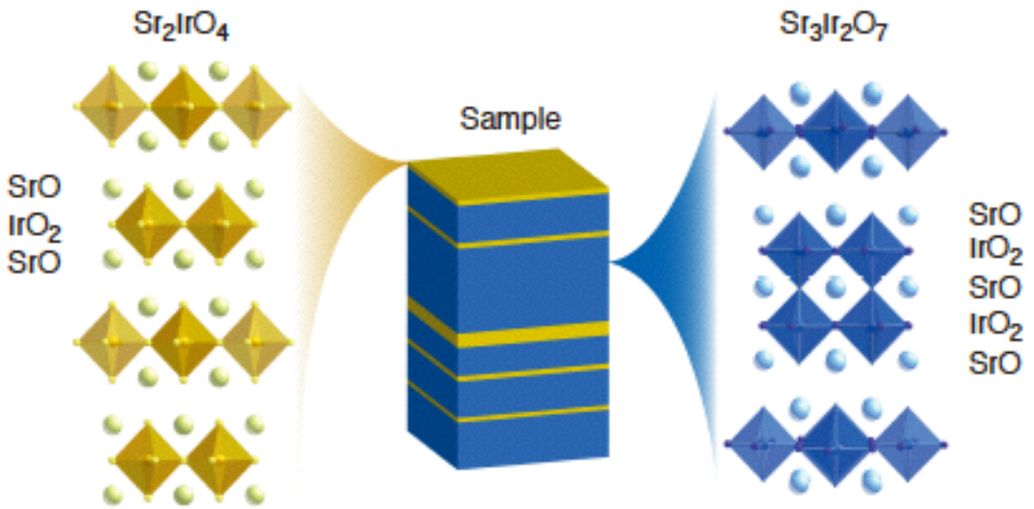


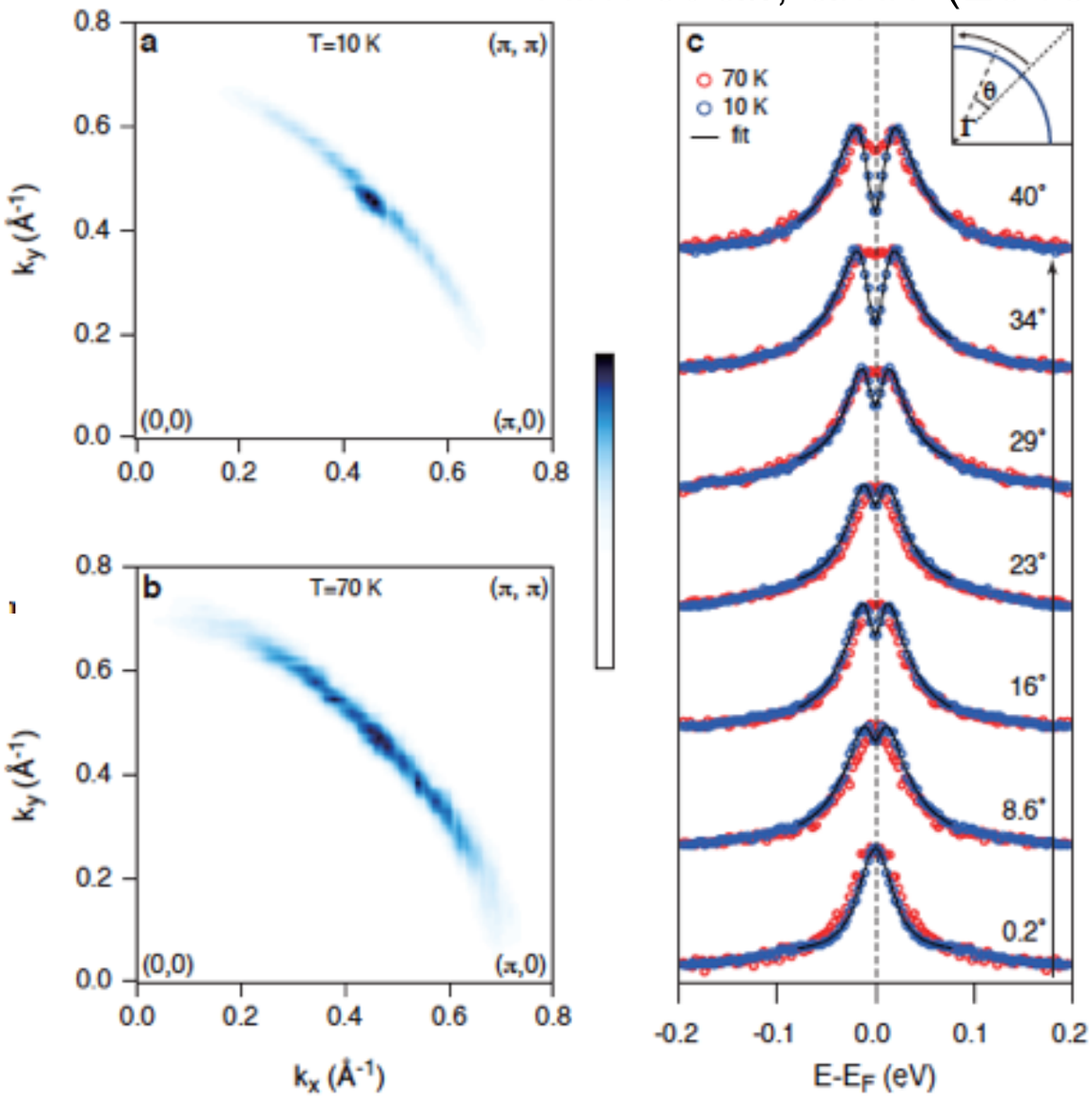
Electron-doped Sr-21

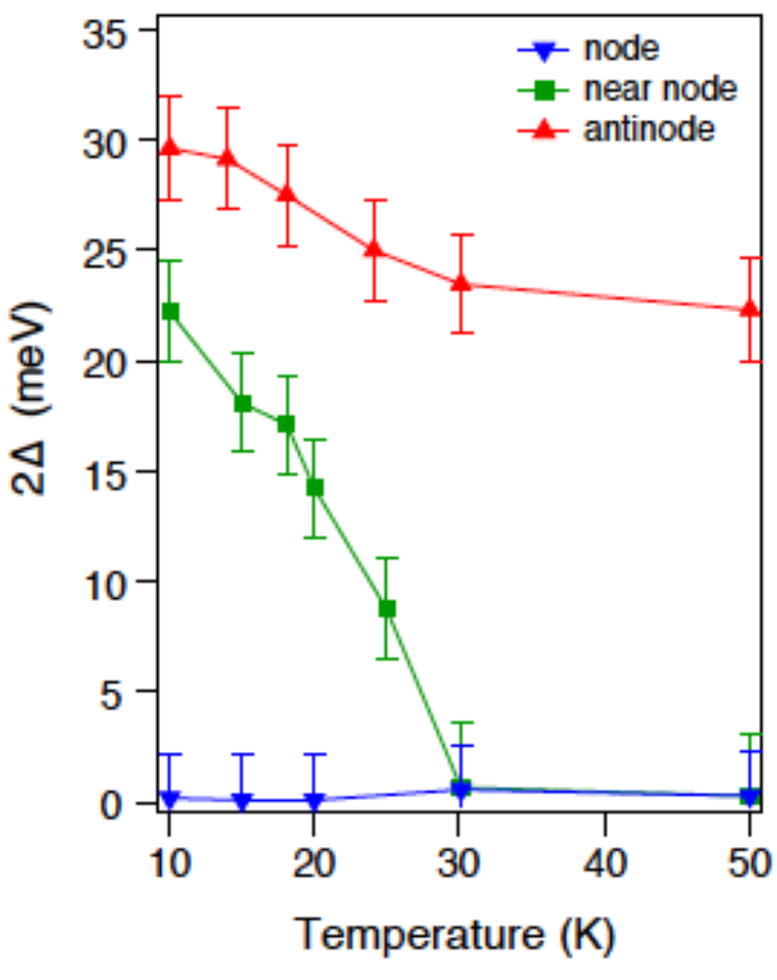
Y. Klein et al. J. Elec. Mat. (2009)

Surface K-doping : ARPES

Kim et al., arxiv (2015)



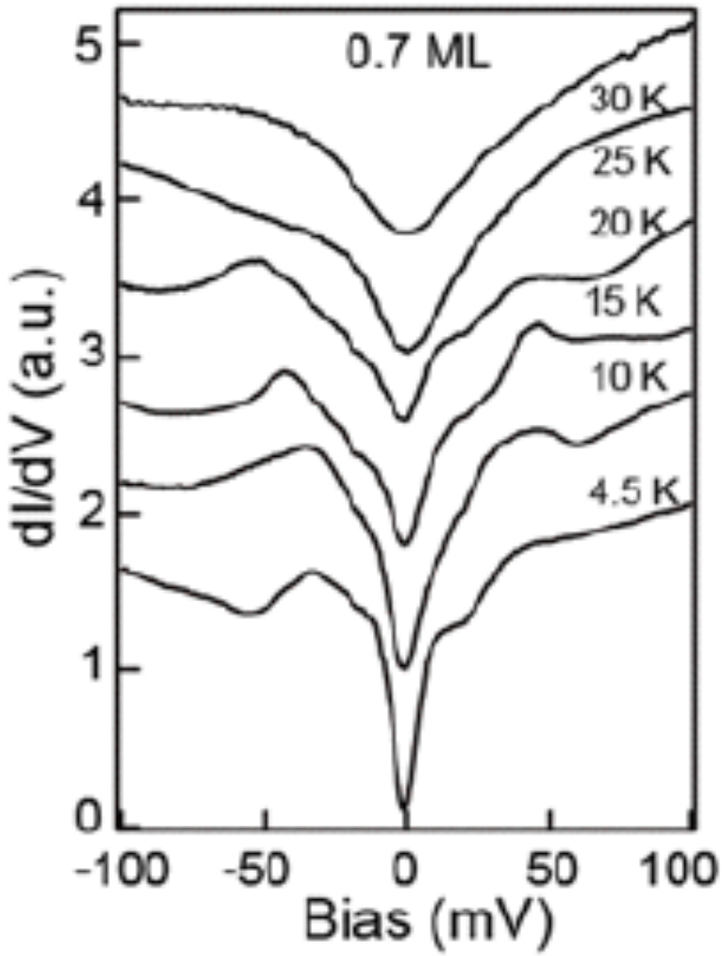




~7% electron-doping

Nodal metal phase opens at 30K

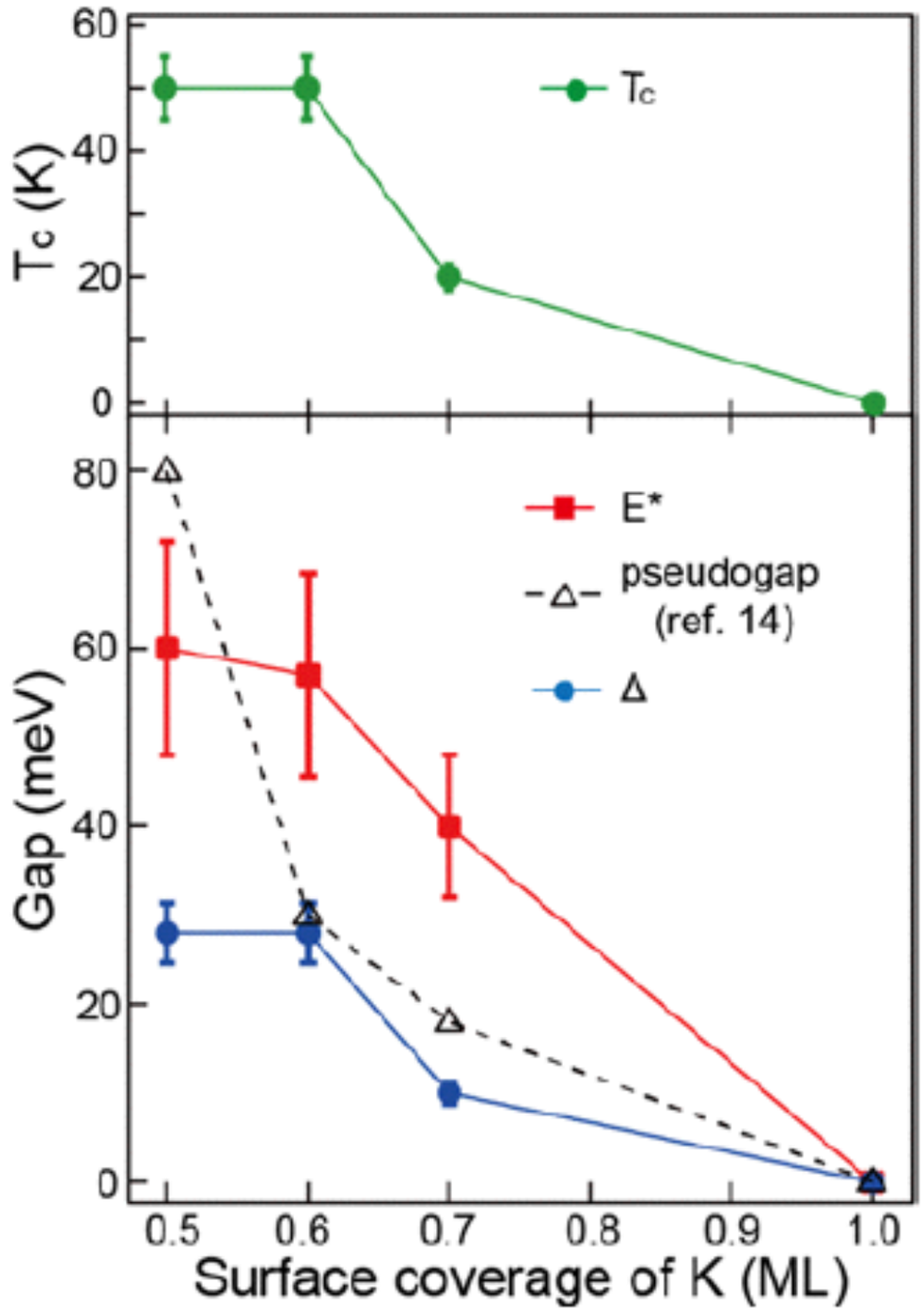
Surface K-doping : STM



~25 meV gap

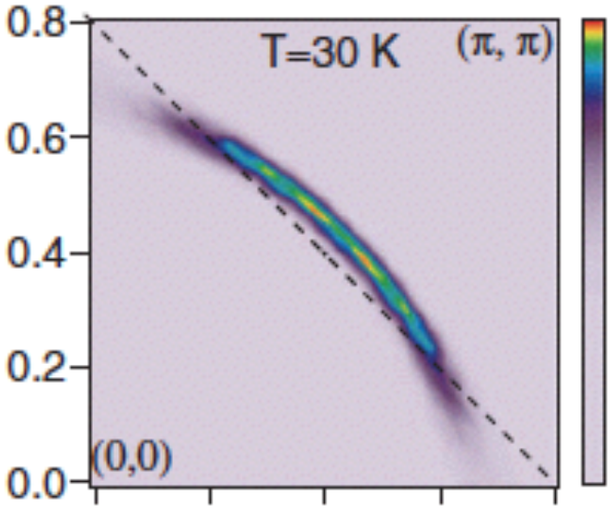
Gap opens at ~50K

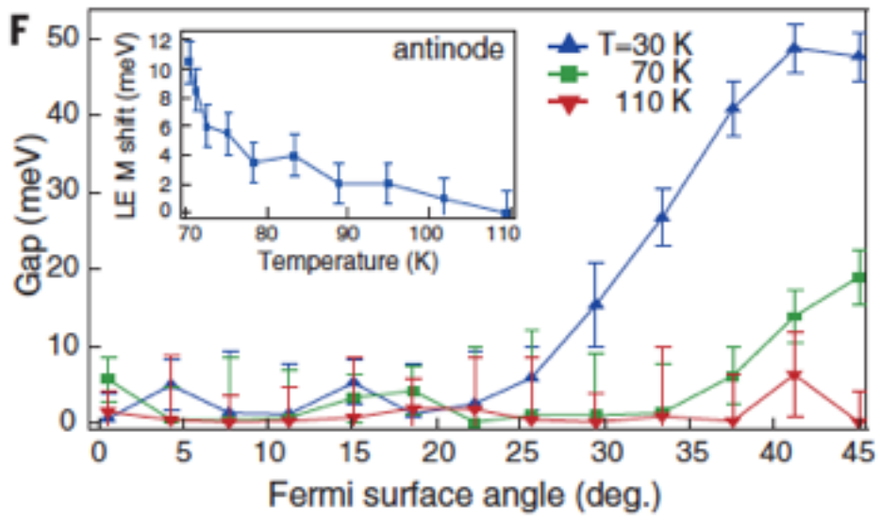
Yan et al., arxiv (2015)



Surface vs. Bulk Doping

Kim et al., Science (2014)



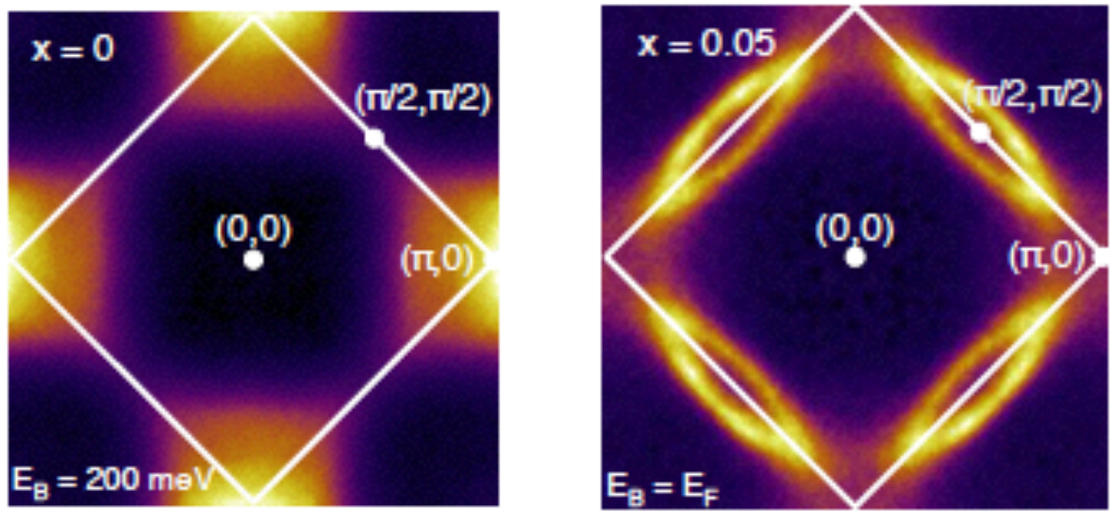


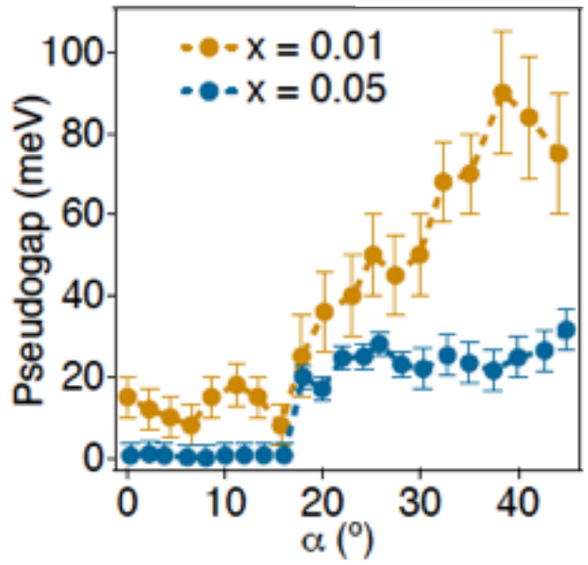
Surface K-doped Sr-214

"Fermi arcs"

PG at antinode vanishes with temperature

de la Torre et al., arxiv (2015)



