Title: Diluted Magnetism in Iridates: Studies of Rh-doped Sr2Ir2O4

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Abstract: The physics of iridium-based 5d transition metal oxides has attracted significant interest due to the potential for exotic magnetic and electronic ground states driven by strong spin-orbit coupling effects. Among the most extensively studied iridates is the layered perovskite Sr2IrO4, which was recently proposed as the first experimental realization of a novel Jeff=1/2 spin-orbital Mott insulating state. Intriguing similarities between Sr2IrO4 and La2CuO4, the parent compound of the high-Tc cuprates, have also led to speculation that it may be possible to induce superconductivity in this system through chemical doping. We have investigated the magnetic properties of the doped system Sr2Ir1-xRhxO4 using a combination of resonant magnetic x-ray scattering (RMXS), resonant inelastic x-ray scattering (RIXS), and x-ray absorption spectroscopy (XAS) techniques. These measurements reveal the effect of Rh-doping on the magnetic structure, phase diagram, and characteristic magnetic excitations of Sr2IrO4, and provide fundamental information about the role of quenched Rh impurities.

Acknowledgements

- **University of Toronto:**
  - Young-June Kim, Hlynur Gretarsson, Andreea Lupascu
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  - Gang Cao (Sample Synthesis)
- **Canadian Light Source:**
  - Yongfeng Hu (XAS)
- **Advanced Photon Source:**
  - Zahir Islam (RMXS), Mary Upton, Jungho Kim (RIXS)
- **Funding:** NSERC, Banting Postdoctoral Fellowship Program, NRC, CIHR, Prov. of Sask., WEDC, Univ. of Sask., U.S. Dept. of Energy

UNIVERSITY OF TORONTO

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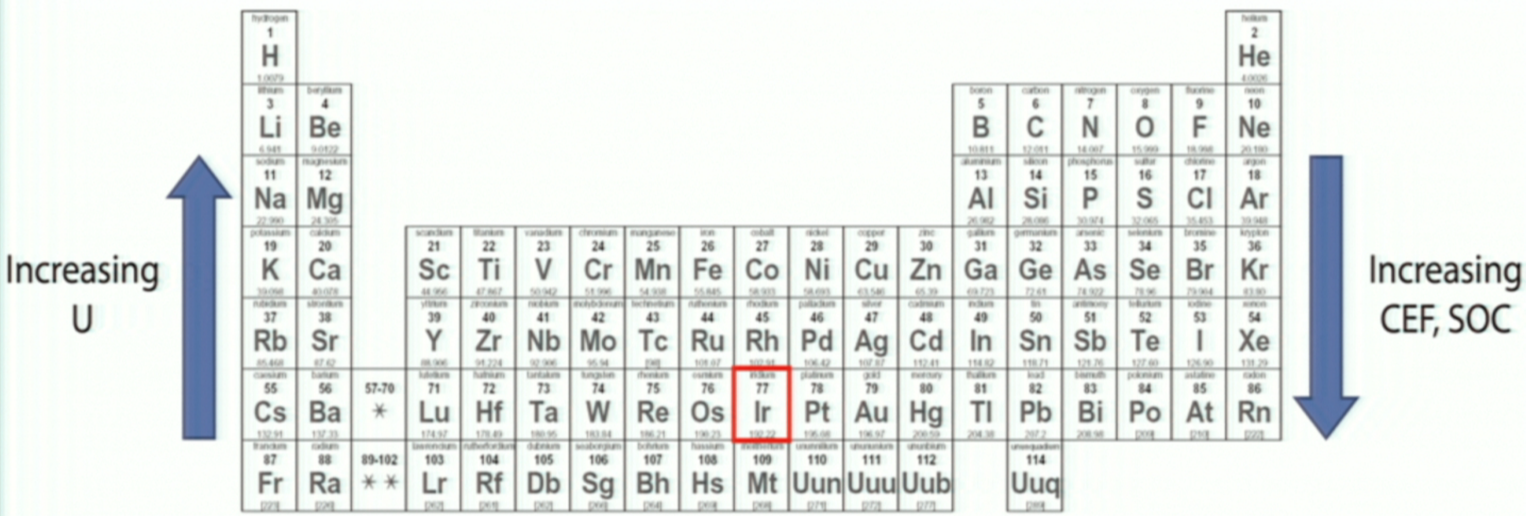
Canadian Light Source Centre canadien de rayonnement synchrotron

UK UNIVERSITY OF KENTUCKY

A Brief Outline...

- Introduction – novel physics in spin-orbit coupled iridates
- **Sr_2IrO_4** – the $j_{eff} = 1/2$ spin-orbital Mott insulator
- **$Sr_2Ir_{1-x}Rh_xO_4$** – investigating the impact of chemical doping
- Experimental Results:
  - What is the role of Rh dopant ions? (**XAS**)
  - Effect of doping on magnetic structure? (**RMXS**)
  - Effect of doping on magnetic excitations? (**RIXS**)
- Summary and conclusions

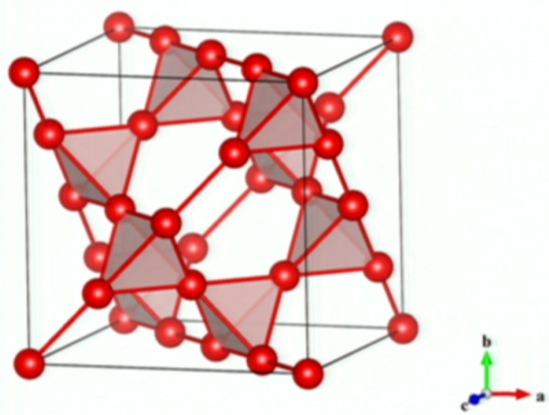
Why Study Iridates?



- Interplay between electronic correlations, crystal electric field, and spin-orbit coupling (U ~ CEF ~ SOC)
- Potential for exotic physics driven by strong SOC (~0.5 eV)

Novel Physics in Iridates

$A_2Ir_2O_7$



Candidate for Topological Insulator, Weyl Semi-Metal, Metallic Spin Liquid:
Yang et al, PRB (2010),
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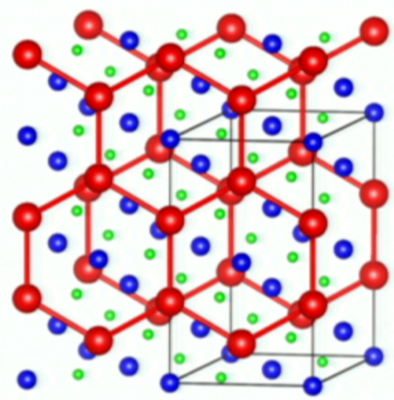
Nakatsuji et al, PRL (2006)

A_2IrO_3



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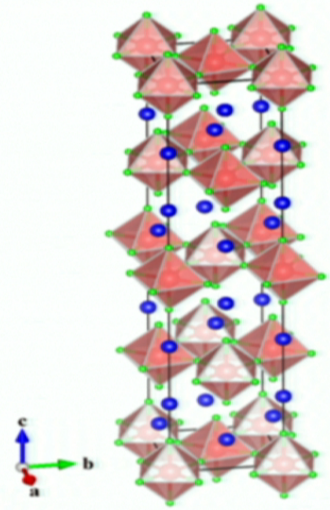
$Sr_{n+1}Ir_nO_{3n+1}$



$J_{eff}=1/2$ spin orbital Mott insulator:
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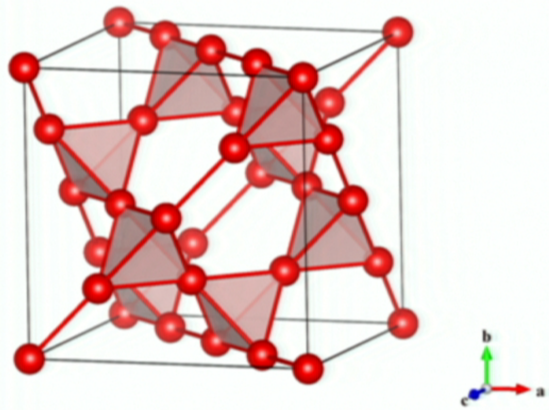

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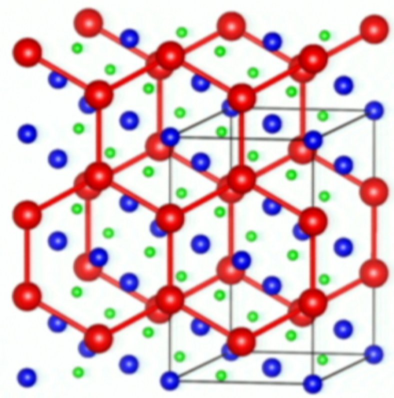

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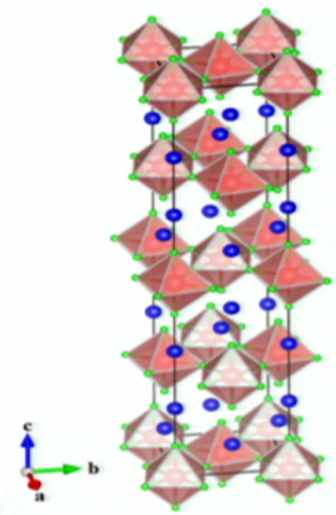

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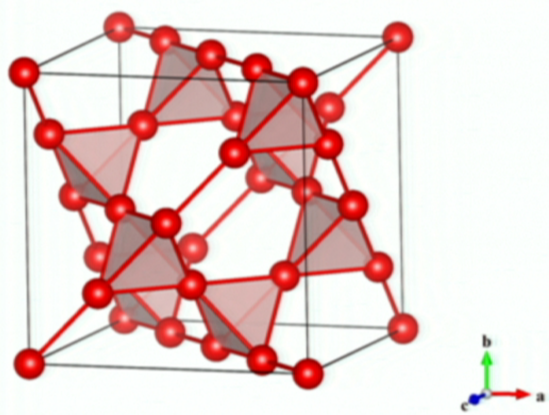

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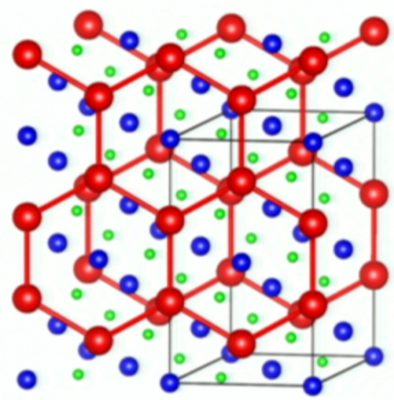

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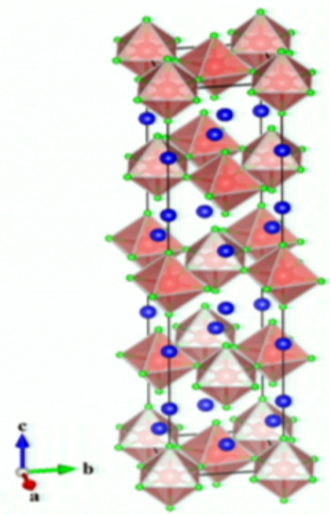

