

Title: Exotic Magnetism on the FCC Lattice of 5dn Double Perovskites

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Abstract: In the search for new exotic quantum states, the impact of strong spin-orbit interaction has been recently underlined with the discovery of the $\text{Jeff} = \frac{1}{2}$ spin orbital Mott state in the 5d5 layered perovskites iridates [1]. The double perovskite structure, where the magnetic ions form a face-centered-cubic (fcc) sublattice, can accommodate a large variety of 5d transition metal elements, and therefore offers an ideal playground for systematic studies of the exotic magnetic and non-magnetic ground states stabilized by strong spin-orbit coupling [2]. Here, we report time-of-flight neutron scattering measurements on the antiferromagnetic, frustrated, cubic double perovskite system Ba_2YOsO_6 . Its non-distorted fcc lattice is decorated with magnetic Os^{5+} (5d3) ions which undergo a magnetic transition to a long range ordered antiferromagnetic state below $T_N = 70$ K, as revealed by magnetic Bragg peaks occurring at the [100] and [110] positions. Our inelastic data reveals a large spin gap to the spin-wave excitations $\hat{\Gamma}' = 19(2)$ meV, unexpected for an orbitally quenched, d3 electronic configuration. We compare this result to the recent observation of a $\hat{\Gamma}'=5$ meV spin gap in the related cubic double perovskite Ba_2YRuO_6 (Ru^{5+} , 4d3) [3], and conclude to a stronger spin-orbit coupling present in the heavier, 5d, osmate system.

[1] B. J. Kim et al., Science 323, 1329 (2009).
[2] G. Chen, R. Pereira and L. Balents, Phys. Rev. B 82, 174440 (2010);
G. Chen and L. Balents, Phys. Rev. B 84, 094420 (2011).
[3] J. P. Carlo et al., Phys. Rev. B 88, 024418 (2013).



Exotic Magnetism on the FCC lattice of the 5d³ double perovskite Ba₂YO₃O₆

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C. A. Marjerrison, C. M. Thompson, J. E. Greedan

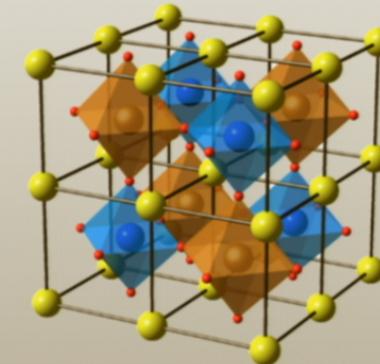
Department of Chemistry, McMaster University

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Quantum Condensed Matter Division, ORNL



4-Corners Symposium – PI Waterloo May 1st, 2014

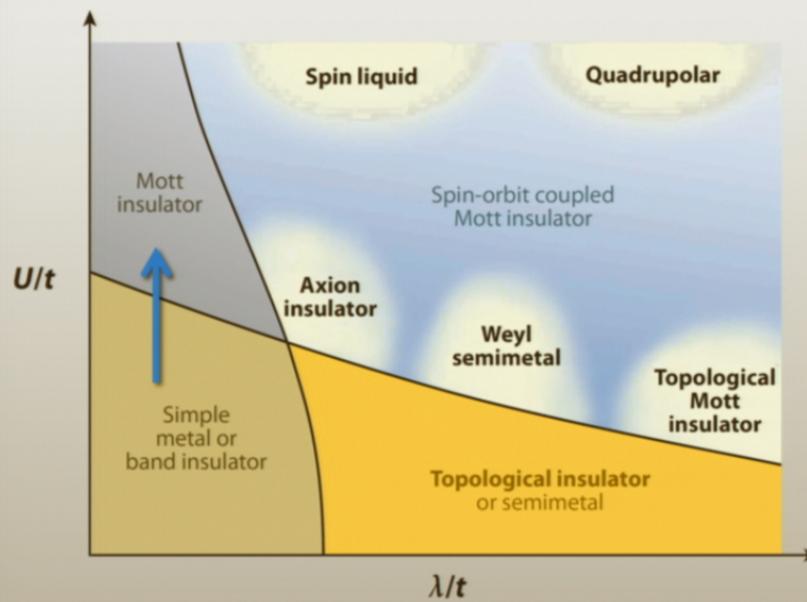


Spin-orbit coupled Mott Insulator

$$H = -t \sum_{i,j}^N (c_i^\dagger c_j + h.c.) + U \sum_i^N n_{i,\uparrow} n_{i,\downarrow} + \lambda \sum_i^N \mathbf{L}_i \cdot \mathbf{S}_i$$

