Experimental Report

Proposal: **4-01-1321** **Council:** 10/2012

Title: Magnetic Correlations of Ba3MTi2O9with M = Ru, Rh and Ir.

This proposal is a new proposal

Researh Area: Physics

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Samples: Ba3RuTi2O9
Ba3RhTi2O9
Ba3IrTi2O9

Instrument	Req. Days	All. Days	From	To
IN4	7	7	01/04/2013	08/04/2013

Abstract:

The triangular spin geometry has been under intense investigation to study the phenomena of spin frustration, primarily on the 3d transition metal-oxides. Recent studies of the S = 1/2 Ba3CuSb2O9 [1], and in the S = 1 Ba3NiSb2O9 materials [2], have attracted great attraction due to their spin frustration. Studying transition metals from the 4d and 5d series introduces an enhanced spin-orbit coupling interaction into the study of magnetism. Here we plan a study magnetic excitations of a series of Ba3MTi2O9 M = Ru (4d S = 1), Rh (4d S = ½) and Ir (5d S = ½) materials in which the spin-orbit interaction will play a prominent role in the magnetism.

Magnetic Correlations of $Ba_3MTi_2O_9$ with M = Ru, Rh and Ir.

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3d transition metal-oxides with lattice symmetries where triangular patterns of magnetic ions are realized, have been under intense investigation to understand the influence of resulting frustration. Notably, systems based on the $Ba_3MX_2O_9$ structure have been attracting great attention due to their large degree of spin frustration, particularly in the S = 1/2 $Ba_3CuSb_2O_9$ [1], and in the S = 1 $Ba_3NiSb_2O_9$ materials [2]. On exchanging the 3d transition metal to a 4d, or a 5d transition metal the spin-orbit interaction strengthens and plays a prominent role in the magnetism of materials. Strong spin-orbit coupling in these systems drives them to an insulating state in spite of a small on-site Coulomb interaction [3], and the materials in this case can be referred to as spin-orbit-driven Mott insulators. Experimental studies of 4d and 5d transition metal oxide materials are therefore crucial in studies of spin frustrated systems.

We have grown polycrystalline 4d and 5d analogs of $Ba_3XY_2O_9$ to investigate the magnetism of this structure in which spin orbit coupling should play a prominent role. Our materials are $Ba_3MTi_2O_9$ with M = Ru (4d S = 1), Rh (4d S = ½) and Ir (5d S = ½). All the three materials crystallize in a hexagonal ($P6_3mc$) structure where M^{4+} (M=Ru,Rh,Ir) spins form a 2D corner shared triangular network. We are currently determining the site disorder in these materials by a joint x-ray and neutron diffraction study. We have measured both the Field-Cooled and Zero-Field-Cooled magnetization of the three samples. The three materials show subtle ordering features in magnetic susceptibility well below their negative Curie-Weiss temperatures, thus indicating frustrated antiferromagnetic interactions. Each material shows ZFC-FC irreversibility indicative of glassy magnetism and sometimes kinks/change of slope possibly associated with a short-ranged frustrated order. This magnetic order is presently being studied by μSR and polarized neutron diffraction. In this proposal we are however interested in determining the spin interactions which drive the magnetism of these materials.

In this proposal we measured the excitation spectrums of $Ba_3MTi_2O_9$ M = Ru, and Ir between E = 5 meV and E = 80 meV on IN4. The measurements of both samples ultimately showed no indications of a magnetic excitation spectrum.

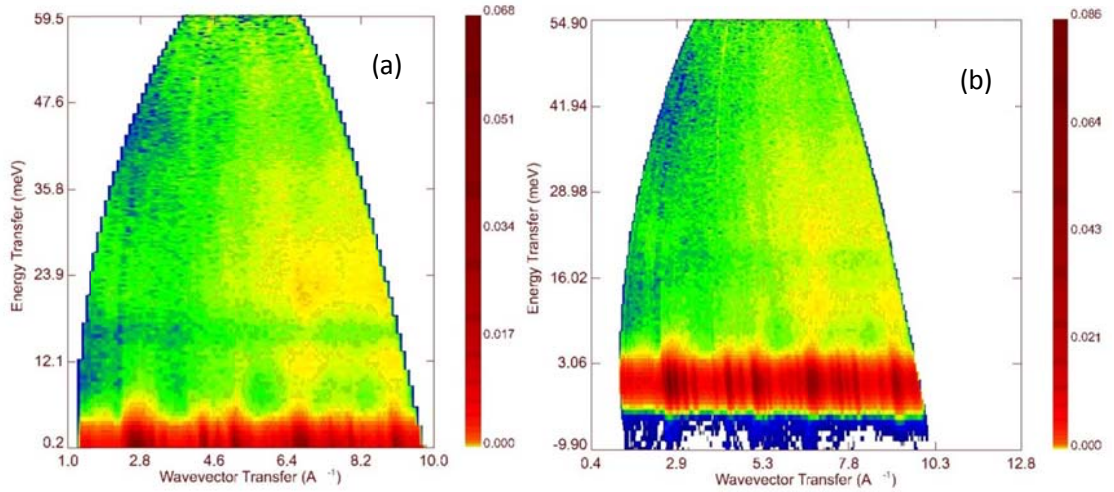

Figure 1: The excitation spectrum of (a) $Ba_3TuTi_2O_9$ and (b) $Ba_3IrTi_2O_9$ measured on IN4 with E_i = 66.4 meV.