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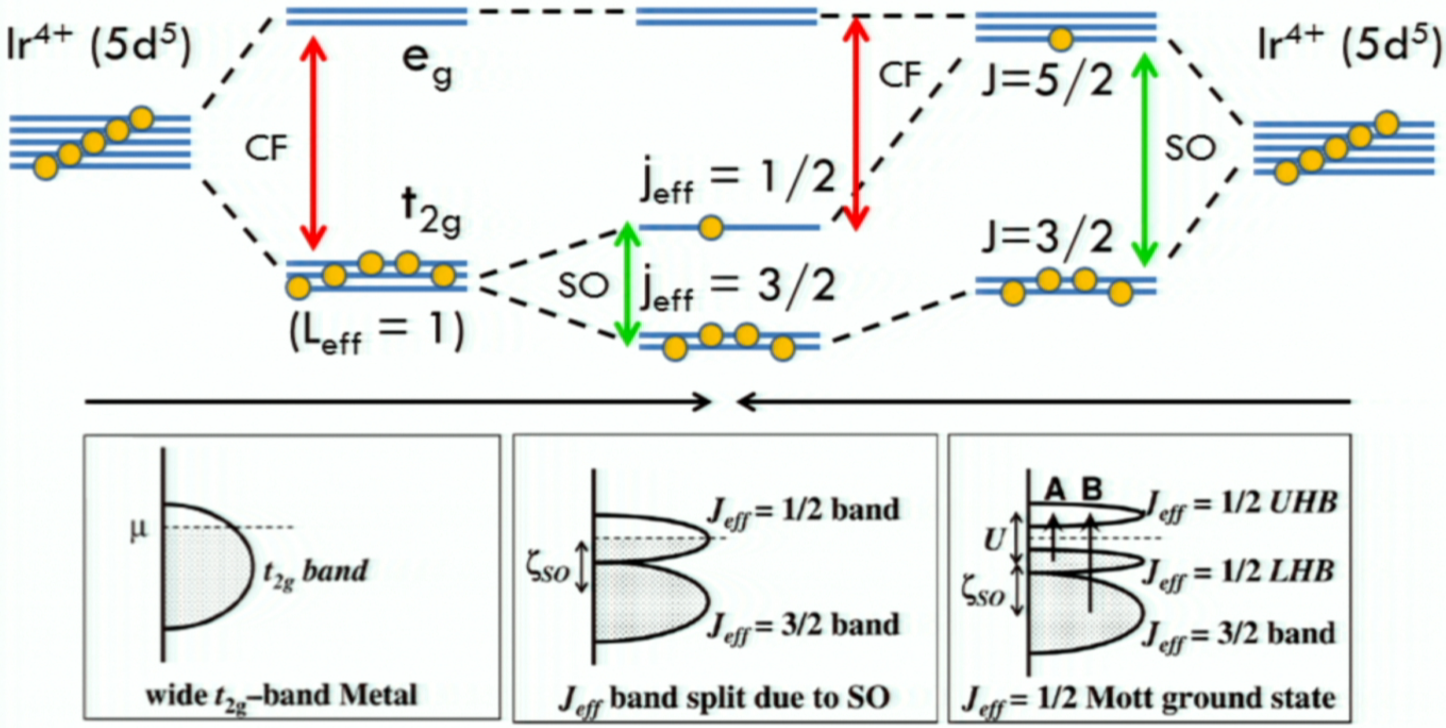
Sr_2IrO_4: the Spin-Orbital Mott Insulator

A surprising number of iridates are insulators - why?



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

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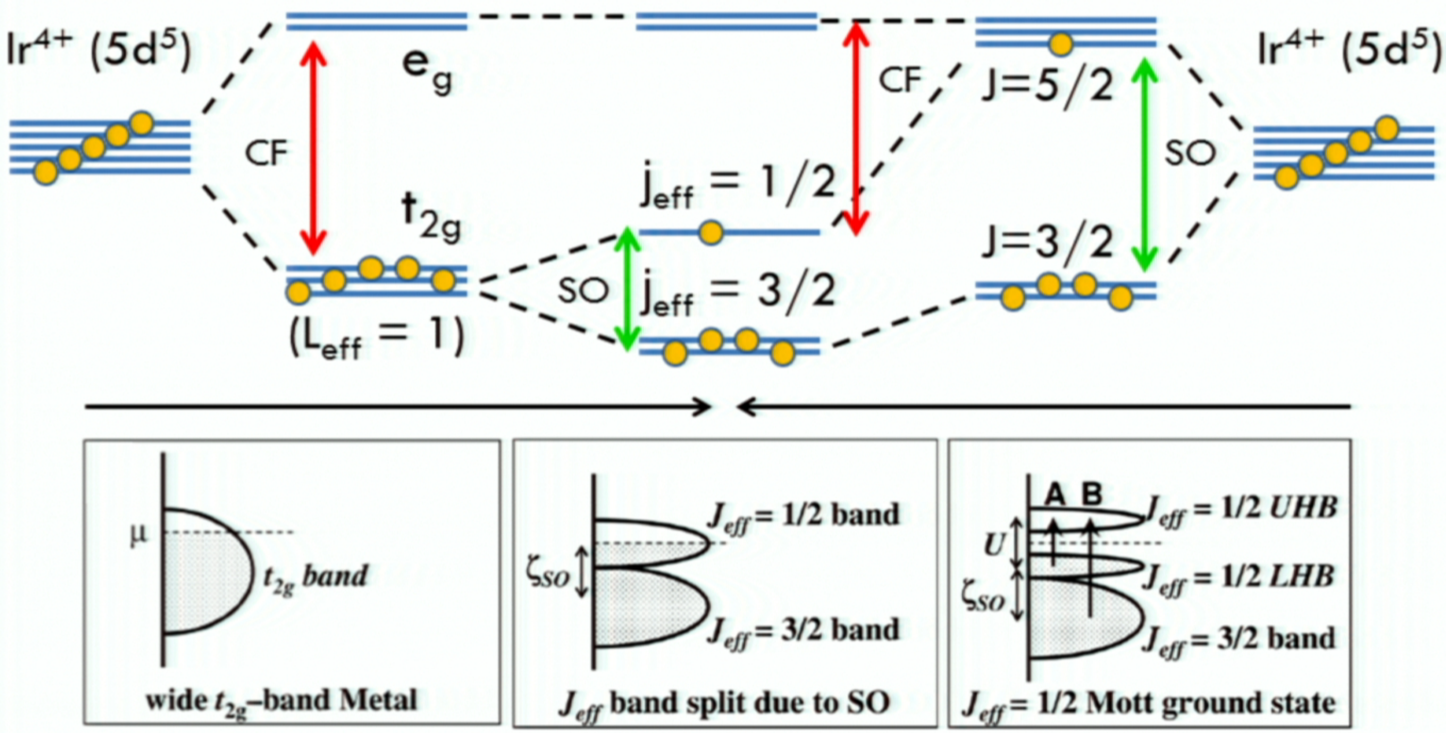


B.J. Kim et al, PRL (2008)

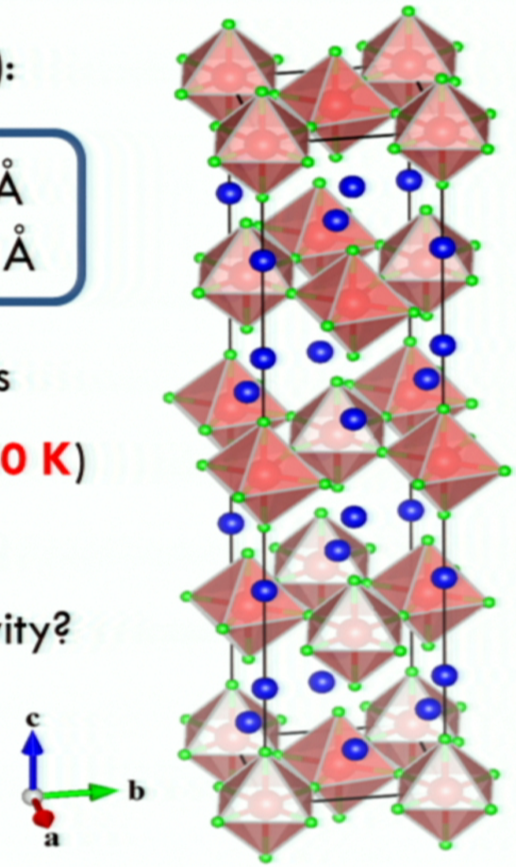
Physical Properties of Sr_2IrO_4

- Member of Ruddlesden-Popper series (n=1):

$Sr_{n+1}Ir_nO_{3n+1}$

$a = 5.50$ Å
$c = 25.80$ Å

- Space Group: ***$I4_1/acd$***
- IrO_6 octahedra rotated by ~11° w.r.t. c-axis
- $J_{eff} = 1/2$ canted antiferromagnet (**T_N ~ 240 K**)
- Similar structure to La_2CuO_4 and Sr_2RuO_4
- Possibility of doping-induced superconductivity?
- *Wang and Senthil, PRL (2011)*
- *Watanabe et al, PRL (2013)*