(SOC term)

Electronic correlations



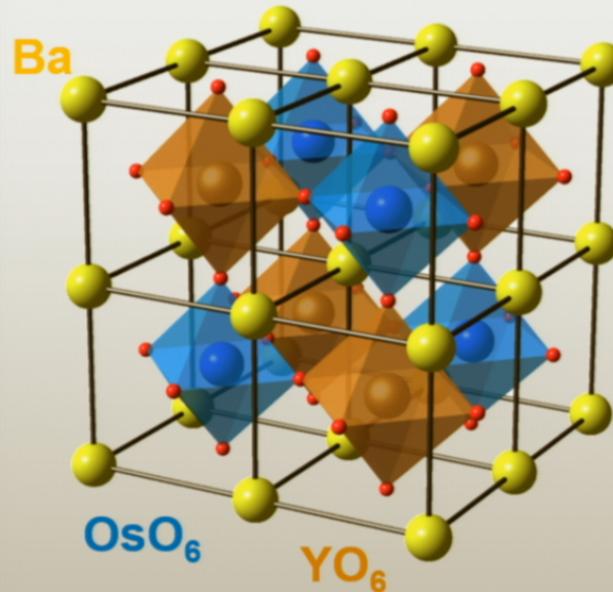
- Weak SOC & $U/t \gg 1$

From W. Witczak-Krempa, G. Chen, Y. B. Kim, and L. Balents,
Annual Review of Condensed Matter Physics, **5**, 57 (2014)

**Spin-orbit coupling
(SOC)**



Double perovskite structure

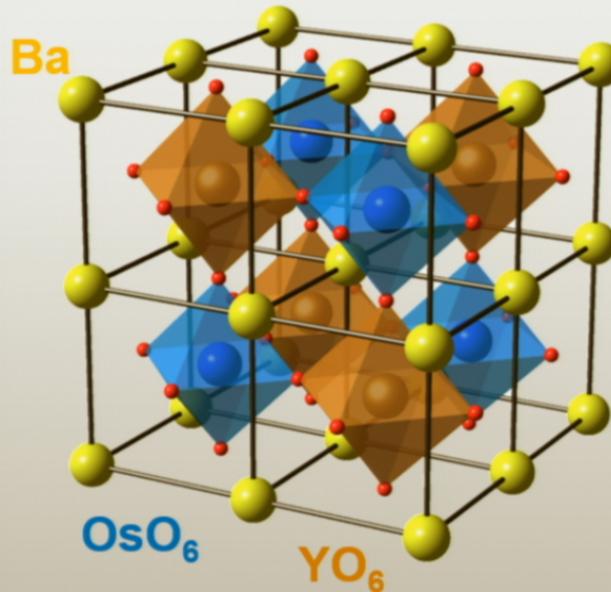


- Ba_2YO_6 Os⁵⁺ 5d³ S=3/2
 - * Ba_2YRuO_6 Ru⁵⁺ 4d³ S=3/2
- “Rock-salt” ordered double perovskite
Large SOC expected ($\lambda \sim Z^4$)

* T. Aharen *et al.*, PRB **80**, 134423 (2009)



