Isospin Dynamics in Iridates revealed by Resonant inelastic x-ray scattering

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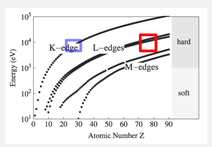
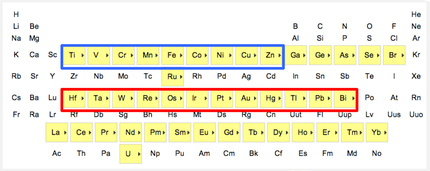
Abstract

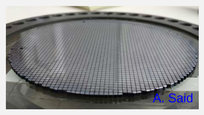
Iridium oxides with strong relativistic spin-orbit coupling have received much attention due to intriguing sets of novel electronic and magnetic phases and phenomena, which were imagined for a long time but not realized in real world. Among many materials, Sr_2IrO_4 and $Sr_3Ir_2O_7$ provide model cases for investigating essential aspects of the magnetic exchange interactions of the so-called J_{eff} isospin states. We present momentum-, and energy-resolved measurements of collective excitations of the spin-orbit coupled composite states in iridium oxide materials by resonant inelastic x-ray scattering (RIXS). The J_{eff}=1/2 isospin dynamics of Sr_2IrO_4 can be well described by the isotropic Heisenberg model. The dynamics of *d-d* excitations across the spin-orbit coupling split levels mirrors the fundamental process of an analogous one-hole propagation in a background of ordered spins which renders the low-energy effective physics of Sr_2IrO_4 much akin to that in superconducting cuprates. The J_{eff}=1/2 isospin dynamics of of $Sr_3Ir_2O_7$ shows a marked departure from the Heisenberg model due to the strongly enhanced bond-directional pseudodipolar interactions, which are essential for a realization of the Kitaev model in a honeycomb lattice with a spin-liquid ground state.

5d transition metal *L* edge RIXS spectroscopy

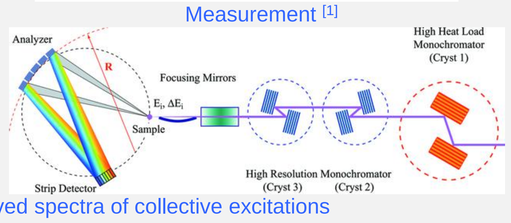




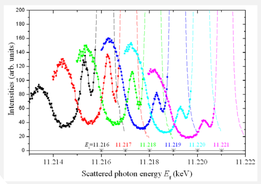
Si 844 Analyzer

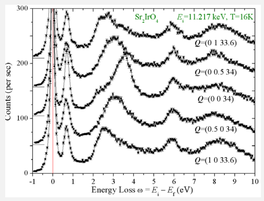


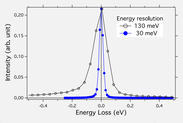
Measurement [1]



Momentum/Energy-resolved spectra of collective excitations



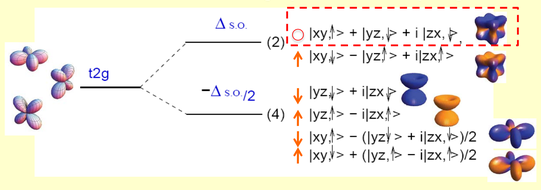




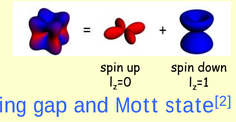
Current energy resolution ~ 30meV

Iridium oxides

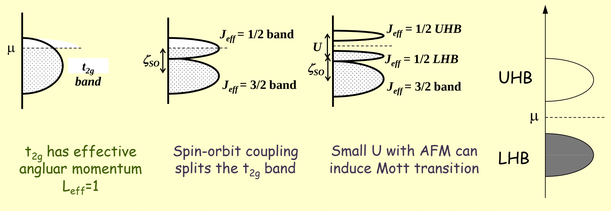
Strong Spin-Orbit couplings



J_{eff}=1/2 Isospin[2]



Nature of Insulating gap and Mott state[2]

