

Probing Excitations in Pyrochlore Iridates with Resonant Inelastic X-ray Scattering

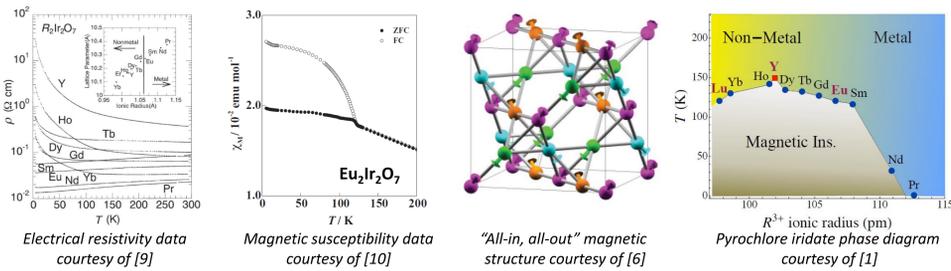
J.P. Clancy¹, H. Gretarsson¹, J. Kim², M.H. Upton², D. Casa², T. Gog², A.H. Said², B.-G. Jeon³, B. Lee³, K.H. Kim³, and Y.J. Kim¹

¹University of Toronto, ²Argonne National Laboratory, ³Seoul National University



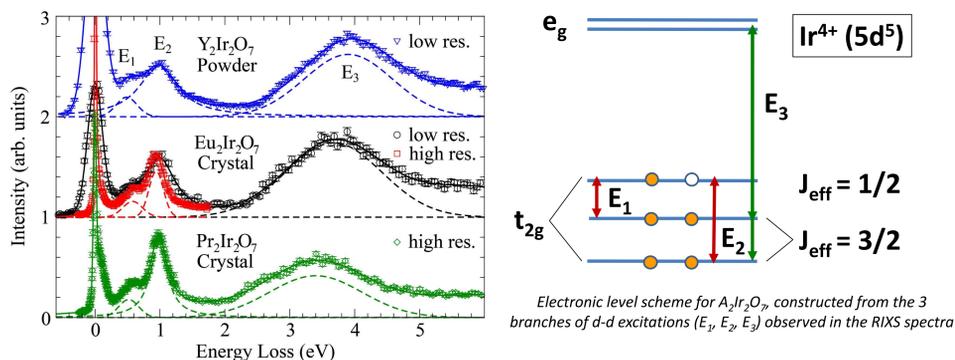
Novel Physics in Pyrochlore Iridates

- The pyrochlore iridates $A_2Ir_2O_7$ ($A = Y$ or lanthanide) have attracted considerable attention due to the potential for exotic physics driven by the interplay between electronic correlations, band topology, geometric frustration, and strong 5d spin-orbit coupling [1].
- Proposed ground states for these materials include: fractional topological insulators/topological Mott insulators [2,3], topological (or Weyl) semi-metals [4-6], axion insulators [6,7], and chiral spin liquids [8].
- Previous experimental work has shown that the electronic and magnetic properties of these materials are very sensitive to A-site cation size. [9-11] Physical behaviour can be tuned via chemical composition.



- The majority of pyrochlore iridates ($A = Dy$ to Nd) display simultaneous magnetic and metal-to-insulator phase transitions at $T_{MI} \sim T_c$. The chief exceptions are $A = Pr$, which remains metallic and paramagnetic down to $T < 30$ mK, and $A = Lu$ to Y , which order at T_c but remain non-metallic up to room temperature.
- The magnetic ground state below T_{MI} has been predicted to display "all-in, all-out" ($Q = 0$) magnetic order [5,6]. Although such a state is difficult to experimentally verify, recent resonant magnetic x-ray scattering [12] and μ SR [13] results are consistent with commensurate $Q = 0$ antiferromagnetic order.
- By measuring the characteristic excitation spectra of the pyrochlore iridates, we hope to:
 - Identify the relevant energy scales associated with magnetic interactions (J), crystal field splitting (CEF , Δ), and spin-orbit coupling (SOC , λ).
 - Shed further light on the nature of the low temperature magnetic ground state.

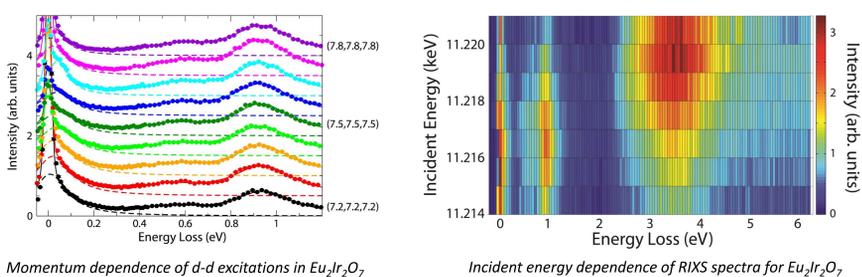
d-d Excitations in $A_2Ir_2O_7$ ($A = Y, Eu, Pr$)



- Compare experimental data with ab initio calculations by L. Hozoi et al [21]:

Compound	E_1 (Exp)	E_1 (Calc)	E_2 (Exp)	E_2 (Calc)	E_3 (Exp)	E_3 (Calc)	λ (SOC)	Δ (CEF)
$Y_2Ir_2O_7$	0.53 eV	0.58 eV	0.98 eV	0.94 eV	3.87 eV	3.48-4.84 eV	0.43 eV	0.56 eV
$Eu_2Ir_2O_7$	0.59 eV	0.60 eV	0.95 eV	0.91 eV	3.68 eV	3.39-4.72 eV	0.46 eV	0.46 eV
$Pr_2Ir_2O_7$	0.52 eV	---	0.98 eV	---	3.40 eV	---	0.42 eV	0.57 eV

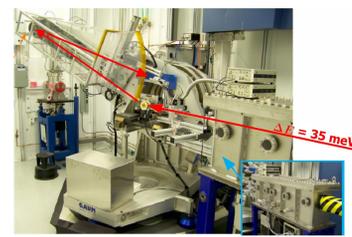
- Multiconfiguration self-consistent field (MCSCF) and multireference configuration interaction (MRCI) calculations performed on finite cluster (6 adjacent IrO_6 octahedra and neighbouring A-site cations).
- Good agreement between experimental and theoretical values.
- Model E_1 and E_2 with simple single-ion Hamiltonian: $H_0 = \lambda \mathbf{L} \cdot \mathbf{s} - \Delta I_z^2$
- Obtain reasonable values for spin-orbit coupling (λ), but surprisingly large trigonal crystal field splitting (Δ).
- Δ remains large, even for calculations with idealized crystal structure/no distortion of IrO_6 octahedra.
- Δ must originate from long-range anisotropy – trigonal field produced by neighbouring A-site ions and IrO_6 .



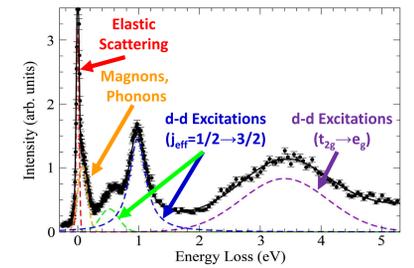
- d-d excitations in $Eu_2Ir_2O_7$ display no obvious dispersion – very different from perovskite iridates [17,18].
- Incident energy dependence demonstrates that these features occur at fixed energy loss (i.e. not fluorescence), and that they resonate near the $Ir L_3$ edge ($E_i \sim 11.216$ keV for t_{2g} , $E_i \sim 11.219$ keV for e_g).

Resonant Inelastic X-ray Scattering

- Resonant Inelastic X-ray Scattering (RIXS)** is a second-order scattering process which can be used to probe elementary excitations involving spin, orbital, charge, and lattice degrees of freedom [16].
- RIXS is particularly well-suited to the study of iridates [17-20]:
 - Element specific probe of magnetic and electronic properties.
 - Small sample volumes required (<10 mg).
 - Large resonant enhancement at $Ir L_3$ absorption edge ($2p_{3/2} \rightarrow 5d_{3/2,5/2}$ at $E_i = 11.215$ keV).
 - These materials are not amenable to conventional inelastic neutron scattering due to strong neutron absorption cross section and difficulty of synthesizing large single crystal samples.
- $Ir L_3$ -edge RIXS measurements carried out using the Advanced Photon Source at Argonne National Lab.



MERIX Spectrometer at Advanced Photon Source

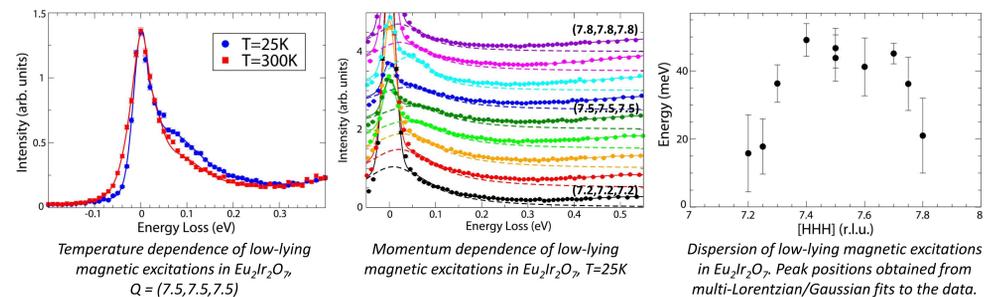


- High resolution setup:** Beamline 30-ID-B (MERIX) – Diamond-(111) primary mono, Si-(844) secondary mono, and spherical (2m) diced Si-(844) analyzer. Overall energy resolution of $\Delta E \sim 35$ meV.
- Low resolution setup:** Beamline 9-ID-B – Si-(111) primary mono, Si-(444) secondary mono, spherical (1m) diced Si-(844) analyzer. Overall energy resolution of $\Delta E \sim 175$ meV.
- Sample Synthesis:** Single crystal samples of $Pr_2Ir_2O_7$ and $Eu_2Ir_2O_7$ ($\sim 0.5 \times 0.5 \times 0.5$ mm³) grown using KF flux methods as described in [15]. Powder sample of $Y_2Ir_2O_7$ synthesized as described in [11].
- Electronic and magnetic properties of these systems appear to quite sensitive to sample stoichiometry [14]. Electron Probe Microanalysis (EPMA) used to determine single crystal sample of $Eu_2Ir_2O_7$ has actual stoichiometry of $Eu_{2(1+x)}Ir_{2(1-x)}O_{7+\delta}$, with $x = 0.09(2)$ and $\delta = 0.06(2)$