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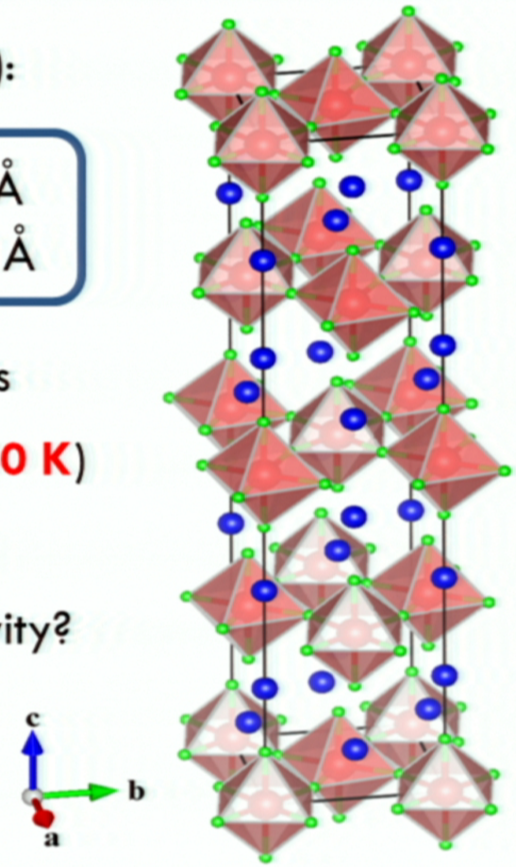
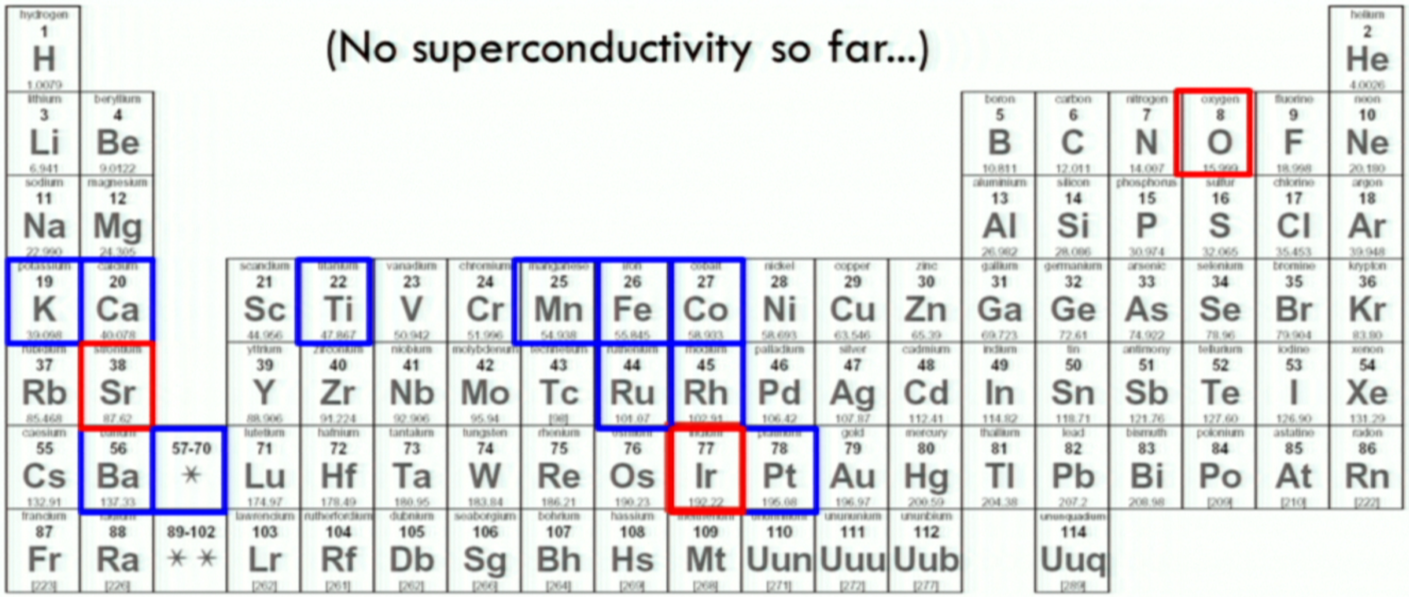
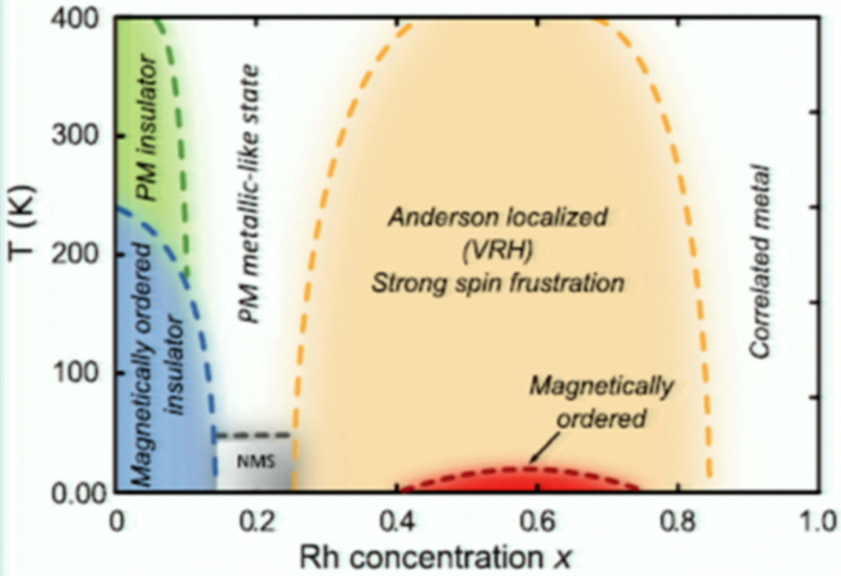


Doping Studies on Sr_2IrO_4

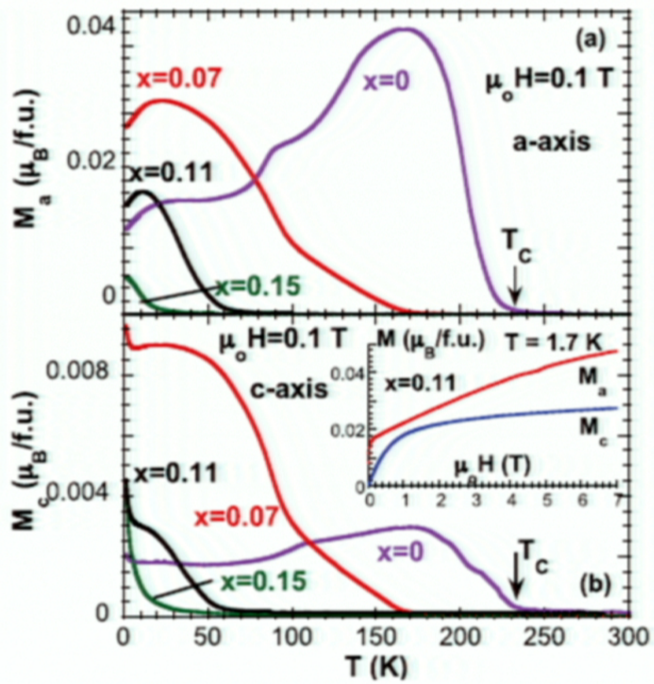


- Isoelectronic: $Sr_{2\text{-}x}A_xIrO_4$(A = Ba, Ca), $Sr_2Ir_{1\text{-}x}TM_xO_4$ (TM = Co, **Rh?**)
- e^--doping: $Sr_2IrO_{4\text{-}\delta}$, $Sr_{2\text{-}x}La_xIrO_4$, $Sr_2Ir_{1\text{-}x}Pt_xO_4$
- Hole-doping: $Sr_{2\text{-}x}K_xIrO_4$, $Sr_2Ir_{1\text{-}x}TM_xO_4$ (TM = Ru, Mn, Fe, Ti, **Rh?**)

What is the effect of Rh-doping?



T. Qi et al, PRB (2012)



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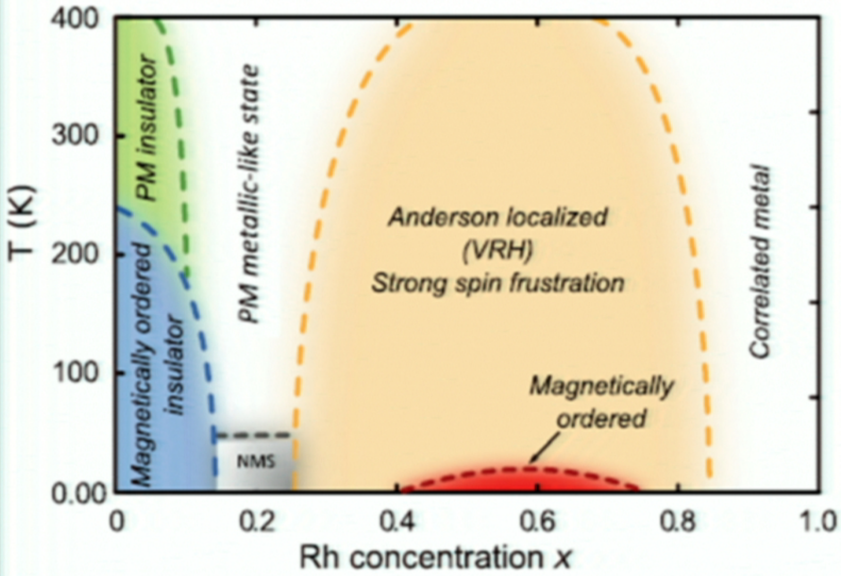

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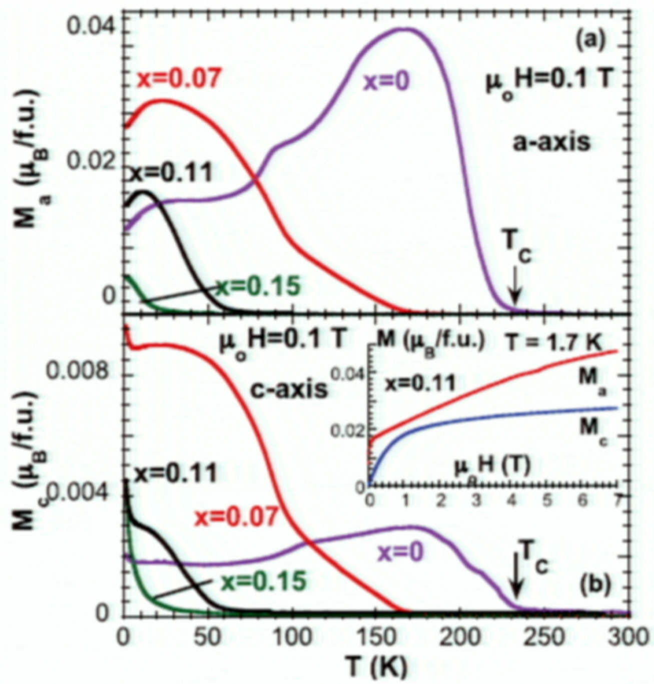

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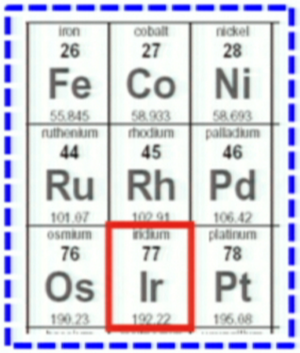
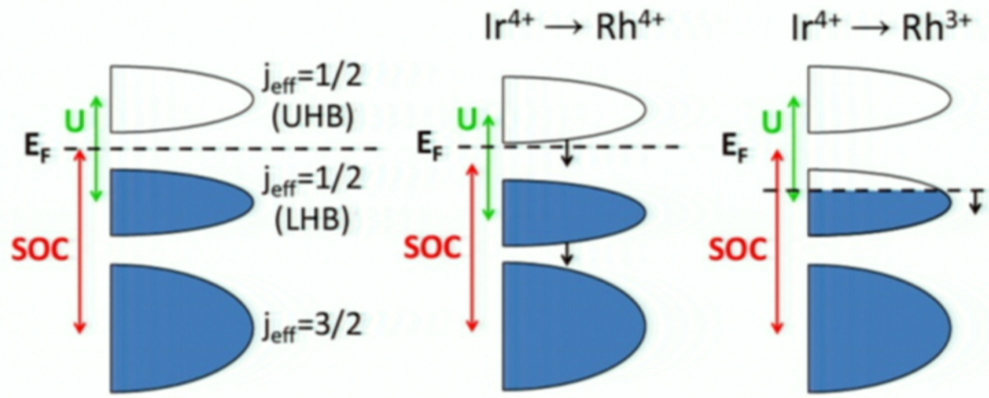
What is the effect of Rh-doping?