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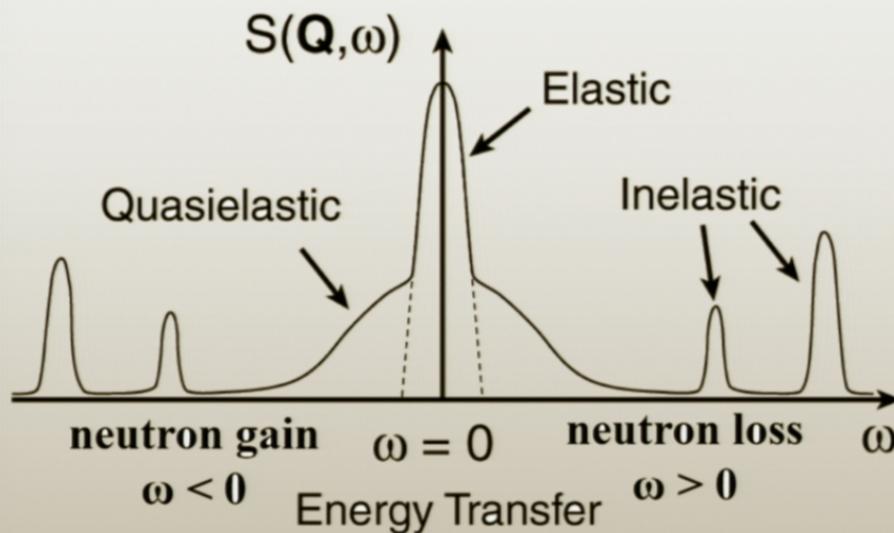


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- "Rock-salt" ordered double perovskite
Large SOC expected ($\lambda \sim Z^4$)
- **Cubic Fm $\bar{3}$ m structure**
No structural change from 290 to 3.5K
- Minute level of intersite mixing Os/Y as evaluated by ⁸⁹Y NMR
 $\text{Os/Y} < 0.5\%$
 $\text{Ru/Y} \sim 1\%$

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TOF inelastic neutron scattering



Elastic scattering

- Long/short range magnetic order
- Magnetic structure

Inelastic scattering

- (Spin-waves) excitations
- Hamiltonian of the system

Adapted from J.R.D. Copley,
NCNR Summer School 2013

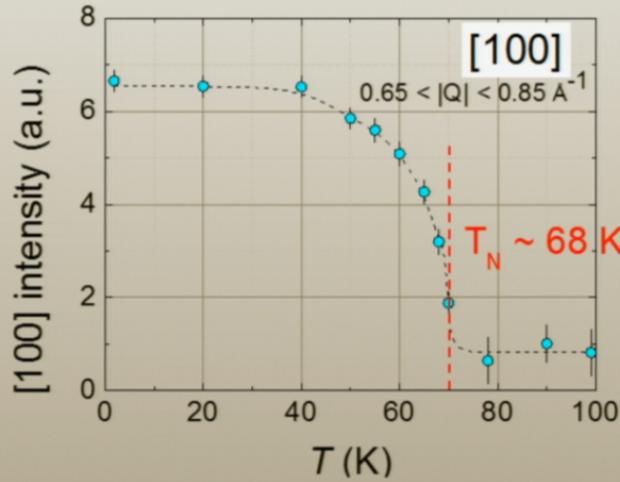
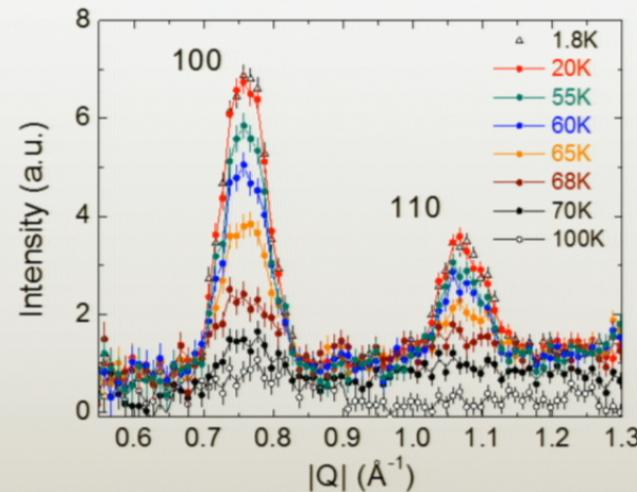