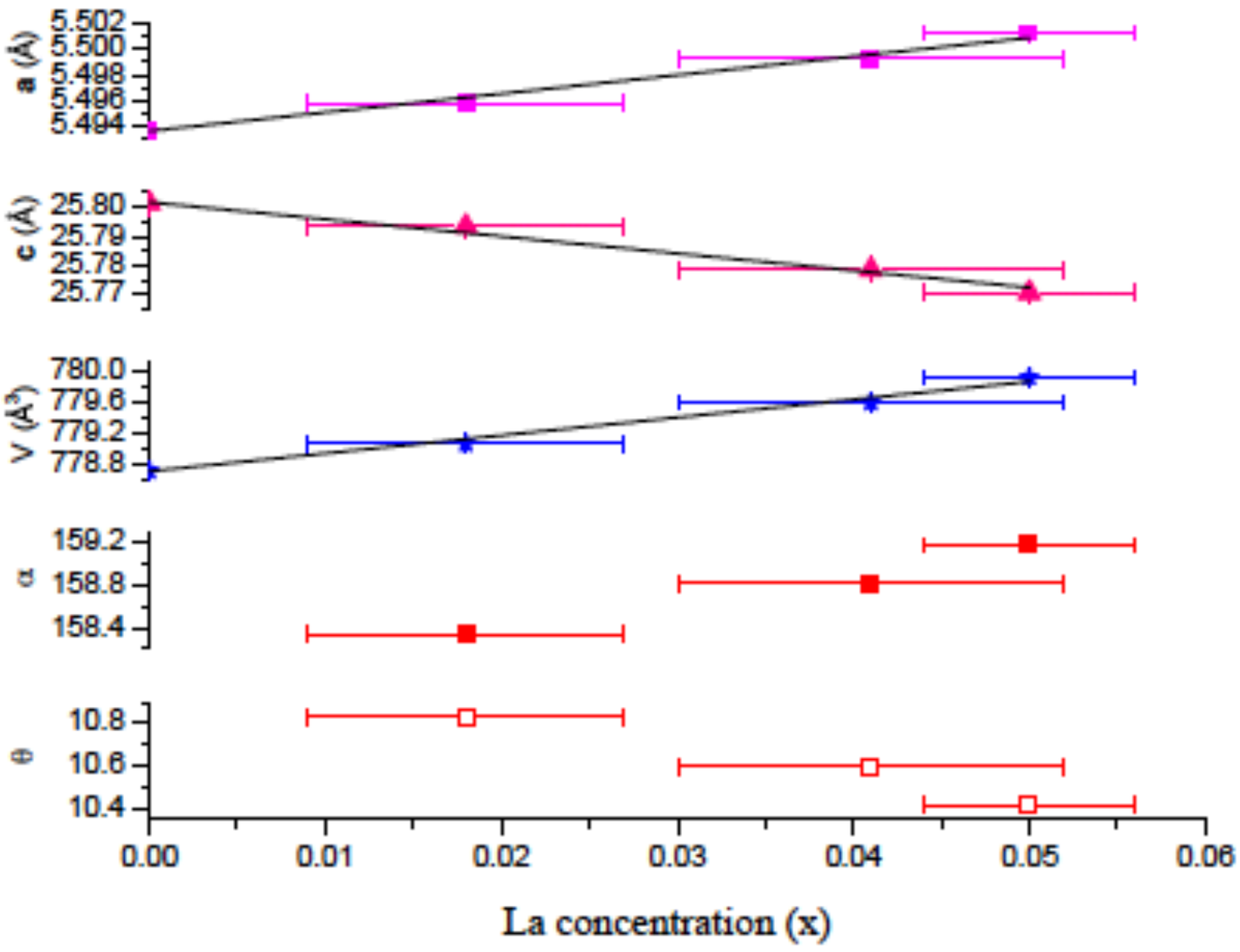


Bulk La-doped Sr-214

Structural back folding into Fermi pockets

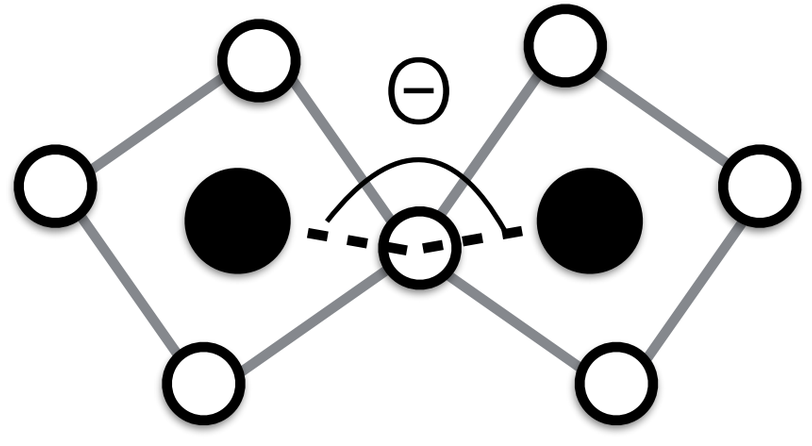
Persistent PG at antinode

Bulk Electron Doping: $(Sr_{1-x}La_x)_2IrO_4$

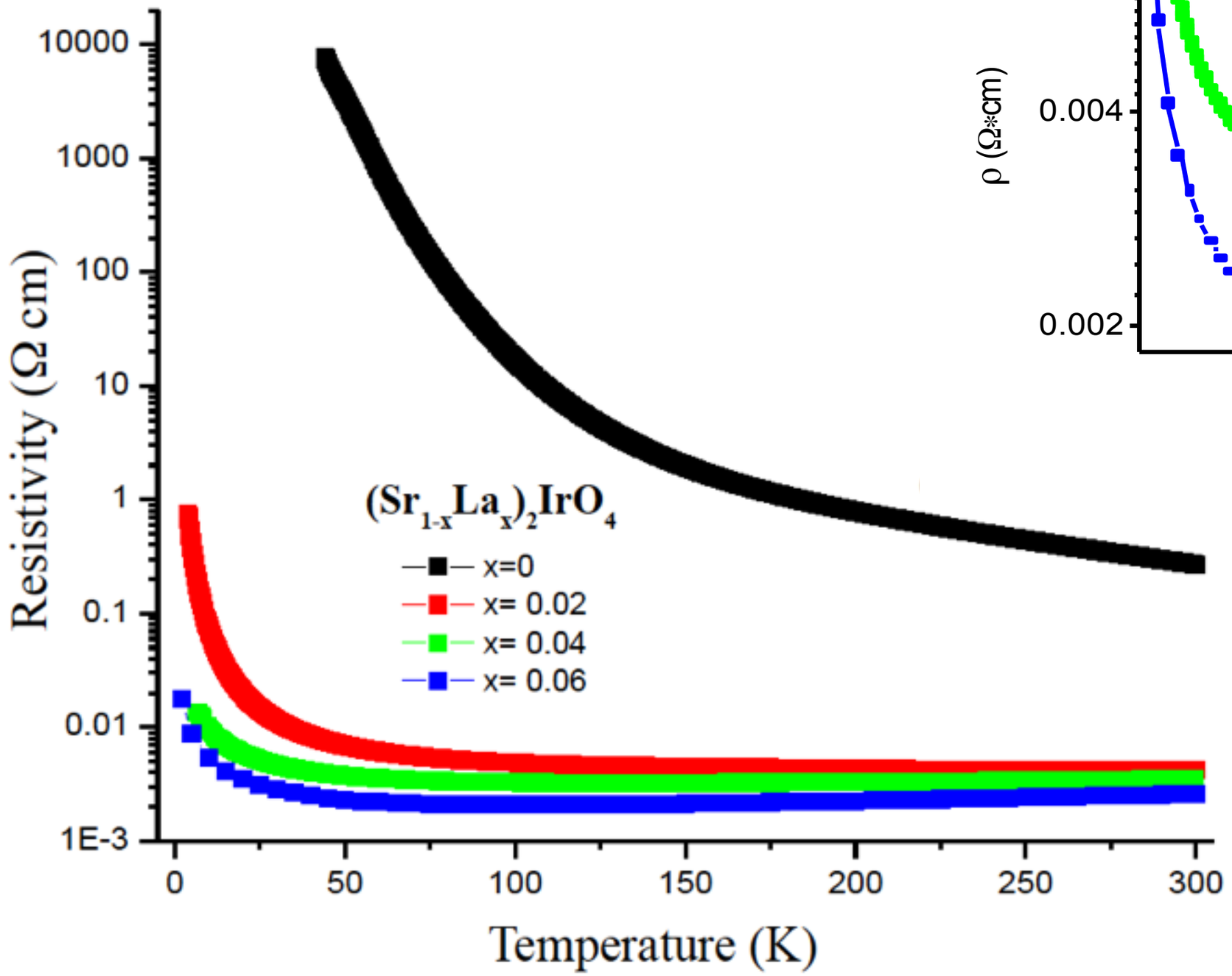


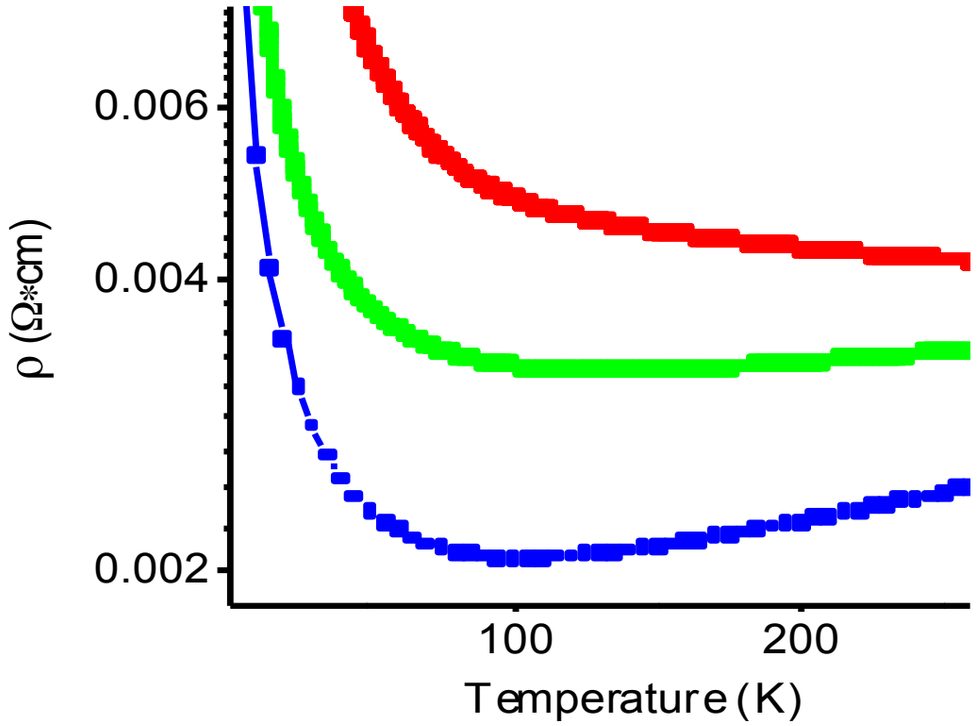
How do structural and electronic symmetries change?

- Easiest way: A-site La substitution
- M. Ge *et al.* PRB (2011)
  - MIT near x=0.04
  - ~8% electrons/Ir
- In-plane Ir-O-Ir bond angle increases
  - Canting slightly relaxed



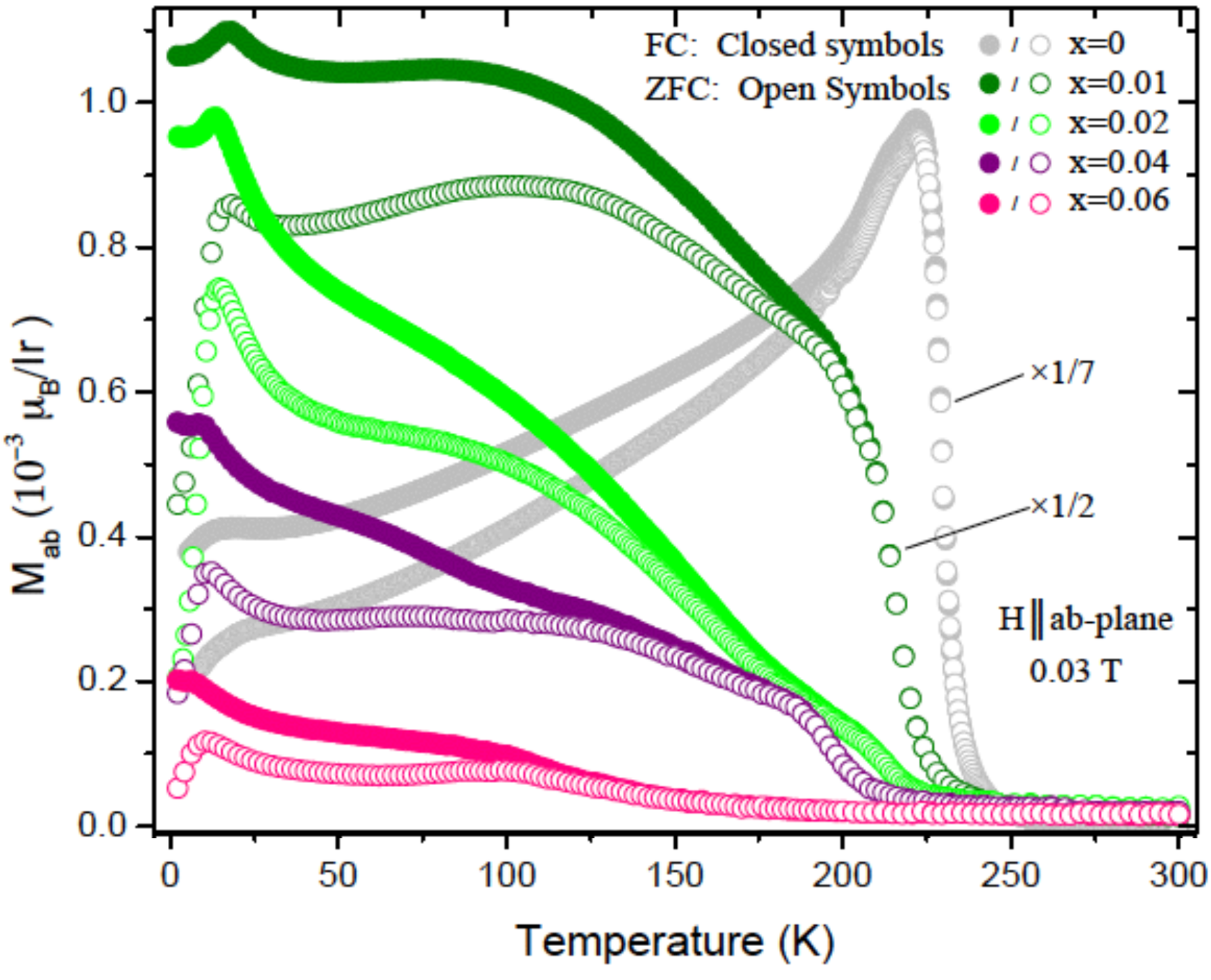
Charge Transport in La-doped Sr-214





- Immediate drop in 300K resistivity
- Persistent low-T upturn in $\boldsymbol{\rho}$(T)
- High-T metallic state for $x \geq 0.04$

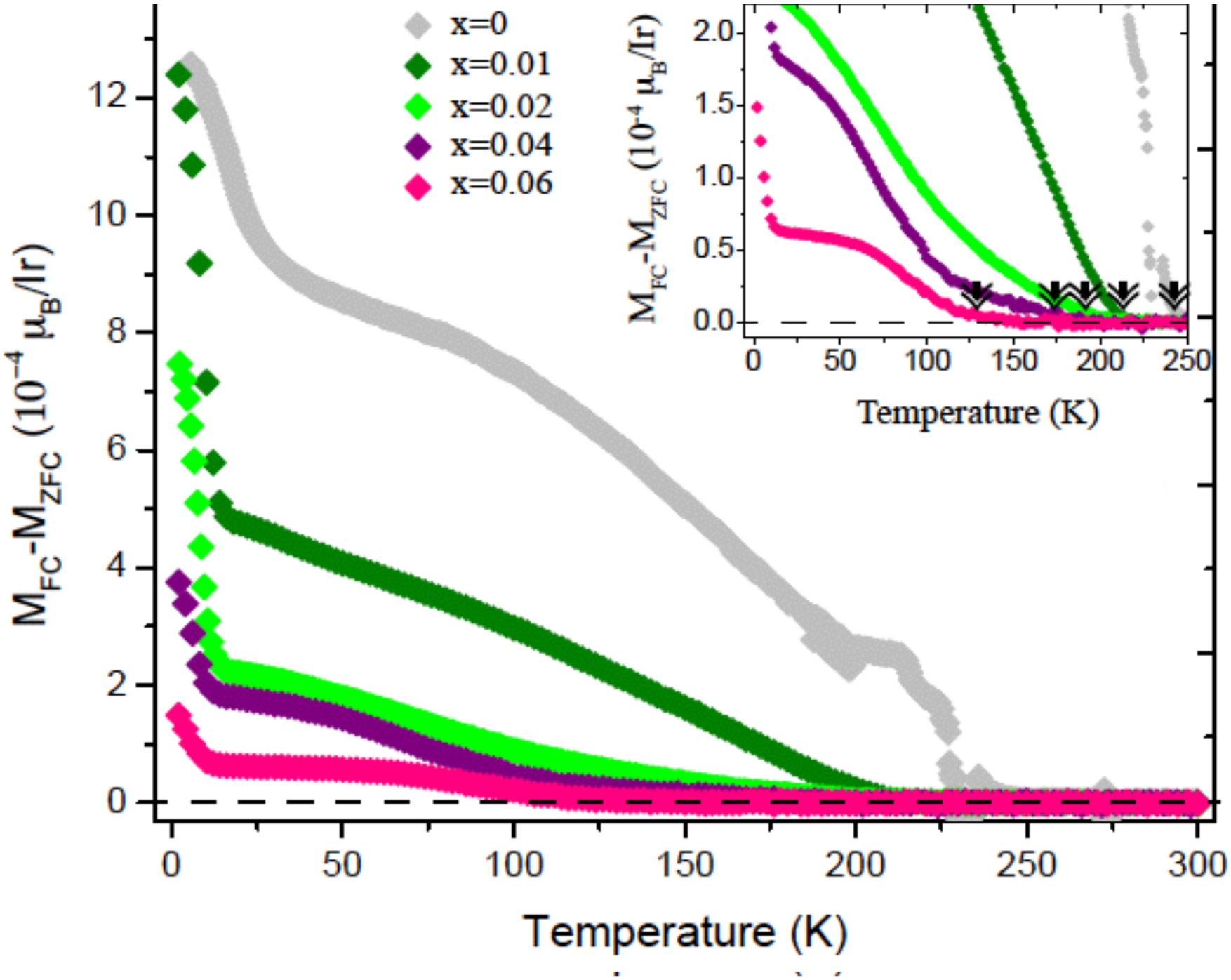
DC Magnetization



X. Chen et al. (in review)

- Two features in irreversibiltiy
  - T_{AF}: CAF
  - T_F: SG
- Both suppressed with La-doping
  - T_{AF} and T_G survive at highest doping

DC Magnetization



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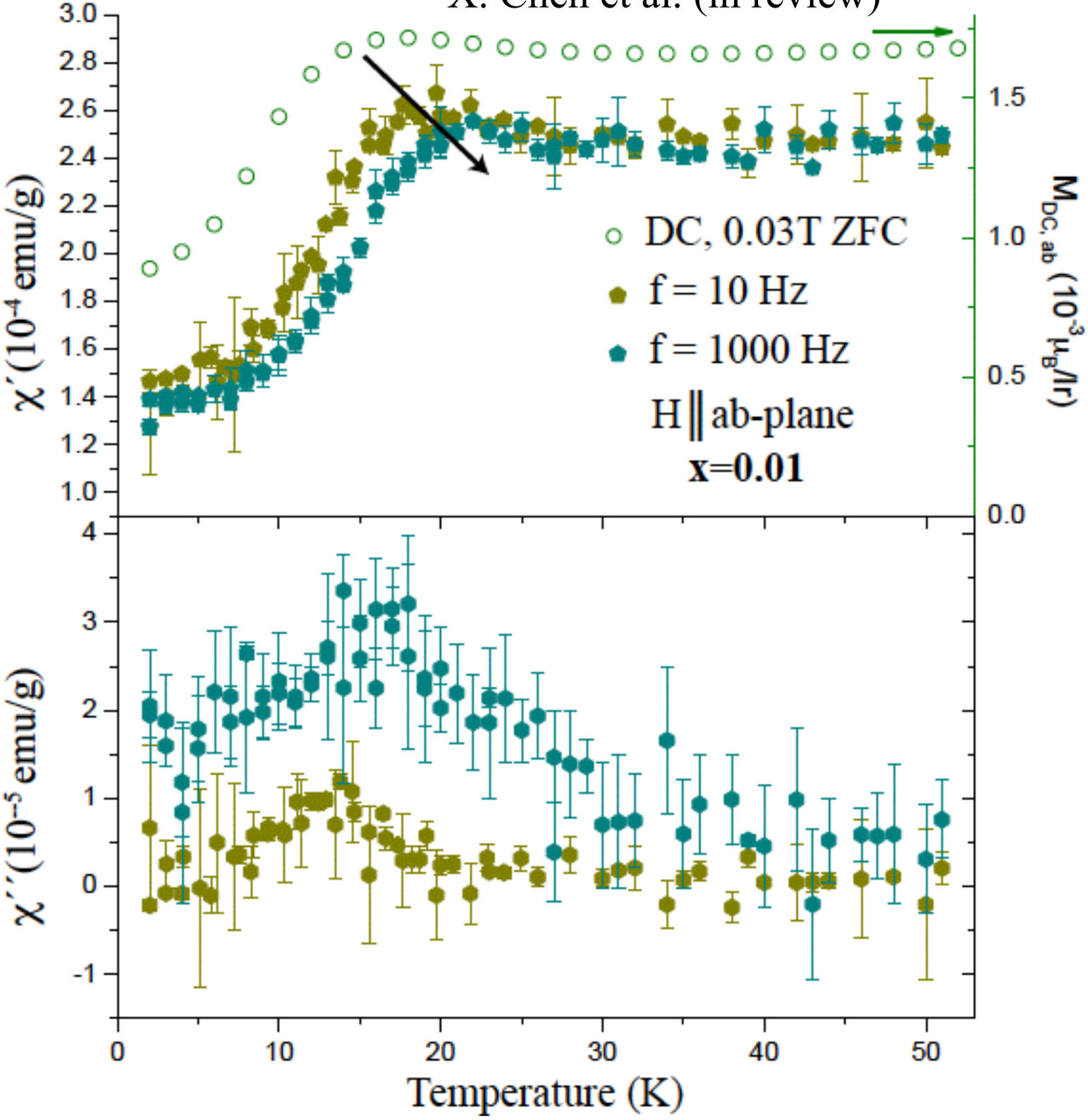
X. Chen et al. (in review)