- **Substitute Rh^{4+} ($4d^5$) for Ir^{4+} ($5d^5$)?** → isoelectronic doping, tune SOC [*Qi et al, PRB (2012), Lee et al, PRB (2012)*]





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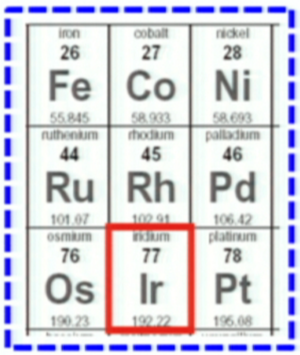

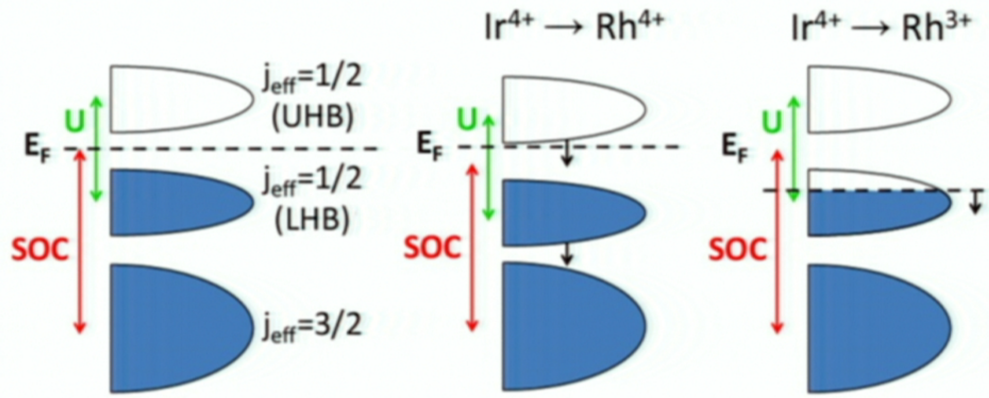

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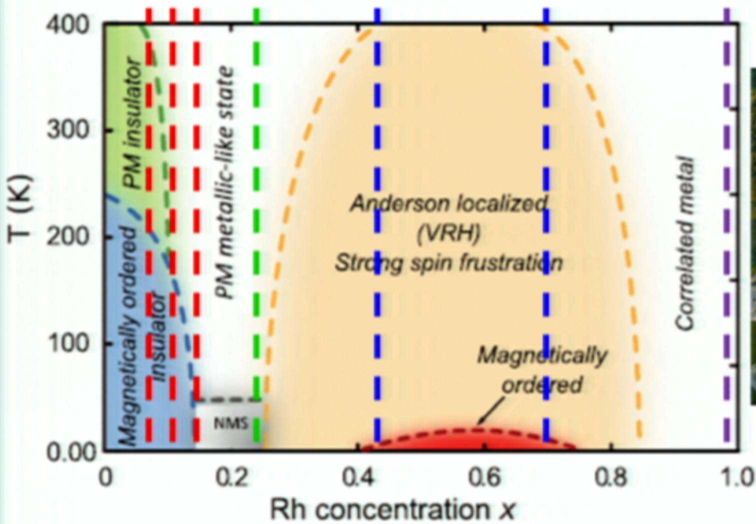
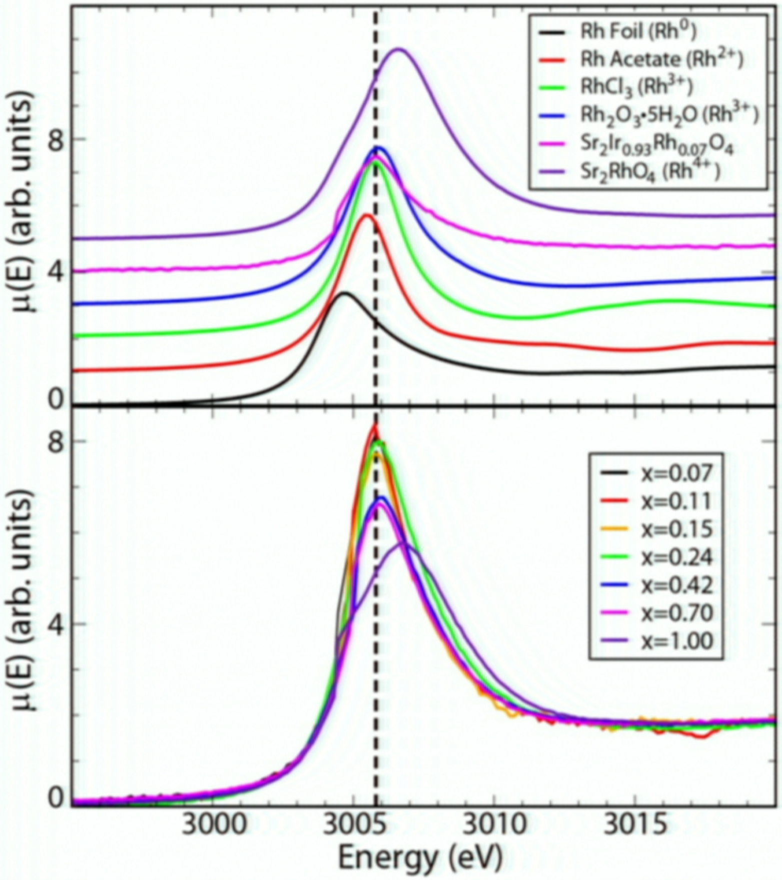
Experimental Details

- Single crystal samples of $Sr_2Ir_{1-x}Rh_xO_4$ with x = **0.07**, **0.11**, **0.15**, **0.24**, **0.42**, **0.70**, and **1.0**



- X-ray Absorption Spectroscopy (**XAS**) – SXRMB at CLS
- Resonant Magnetic X-ray Scattering (**RMXS**) – 6-ID-B at APS
- Resonant Inelastic X-ray Scattering (**RIXS**) – MERIX at APS

XAS: Oxidation State of $Sr_2Ir_{1-x}Rh_xO_4$



- Compare XAS white-line features at Rh L_3-edge:
- Suggests Rh dopant ions adopt a **Rh^{3+} state** for x = 0.07 to x = 0.70
- $Sr_2Ir_{1-x}Rh_xO_4$ appears to be a hole-doped system

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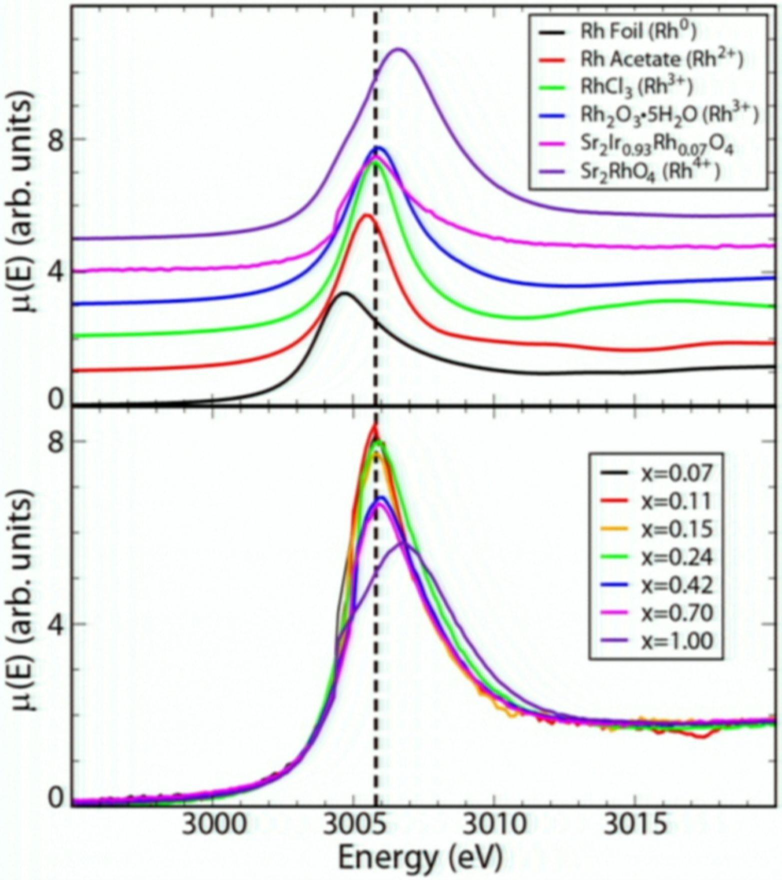

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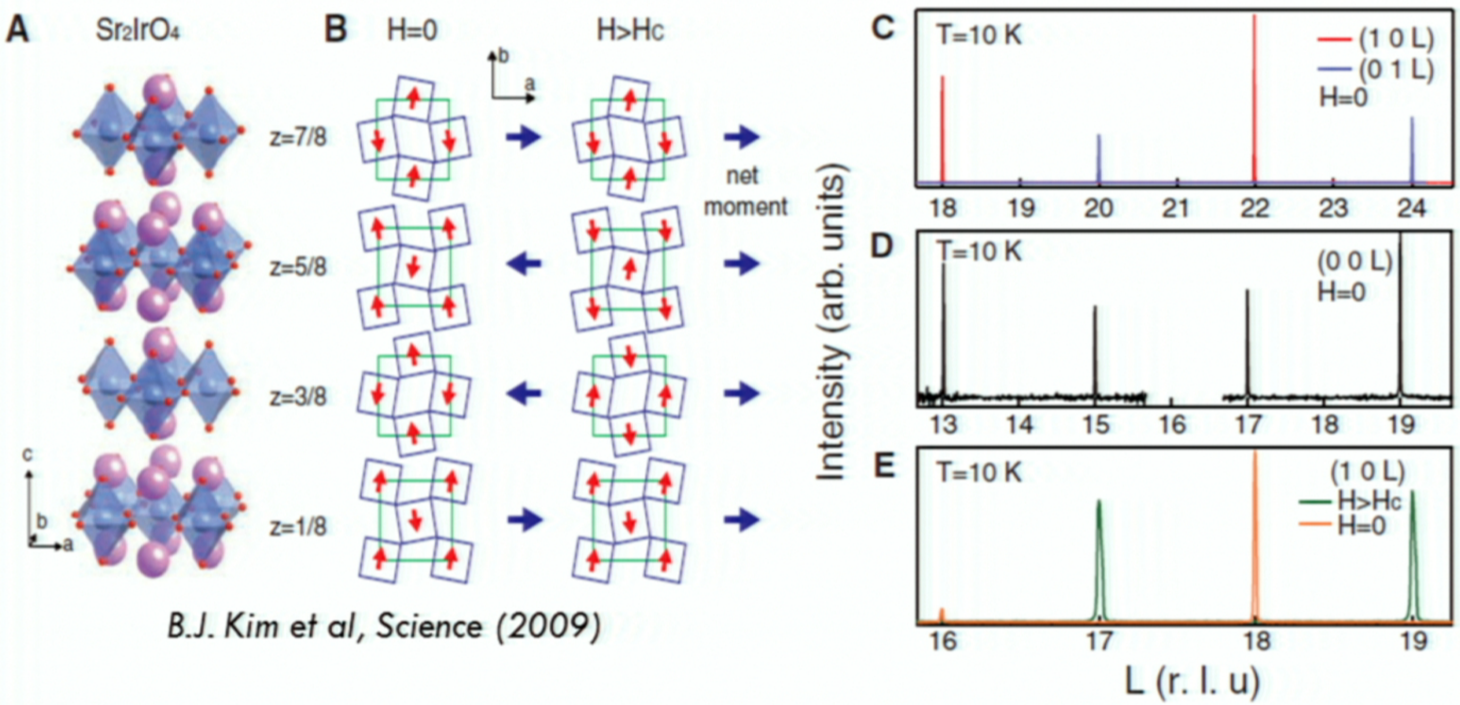
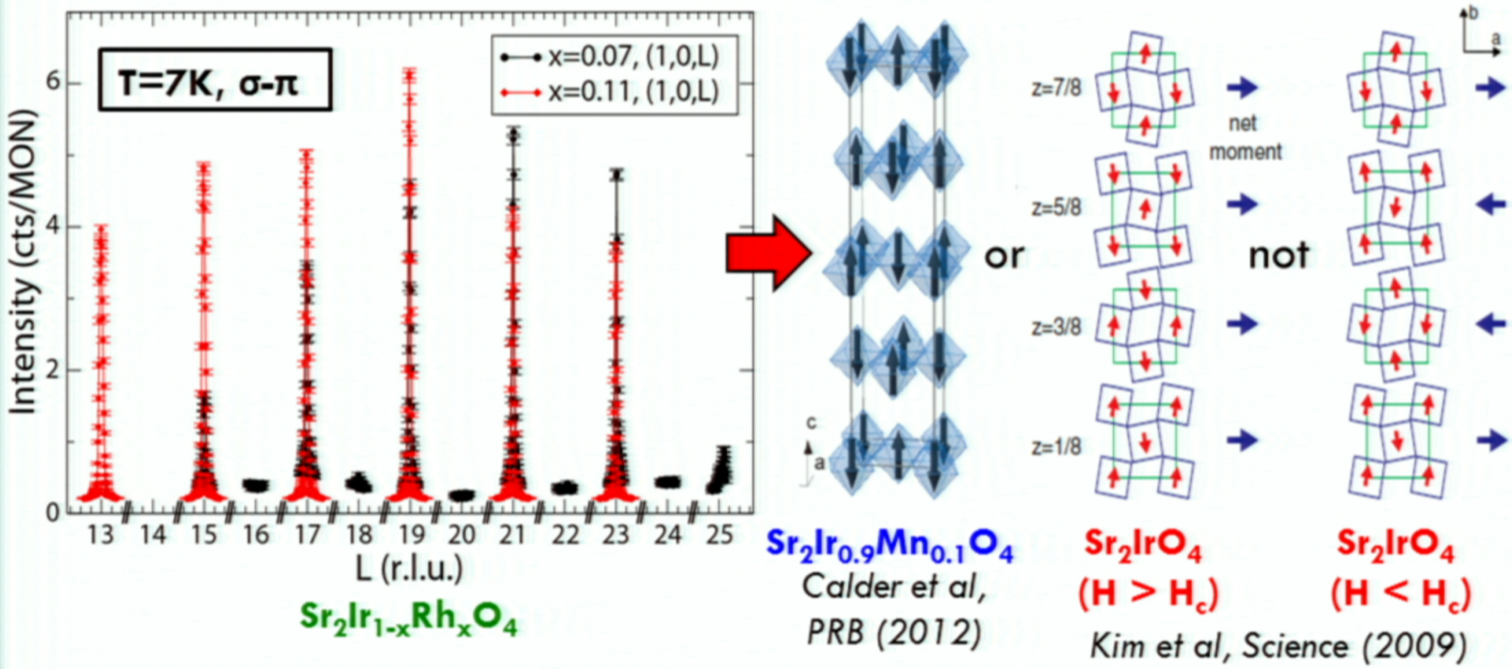
RMXS: Magnetic Structure of Sr_2IrO_4