The first sample we studied was the Ru sample, for which magnetic susceptibility indicates a sizable Ru S = 1 moment. For the Ru sample a well-defined excitation spectrum was observed, but after subtraction of the instrument background the temperature dependence of the excitations indicated all modes were phonons in origin. Different neutron energies were used (115 meV, 66.4 meV, 16.75 meV, 14.2 meV) for the measurements to access different regions of reciprocal space, but no candidate magnetic excitations were observed.

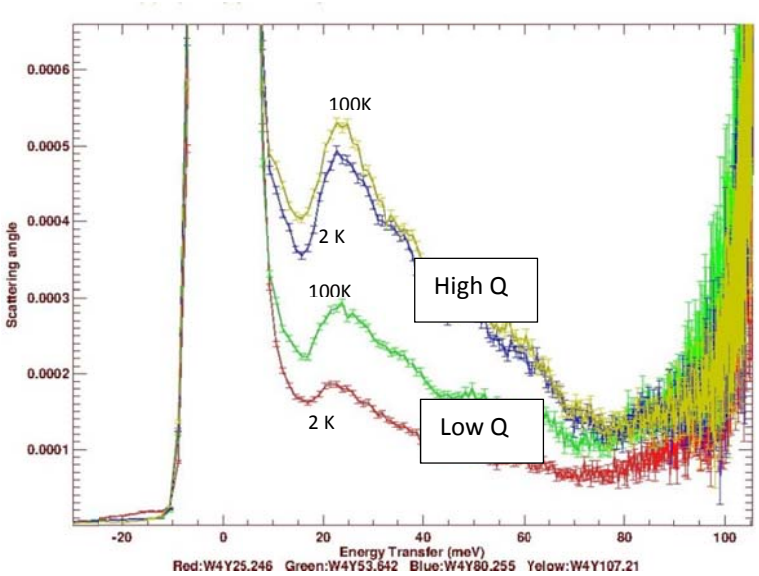

Figure 2: The temperature dependence of the excitation spectrum of $Ba_3RuTi_2O_9$, measured at 2 K and 100 K. The excitations are seen to increase in intensity with increasing temperature for both large and small wavevectors, indicating all excitations to be phonon in origin.

Next we studied the excitation spectrum of the Ir sample. The 2.5g sample was spread across the full incident neutron beam, and despite of the high neutron absorption of Ir use of short wavelength neutrons allowed the full excitation spectrum be measured over the same energy range as the Ru sample. Again in the Ir sample all observed modes were phonon in origin.

Our data on the M = Ir and Ru materials observed no magnetic excitation spectrum from the spin moments in these materials. The question this raises, is where is the magnetic excitation spectrum located in these two materials? There are a number of possibilities to the answer this question. In the two materials the Ir and Ru are in a frustrated spin lattice, a 2D corner shared triangular network, and there is a degree of site disorder with the Ti. It is possible that through a spin frustration, or through glassy behavior due to site disorder, the spin correlations occur with a large periodicity. In this case the magnetic excitations would occur at smaller wavevectors than IN4 can access. A second possibility is that the spin interaction is far weaker than we postulated from susceptibility measurements. In this case the magnetic excitation spectrum would be below 3 meV indicating a main interaction strength less than 1 meV. Alternatively the magnetic excitation may be highly diffusive and spread across a wide region of reciprocal space in both energy and wavevector, which would be highly challenging to see with inelastic neutron scattering.

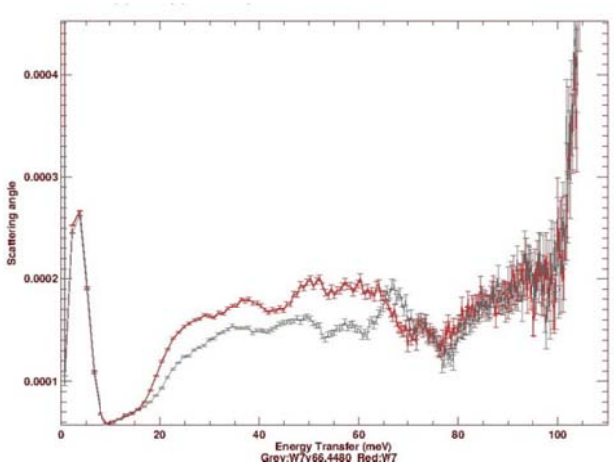

Figure 3: The phonon density of states $Ba_3MTi_2O_9$ with M = Ru (grey) and M = Ir (red). The strong phonon mode observed in Ru at 67 meV is observed to soften in the Ir material. The background signal increases towards 100 meV in due to known experimental reasons.

To conclude the excitation spectrums of $Ba_3MTi_2O_9$ with M = Ru, and Ir were measured by inelastic neutron scattering. All the excitation modes observed were phonon in origin. The data obtained on IN4 will be insightful into understanding the structural properties of these two materials, with both the energy and wavevector dependences, and density of states obtained. The strategy to understand the magnetism of these materials requires further thought, and consideration.

Refernces:

[1] S. Nakatsuji *et al.*, Science **336**, 559 (2012)
[2] J. G. Cheng *et. al.* Phys. Rev. Lett. 107, 197204 (2011)
[3] B. J. Kim *et al.*, Phys. Rev. Lett. **101**, 076402 (2008)