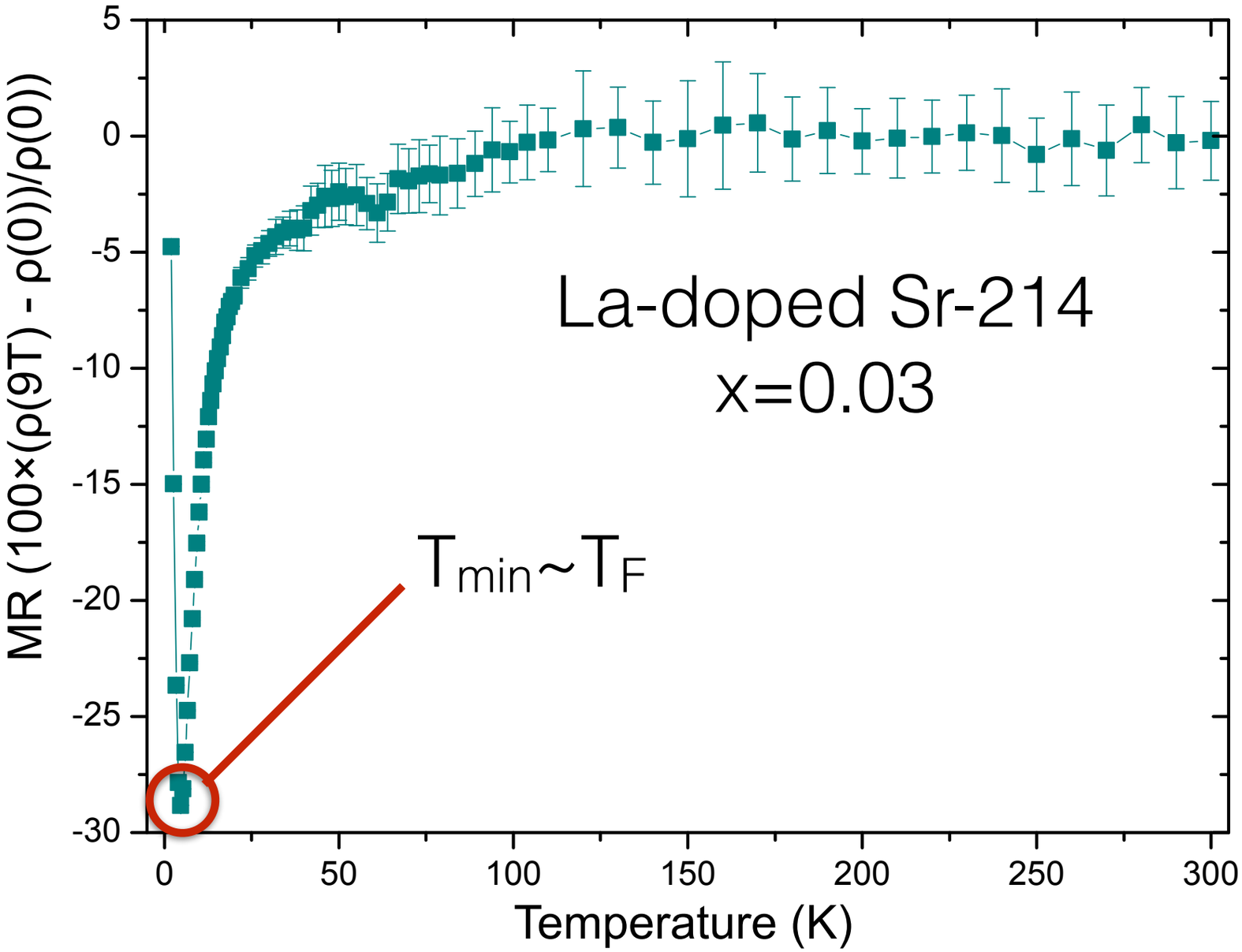
Origin of T_F

X. Chen et al. (in review)



- Frequency dependent shift in T_F cusp
- Accompanying peak in disspation
- Consistent with low-T SG state

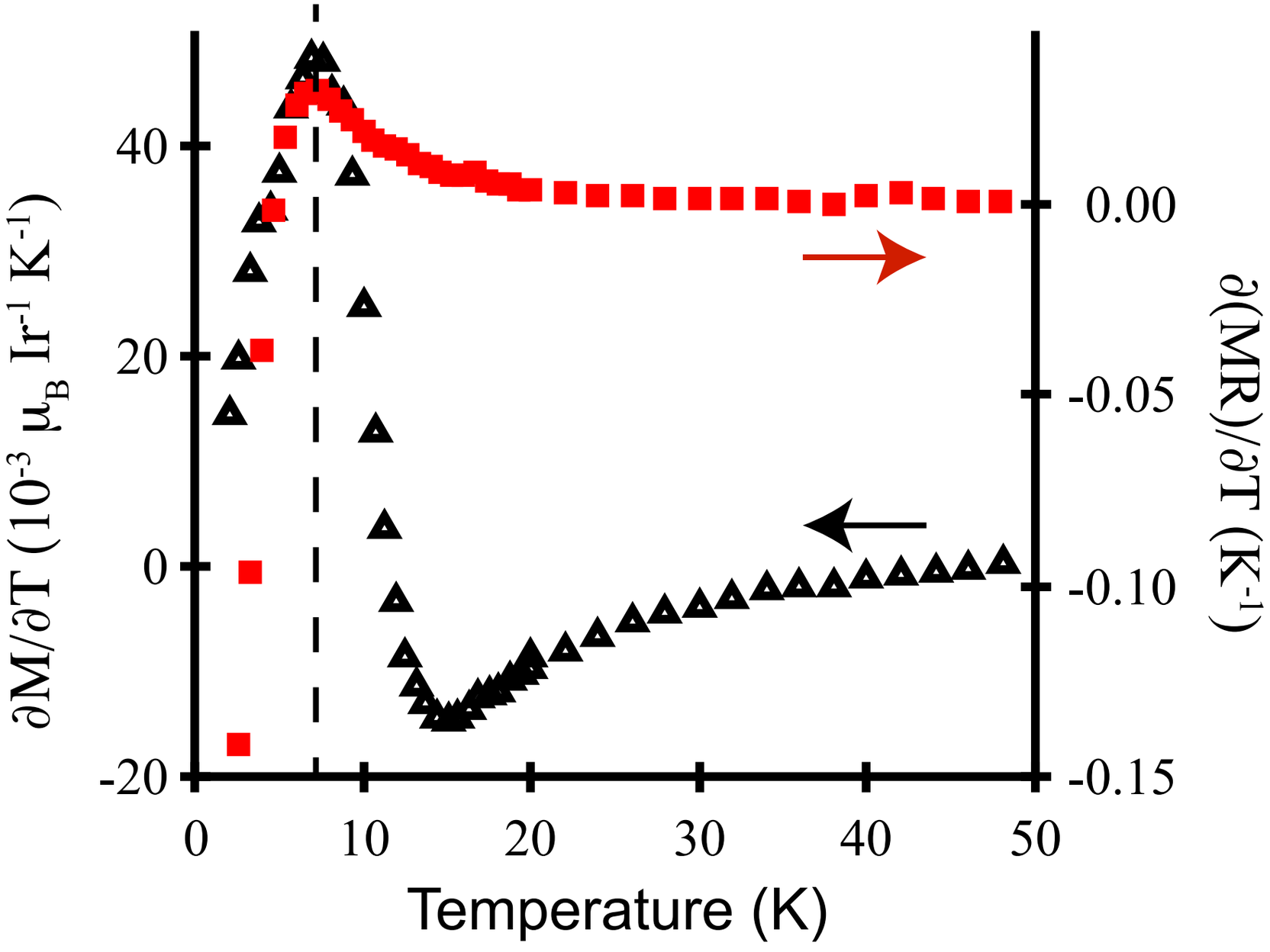
MR in lightly doped Sr-214



- Transverse MR (H,I||ab-plane)
- Charge couples strongly to T_G

X. Chen et al. (in review)

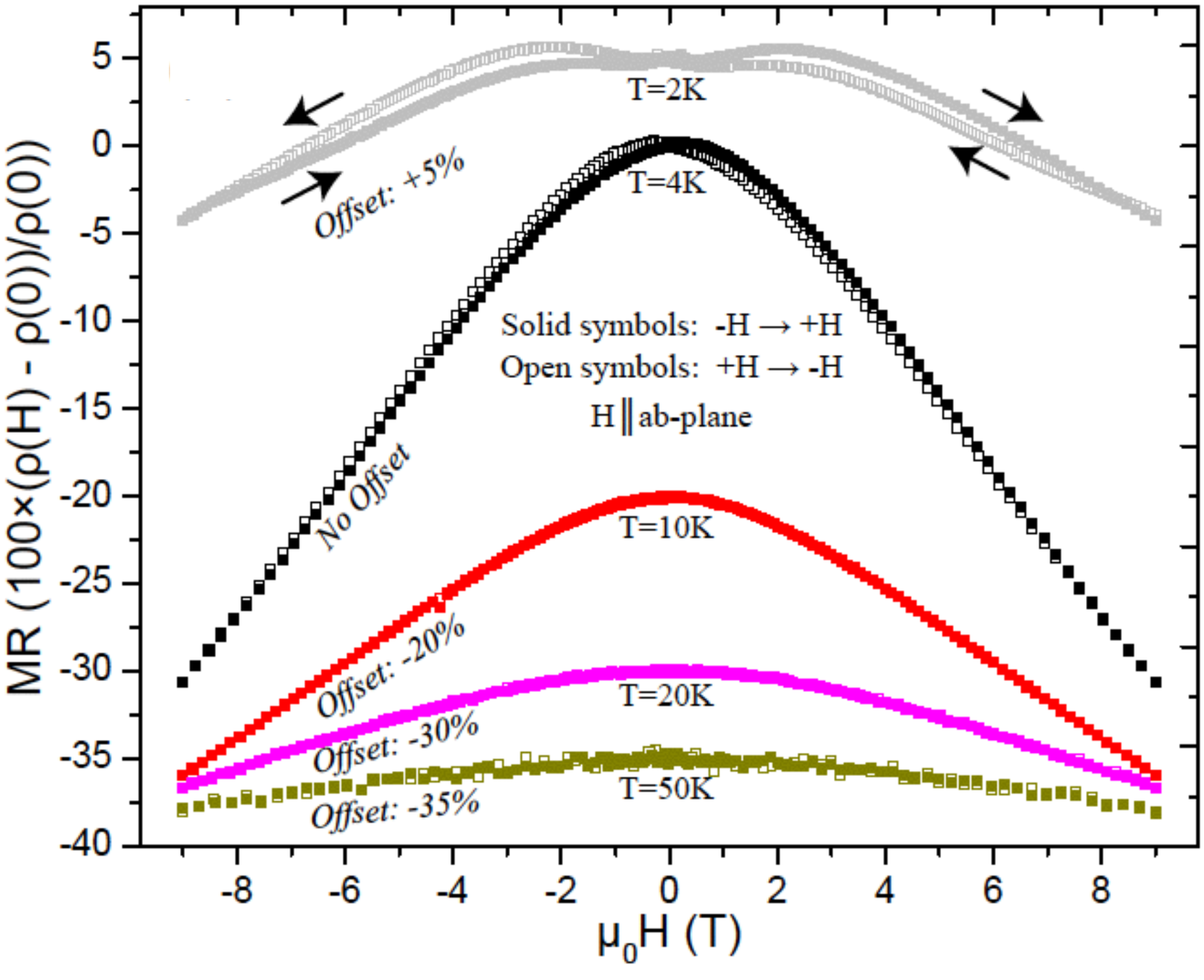
MR in lightly doped Sr-214



- Transverse MR (H,I||ab-plane)
- Charge couples strongly to T_G

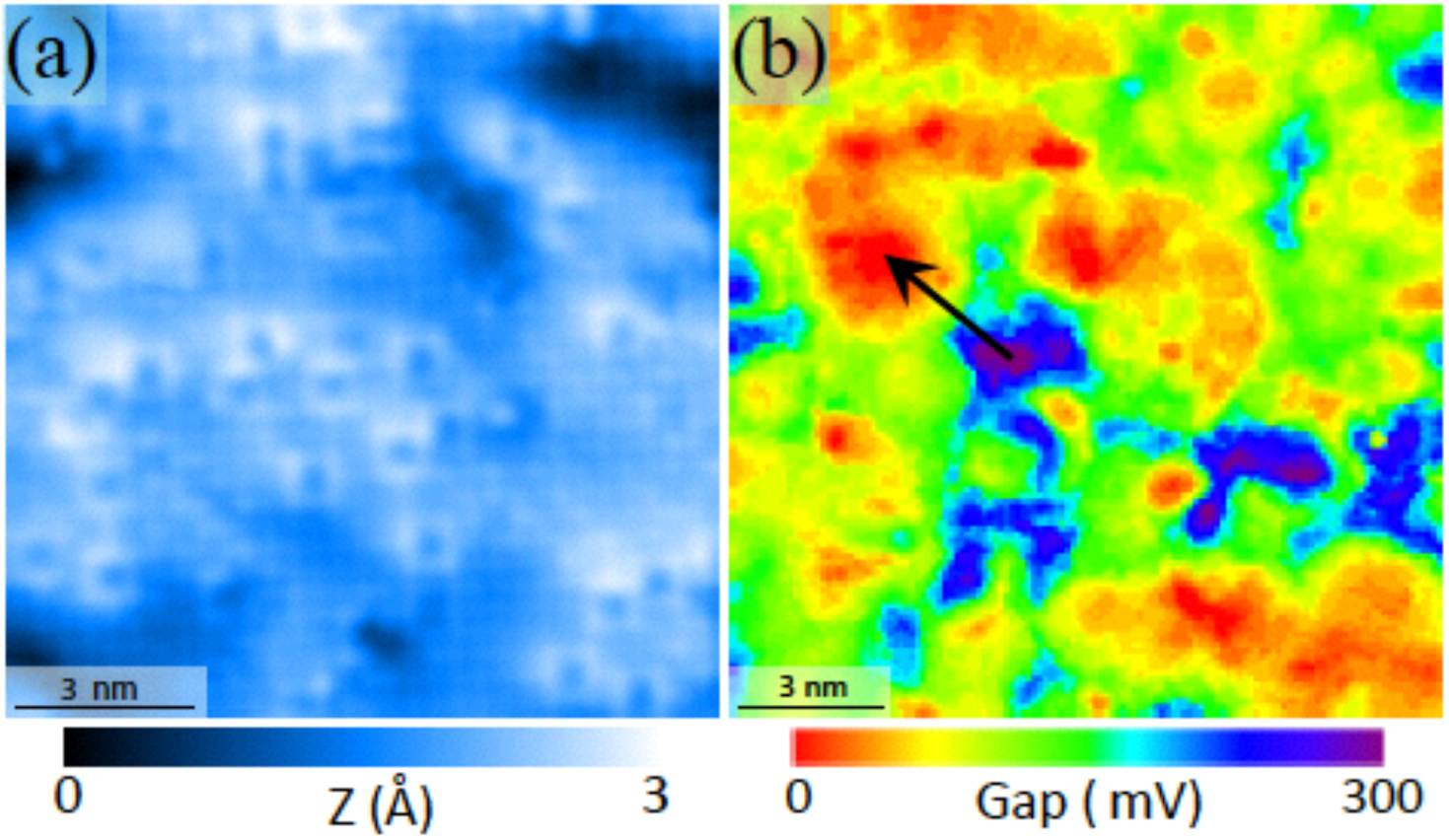
X. Chen et al. (in review)

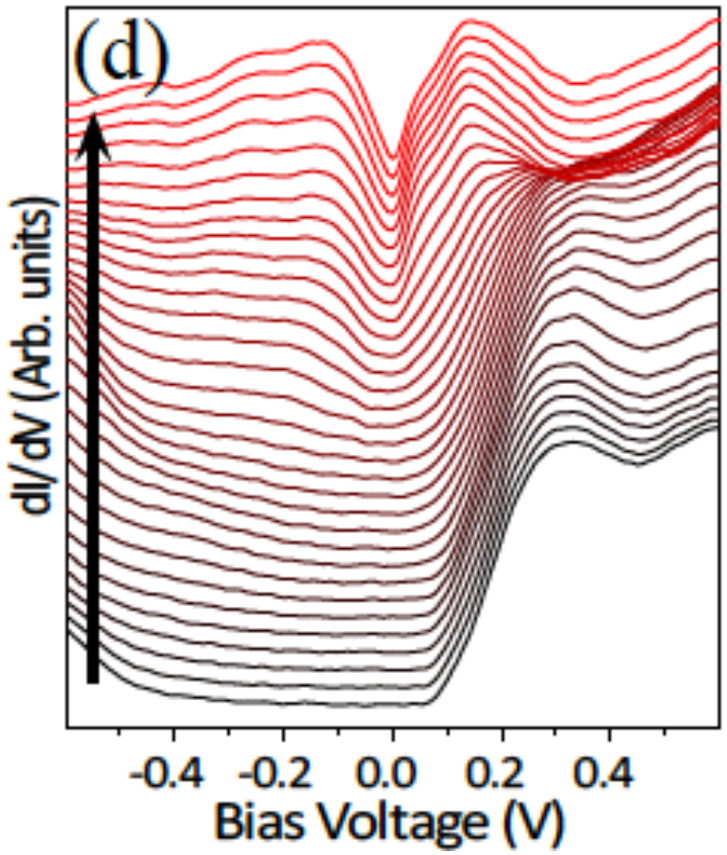
MR in La-doped Sr-214 cont.



- Transverse MR (H,I||ab-plane)
- For $T<T_F$:
  - Low-field MR turns positive
  - Hysteresis in MR
- Seen in SG systems with coexisting FM
  - NiMn alloys: Low-field "unlocks" spins from frozen state
- Also in PS TMOs: hole-doped $La_{1-x}Sr_xCoO_3$

Persistent Nanoscale Phase separation

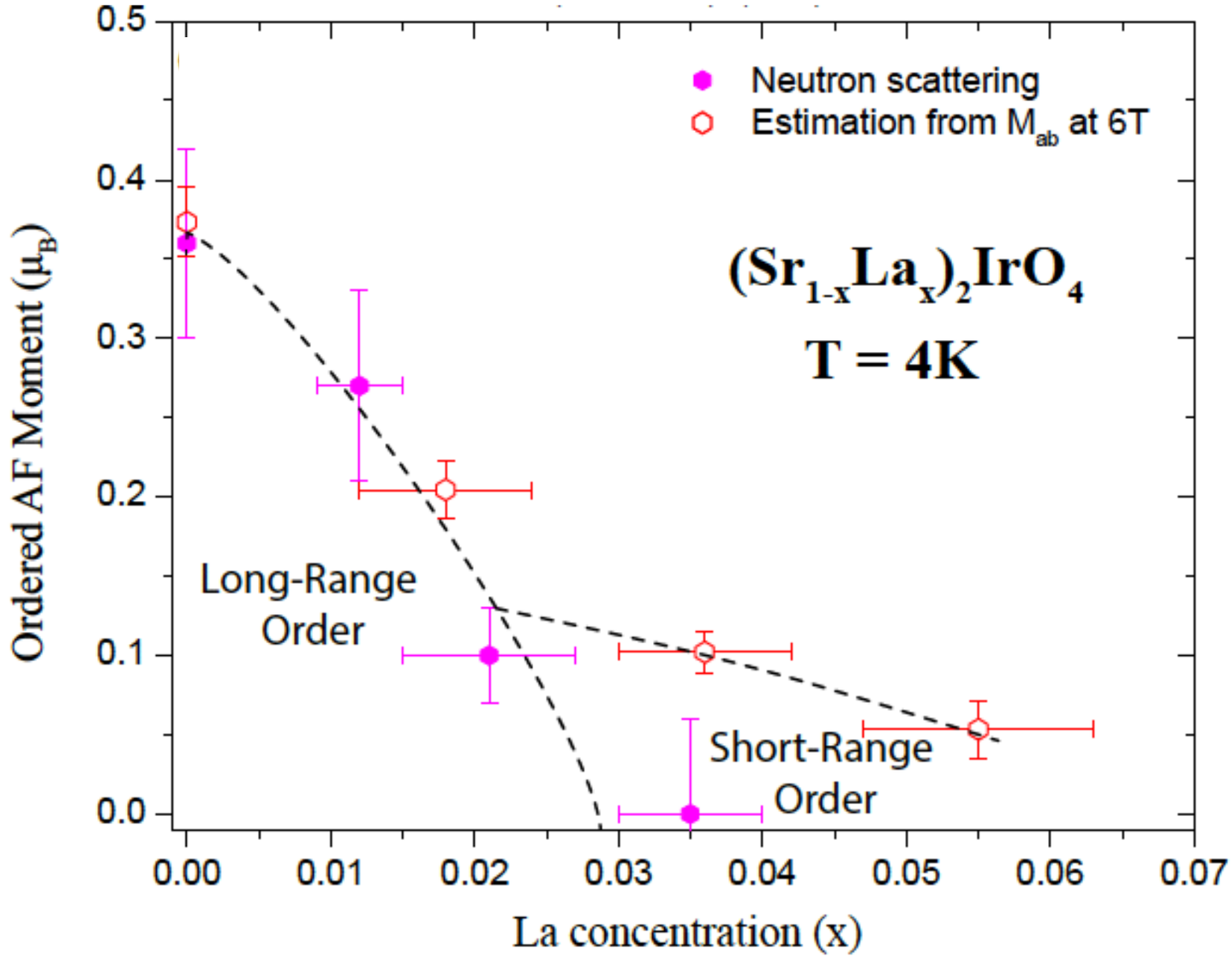




- Near limit of La-substitution (x=0.05)
  - Gapped and metallic regions coexist
  - Mott state not completely destabilized