B.J. Kim et al, Science (2009)

- Parent compound displays canted AF ground state
- Field-induced magnetic phase transition above H_C ~ 0.25 T

Magnetic Structure of $Sr_2Ir_{1-x}Rh_xO_4$



- Magnetic Bragg peaks observed at (0,1,L)/(1,0,L) for L=odd
- No magnetic peaks for (0,0,L), (1,1,L), (1/2,1/2,L), or (1/2,0,L)
- Doping-induced change in magnetic structure for $x \leq 0.07$

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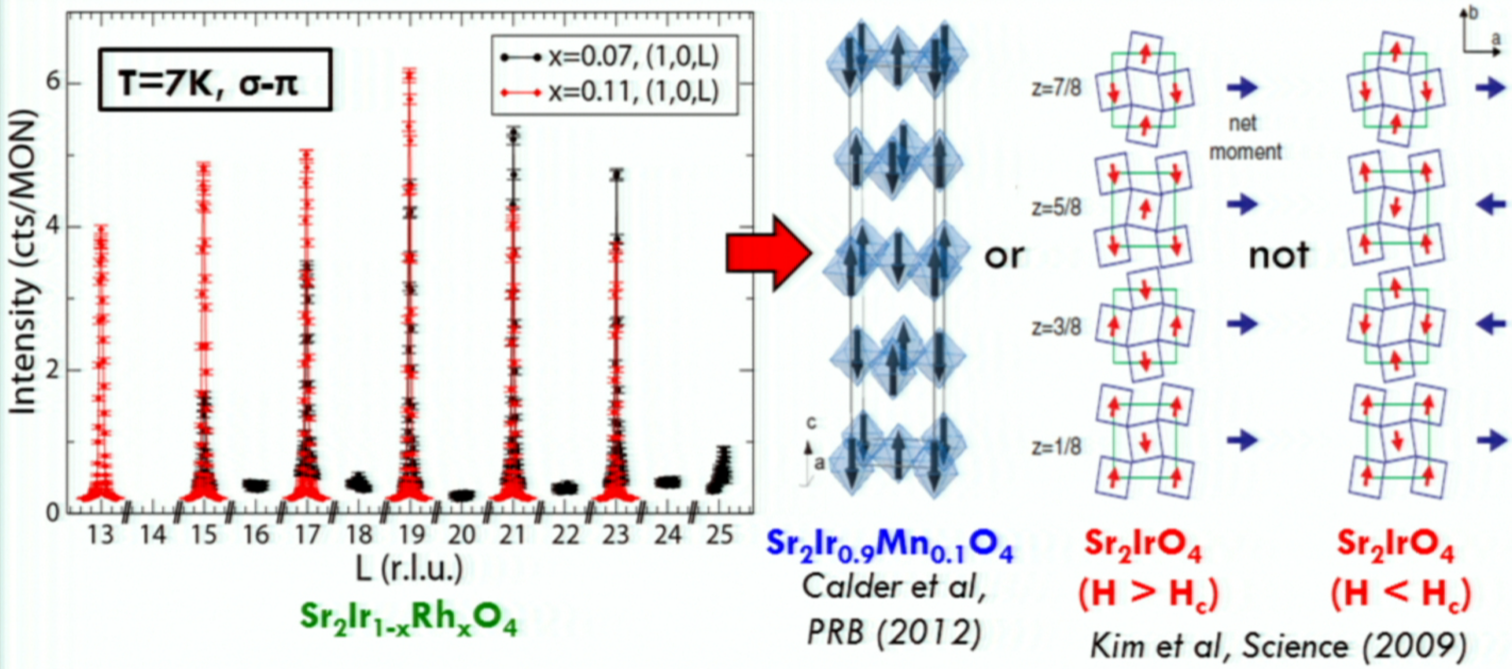

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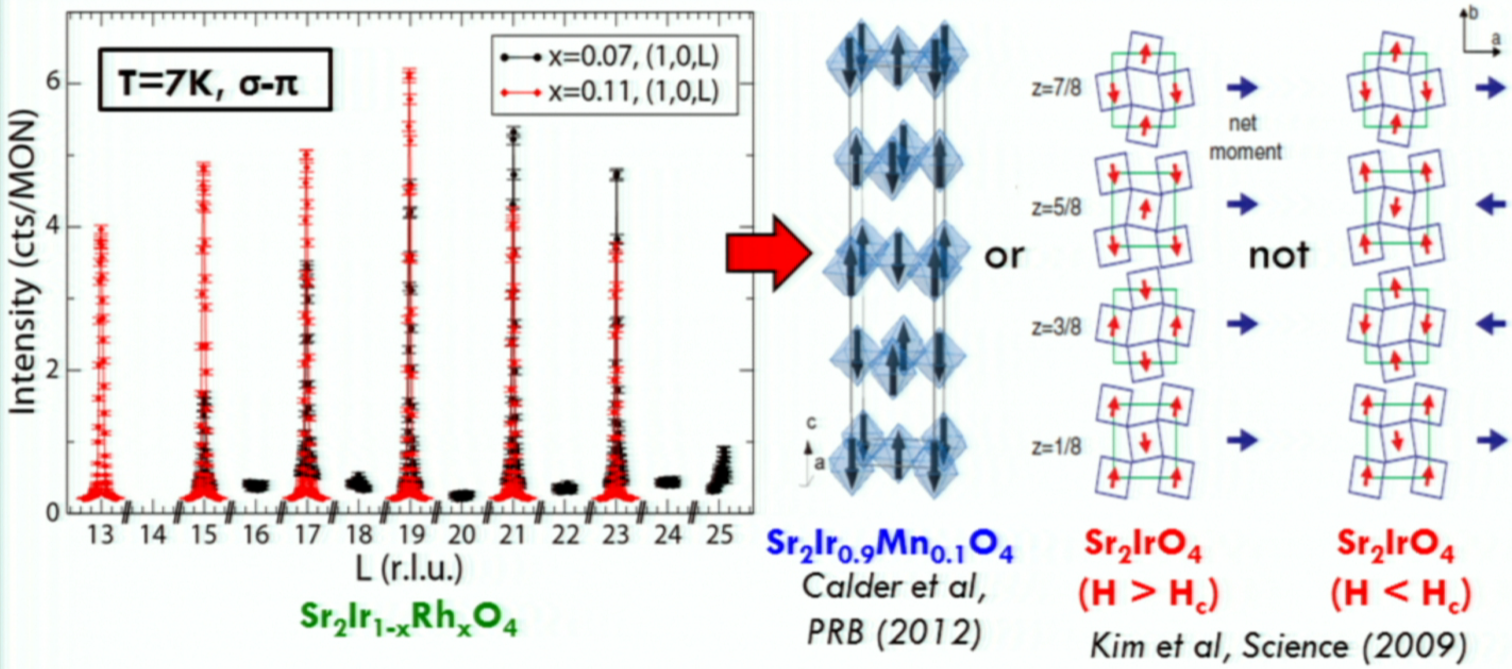

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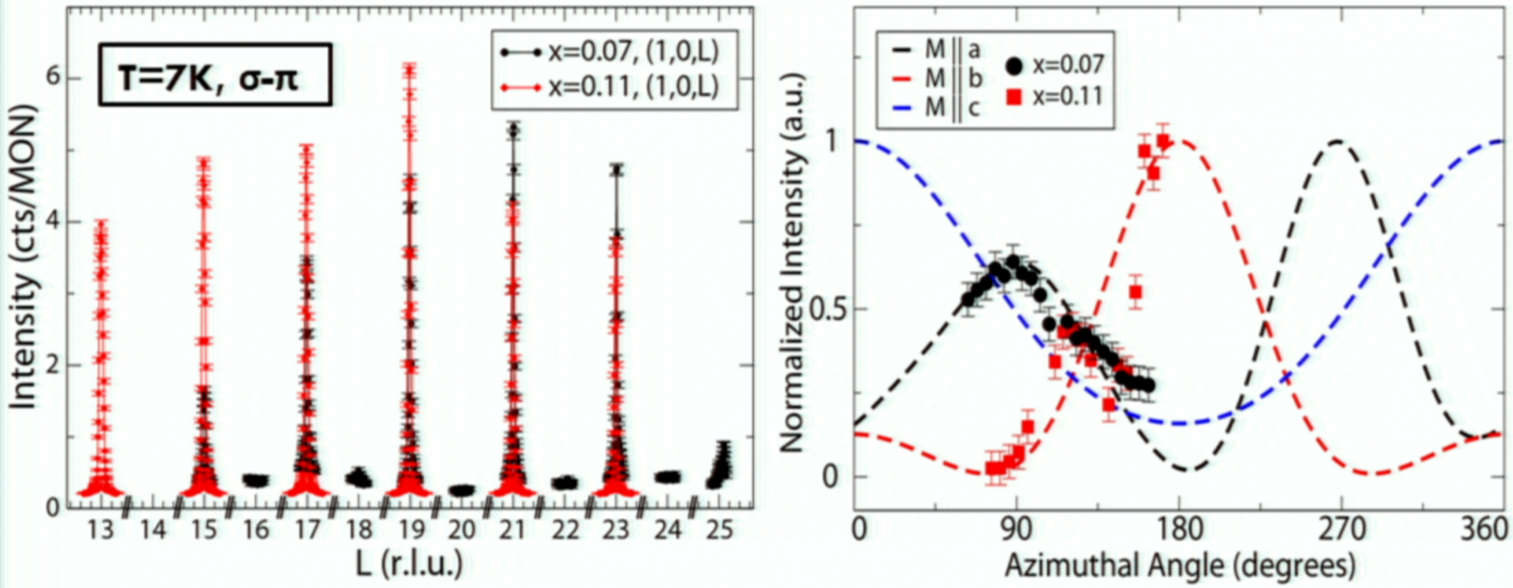
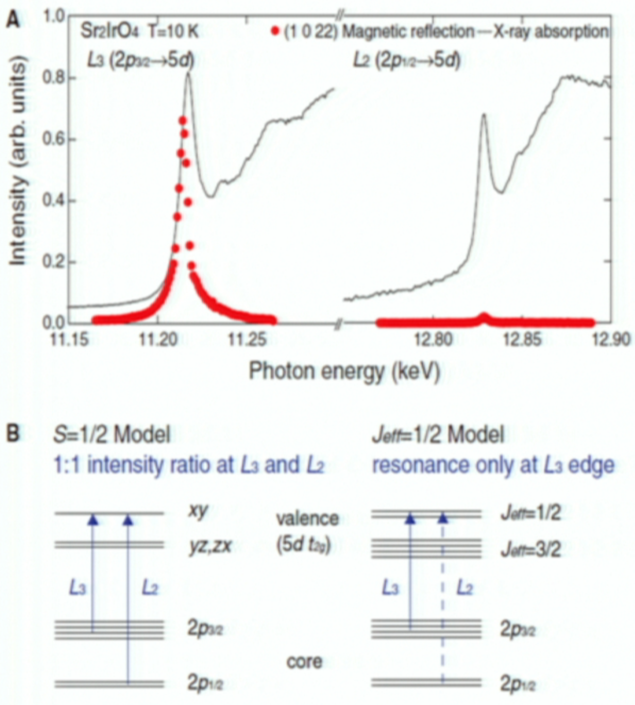


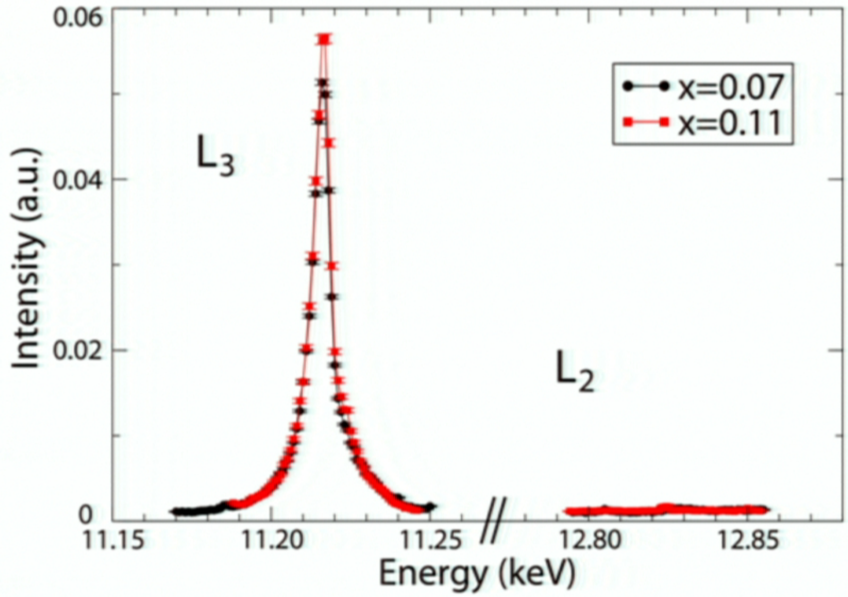
- Magnetic structure factor and azimuthal dependence confirm magnetic moments lie within the *ab*-plane
- Rh-doped Sr_2IrO_4 displays same magnetic structure as Sr_2IrO_4 in applied field ($H > H_c$)

Robustness of the $J_{eff}=1/2$ State

- Large I_{L3}/I_{L2} ratio is a defining signature of the $J_{eff} = 1/2$ state

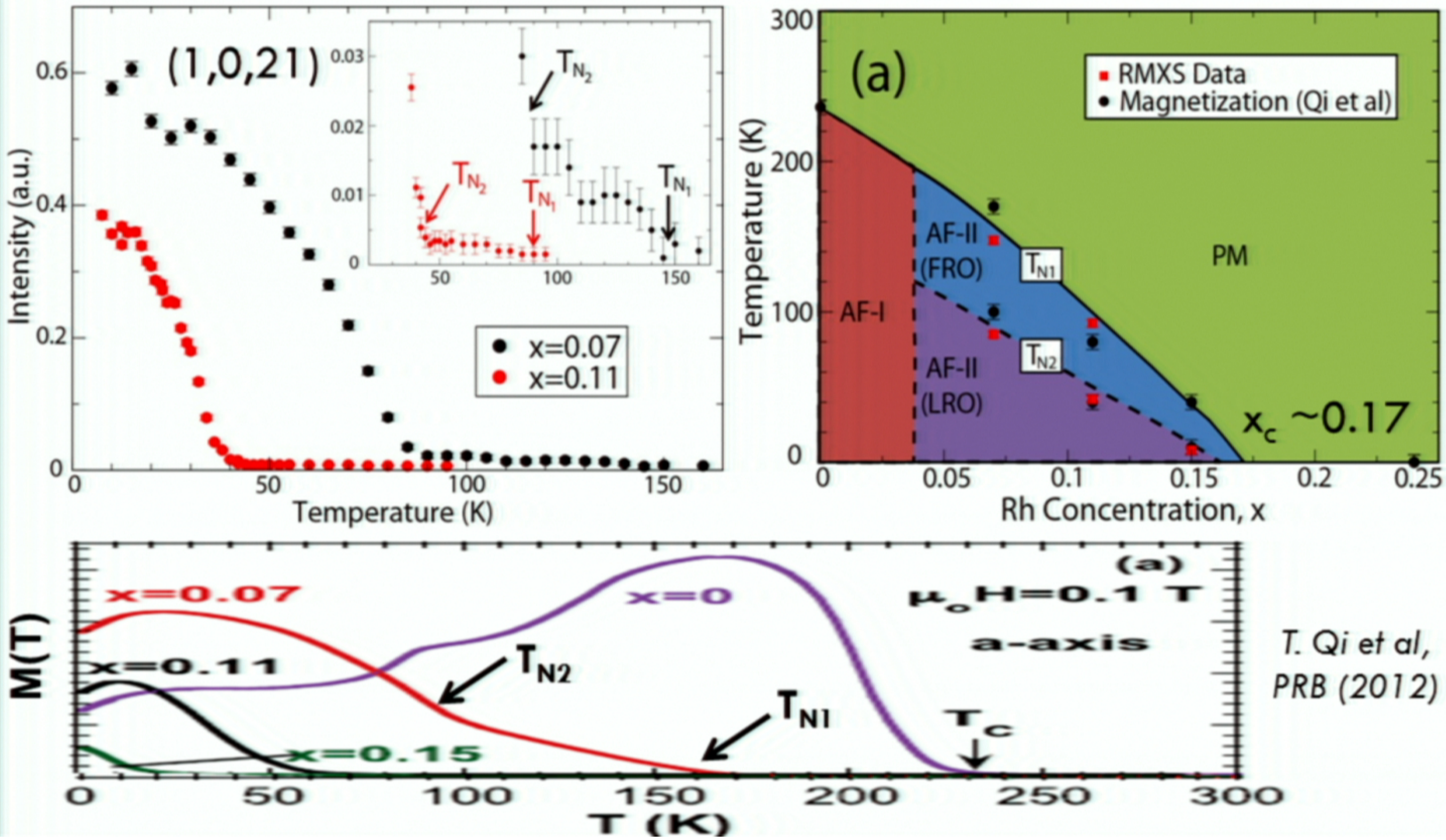


B.J. Kim et al, Science (2009)



- Minimum intensity ratio for Rh-doped Sr_2IrO_4 is $I_{L3}/I_{L2} > 200$
- Similar result observed in $Sr_2Ir_{0.9}Mn_{0.1}O_4$ (*Calder et al, PRB [2012]*)

Magnetic Phase Diagram of $Sr_2Ir_{1-x}Rh_xO_4$



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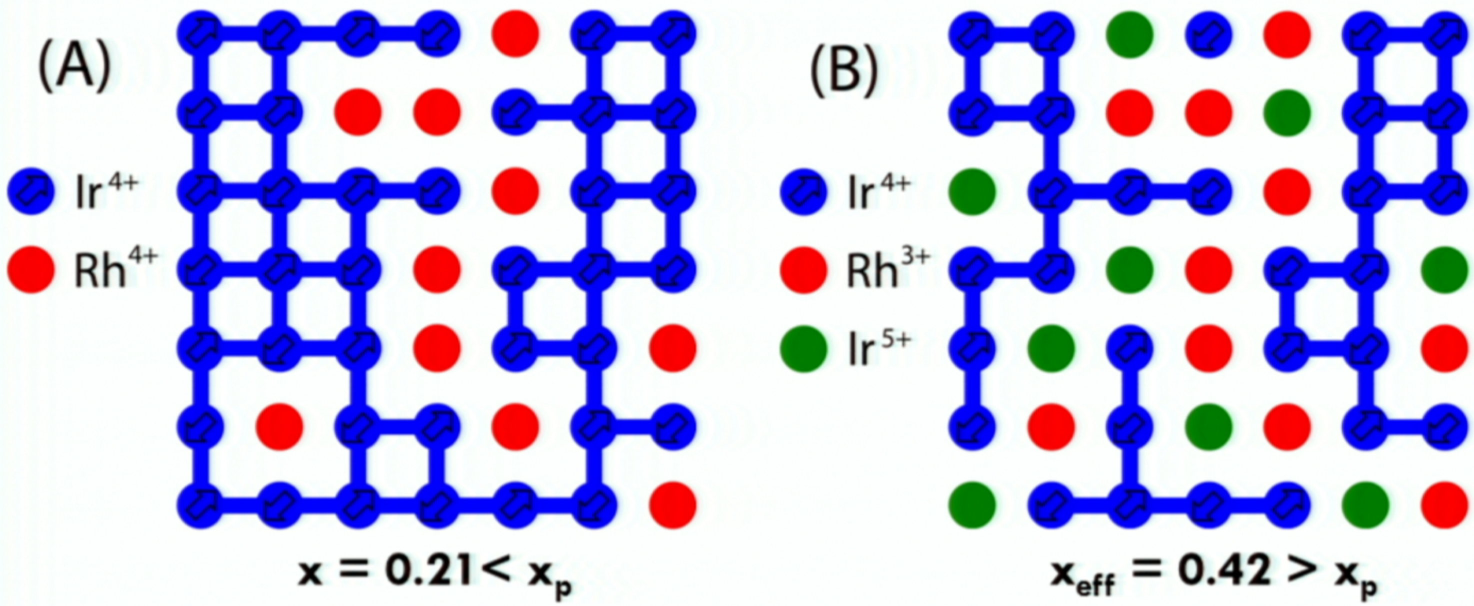
Implications of the Phase Diagram

