Experimental report

08/07/2016

Proposal: 4-01-1423 **Council:** 10/2014

Title: Investigation of a bilayer square lattice Jeff=1/2 iridate Mott insulator

Research area: Physics

This proposal is a new proposal

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Samples: Sr3Ir2O7

Instrument	Requested days	Allocated days	From	To
IN20	4	4	13/07/2015	17/07/2015
IN8	7	7	27/07/2015	03/08/2015

Abstract:

We propose a neutron scattering study of the bilayer 2D Mott insulator Sr3Ir2O7. This study is required to resolve both the magnetic structure and interactions and the striking difference of the magnetism of Sr3Ir2O7 in comparison to the monolayer Sr2IrO4. A recent neutron diffraction study has identified high temperature Bragg reflections that they associate with a newly observed high temperature magnetic phase, without definite proof of the magnetic nature of these reflections, or whether this structure causes canting of the low temperature magnetic order. Our Iridium L-edge resonant inelastic x-ray scattering (RIXS) data have indicated a large spin gap with apparent absence of an acoustic mode. Similar data on the same compound has been interpreted in terms of a spin-Hamiltonian very different from its monolayer version in Sr2IrO4, introducing a very large magnetic anisotropy. An alternative explanation is that the acoustic Sr3Ir2O7 mode has no RIXS matrix element at the Iridium L-edge. Our neutron scattering proposal is designed to resolve the magnetic nature of Sr3Ir2O7.

Investigation of a bilayer square lattice Jeff=1/2 iridate Mott insulator

Owing to a rich variation of ground states, the iridium oxide materials (iridates) have received considerable attention in recent years. Novel physics, originating from the combination of strong electron correlations, strong spin-orbit coupling and in some cases also geometrical frustration, have been realized or proposed in these materials. A novel Mott state induced by spin-orbit coupling was argued from spectroscopy experiments on Sr2IrO4 [1] which crystallizes in the layered perovskite structure similar to cuprates. It has been further argued that a Jeff=1/2 one-band Hubbard model could account for the low-energy physics of Sr2IrO4 [2], thus realizing, as in the case of cuprates, a spin-1/2 square lattice antiferromagnet.

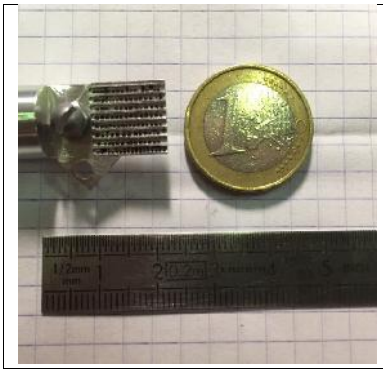

Figure 1: The 100+ x-ray co-aligned multi-crystal sample of $Sr_3I2_2O_7$, of 50 mg weight.

Ir L-edge RIXS has been reported on Sr2IrO4 [3] showing an isotropic spin-wave-like dispersion which resembles La2CuO4, and orbital excitations at higher energy. In contrast to this, for the bilayer compound $Sr_3I2_2O_7$ an acoustic and an optic branch would be expected, but at the Ir L-edge we observe a gapped branch with a spin gap of 90 meV. The difference between RIXS spectra of Sr2IrO4 and $Sr_3I2_2O_7$ contrast dramatically to the universal spectra seen in mono-, bi- and tri-layer cuprates In $Sr_3I2_2O_7$, the addition of an extra IrO6 layer introduces further interactions which were modelled on the basis of a highly anisotropic spin model by another group [4]. In this scenario the optical and acoustic modes are perfectly degenerate and gapped by 90 meV. It is however highly coincidental that the numerous magnetic interactions give rise to almost perfectly degenerate modes and such an high energy acoustic mode. In an alternative explanation, the gapped mode of Sr3Ir2O7 is due to dimerization of the Ir spins along the out of plane direction, resulting in a singlet to triplet spin gap. In this way the magnetism of $Sr_3I2_2O_7$ is highly controversial. We alternatively propose that an acoustic magnon mode in $Sr_3I2_2O_7$ exists at low energies that the RIXS measurements are insensitive to or cannot resolve. There are other examples of very weak goldstone modes in quantum magnets, for instance in the material Cu2Te2O5Cl2[5].

The magnetic structure of $Sr_3I2_2O_7$ is complicated by the presence of two closely related orthorombic twins, with Bbcb and Acaa structures, which result in antiferromagnetic Bragg reflections at l = even and l = odd respectively. Resonant x-ray diffraction determined that below 280 K the spins order antiferromagnetically between the bilayers with the spins pointing along the c-axis[6]. Whereas neutron diffraction additionally sees an additional peak above 280K for the Acaa twin, that was confirmed to be non-magnetic at 300 K by polarized neutron diffraction[8]. In this proposal aswell as search for low energy acoustic magnons we employed polarized neutron analysis to determine the magnetic structure and track the temperature evolution of the (0.5,0.5,l) l = odd structural peak.

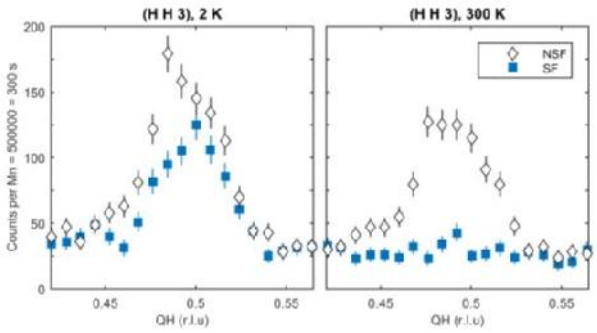

Figure 2: Polarized neutron diffraction of the (0.5, 0.5, 3) position in $Sr_3I2_2O_7$ taken on IN20. The l = 2 magnetic peak has an additional non-magnetic component that is responsible for all of the intensity of this Bragg reflection above 280 K [7,8]. No significant variation in the non-magnetic component was observed across the 2-300 K temperature range.

On In20 we maintained the neutron spin polarization to be parallel to the scattering wavevector, **P**||**Q**, to split the magnetic and structural components of the Brag peaks, and studied the temperature evolution of both components. The structural component of the Bragg reflection at l = 3 was observed to have little significant temperature dependence, so that additional intensity of the reflection observed below 280 K is purely magnetic. Using longitudinal polarized neutron diffraction we determined the spin orientation for both twins, at different temperatures across the magnetically ordered phase. Preliminary analysis of this data indicates that the spins are orientated along the c-axis for both twins within experimental uncertainty, in agreement with the resonant x-ray diffraction study [6].

Our unpolarised neutron scattering of the excitation spectrum was hindered by minor technical problem with the cryostat, that lead to a time dependent background, and our initial data suffered from irreproducibility problems. Constant energy scans of the (0.5, 0.5, l) l = 1,2 and 3 positions showed significant structure in the background signal. Using IN8's high flux we studied the diffraction signal along (0,0,l), that revealed broad structural peaks across the range l = 1 to 4.5. The background structure observed in the low energy excitation spectrum could be accounted as from phonons from these structural peaks. The crystal mosaic of our multicrystal sample, does not account for the presence of all the broad Bragg peaks away from l = 2 and 4. These additional Bragg peaks will be further studied on a single crystal using synchrotron radiation.

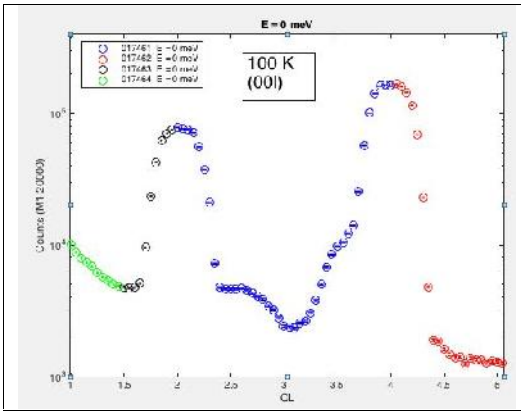

Figure 3: Logarithmic plot of the diffraction signal scanned along (0,0,l) of our in $Sr_3I2_2O_7$ multicrystal.

Having found no evidence for acoustic magnons at low energies, we performed a constant wavevector scan of the excitation spectrum at (0.5, 0.5, 2) at K. Modes were observed at 12 meV, 16 meV, 19 meV and 24 meV. The 16 meV mode may have been previously observed in ARPES data (King PRB 87, 241106(R) (2013)), and was suggested to be a signature of electron-boson coupling. The 19 meV and 24 meV modes by Raman studies, Cetin PhD thesis, http://bit.ly/1JofRN5. The only mode that we have not identified in our preliminary analysis is the weak 12 meV.

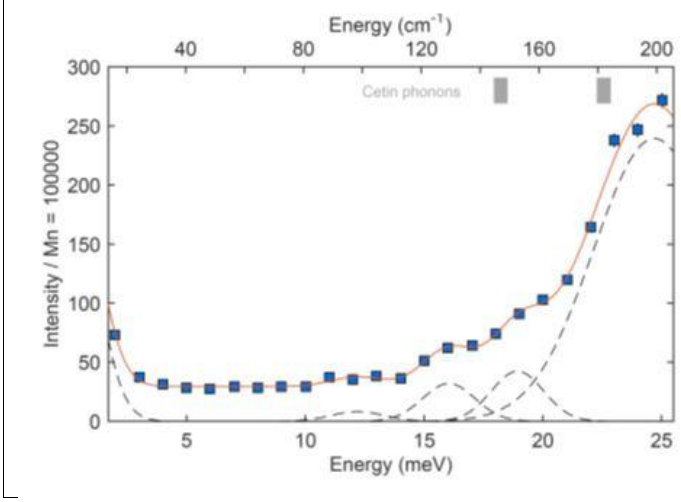

Figure 4: Constant wavevector scan of $Sr_3I2_2O_7$ at the position (0.5,0.5, 5), measured at 2K. Excitation modes are observed at 12 meV, 16 meV, 19 meV and 24 meV.

To conclude our detailed polarization study of the magnetic order in $Sr_3I2_2O_7$ is consistent with published work, and provides greater detail. No low energy magnon modes were observed by our inelastic neutron scattering, implying any acoustic magnon modes in $Sr_3I2_2O_7$ must be weak. A weak 12 meV excitation needs to identified. Potential new structural reflections along (00l) were identified, and need to be examined in a single crystal by x-ray diffraction.

References:

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