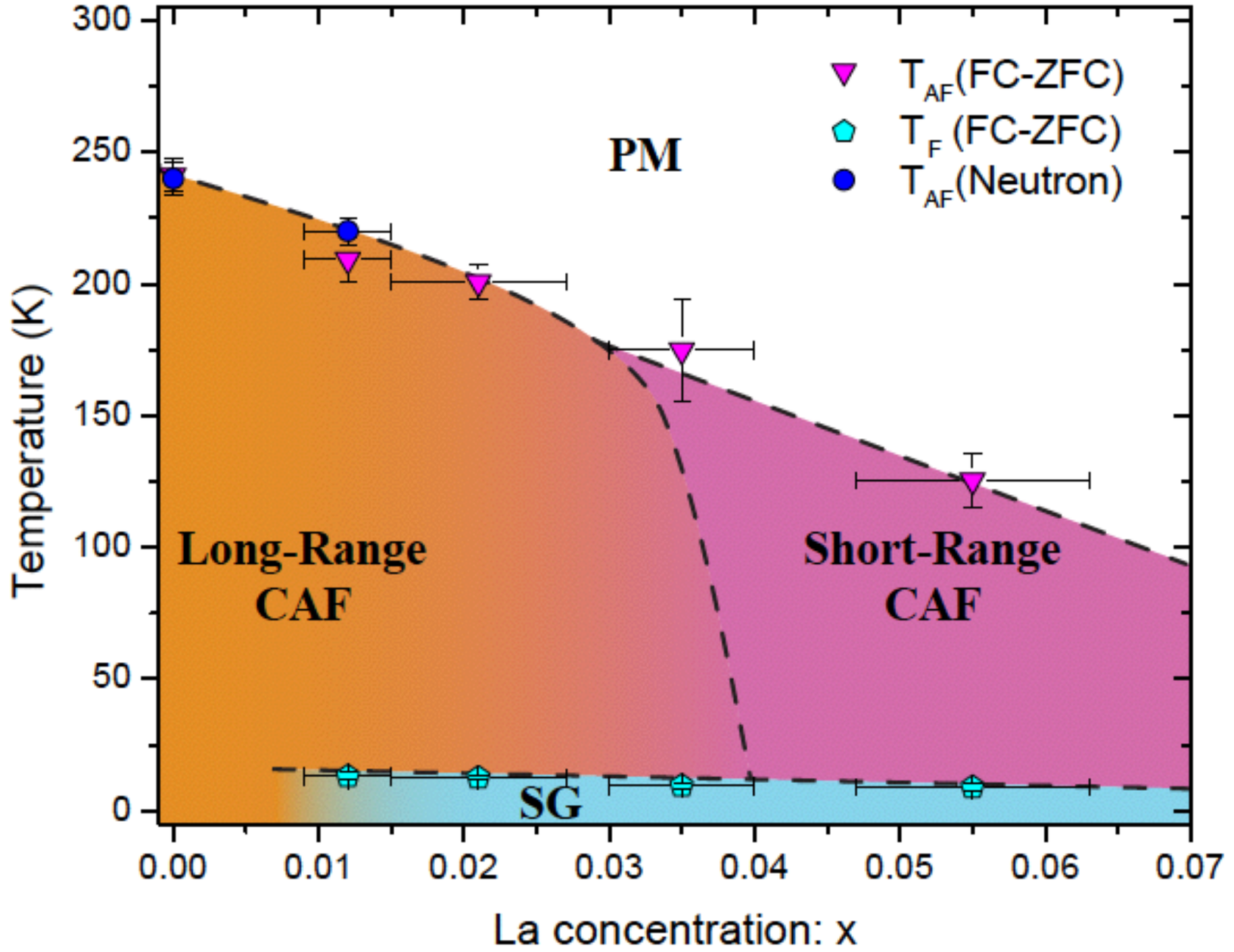
Transport is percolative

Collapse of L.R. AF order



- Long-range AF order collapses near x=0.03
- Short-range static order remains up to doping limit
  - Likely inside of local insulating clusters

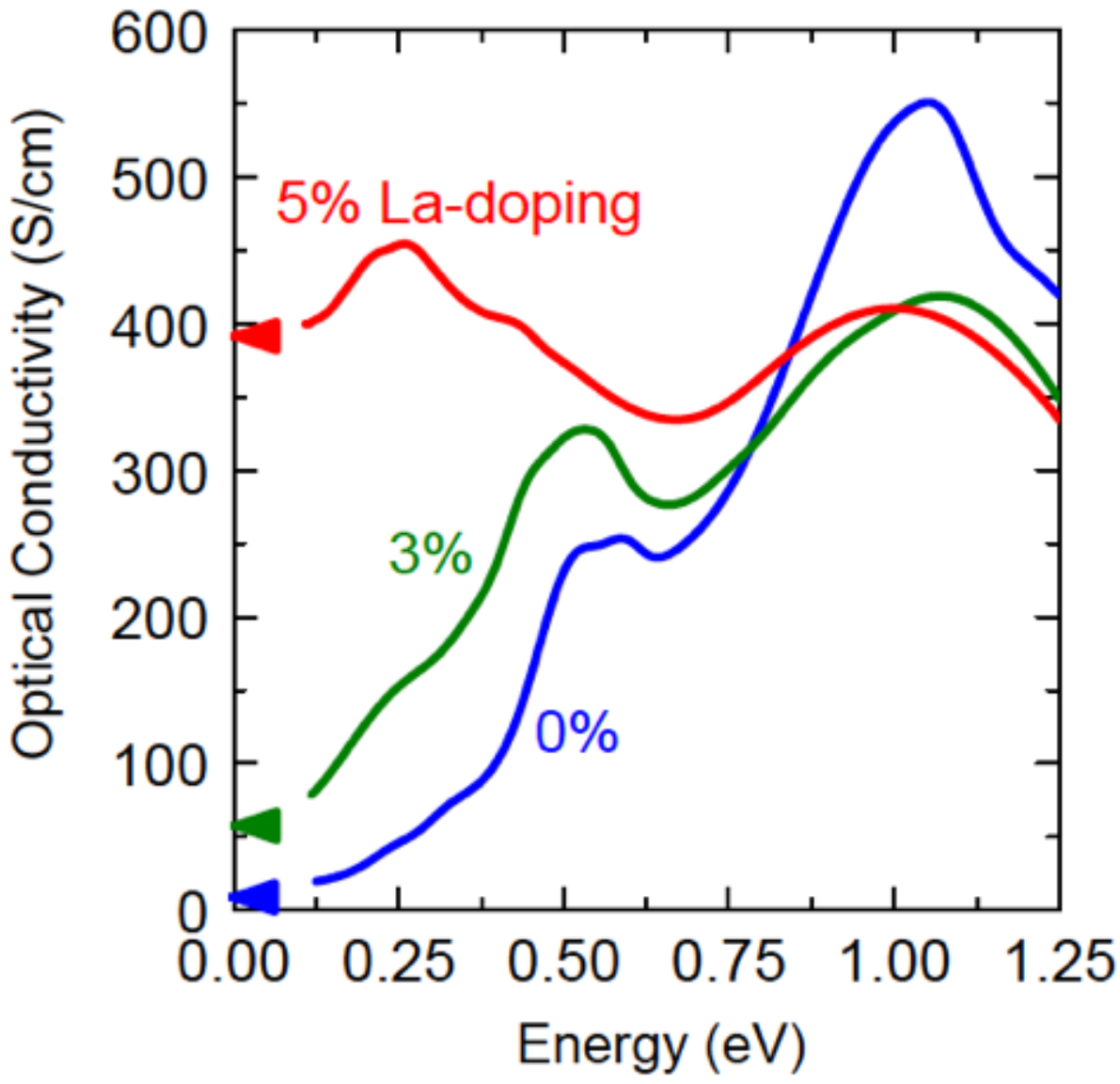
Collapse of L.R. AF order



- Long-range AF order collapses near x=0.03
- Short-range static order remains up to doping limit
  - Likely inside of local insulating clusters

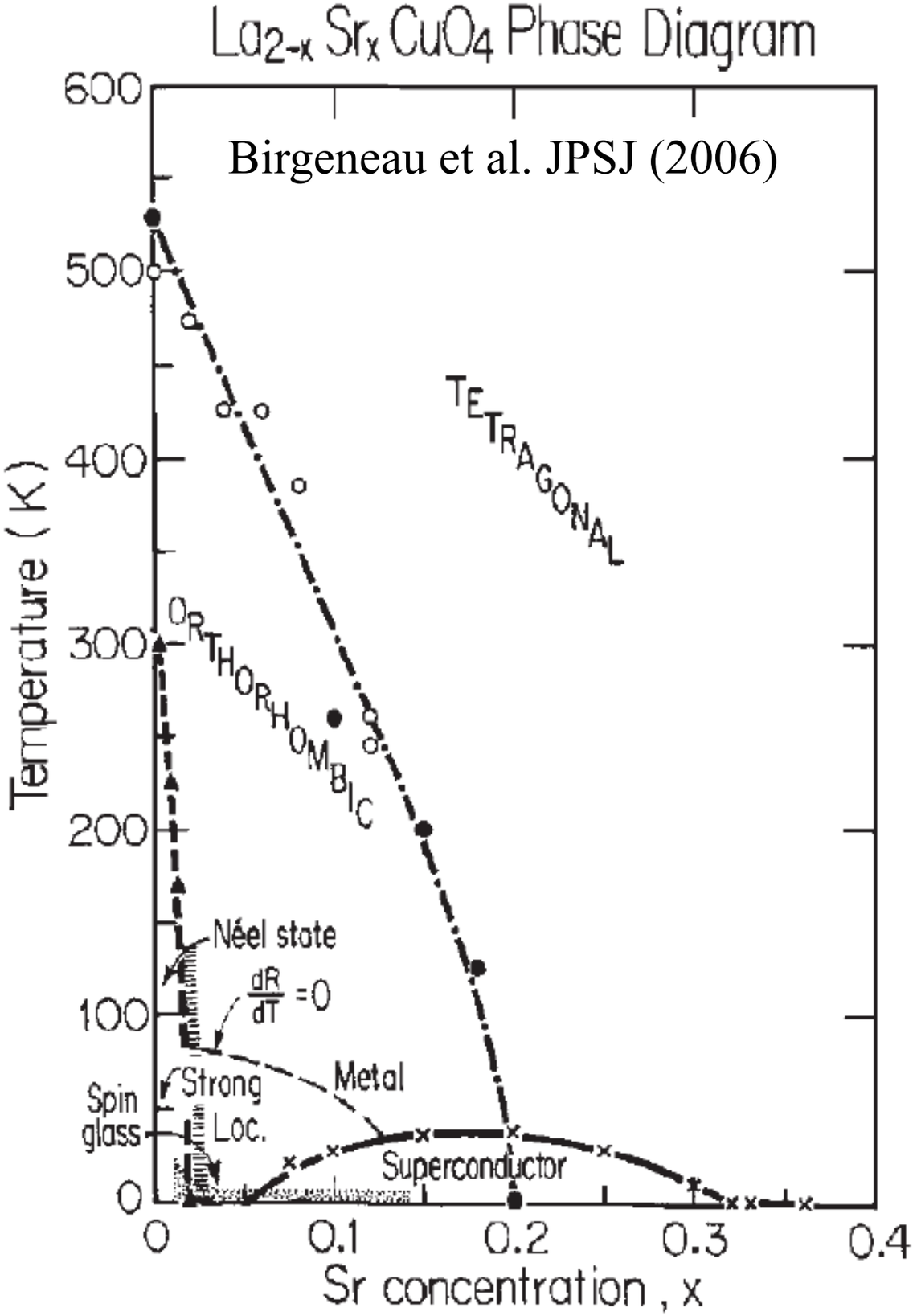
Takeaway: La-doped Sr-214

- ~6% La doping not enough to globally quench Mott state
  - ~12% electrons/Ir
- Nanoscale phase separation between gapped AF clusters and percolating metallic network
  - Consistent with recent ARPES work Brouet et al., arxiv (2015)
- Long-range AF order vanishes at 5-6% electrons/Ir
- Glass-like spin freezing below T_F appears with La-doping



A. Hauser, J. Allen (unpublished)

Hole-doped 214 Cuprates vs 214 Electron-doped Iridates



- Common features:
  - Rapid collapse of LR AF order
  - Electronic phase separation under light doping
  - Intermediate SG phase
- Differences:
  - Mott state more robust to doping in n-type Sr-214
  - Persistent SR AF order

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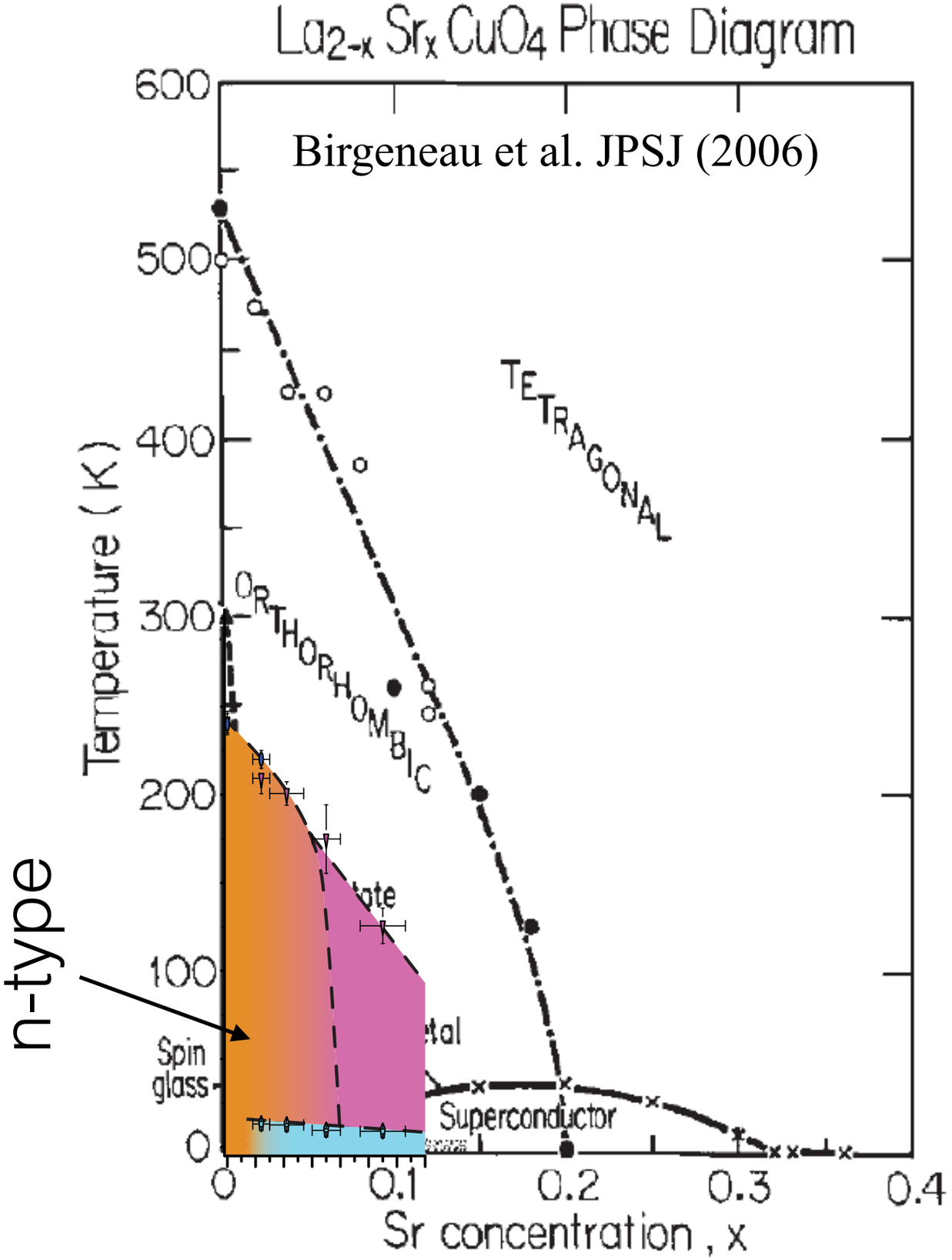

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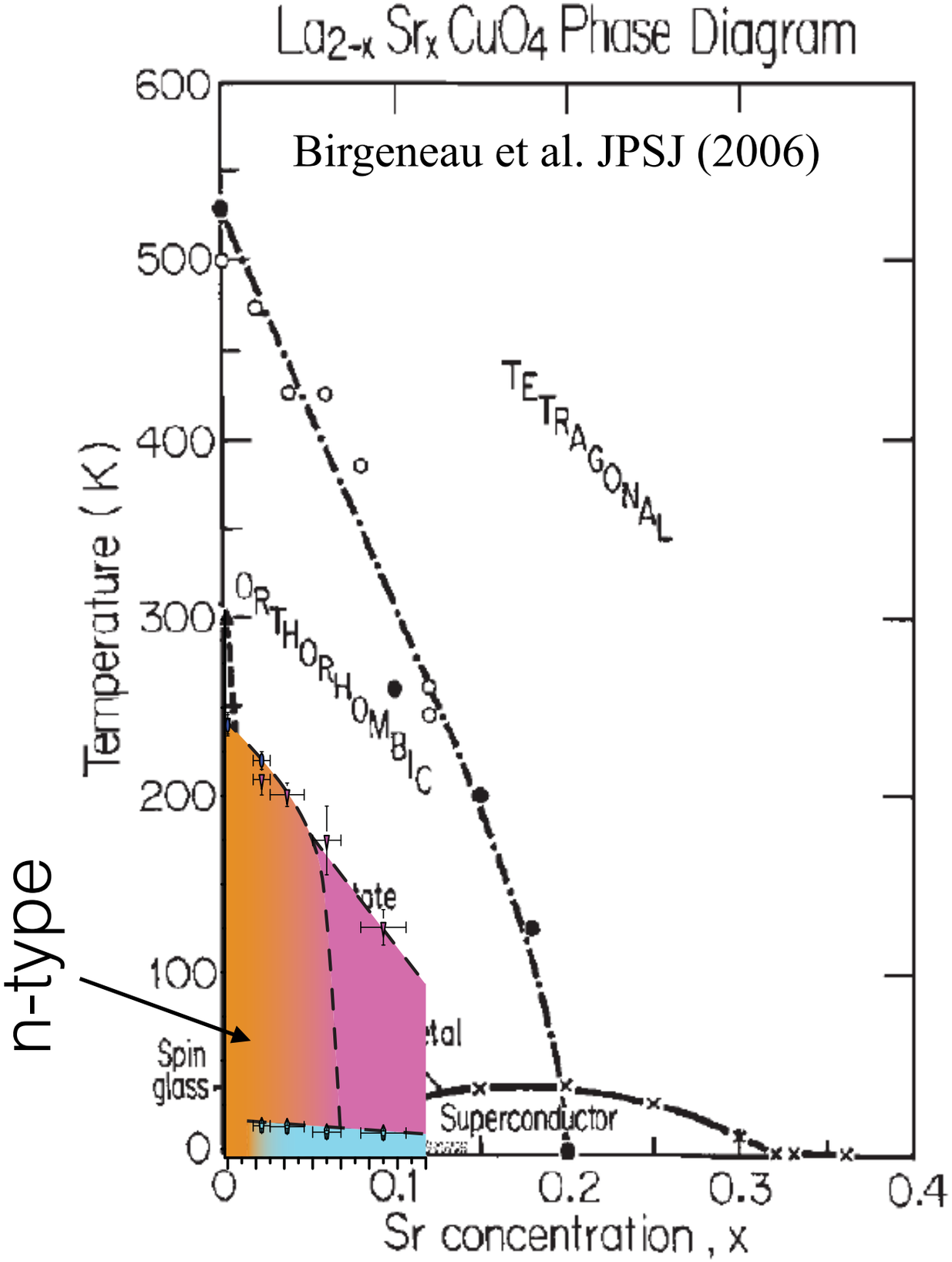
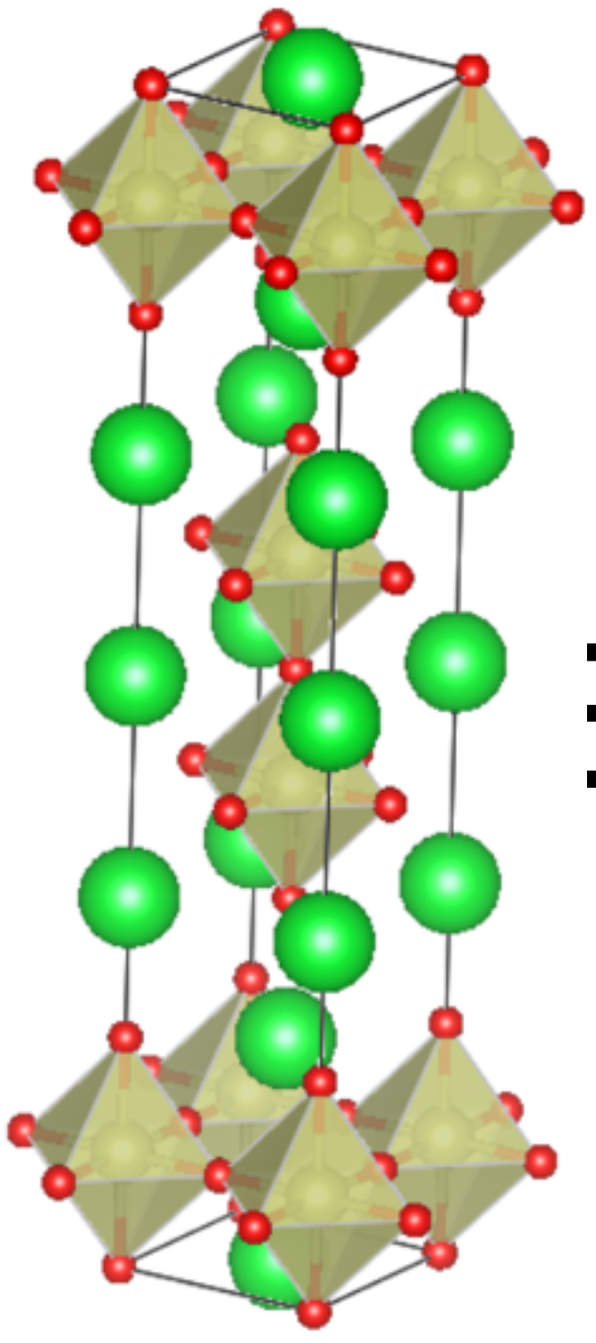


We need more doping!