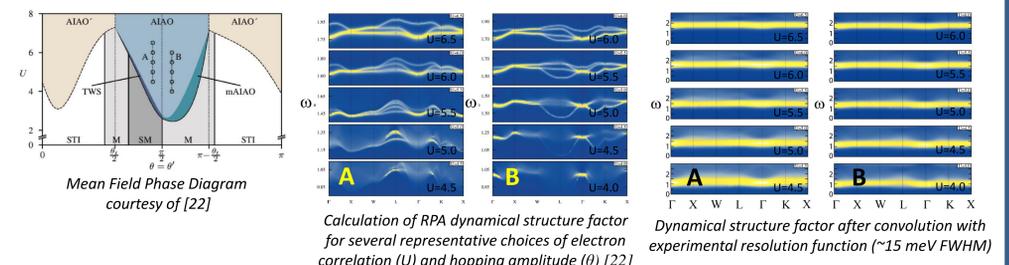
Compound	a (Å)	x	Electronic	Magnetic
$Y_2Ir_2O_7$	10.18	0.333	Always Non-Metallic	$T_c \sim 150$ K
$Eu_2Ir_2O_7$	10.28	0.331	$T_{MI} \sim 120$ K	$T_c \sim 120$ K
$Pr_2Ir_2O_7$	10.40	0.329	Always Metallic	$T_c < 30$ mK

Magnetic Excitations in $A_2Ir_2O_7$ ($A = Eu$)

- Investigate low-lying inelastic scattering in single crystal $Eu_2Ir_2O_7$ using high-resolution experimental set-up ($\Delta E \sim 35$ meV). Observe broad, dispersive feature at ~ 45 meV.
- Temperature dependence, incident energy dependence, and Q-dependence indicate that this feature is magnetic in origin.



- Dispersion of magnetic excitation along [HHH] is consistent with $Q = 0$ or all-in, all out ordering.
- Width of excitation is much broader than experimental resolution (FWHM ~ 150 -200 meV). Are finite lifetimes intrinsic or due to deviation from ideal stoichiometry?
- Compare observed magnetic excitation spectrum with theoretical calculations by E.K.H. Lee et al [22]:



- Underscores need for higher experimental resolution and further development of RIXS instrumentation.

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[1] W. Witczak-Krempa et al, arXiv:1305.2193 (2013).
 [2] D.A. Pesin and L. Balents, Nat. Phys. **6**, 376 (2010).
 [3] M. Kargarian et al, Phys. Rev. B **83**, 165112 (2011).
 [4] B.J. Yang et al, Phys. Rev. B **82**, 085111 (2010).
 [5] W. Witczak-Krempa et al, Phys. Rev. B **85**, 045124 (2012).
 [6] X. Wan et al, Phys. Rev. B **83**, 205101 (2011).
 [7] A. Go et al, Phys. Rev. Lett. **109**, 066401 (2012).
 [8] Y. Machida et al, Nature **463**, 210 (2010).
 [9] D. Yanagishima et al, J. Phys. Soc. Jpn. **70**, 2880 (2001).
 [10] N. Taira et al, J. Phys.: Condens. Matt. **13**, 5527 (2001).
 [11] K. Matsuhira et al, J. Phys. Soc. Jpn. **76**, 043706 (2005).

[12] H. Sagayama et al, Phys. Rev. B **87**, 100403(R) (2013).
 [13] S. Zhao et al, Phys. Rev. B **83**, 180402(R) (2011).
 [14] J.J. Ishikawa et al, Phys. Rev. B **85**, 245109 (2012).
 [15] J.N. Millican et al, Mater. Res. Bull. **42**, 928 (2007).
 [16] L.J.P. Ament et al, Rev. Mod. Phys. **83**, 705 (2011).
 [17] J. Kim et al, Phys. Rev. Lett. **108**, 177003 (2012).
 [18] J. Kim et al, Phys. Rev. Lett. **109**, 157402 (2012).
 [19] H. Gretarsson et al, Phys. Rev. B **87**, 220407 (2013).
 [20] H. Gretarsson et al, Phys. Rev. Lett. **110**, 076402 (2013).
 [21] L. Hozoi et al, arXiv:1212.4009 (2012).
 [22] E.K.H. Lee et al, Phys. Rev. B **87**, 214416 (2013).