Elastic scattering

- Magnetic Bragg peaks

[100] $|Q| = 0.76 \text{ \AA}^{-1}$

[110] $|Q| = 1.06 \text{ \AA}^{-1}$





Introduction

Elastic scattering

- Magnetic Bragg peaks

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3D long range magnetic order

- Magnetic structure

Type I f.c.c.

magnetic moment $\mu = 1.65(6) \mu_B / \text{Os}^{5+}$

spin-only moment $\mu = 3.87 \mu_B / \text{Os}^{5+}$

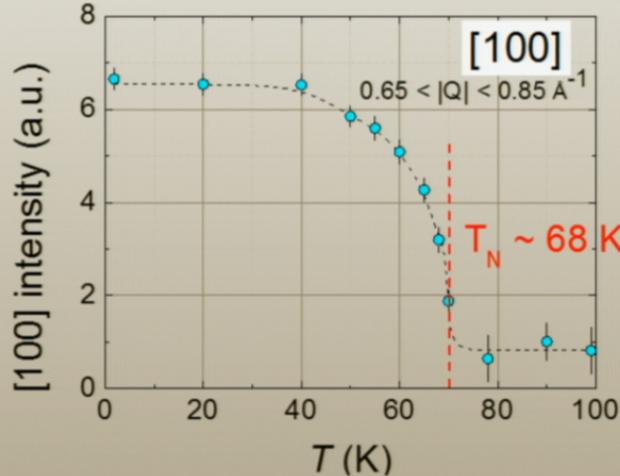
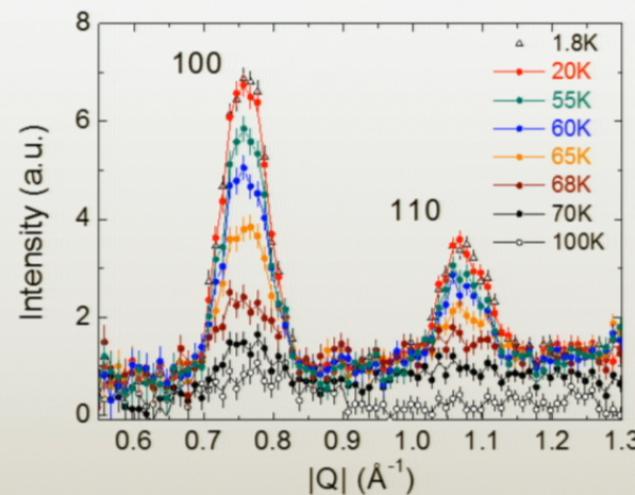
- Order parameter