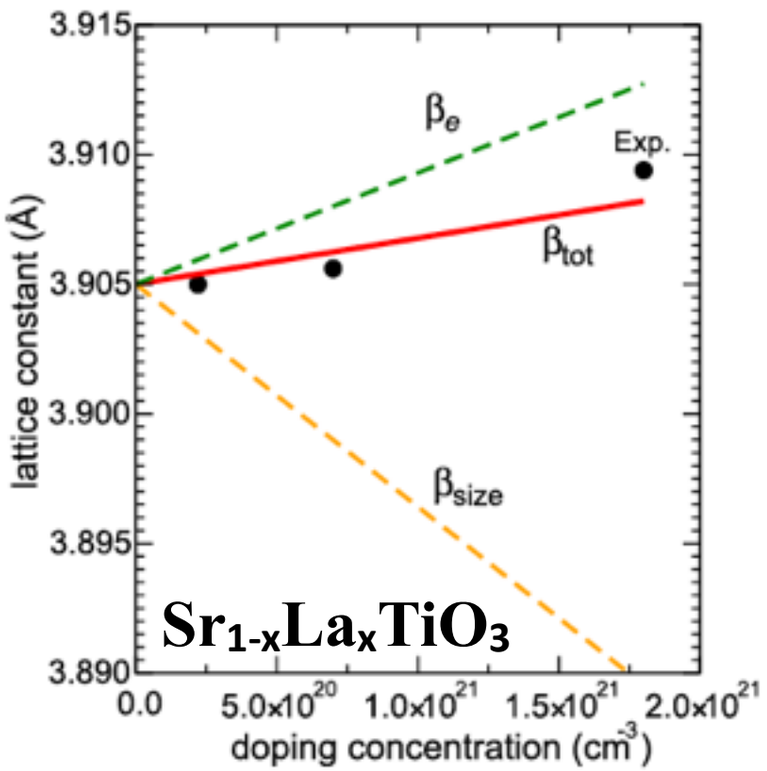


Electron-doped Sr-327

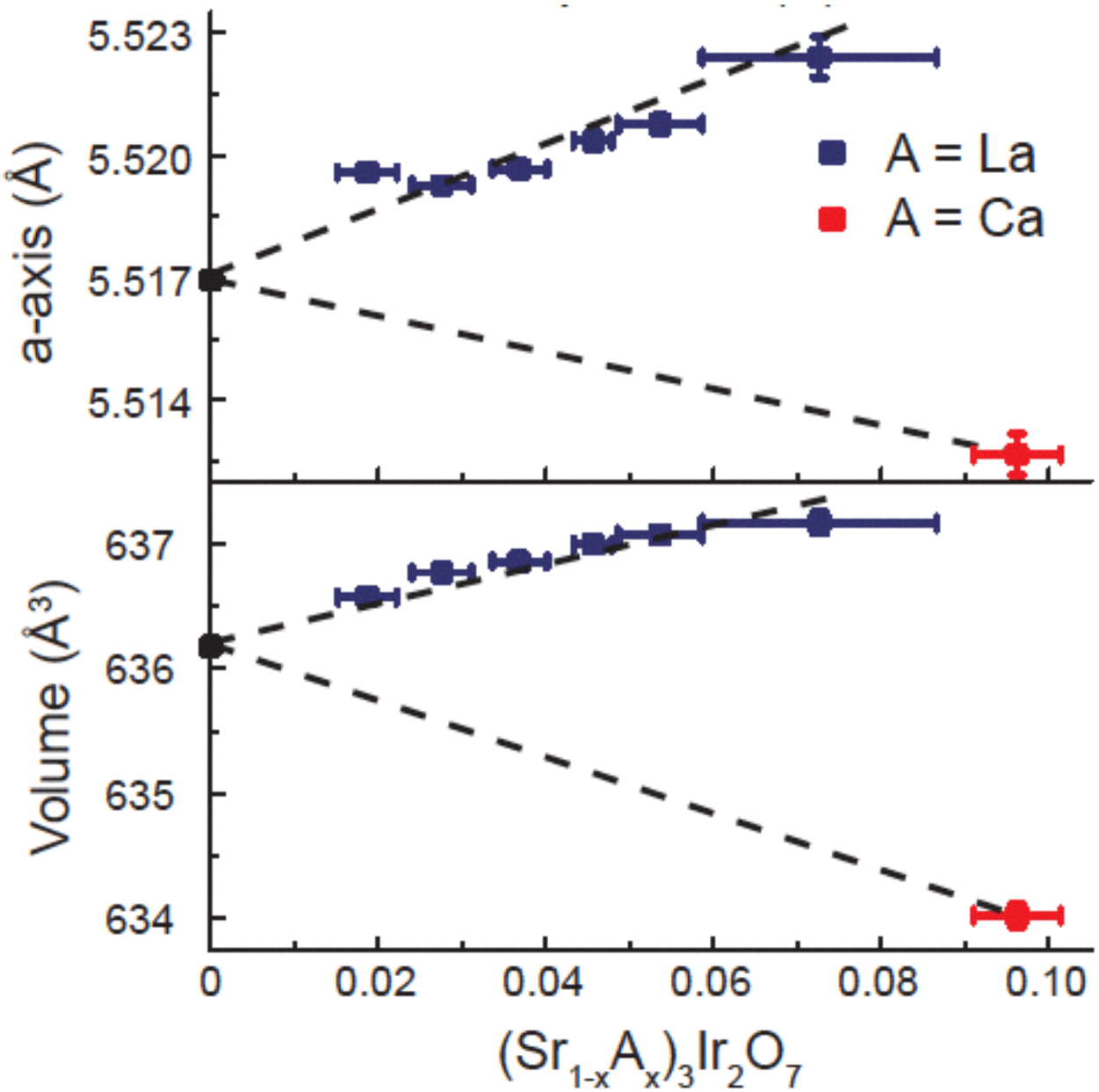
Bulk Electron Doping: $(Sr_{1-x}La_x)_3Ir_2O_7$

- 300K lattice expands with La substitution
  - Electronic deformation dominates steric effects
- Similar to La-doped $SrTiO_3$

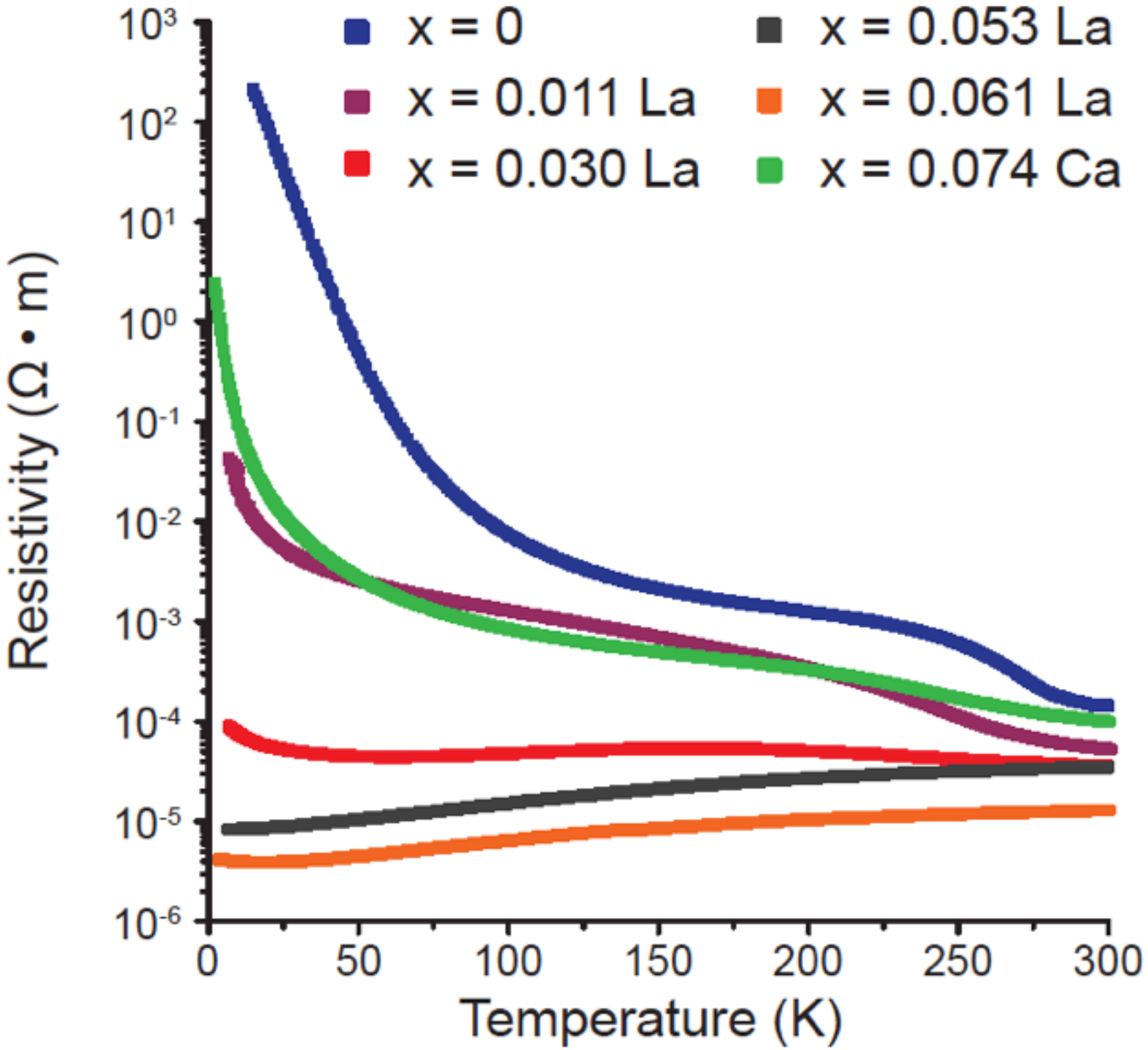
A. Janotti et al. APL (2012)



T. Hogan et al. PRL (2015)



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

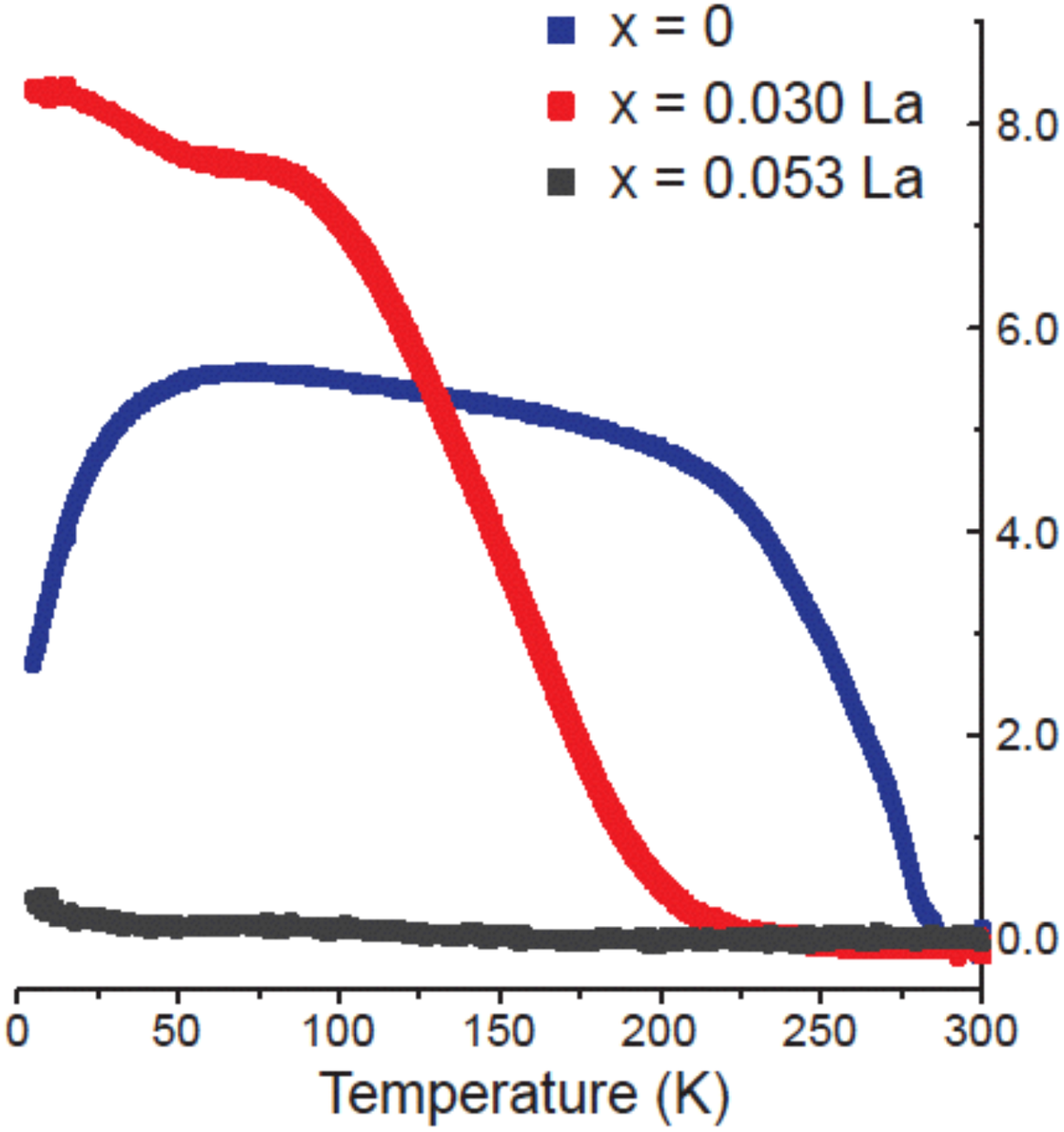


T. Hogan et al. PRL (2015)

- MIT near x~0.04
- Consistent with Li *et al.*

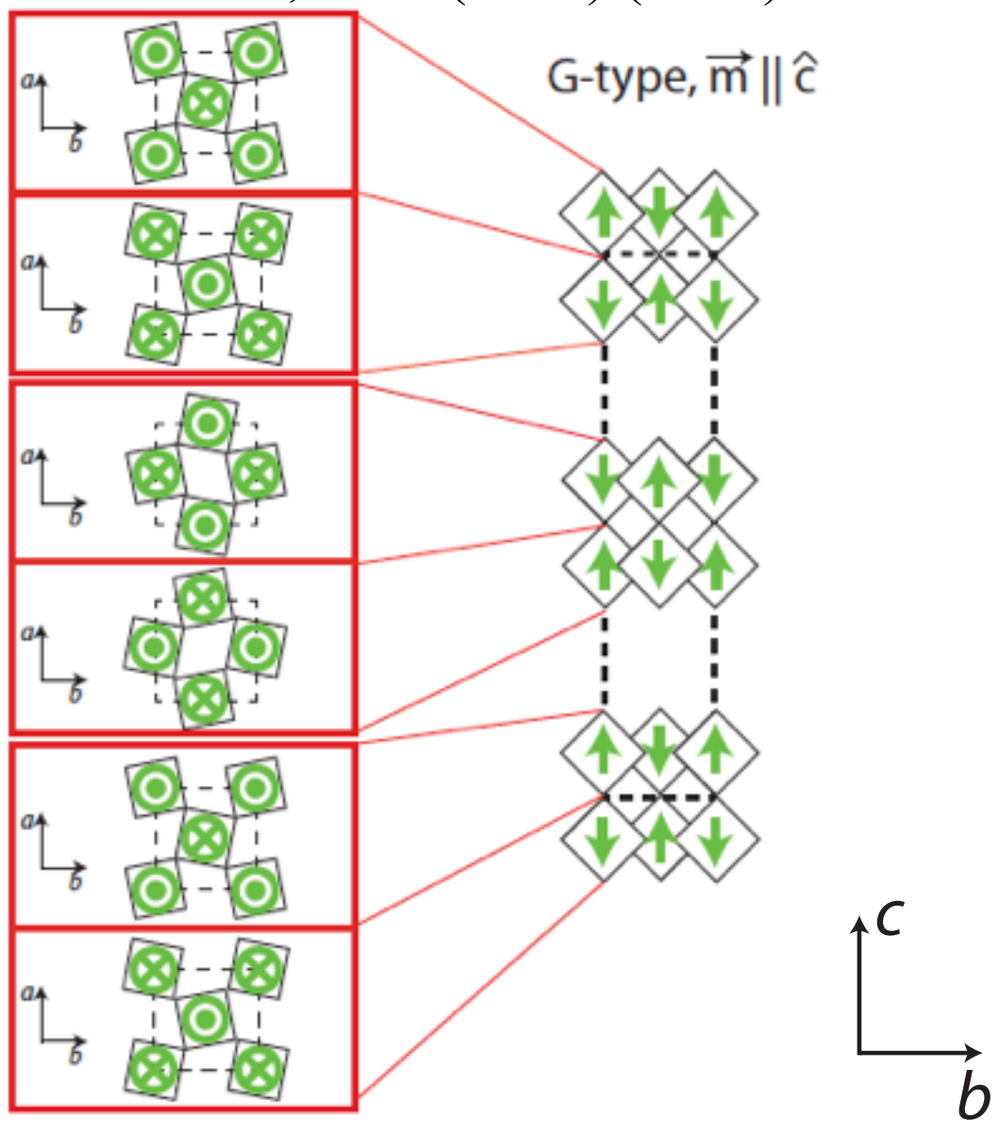
  Li et al. PRB (2013)
- La limit near x~0.06
- Equivalent x_{Ca} only slightly affects SOM

DC Magnetization



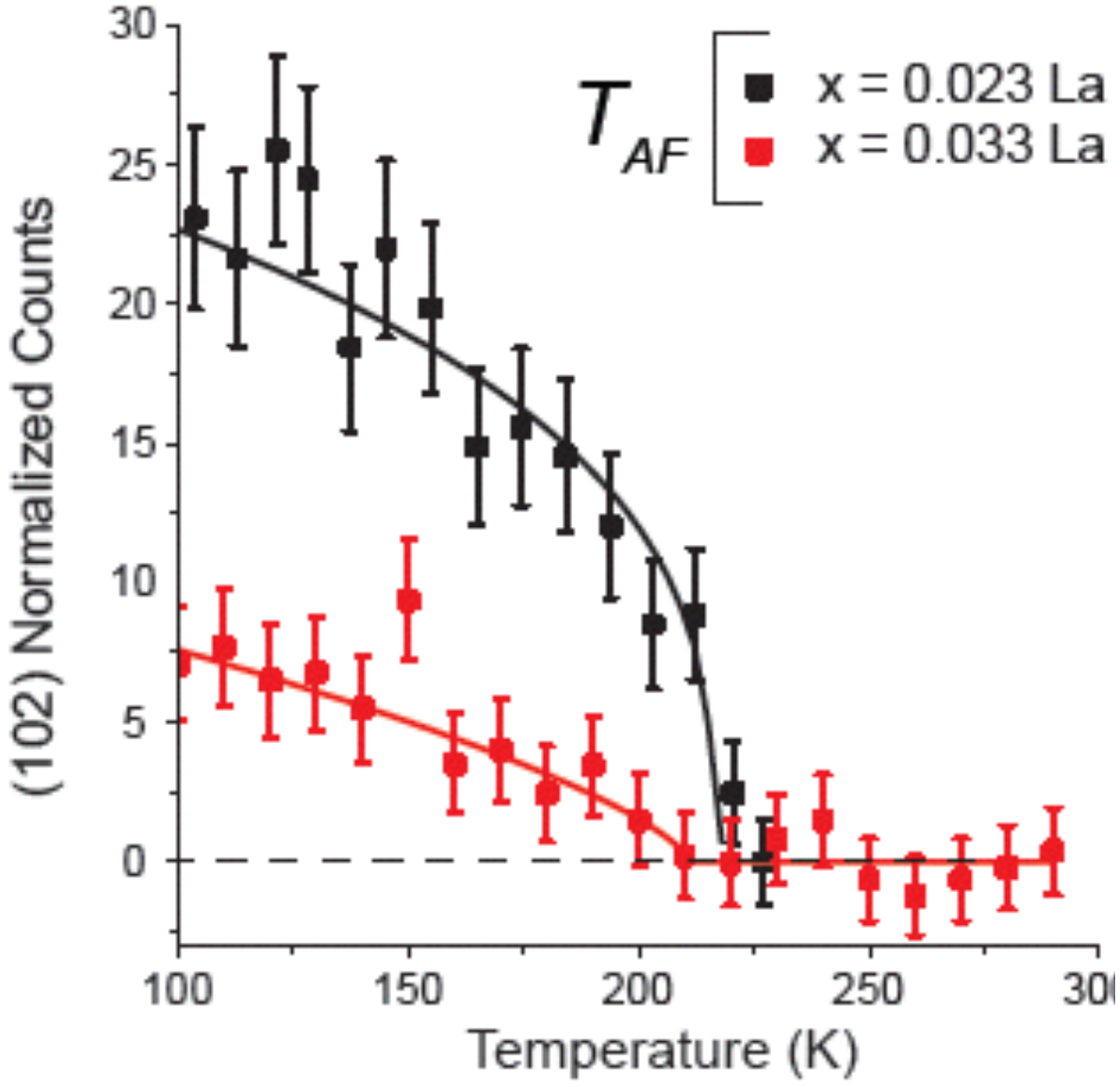
T. Hogan et al. PRL (2015)

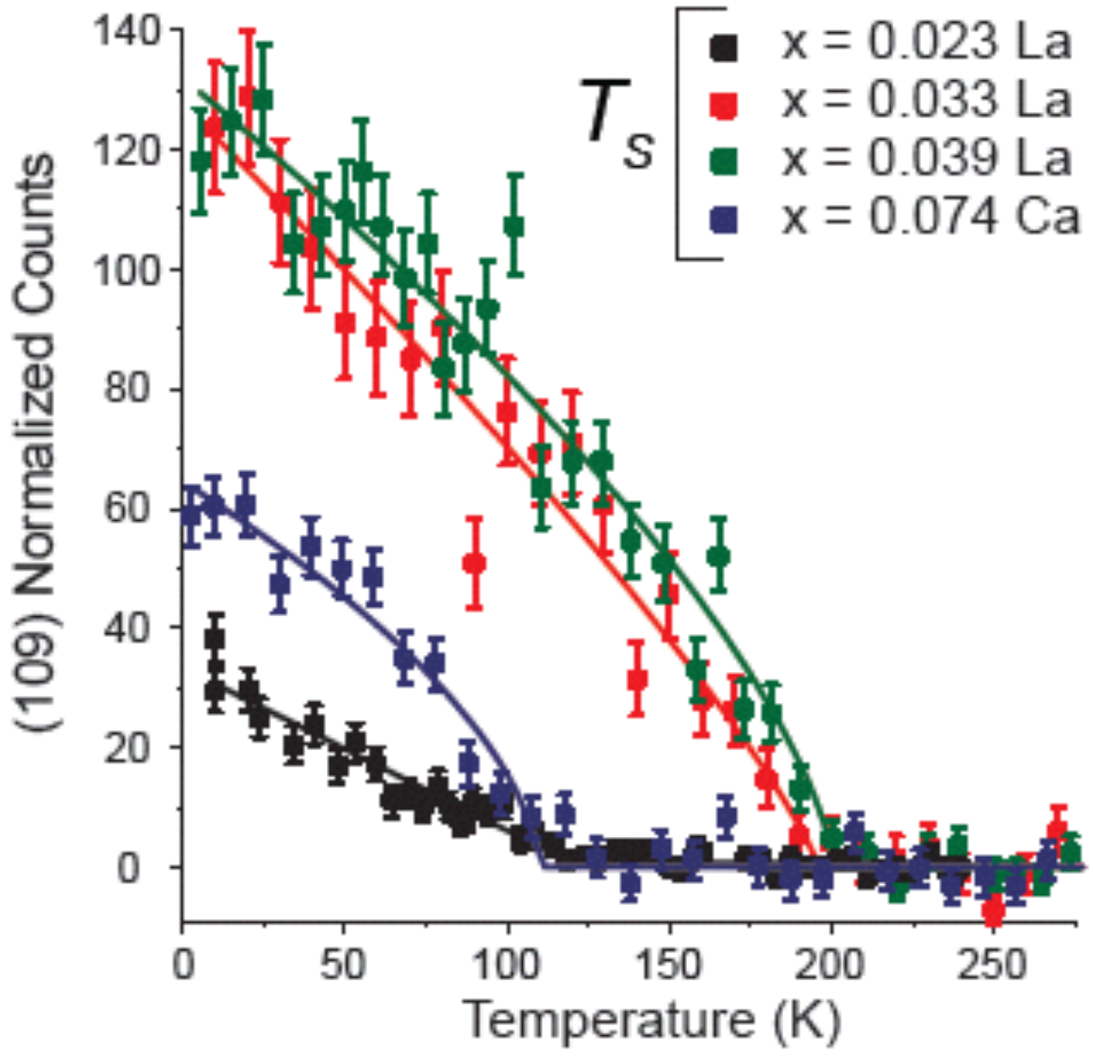
Dhital et al., PRB (2013) (2013)



- Irreversibilty vanishes beyond MIT

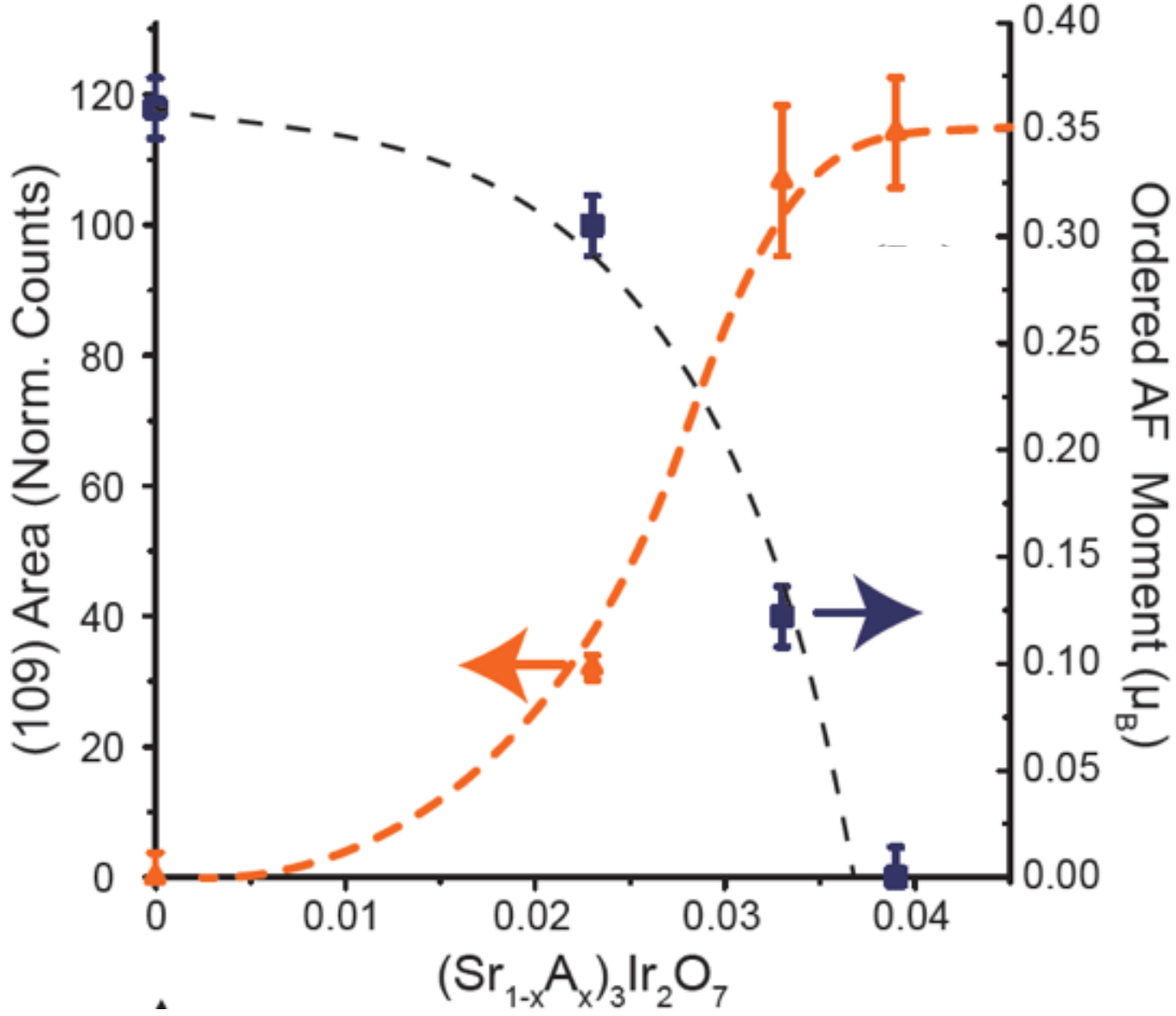
Neutron Diffraction Data





- T_S: Structural distortion at (*odd, 0, odd*)
- T_{AF} slightly suppressed, AF moment collapses
- Scattering weight increases for T_S

Neutron Diffraction Data



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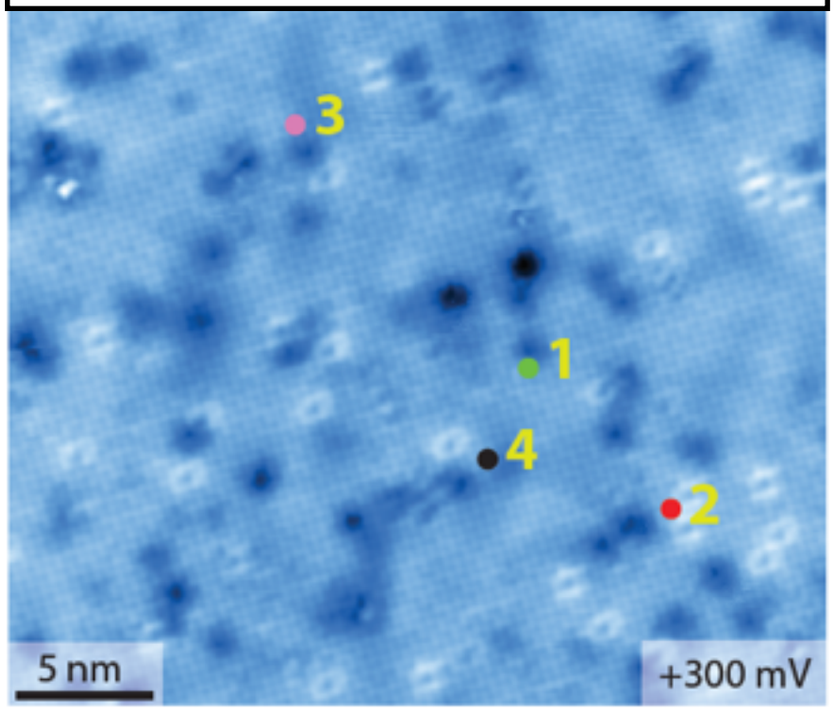
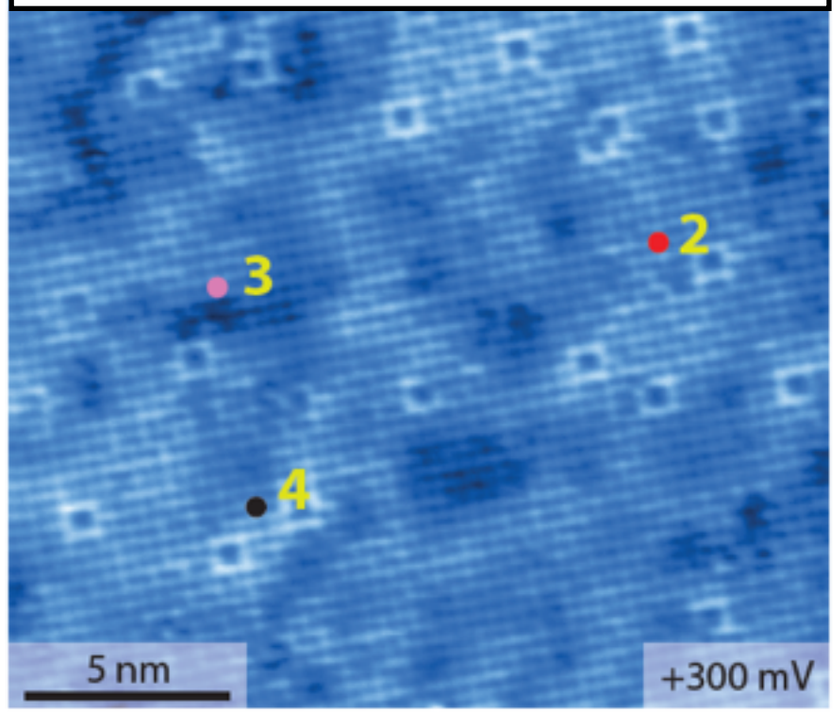
STM Data

x=0.035 “insulator”



x=0.048 “metal”



T. Hogan et al. PRL (2015)

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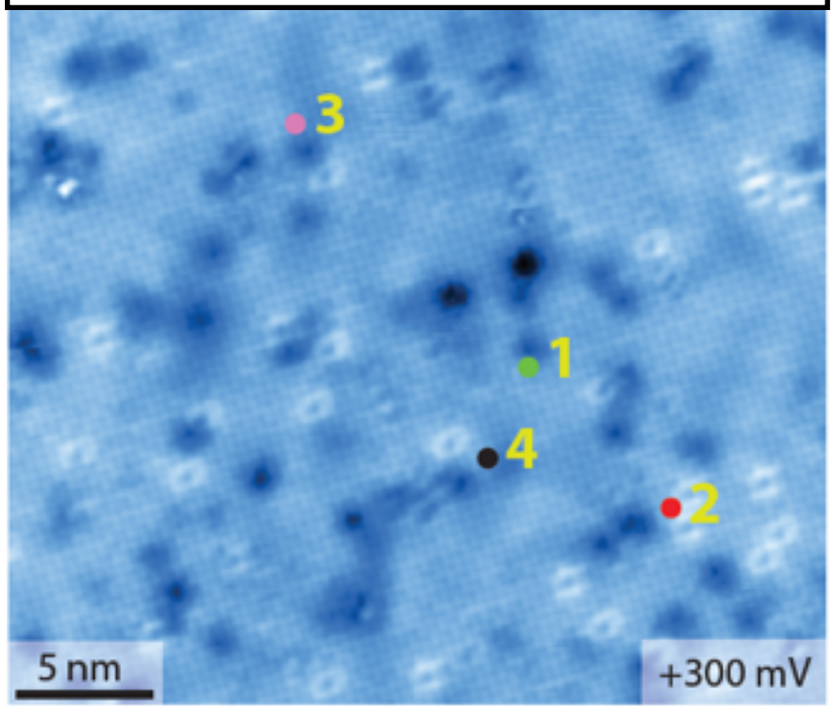

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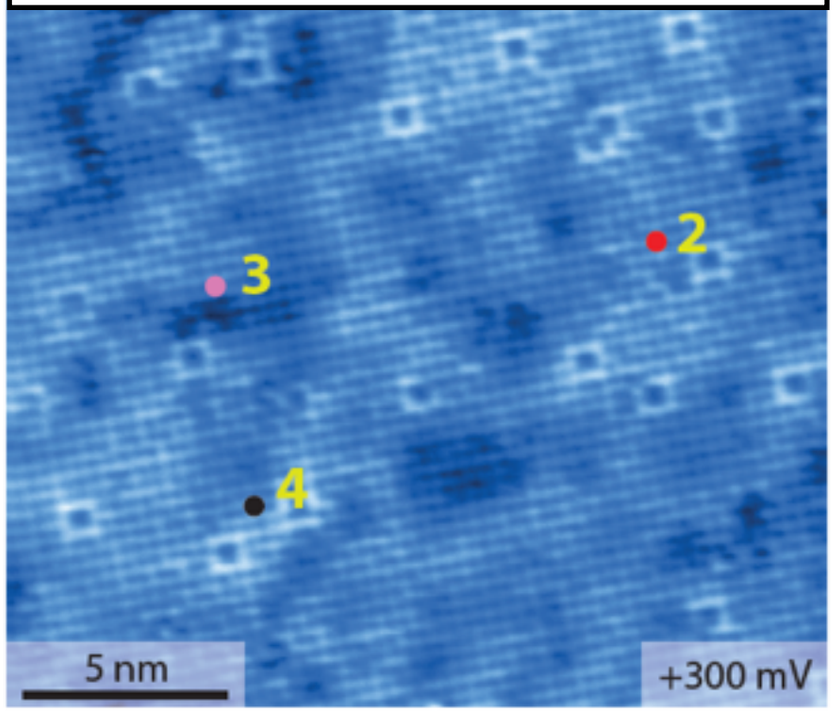

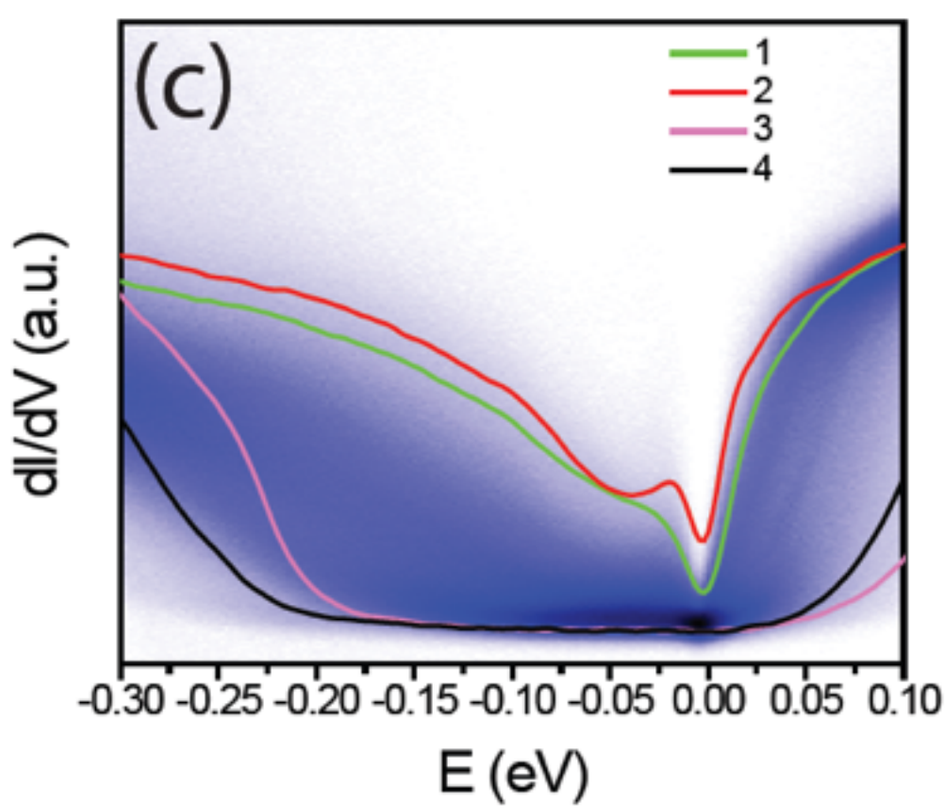

T. Hogan et al. PRL (2015)

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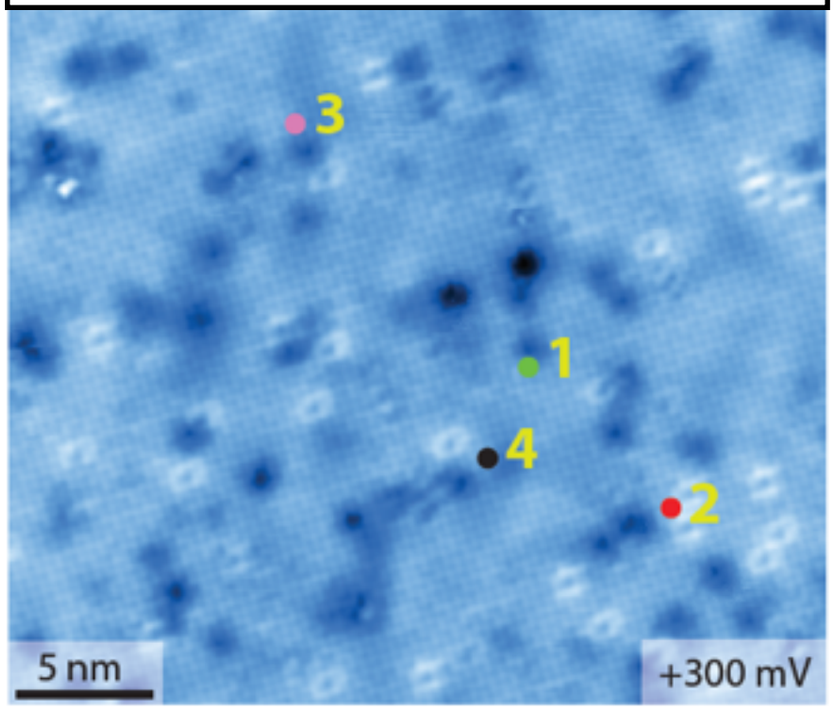

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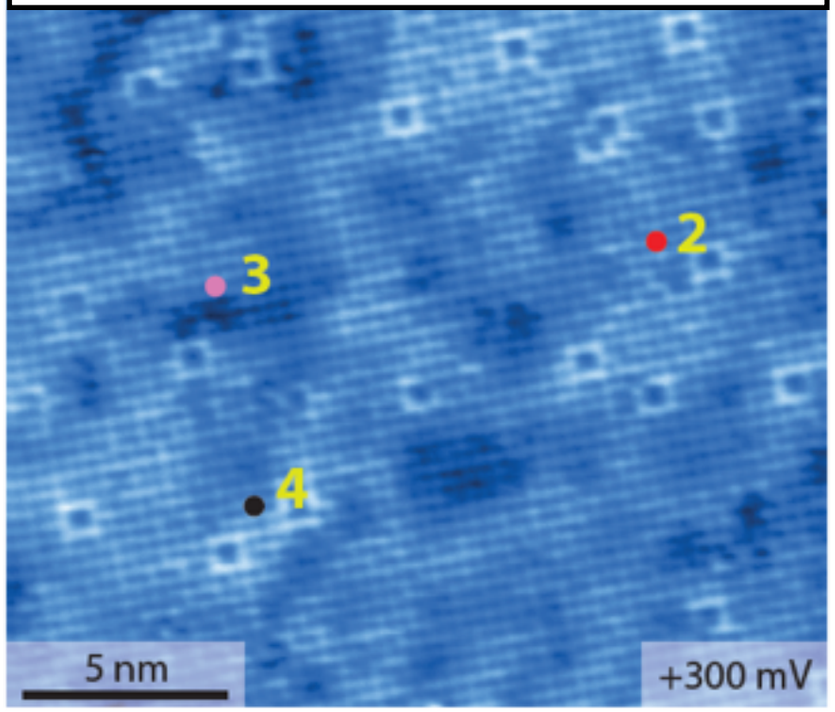
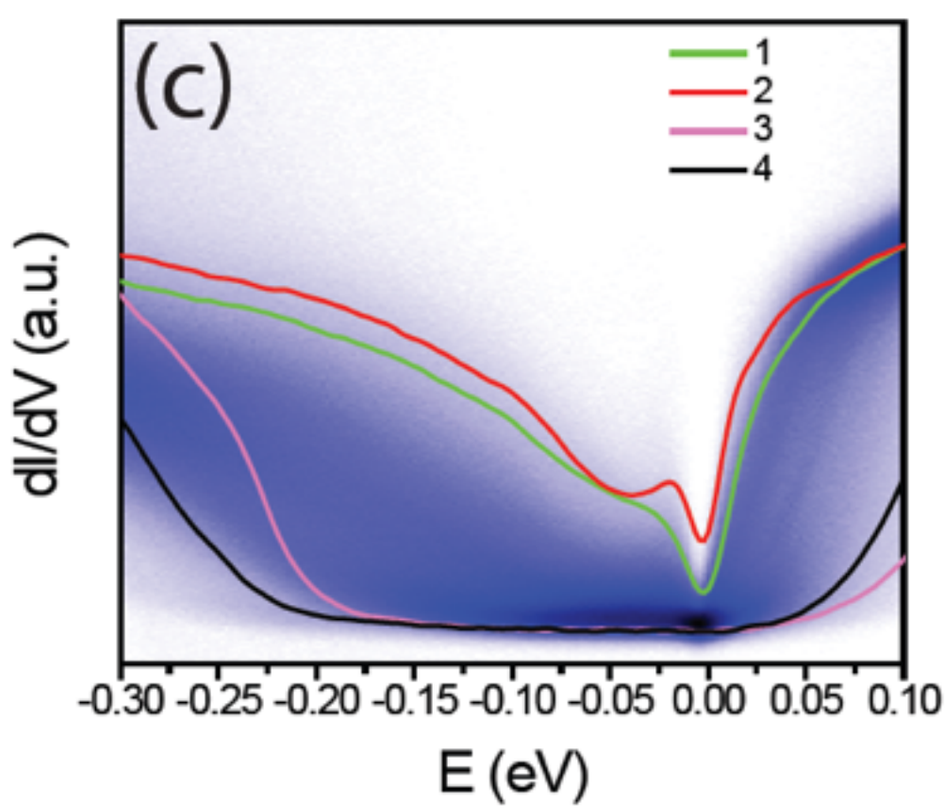
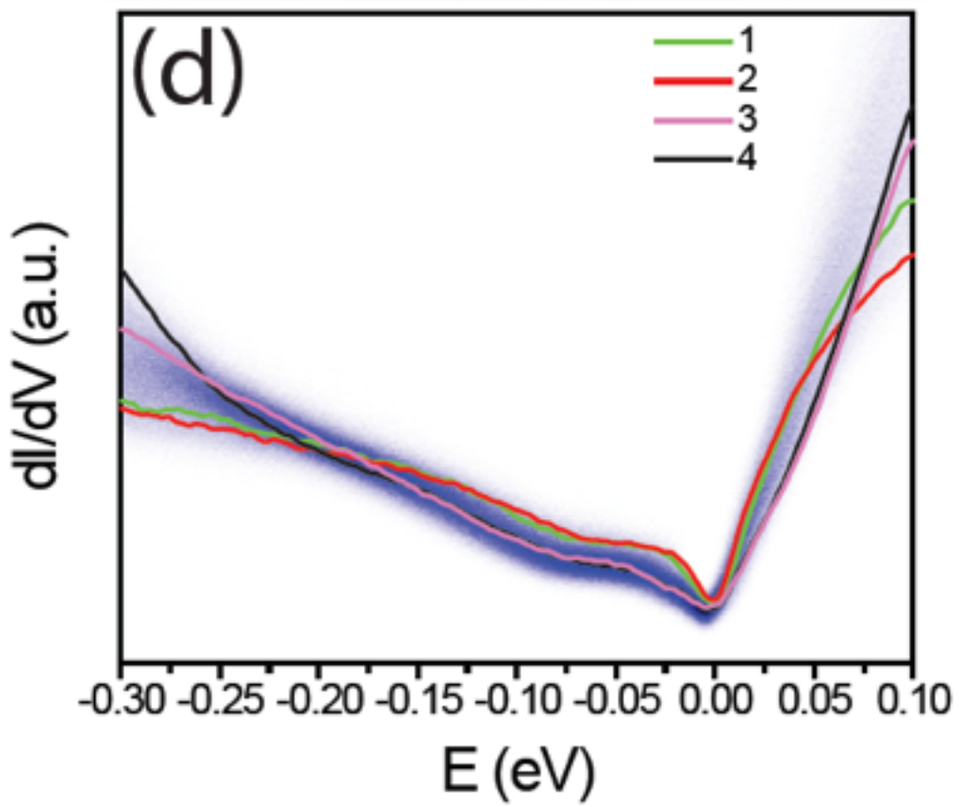




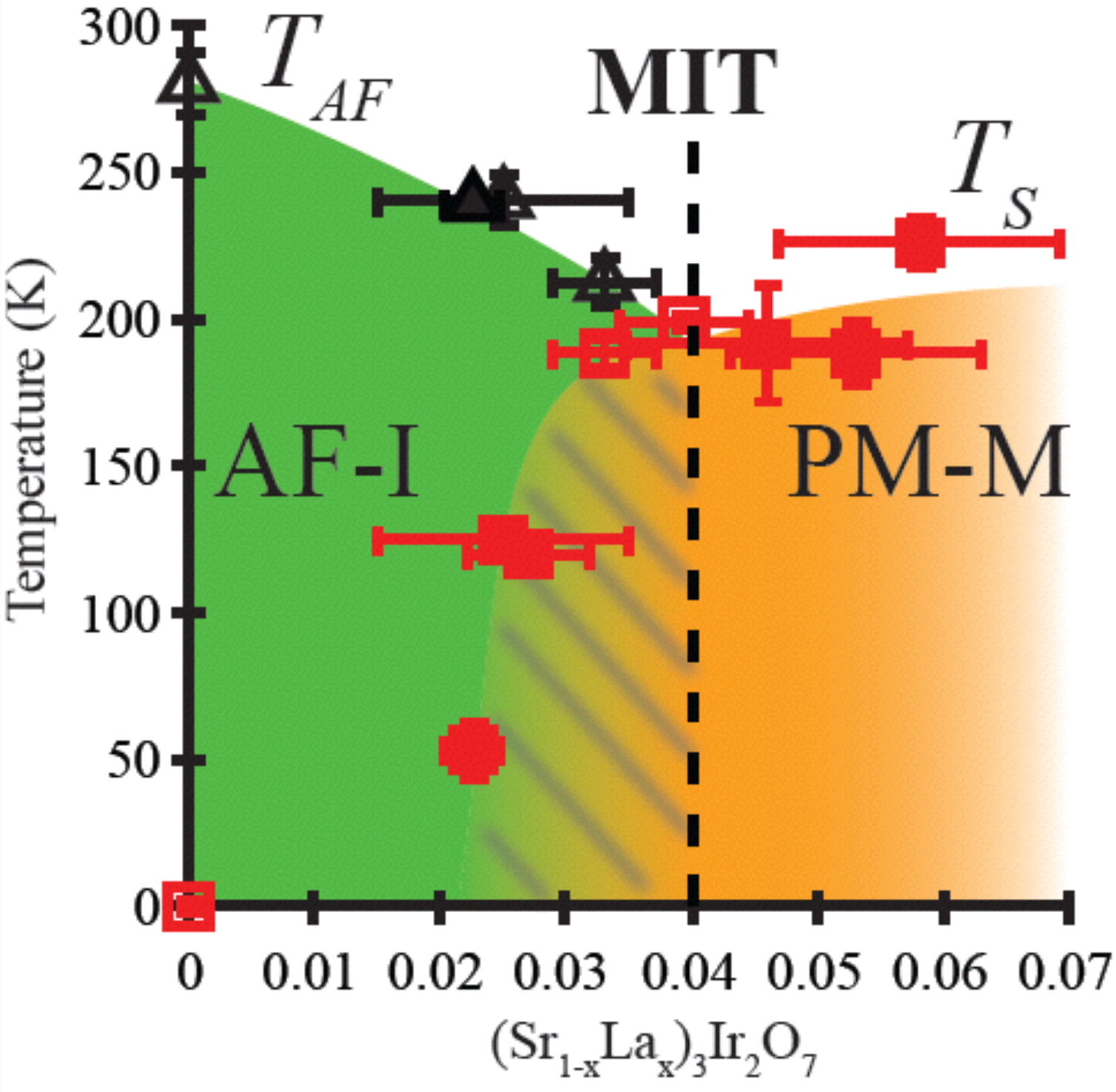


T. Hogan et al. PRL (2015)

La-doped Sr-327 Phase Diagram

- 1st order MIT
- AF order gone at MIT
- Global metallic state
- Suggestive of competing instability T_S
- What type of metal is realized?

T. Hogan et al. PRL (2015)



Correlated metal beyond MIT:

- CW metal:

  $\Theta = -70K$

  $m_{eff}=0.5\ \mu_B$

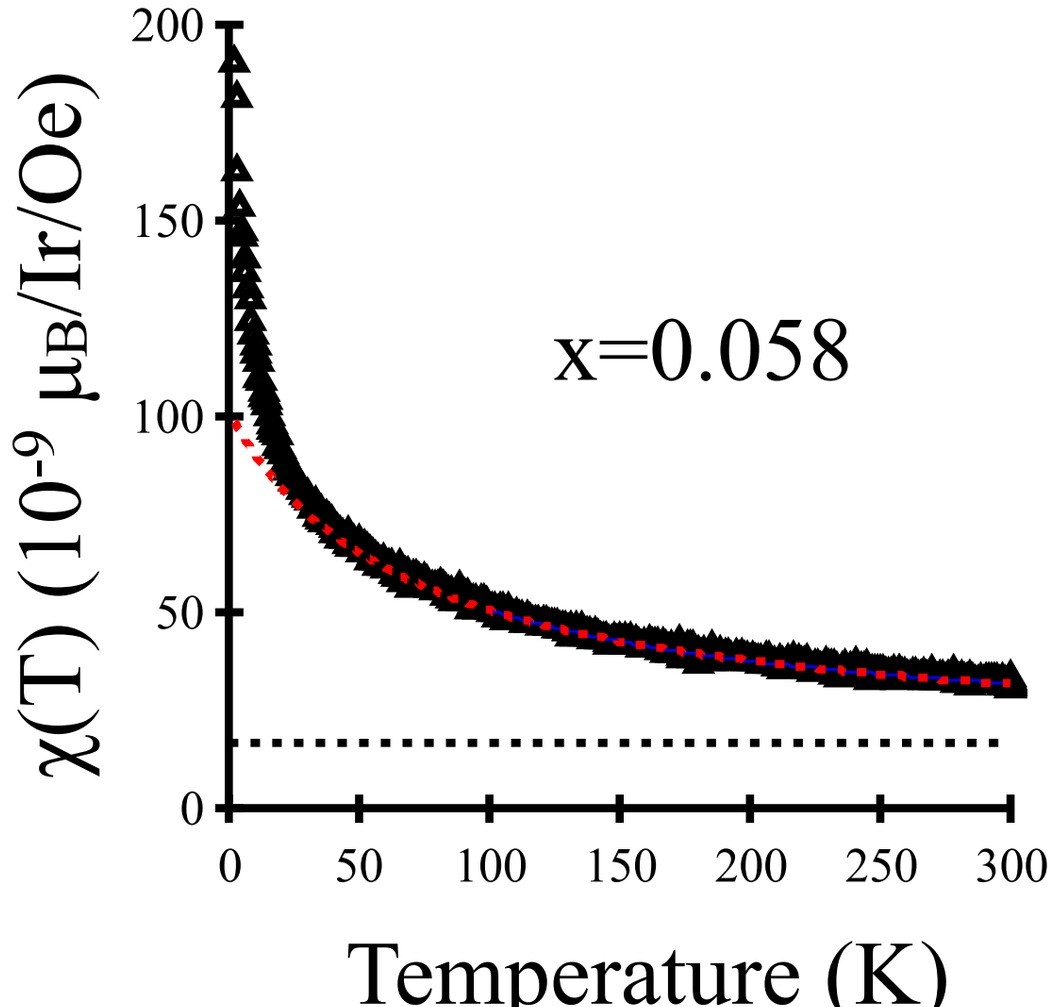
- Enhanced R_W:

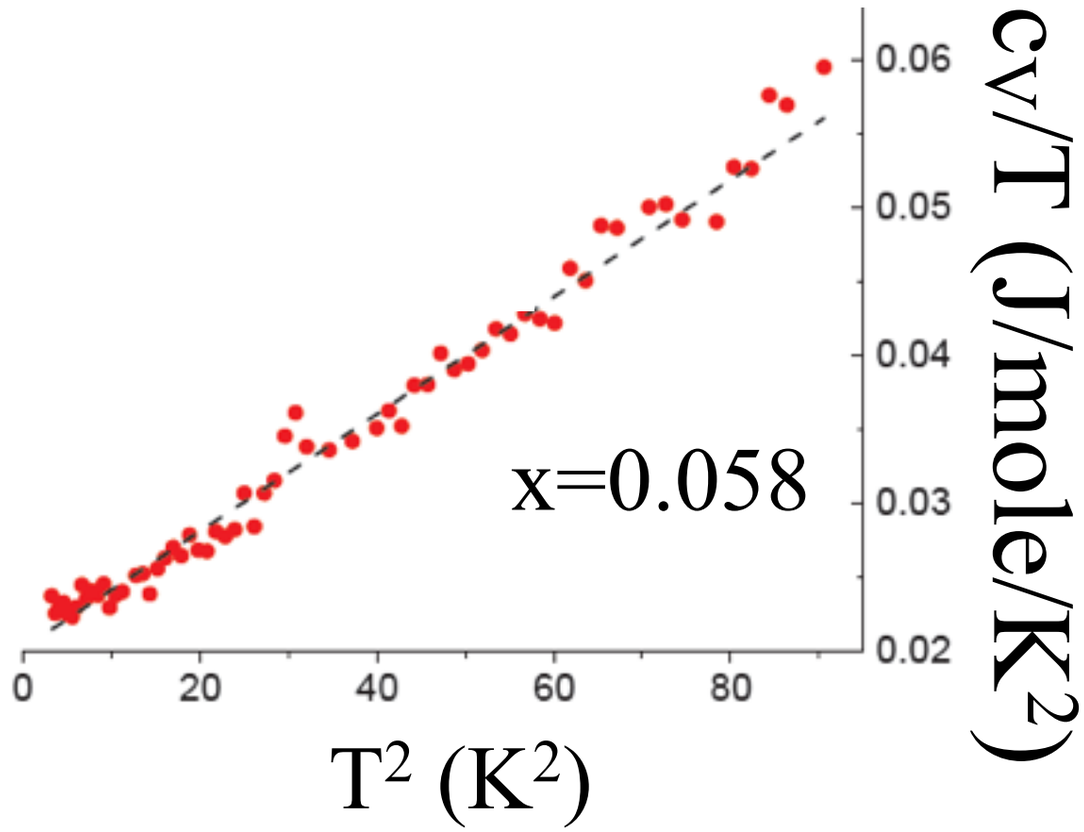
  $\gamma = 20$ (mJ/mole/K^2)

$$R_W = \frac{\pi^2 k_B^2 \chi}{3\mu_B^2 \gamma} \approx 8.4$$

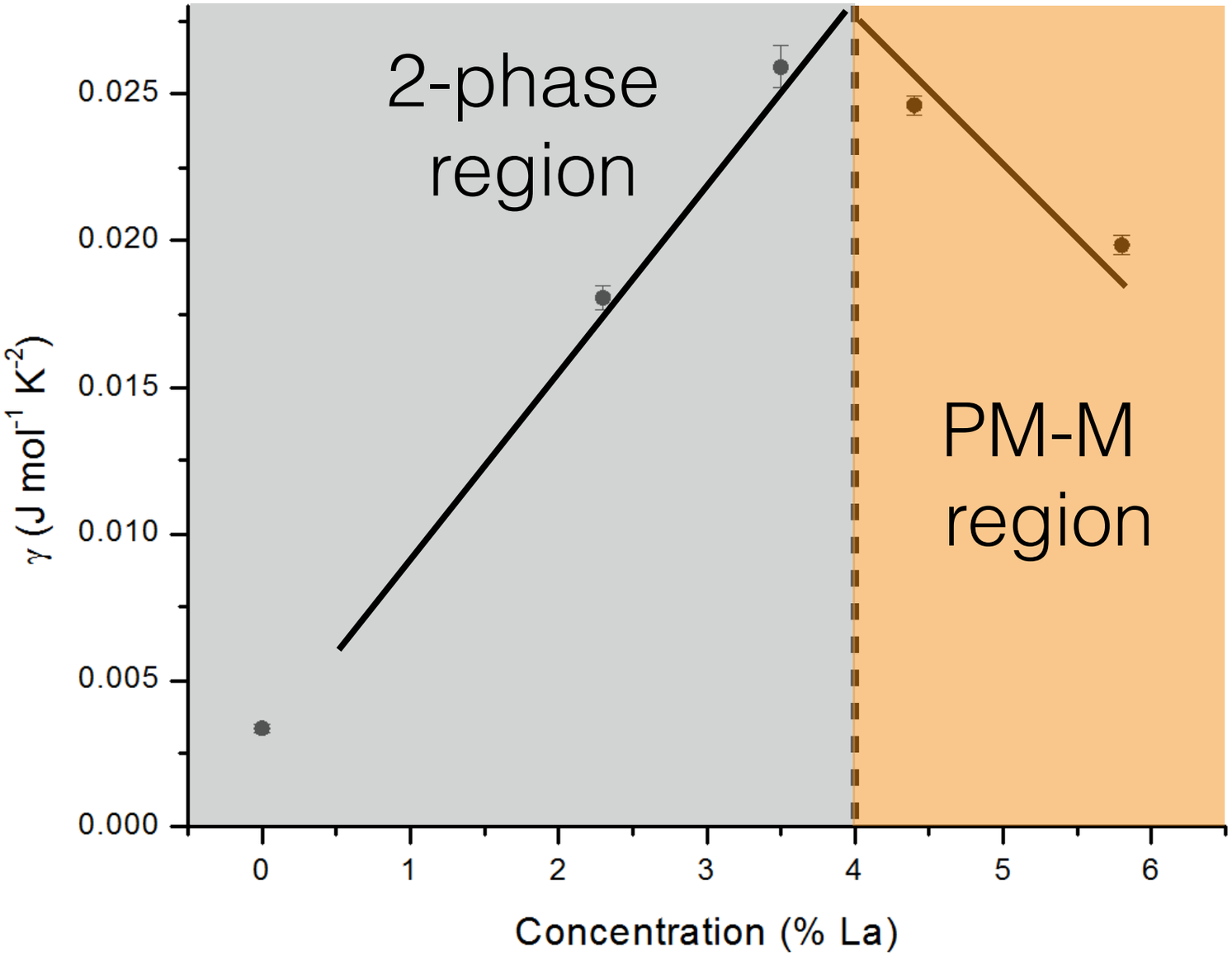
- Metal near magnetic instability

Remnant correlations from SOM parent





γ across MIT

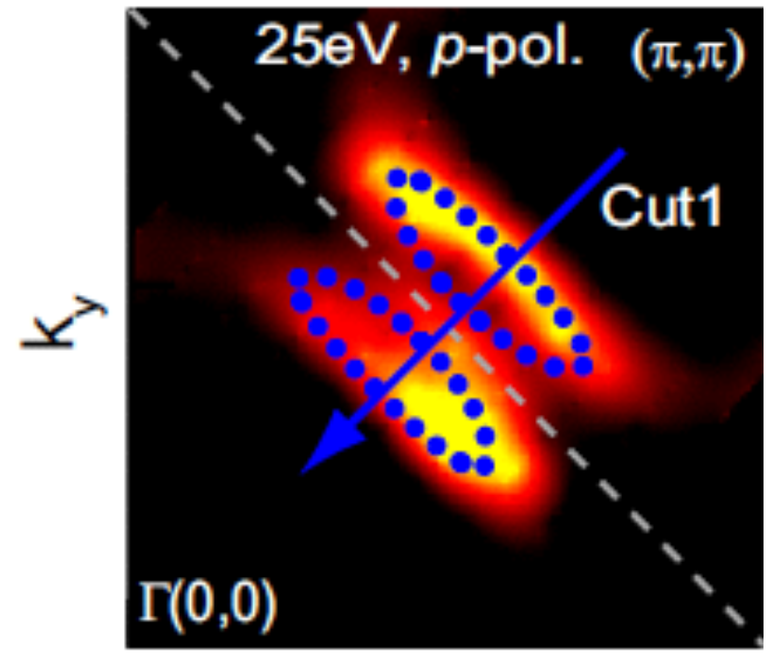


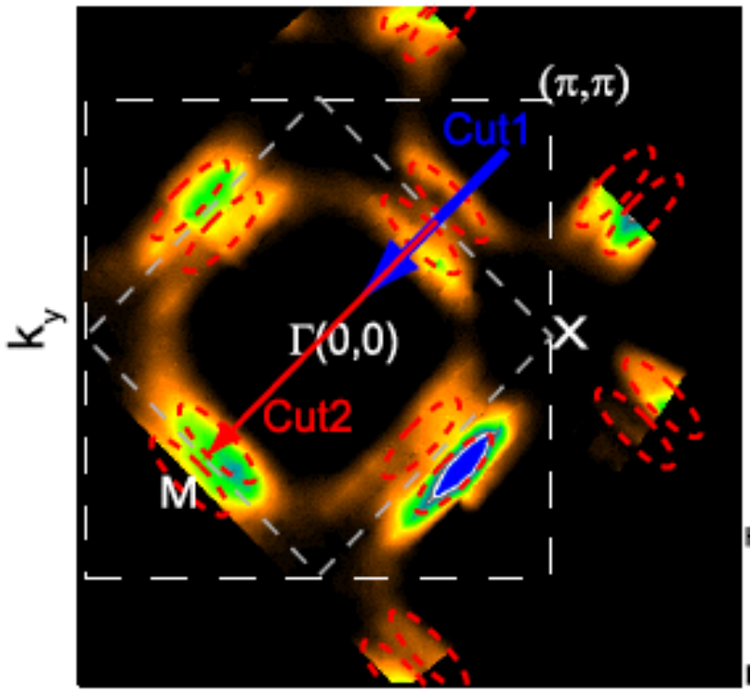
$$\gamma \sim \boldsymbol{\rho}(\varepsilon_F)$$

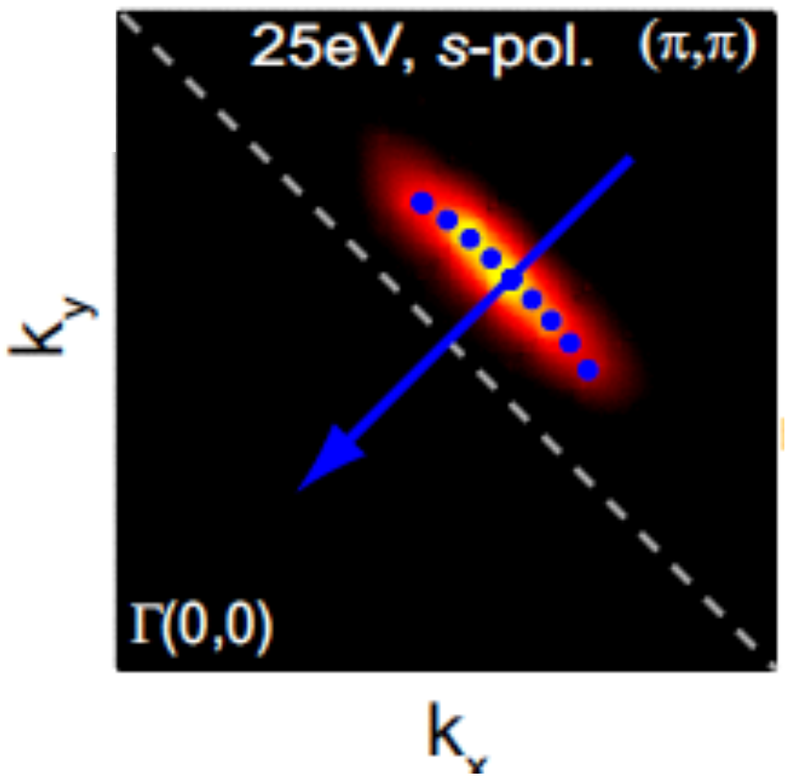
$$\gamma \sim m^*$$

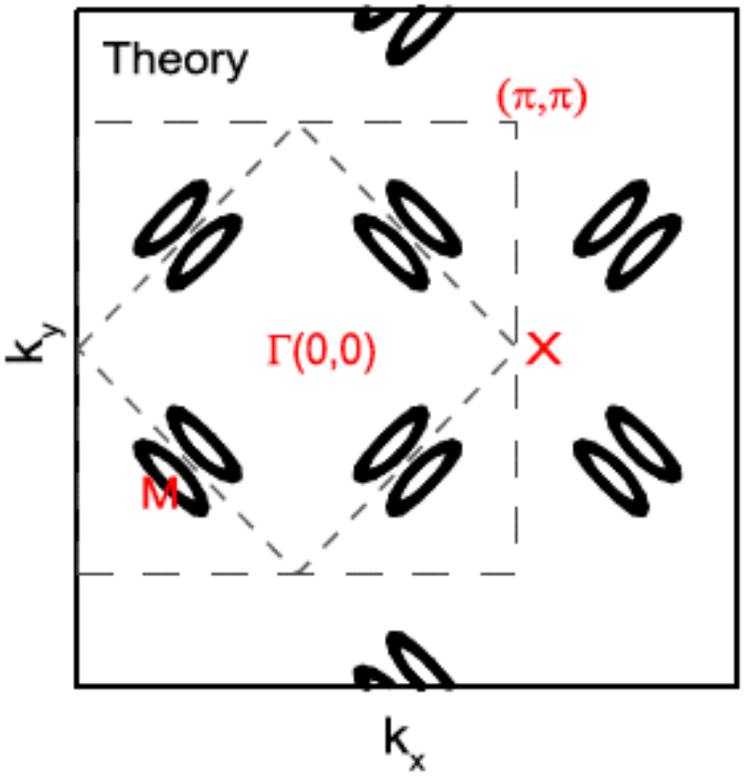
T. Hogan et al. unpublished

La-doped Sr-327: ARPES



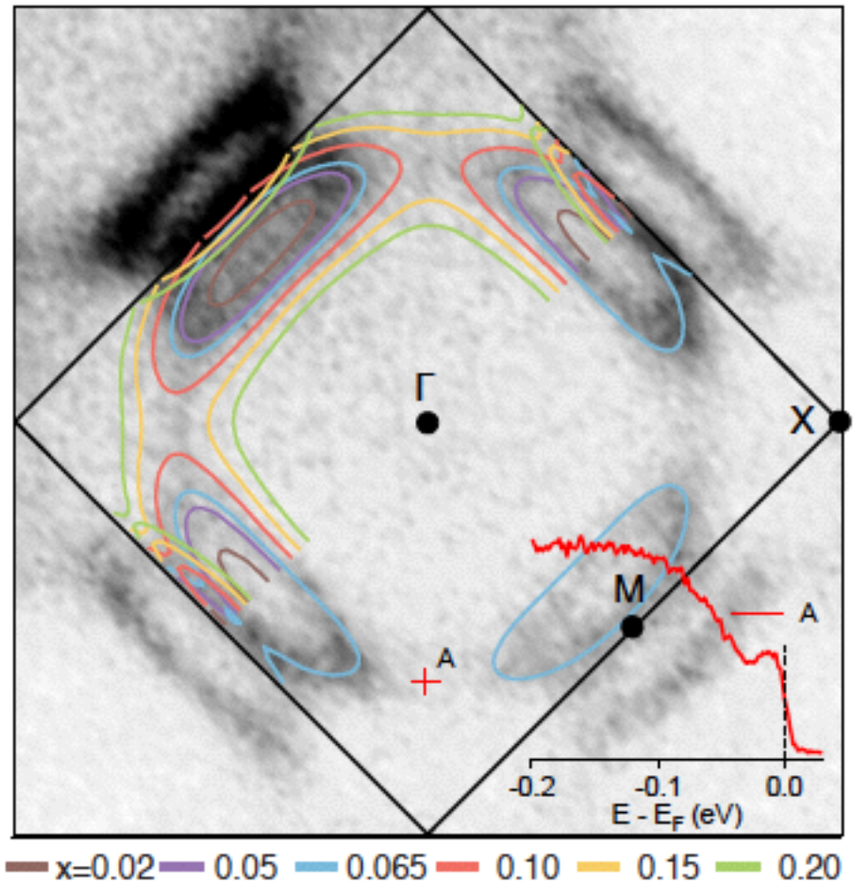






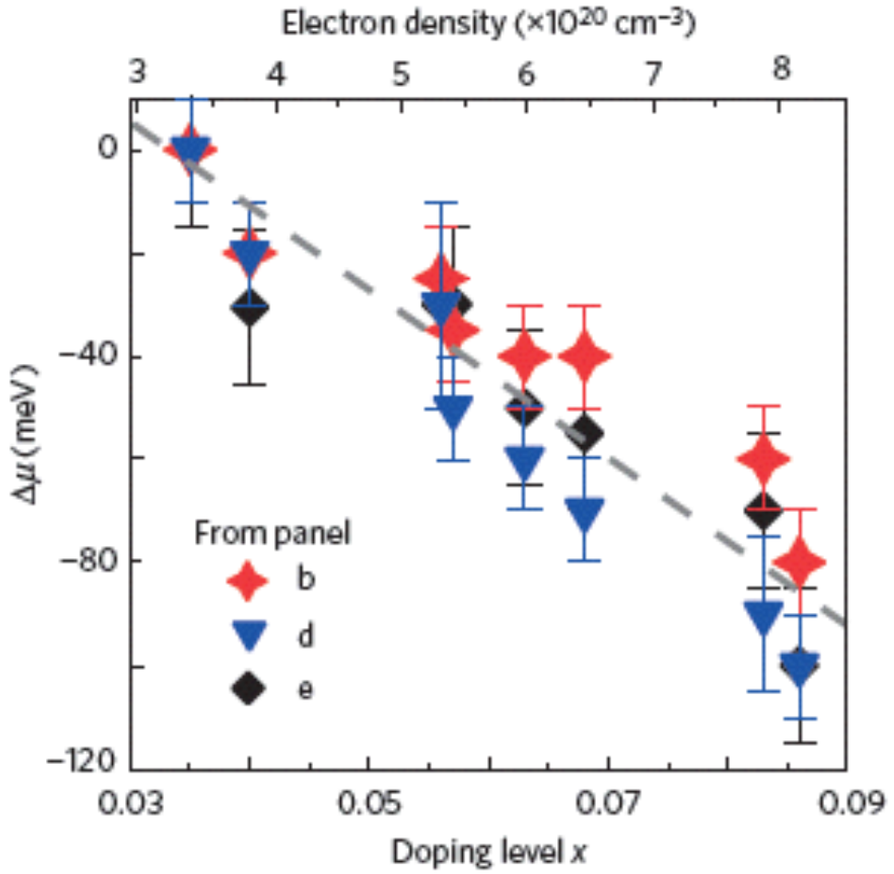
J. He et al. Nat. Sci. Rep. (2014)

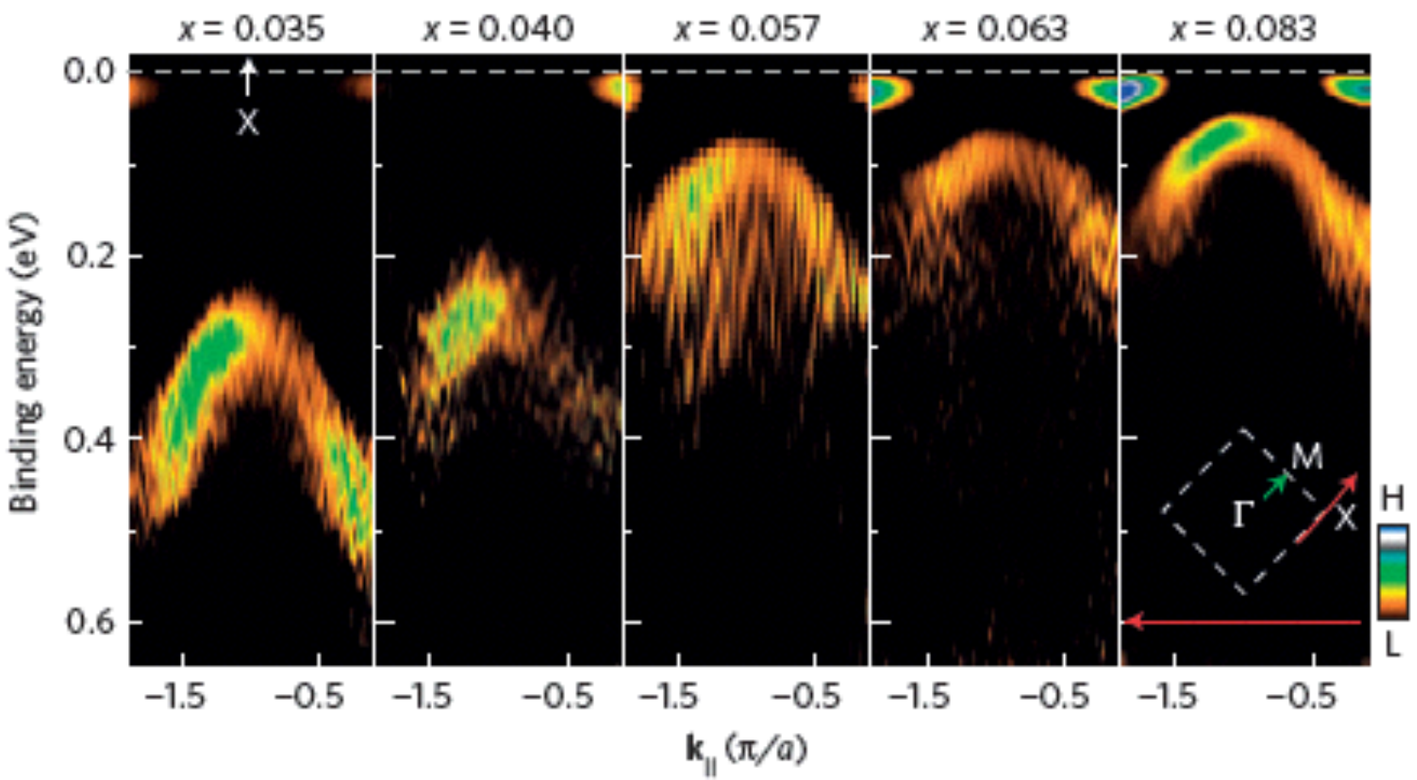
de la Torre et al. PRL (2015)



Negative Compressibility

J. He et al. Nat. Mat. (2015)





- $\delta\mu/\delta n < 0$
  - μ tracked relative to O *2p*
- Local density fluctuations drive electronic phase separation
- Likely related to deformation potential that swells lattice

Comparison: La-doped Sr-214 & Sr-327

- Phase separation (PS) under light electron doping

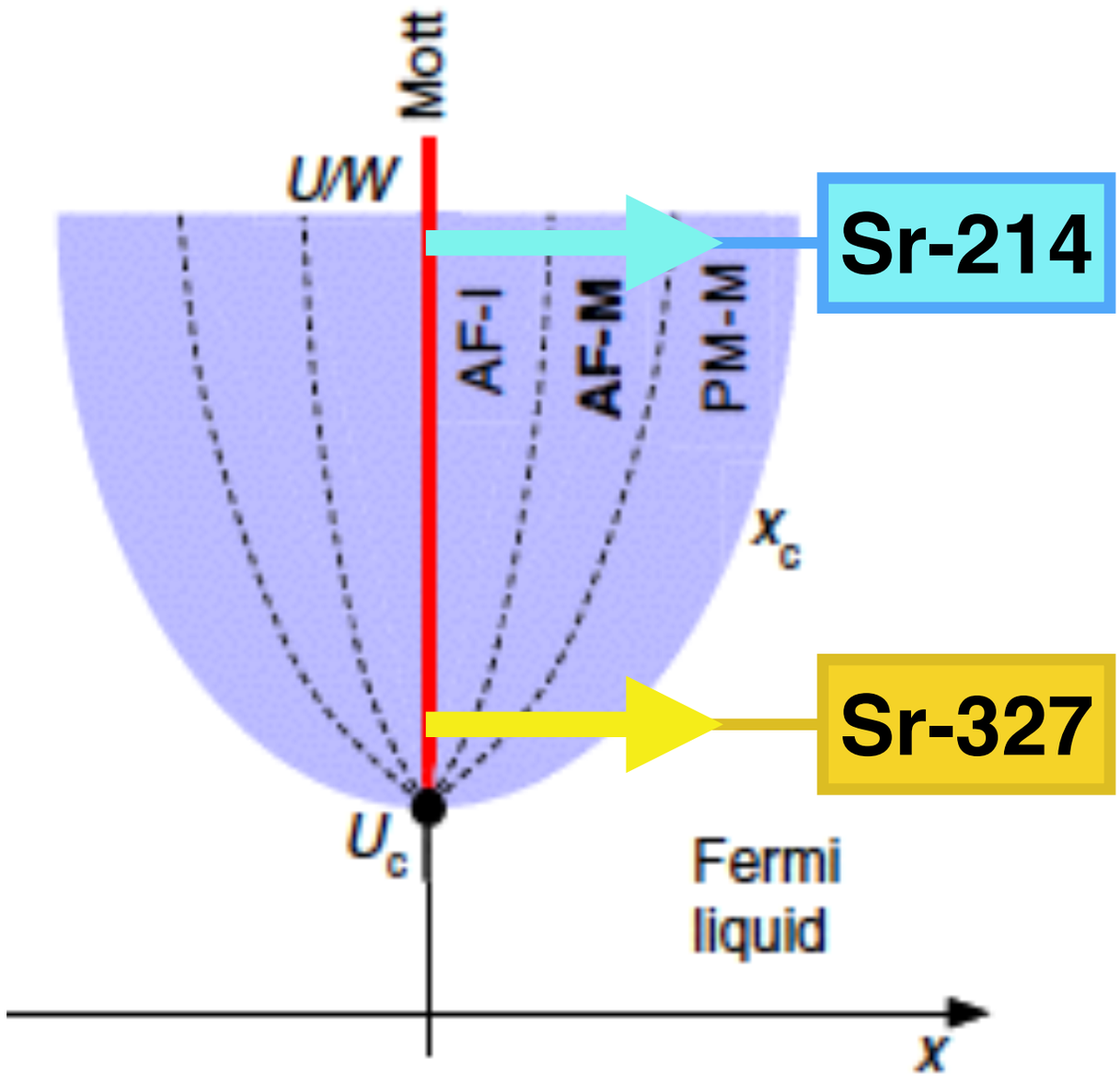
  1st order Mott MITs

- Long-range AF Order collapses at 5-6% electrons/Ir

  Short-range order survives in Sr-214

- Persistent PS in Sr-214 up to limit of La in matrix

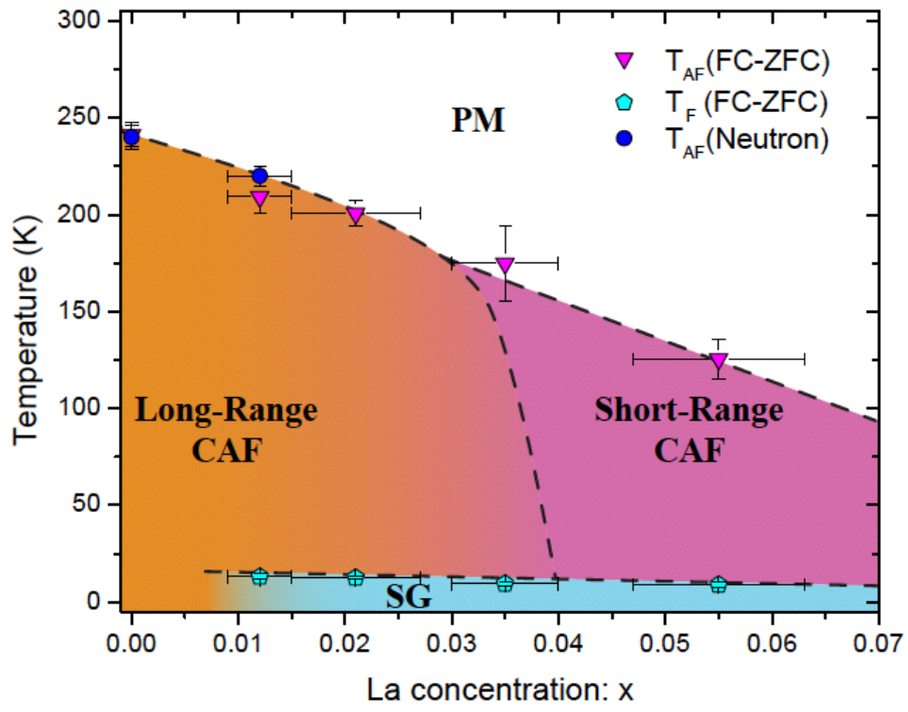
- Consistent with thermodynamic arguments in model by Yee and Balents



C. Yee et al. PRX (2014)

Summary

- Explored the MIT in La-doped Sr-214
  - PS + SR AF order persists to highest doping levels currently possible
  - Transport remains percolative
  - Low temperature spin glass state appears on light electron substitution



- Explored the MIT in La-doped Sr-327
  - Global MIT into correlated metal
  - T_S suggestive of competing instability
  - 1st order collapse of SOM state