- Can magnetic properties be explained using a percolation picture?
- S=1/2 square lattice Heisenberg AF has $x_p = 0.407$
- Appears to provide good description of highly doped cuprates
- ex. $La_2Cu_{1-x}(Mg,Zn)_xO_4$ [*Vajk et al, Science (2002)*]



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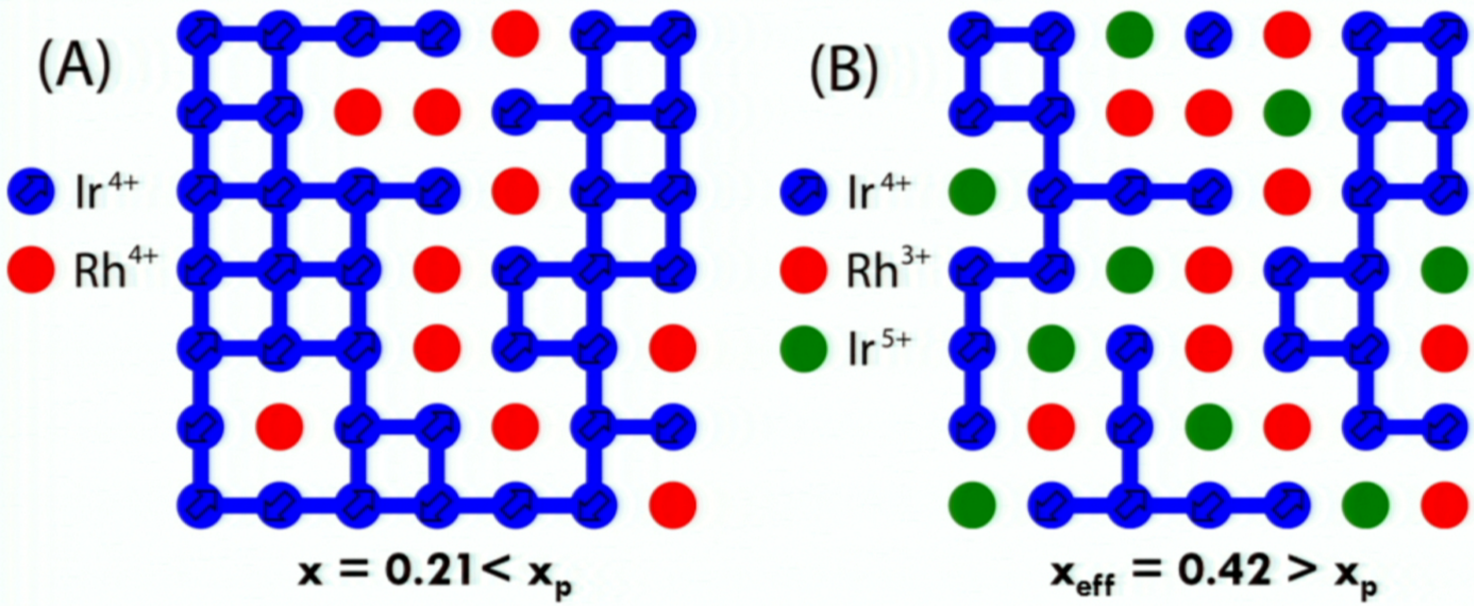
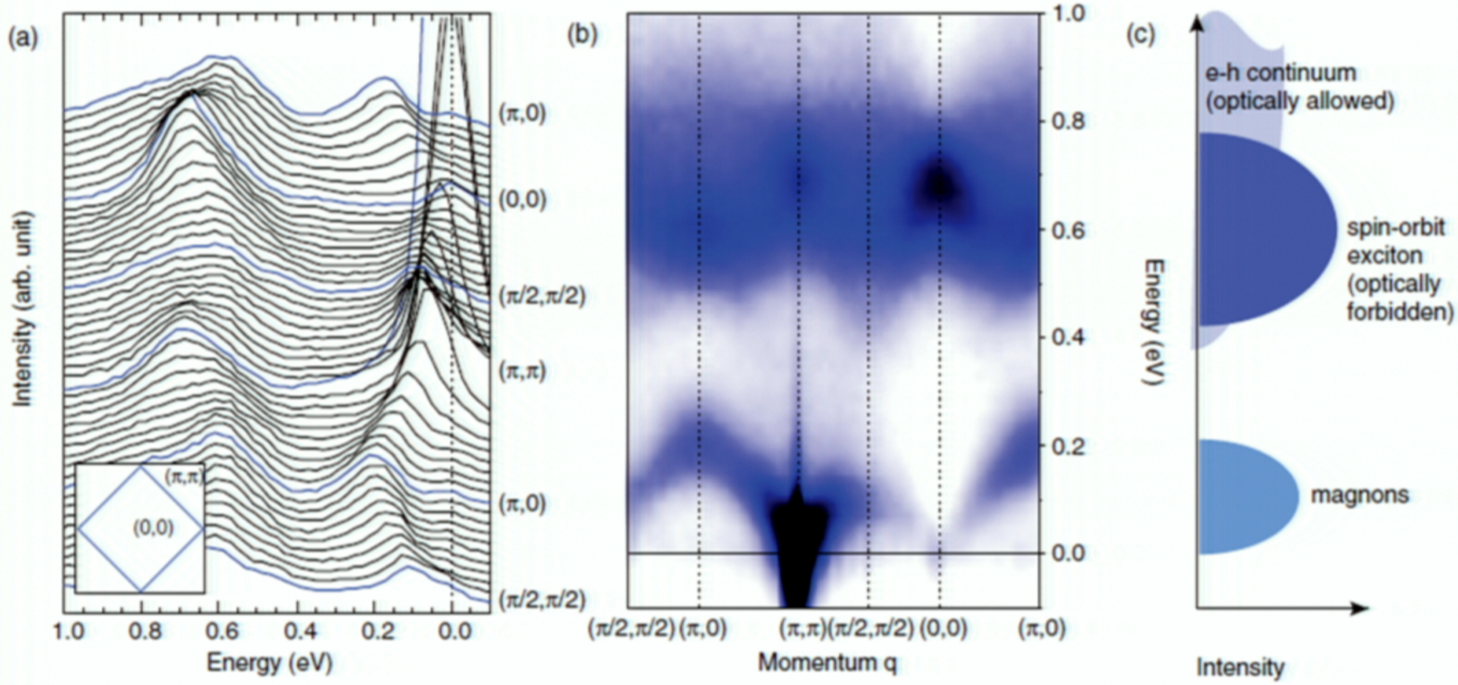


RIXS: Magnetic Excitations in Sr_2IrO_4

- Ir L_3-edge RIXS measurements on the parent compound:



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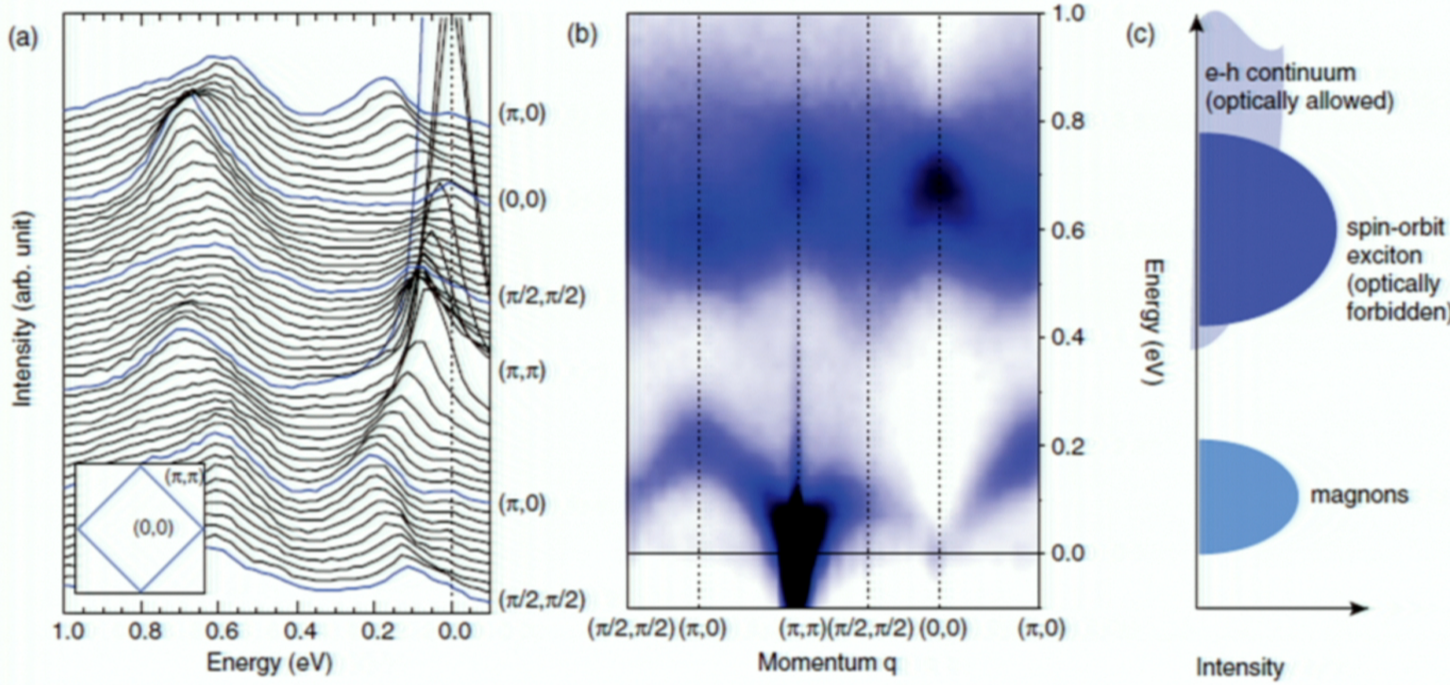

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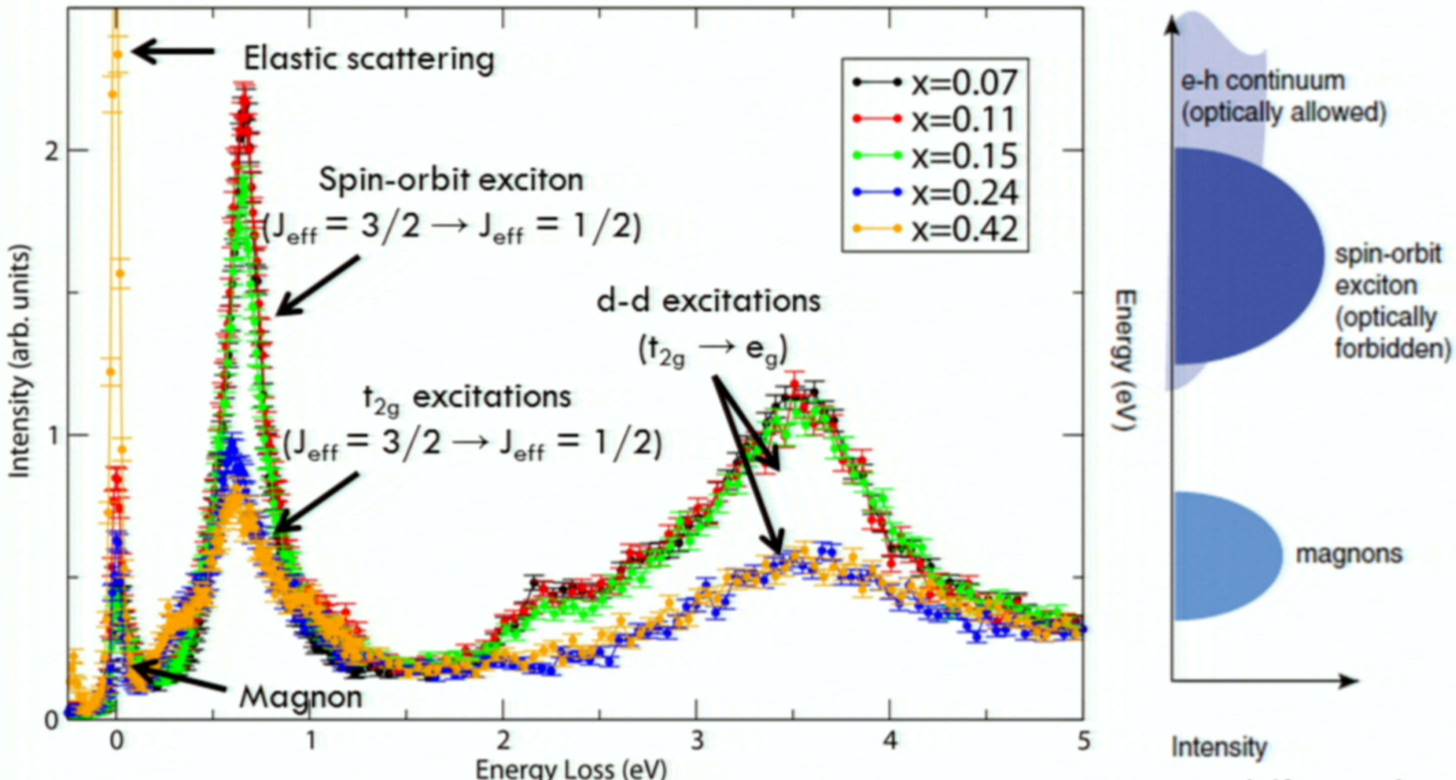
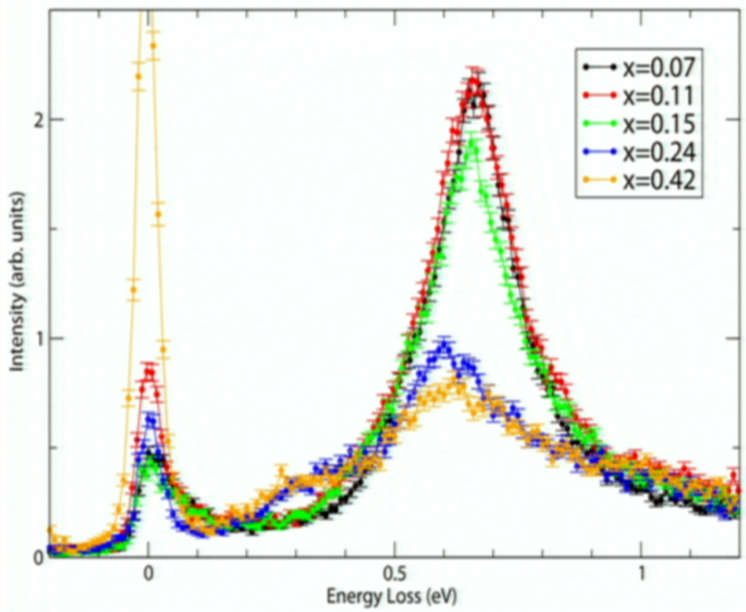
RIXS Measurements on $Sr_2Ir_{1-x}Rh_xO_4$

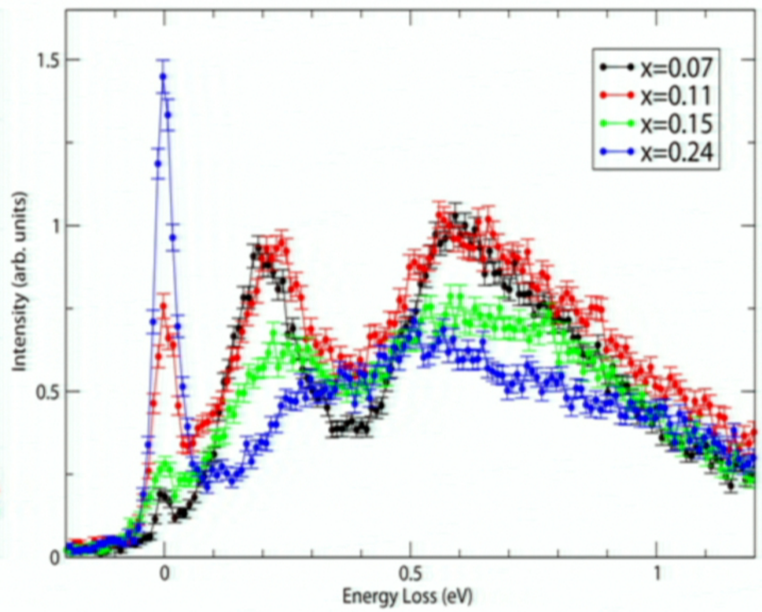


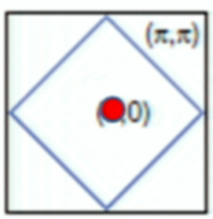
$Q = (0,0,33)$, $E_i = 11.216$ keV, $T \sim 10K$

J. Kim et al, PRL (2012)

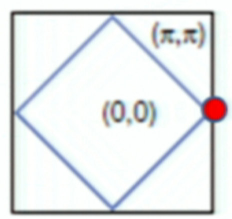
Magnetic Excitations in $Sr_2Ir_{1-x}Rh_xO_4$





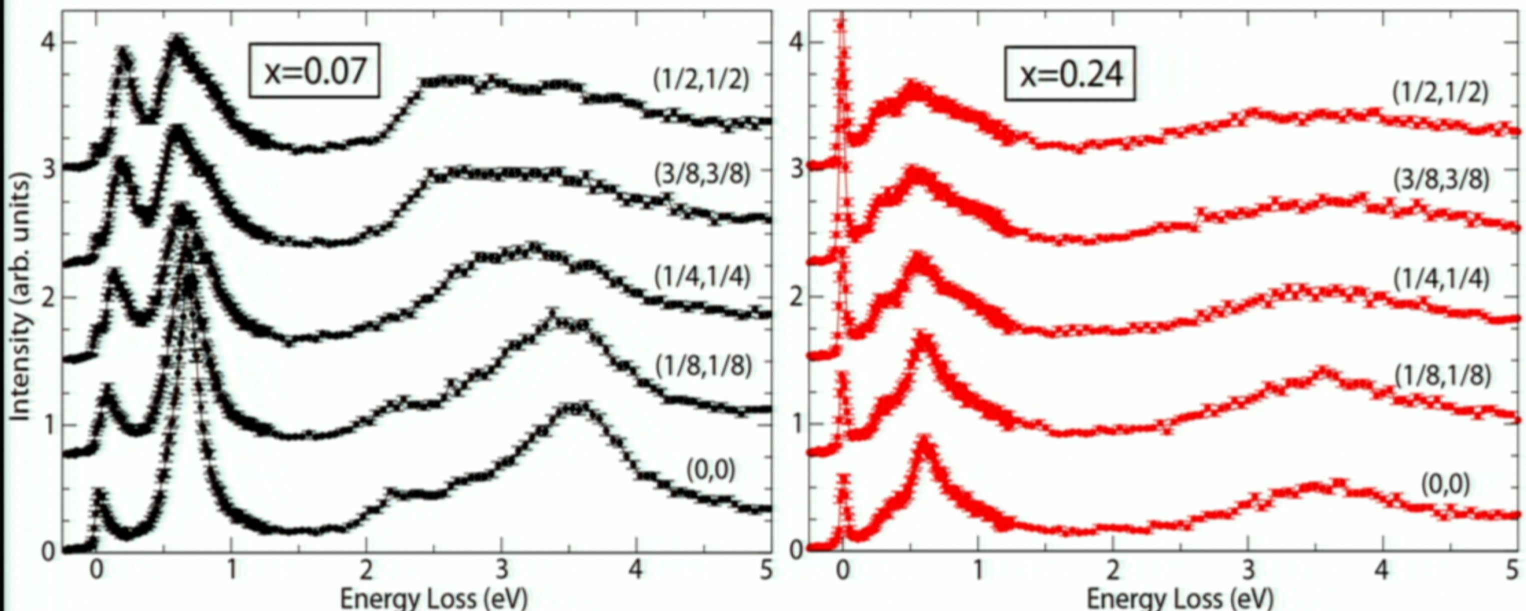


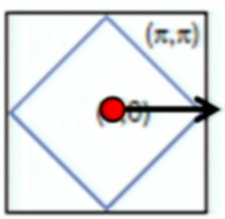
Q=(0,0,33)
Zone Center



Q=(1/2,1/2,33)
Zone Boundary

Orbital Excitations in $Sr_2Ir_{1-x}Rh_xO_4$





- Within the magnetically ordered state ($x < x_c$), orbital excitations exhibit significant dispersion
- Intensity and dispersion of these modes is dramatically reduced above x_c

Summary and Conclusions

- **$Sr_2Ir_{1-x}Rh_xO_4$** – a doped spin-orbital Mott insulator
- Rh dopants adopt 3+ oxidation state (hole-doping) not 4+ (isoelectronic)
- Doping-induced change in magnetic structure ($x \leq 0.07$)
- Magnetic order suppressed above $x_c \sim 0.17$
- Doping does not appear to disrupt the $j_{eff}=1/2$ state
- Well-defined magnons and spin-orbit excitons for $x < x_c$
- Magnetic excitations broaden/harden with increasing x
- Orbital excitations strongly coupled to magnetic ground state