$T_N \sim 68 \text{ K}$

2nd order phase transition

INS results

Conclusion





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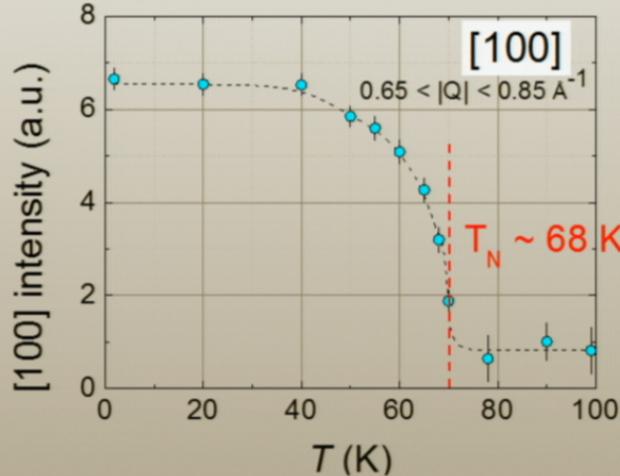
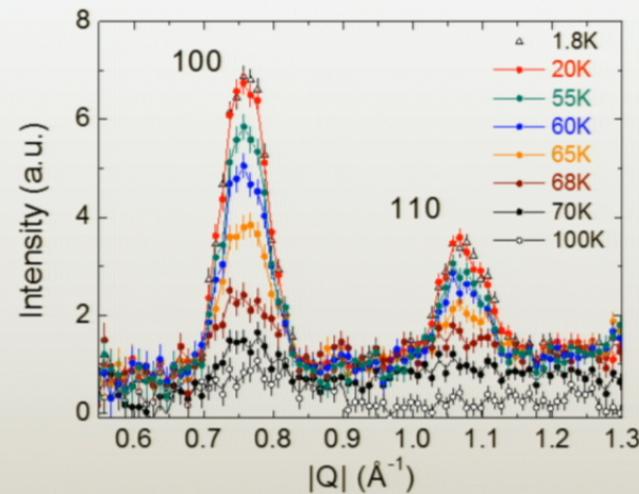
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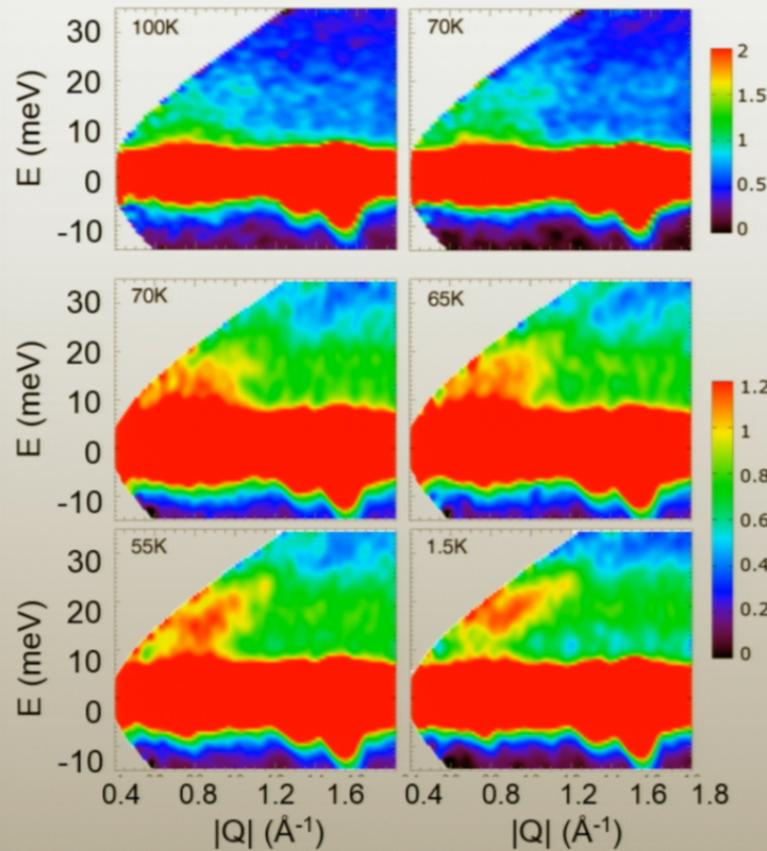
INS results

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Inelastic scattering: spin gap



- $E_i = 120\text{meV}$
Energy resolution $\pm 7 \text{ meV}$
polycrystalline sample