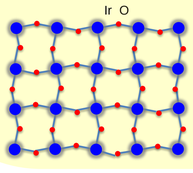


t_{2g} has effective angluar momentum L_{eff}=1

Spin-orbit coupling splits the t_{2g} band

Small U with AFM can induce Mott transition

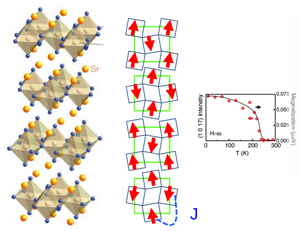
Exchange interactions in square lattice[2]



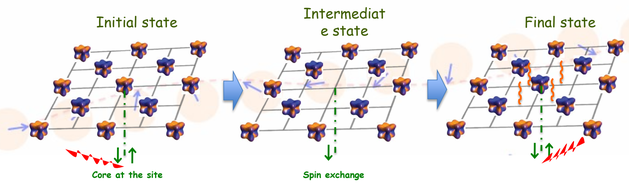
$$\mathcal{H}_{ij} = J_1 \vec{S}_i \cdot \vec{S}_j + J_2 (\vec{S}_i \cdot \vec{r}_{ij})(\vec{r}_{ij} \cdot \vec{S}_j)$$

Heisenberg Pseudodipolar

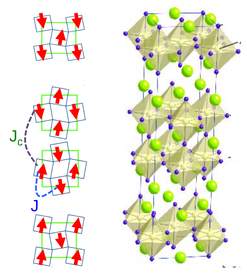
Sr_2IrO_4

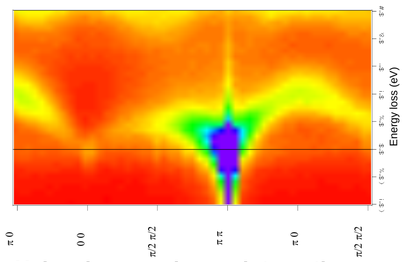


Magnon creation after the RIXS process

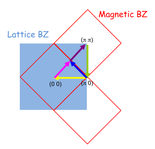


$Sr_3Ir_2O_7$

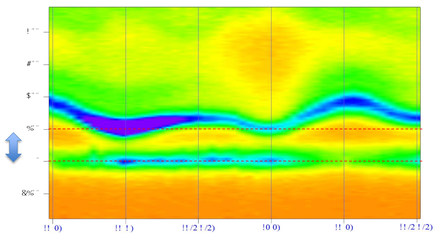




Full dynamics of J_{eff}=1/2 isospin revealed by RIXS



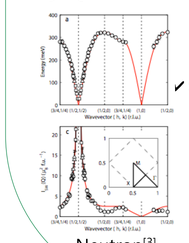
Large magnon gap



Heisenberg exchange interaction

La_2CuO_4 (J=60, J_1=-20, J_2=15meV) Sr_2IrO_4

: Parent compound of High-T_C Superconductors

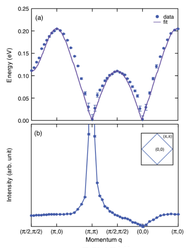


✔ Single band Mott Hubbard

✔ 2D square lattice

✔Spin ½

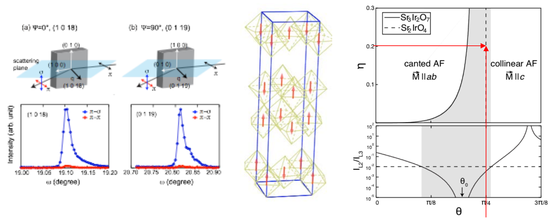
Neutron[3]

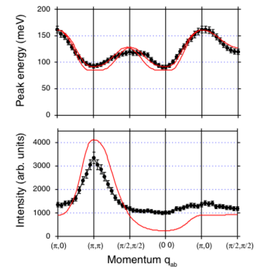


RIXS[4]

Strongly enhanced bond-directional pseudodipolar interactions

c-axis collinear antiferromagnetic structure[5]



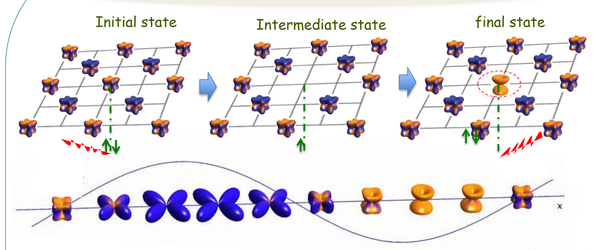


Larger window for the collinear AF in $Sr_3Ir_2O_7$ due to the isotropic wavefunction of isospins.
Large magnon gap due to large $\eta=J_H/U$ with large θ.

