Experimental report

12/04/2016

Proposal: 5-32-807 **Council:** 10/2014

Title: The Frustrated Magnetic Order of the Dimer Spin System Ba3B´Ir2O9, B´ = Sc or Y.

Research area: Physics

This proposal is a continuation of 5-32-782

Main proposer: **Paul FREEMAN**

Experimental team: Katharina ROLFS
Paul FREEMAN

Local contacts: Goran NILSEN

Samples: Ba3YIr2O9
Ba3ScIr2O9

Instrument	Requested days	Allocated days	From	To
D7	7	5	15/07/2015	20/07/2015

Abstract:

Studies of iridium-based compounds are at the forefront of solid state physics research. In these materials the electronic configurations are driven by strong spin-orbit coupling. We are studying the spin frustration in Ba3B´Ir2O9 B´= Y, or Sc materials, where one lone electron is shared over the structural Ir dimer. Due to the weak moment of these materials antiferromagnetic Ir dimers have been proposed, that have frustrated antiferromagnetic interactions on the hexagonal lattice. Like in the isostructural Ru materials changing B´ dramatically changes the magnitude of the moment, with Ru forming ferromagnetic dimers. We plan to investigate the role of increased spin-orbit coupling on the frustrated magnetism in this structural series of materials.

The Frustrated Magnetic Order of the Dimer Spin System Ba3B´Ir2O9, B´ = Sc or Y.

Research into the magnetic properties of 5d-transition metal based oxide systems is of significant interest due to the strength of spin-orbit coupling in these materials which can drive these materials towards a Mott insulating state in spite of large covalency, and a small on-site Coulomb repulsion energy U[1]. The theoretical possibilities of the magnetic states of these systems is identifying new novel phases such as the quantum compass phase[2]. Until recently the search for a quantum spin liquid (QSL) phase in which quantum fluctuations prevent magnetic ordering, has focused on 3d transition metal-oxide materials such as as in the S = 1/2 Ba3CuSb2O9 [3]. In our research we have taken the approach that spin-orbit coupling of Ir based 5d-transition metal oxide materials will enhance quantum magnetism leading to novel magnetic phases.

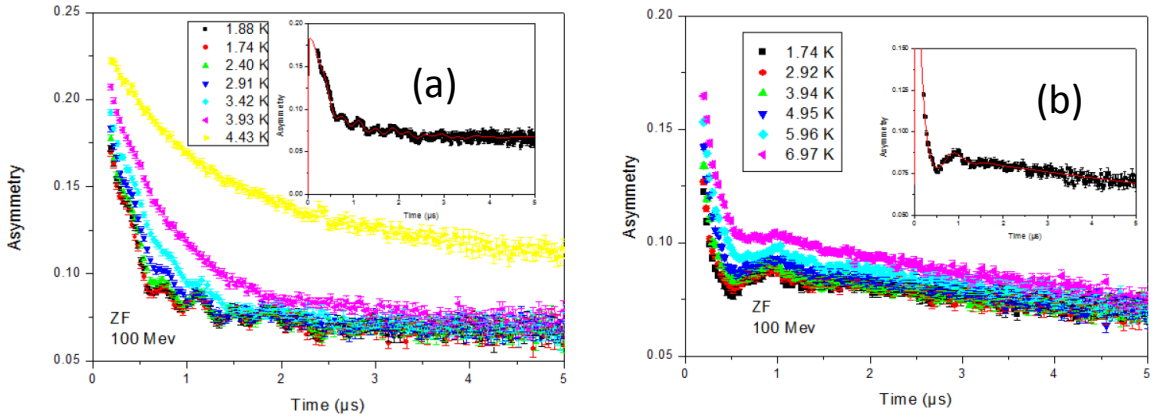

Figure 1: The μSR spectra of (a) $Ba_3YIr_2O_9$ and (b) $Ba_3ScIr_2O_9$. In the inset of (a) 1.88K spectrum of the Y material shows clear numerous oscillations, were the oscillations are observed to be smeared out by disorder in the Sc material as seen at 1.74 K in the inset of (b).

One system we have been studying is Ba3BIr2O9 B = Y, or Sc, which crystallizes in a hexagonal (P63mmc) structure with Ir-Ir structural dimers forming an edge-shared triangular network, with the dimers aligned along the crystal c-axis. Ir occupies a unique crystallographic site and has a fractional (+4.5) oxidation state in a purely ionic picture thereby suggesting metallicity. However the materials are likely spin-orbit driven Mott insulators. Reports of reduced moments in these materials have led to the postulation of antiferromagnetic (AFM) Ir dimers from the single electron across the dimer[4]. While our structures are similar to Ba3BRu2O9 B´ = La, Y[5], a recent diffraction study has determined ferromagnetic (FM) Ru-dimers interacting antiferromagnetically on a hexagonal lattice, with dramatic differences in size of the ordered moment with B´ [6] . We wish to determine whether we have FM or AFM dimers with frustrated spin interactions on a hexagonal lattice in our Ir based materials.

After performing detail studies of Ba3BIr2O9 B = Y, or Sc we performed both μSR spectroscopy and polarized neutron diffraction on D7. The original D7 experiment suffered from the presence of a small quantity of water (< 5 mg) in the material, this hydrogen causing significant scattering and effectively half the quality of data due to the need for high precision to subtract the scattering components. In the original experiment a weak broad magnetic

Bragg peak was observed at 0.5 $Å^{-1}$. This magnetic Bragg peak would be consistent with only a small fraction of the Ir spin ordering. µSR spectroscopy on both samples showed clear oscillations consistent with small moment magnetic ordering below 4.5 K in the Y material, and below 8 K in the Sc material. Consistent with the cite disorder observed in the crystal structure of the Sc material, the magnetic ordering is enhanced due to pinning to the disorder. We proposed in this experiment to study larger samples of Ba3BIr2O9 B = Y, and Sc, which had been baked dry to remove the presence of water, on D7 by polarized neutron diffraction. The higher quality data would allow determination of whether the magnetic Bragg peak was observed at 0.5 $Å^{-1}$ is a single or double peak, that would enable a proposal of the magnetic structure, and we could compare the magnetic structure of the two materials.

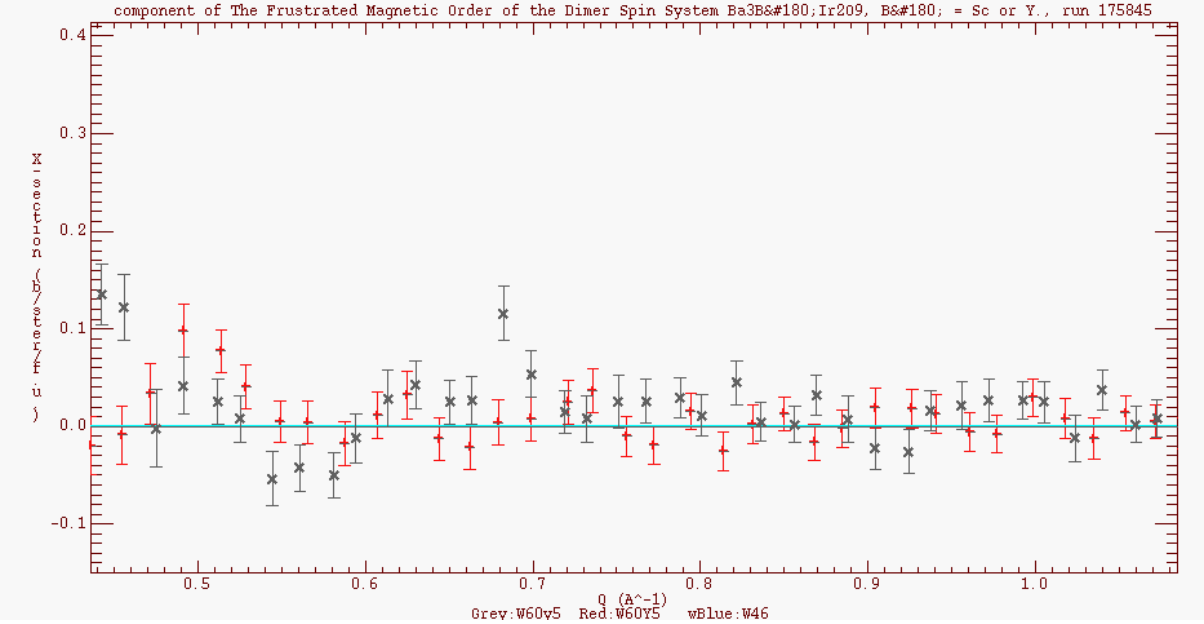

Figure 2: Polarized neutron diffraction of $Ba_3YIr_2O_9$ taken on D7, showing only the coherent magnetic scattering at 2 K. The red data was the initial measurements of proposal 5-32-782 on a 2.5 g sample which contained <5 mg of water. The Grey data points are 2 days of new data taken in this experiment on a 4.5 g sample that was baked dry, with no indication of the presence in the sample. No repeatable magnetic signal is observed in the material.

In our new measurements of Ba3YIr2O9 B = Y at 2 K the magnetic Bragg peak at 0.5 $Å^{-1}$ was not reproduced, and no magnetic Bragg peak was observed across the wave vector range studied. We concentrated our beam time on the Y based material, counting for >3.5 days and this data produces a lower limit on the maximum possible ordered moment in this material. A shorter measurement on Ba3ScIr2O9 was compared to a measurement on Ba3ScIr2O9 taken on DNS. The DNS data suffered from an unknown uncertainty in data subtraction. Comparison of the two data sets reveal no consistent magnetic Bragg peak.

Due to the high neutron absorption cross section of Ir the possibility to further increase the quality of polarized neutron diffraction data on polycrystalline samples of Ba3BIr2O9 B = Y, or Sc. Determination of the magnetic structure of these materials by neutron diffraction may only be possible if large single crystals can be obtained.

The fact that the ordered magnetic moment is so small in these materials indicates that they are close to the full realization of an electron antiferromagnetically dimerizing across the two Ir sites. Our D7 study will be used to determine a limit on the size of this magnetic moment.