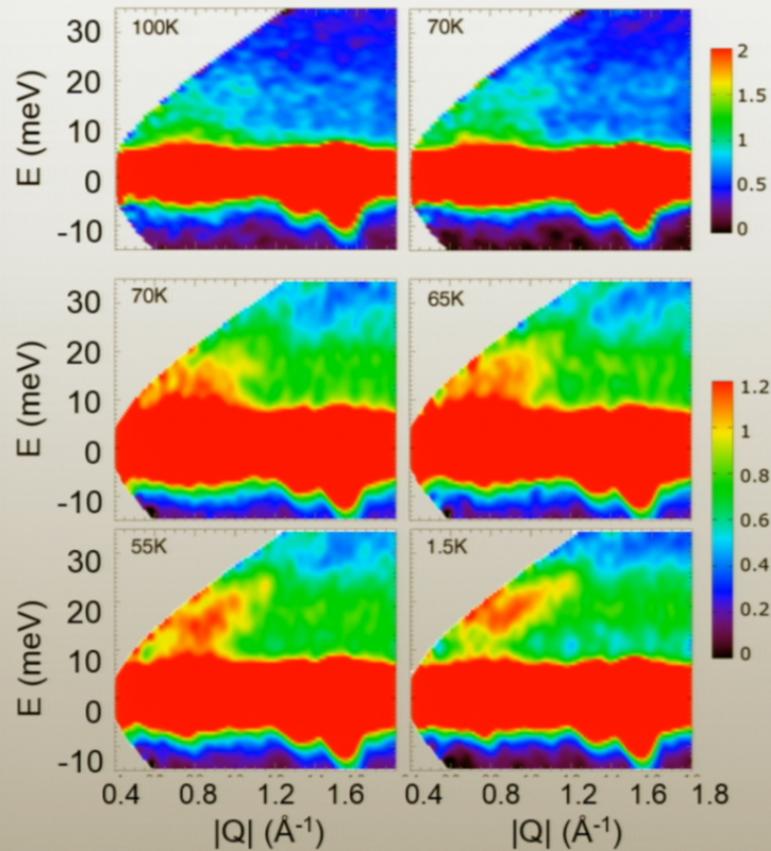
- Temperature dependence of $S(|Q|, E)$

$T \geq 70 \text{ K}$: quasi-elastic intensity at $|Q|=0.76 \text{ \AA}^{-1}$

$T < 70 \text{ K}$: opening of a large gap to the inelastic spin bandwidth



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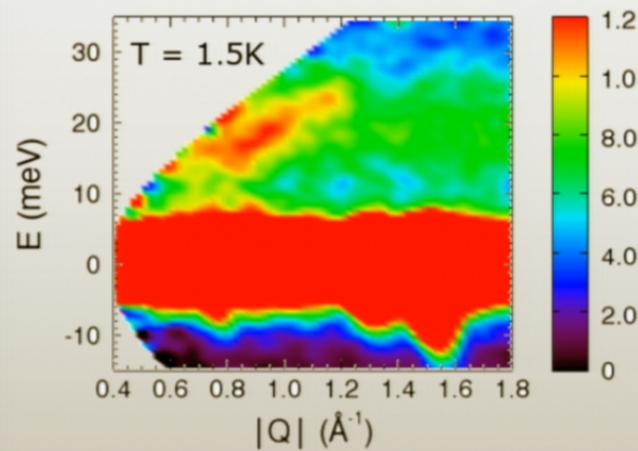
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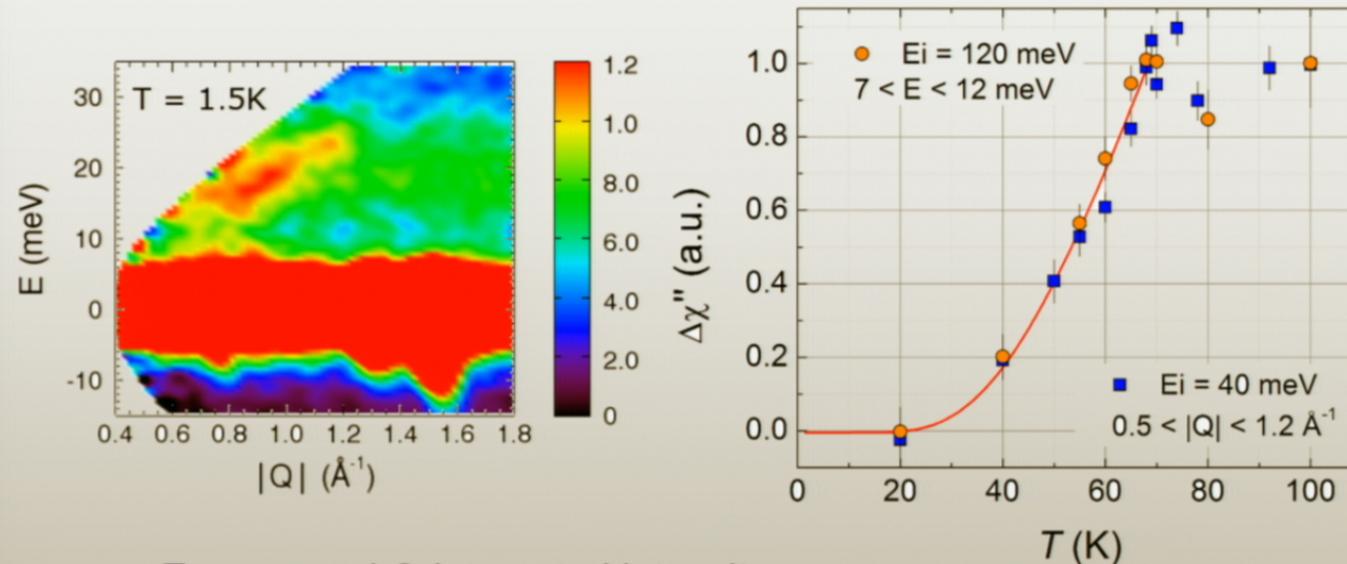
Inelastic scattering: spin gap



- Q-integrated cuts



Inelastic scattering: spin gap

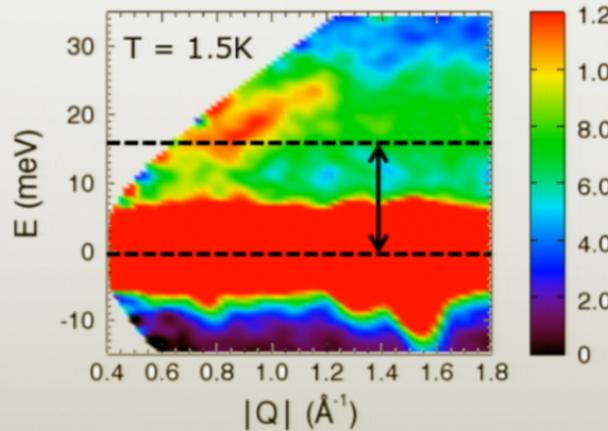


- Energy- and Q-integrated intensity

$$\Delta\chi''(T) \sim e^{-\Delta/k_B T} \quad \Delta = 16(2) \text{ meV}$$



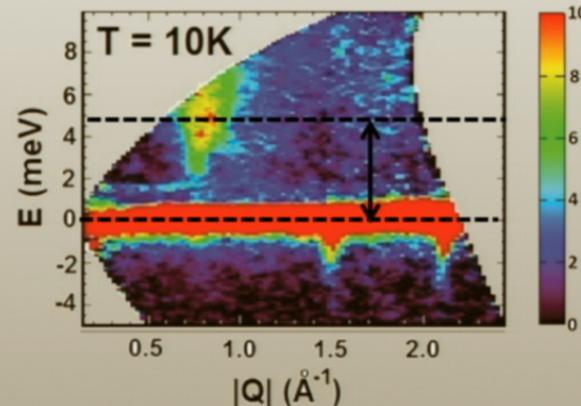
Comparison between Ru⁵⁺ (4d³) and Os⁵⁺ (5d³)



Ba₂YO₃O₆ Os⁵⁺ 5d³

$$\Delta_{\text{Os}} \sim 17 \text{ meV}$$

* SOC $\lambda_{\text{Os}5+} \sim 186 \text{ meV}$



* Ba₂YRuO₆ Ru⁵⁺ 4d³

$$\Delta_{\text{Ru}} \sim 5 \text{ meV}$$

* SOC $\lambda_{\text{Ru}5+} \sim 55 \text{ meV}$

$$\Delta_{\text{Os}} / \Delta_{\text{Ru}} \sim 3.4$$

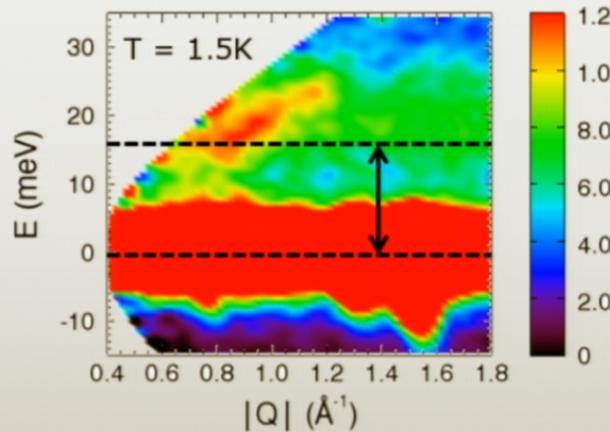
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* J. P. Carlo *et al.*, PRB **88**, 024418 (2013)

* C.-G. Ma and M. G. Brik, J. Lumin. **145**, 402 (2014)



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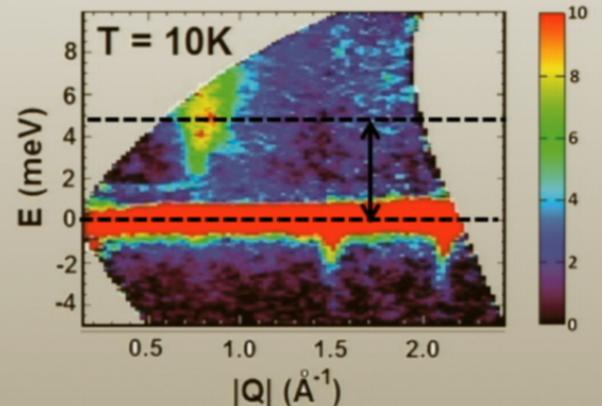
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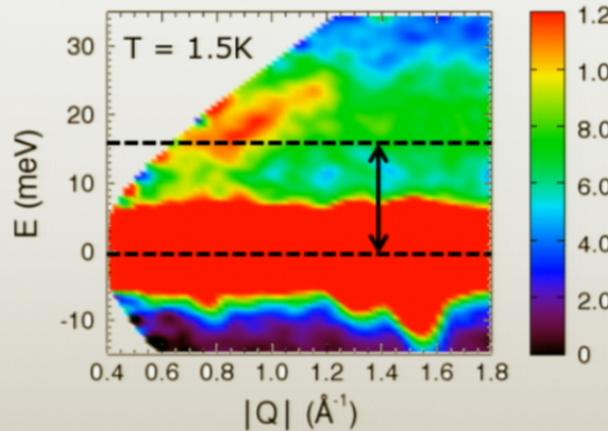
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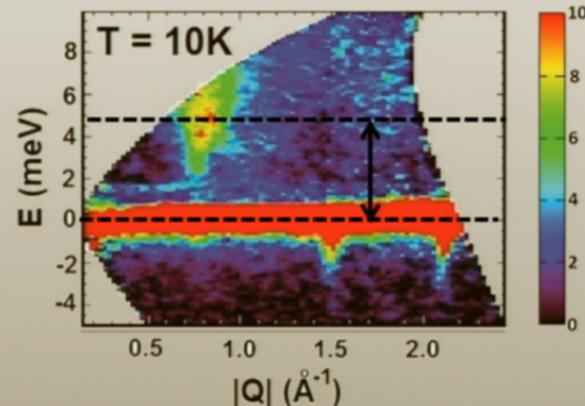
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Summary

- The 5d³ based cubic double perovskite, Ba₂YO₃O₆, has been characterized and studied through inelastic neutron scattering.
- Long range antiferromagnetic order below T_N = 68 K.
Magnetic frustration f ~ 10
- Effects of strong spin-orbit coupling:
 - Reduced ordered magnetic moment $\mu = 1.65(6) \mu_B / Os^{5+}$
(compared to spin-only moment $\mu = 3.87 \mu_B / Os^{5+}$)
 - Large spin gap (no gap expected for t_{2g}³ configuration) $\Delta_{Os} \sim 17 \text{ meV}$
The ratio of the gaps for the osmate and the ruthenate $\Delta_{Os} / \Delta_{Ru} \sim 3.4$ is essentially the same as the ratio of the free ion SOC constants for Os⁵⁺ and Ru⁵⁺.
- Work in progress to investigate the magnetic properties of 5d¹ and 5d² systems, where exotic ground-states have been predicted.
(G. Chen et al., PRB **82**, 174440 (2010) and PRB **84**, 094420 (2011))