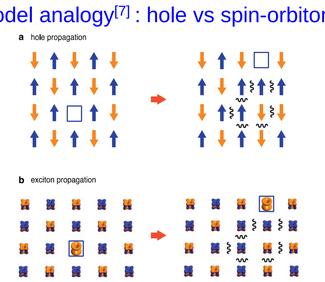
$\theta = 0.26\pi$ and $\eta = 0.24$.

J	J_c	J_2	J_3	J_{2c}	Γ	Γ_c	D	D_c
93	25.2	11.9	14.6	6.2	4.4	34.3	24.5	28.1

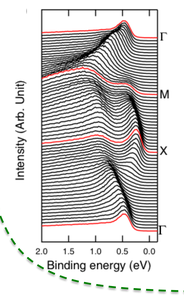
Spin-orbiton creation after the RIXS process

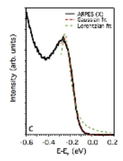


t-J model analogy[7] : hole vs spin-orbiton



Missing quasiparticles in ARPES of Sr_2IrO_4[6]

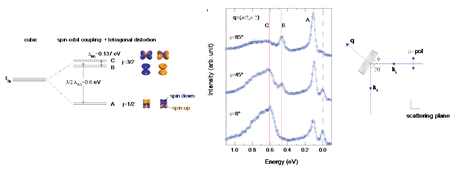




Two scenarios

- Franck-Condon broadening of the spectral functions the strong interactions between holes and optical phonons.
- Charged (Coulomb) defects

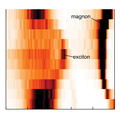
RIXS matrix elememts and selective probe

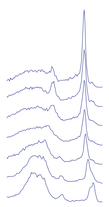


Selective mapping of the two exciton modes and their comparison to SCBA calculations. [7]

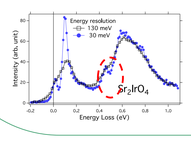
Asymmetric quasiparticle intensity[8]

: A generic feature predicted for a hole in a *t-J* model and indirectly inferred from ARPES measurements on cuprates, which possibly accounts for the Fermi arc observed in the doped case as due to the strongly suppressed shadow band intensity.



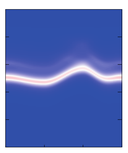


Resolution improvement and observation of sharp exciton



Orbital exciton by RIXS is a better realization of the *t-J* model than ARPES

✔ Charge neutral
→ Less lattice deformation and so less Frank-Condon broadening and no effects from Coulomb defects.
✔ Bulk-sensitive
→Immune to the surface defects



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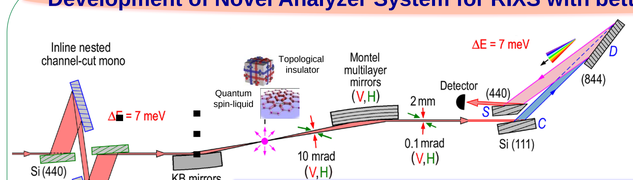
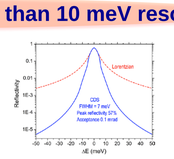
Development of Novel Analyzer System for RIXS with better than 10 meV resolution





Greatly improved tails of resolution function compared to conventional spherical analyzers.

Si-based *collimation-dispersion-selection* (CDS) analyzer combined with collimating multilayer mirrors

- Achieve energy resolution 2-20 meV for any hard x-ray energy and balance the resolution with flux to provide practical counts.
- Take RIXS energy resolution to a level competitive with neutron spectrometers, the current 'gold standard' for inelastic scattering with a capability to study far smaller (sub-mm) or thin-film samples .
- Compatible with a conventional scattered photon polarization analyzer.
- Augment the APS upgrade project, which is to provide an unprecedented combination of high-energy, high-brilliance, and nano-scale focal spots for modern scientific and technological research.

The Advanced Photon Source is funded by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences