Experimental Report

Proposal: **5-32-774** **Council:** 4/2012

Title: Magnetic excitations and short range order in frustrated spin-orbitalsystem FeSc2S4

This proposal is a new proposal

Researh Area: Physics

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Samples: Fe Sc2 S4

Instrument	Req. Days	All. Days	From	To
D7 He3 Spin Filter	8	11	28/09/2012	05/10/2012
			22/02/2013	26/02/2013

Abstract:

This proposal is part of our research programme to investigate novel forms of spin and orbital order stabilized by quantum fluctuations and geometric frustration effects. The antiferromagnetic spinel FeSc2S4 where electronically-active Fe2+ ions occupy a diamond lattice has been proposed to realize an unusual “spin-orbital liquid” ground state with no long-range magnetic or orbital order down to temperatures several orders of magnitude lower than the strength of the inter-site exchange interactions. We have recently observed the full bandwidth of the magnetic excitations and also studied the field dependence of the excitations and observed good agreement with a recent theoretical model proposing strongly-dispersive spin-orbital triplons propagating on a spin-orbital singlet ground state stabilized by the strong spin-orbit coupling at the Fe2+ ions. Here we propose to use D7 with energy analysis and polarization setup to determine the temperature-dependence and polarization of low-energy features in the scattering.

Magnetic excitations and short range order in frustrated spin-orbital system $FeSc_2S_4$

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The A-site spinel $FeSc_2S_4$ is believed to form a novel, spin-orbital liquid (SOL) state, where spin and orbital moments remain disordered down to the lowest measurable temperatures, even in the presence of strong exchange between magnetic Fe^{2+} ions. Here we report on polarized, inelastic neutron scattering measurments conducted on a powder sample of $FeSc_2S_4$ on the D7 instrument at ILL.

$FeSc_2S_4$ forms in the spinel structure, (space group $Fd\bar{3}m$, no. 227) and is characterized by a dominant next-nearest neighbour (NNN) exchange, J_2 between Fe^{2+} ($S = 2$, $L = 2$) ions. No transition is observed in susceptibility measurements even though the C-W temperature derived from these measuremets suggest substantial exchange, moreover elastic neutron scattering has shown no sign of structural distorion, even though the system is J-T active, having a hole in the lower e-doublet [1]. Both these results indicate that neither the spin nor orbital degrees of freedom order, and instead continue to fluctuate down to the lowest measureable temperatures. The term '*Spin-Orbital Liquid*' has been coined to describe this state, in analogy with the 'typical' Spin Liquid [2].

We have proposed the following single ion Hamiltonian for Fe^{2+} ions in $FeSc_2S_4$:

$$H = H_{cf} + H_{SO} \tag{1}$$

where H_{cf} and H_{SO} are the crystal field and spin-orbit coupling contributions, respectively.

Diagonalization of (1) yields a non-magnetic, singlet groundstate which is a complex admixture of spin and orbital states - termed a 'Spin Orbital Singlet' (SOS) - and a triplet as the 1^{st} excited state. Excitations into the triplet state can be thought of as the creation of a triplon, and these triplons are able to hop throughout the FCC lattice of NNNs due to the strong J_2 exchange, so in inelastic neutron experiments we observe a highly k-dependent, gapped mode [3]. Such experiments have been carried out on a powder sample, the results of these measurements agreed with our model and we were able to estimate the microscopic parameters contributing to (1).

However, one feature which could not be explained by our model, but seemed to be present in neutron scattering data measured on both FOCUS - at the Paul Scherrer Institut (PSI) - and MERLIN - at ISIS,

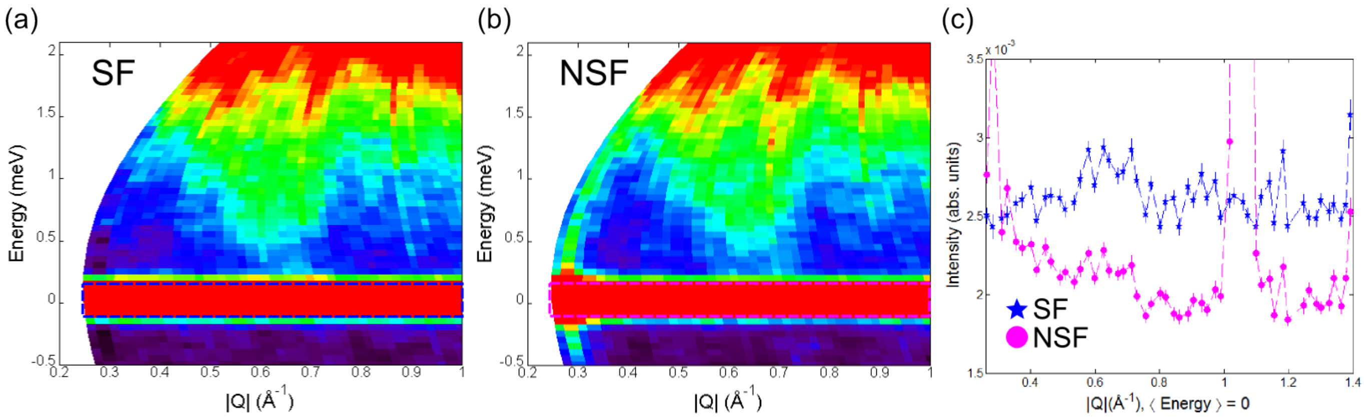

Figure 1: $S(Q, \omega)$ plots for (a) Spin Flip (SF) and (b) Non-Spin Flip (NSF) neutron scattering channels taken at D7. (c) Cut along the elastic line for the SF (blue pentagrams) and NSF (magenta circles) data shown in (a) and (b).

Rutherford Appleton Laboratory - was a top up of intensity on the elastic line at around 0.6Å^{-1} ($\approx \frac{2\pi}{a}$). Such an effect was obviously quite an unusual result, given that $FeSc_2S_4$ is not expected to order nor become structurally distorted, and so no extra magnetic or structural Bragg peaks should be visible at low temperatures.

The difficulty in characterizing this feature is two fold; firstly, the inelastic instruments previously used to probe this powder sample are not optimized to resolve elastic/quasi-elastic effects, and, secondly, $\frac{2\pi}{a}$ is the wavevector where the dispersive triplon mode softens, so in elastic neutron scattering measurements - optimized for exploring $\Delta E = 0$ - this inelastic signal is integrated over. Therefore, even though a top up of intensity is observed at around 0.6Å^{-1} in elastic neutron scattering experiments measured at HRPT (PSI), this can equally well be attributed to the known inelastic signal occuring at low energies at this wavevector. An experiment on D7 then, provided us with a unique opportunity not only to resolve both quasi-elastic and elastic signal in $S(Q, \omega)$ but also determine, with the use of polarized neutrons, whether such signals were magnetic or structural in origin.

4g of high-quality powder were placed in an aluminium can with annular geometry, before being lowered into an orange cryostat. We ran the experiment in D7's time-of-flight (TOF) mode with incident neutrons of wavelength $\lambda = 4.84\text{Å}$, and typical results are shown in Fig 1. The expected dispersive mode which softens at around 0.6Å^{-1} is clearly visible in both Fig 1(a) and Fig 1(b), exemplifying the magnetic nature of this excitation.

Our real interest however is in the elastic region below this excitation, and Fig 1(c) shows a cut along Q in this area for both SF and NSF channels. This cut shows the typical results expected from a polarized neutron scattering experiment; i.e. the asymmetric distribution of incoherent scattering between SF and NSF scattering channels (and indeed, upon background subtraction, this is found to be in the ratio 2:1 as expected) and appearance of nuclear Bragg peaks in the NSF channel only (for example here the (111) peak at around 1.1Å^{-1}), but closer inspection also reveals a top up of intensity peaked at the expected $\frac{2\pi}{a}$ wavevector in both the SF and NSF channels (although a low Q spurion in the latter makes this less clear). Our present understanding is that this feature is of magnetic origin, though difficulties in D7 TOF data reduction through LAMP have made the process of characterising this signal rather complex.

It is hoped that with a few enhancements of the IDL scripting for TOF data reduction on D7, as well as the addition of supplementary high T data taken on D7, that this intriguing feature's origin can be conclusively determined, thus solving one of the last remaining mysteries of $FeSc_2S_4$'s intriguing ground state.

References

[1] V. Fritsch et al, Phys. Rev. Lett. **92**, 116401 (2004)

[2] L. Balents, Nature **464**, 199 (2010)

[3] A. Biffin et al, (*in progress*) (2014)