Experimental report

14/02/2017

Proposal: 5-32-833 **Council:** 4/2016

Title: An investigation on the role of spin fluctuations in the superconductivity of the spinel oxide LiTi2O4

Research area: Physics

This proposal is a new proposal

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Samples: LiTi2O4

Instrument	Requested days	Allocated days	From	To
D7	7	7	15/12/2016	22/12/2016

Abstract:

Among the over 300 known spinel compounds, only four exhibit superconducting properties with LiTi2O4 (LTO) being the only nondisordered transition metal oxide. Despite its superconductivity (SC) identified in 1973, understanding of its SC mechanism has seen slow progress due to sample reproducibility. A recent renewed interest concerning the role of spin correlations in LTO began with an investigation of applying a quantum site percolation model (QSPM) and was supported by extensive charge transport/tunneling measurements on LTO films, revealing anomalous magnetoresistance behaviour. Despite evidence provided for the crucial role of spin fluctuations for SC in LTO, neutron studies have been limited to diffraction. We propose to use 7 days on D7 to measure the temperature-dependence of the magnetic diffuse scattering with the objective of observing significant changes in the magnetic diffuse scattering, supporting the hypothesis that these fluctuations play a key role in the SC of LTO. Since these fluctuations are known to play a crucial role in the high Tc's, if the role of fluctuations in LTO is confirmed, the proposed analogy between LTO and the high Tc's will be strengthened.

An investigation on the role of spin fluctuations in the superconductivity of the spinel oxide $LiTi_2O_4$ on D7: Experimental Report

Objective: The original objective of the experiment was to (i) isolate the magnetic component of the scattering using the polarisation analysis provided by D7 and to (ii) determine the temperature dependence of the aforementioned magnetic component. The overall objective of the experiment on D7 was to establish a clear experimental connection between magnetic fluctuations *via* magnetic diffuse scattering and the superconductivity mechanism [1,2] *via* the superconducting temperature T_c.

Experimental Details: The XYZ polarisation analysis option was selected instead of the 10 point method because additional information concerning off planar contributions (i.e. defined *via* α) was deemed not necessary versus the additional counting time that could be available by performing the six point method. A ratio of 1:4 for NSF:SF was used to improve the statistics for the isolation of magnetic signal through SF. In order to improve (i) the relative |**Q**|-resolution and (ii) the relative statistics, 10 points along 2θ were collected ($\Delta 2\theta = 0.5^o$) per temperature. In order to achieve a |**Q**|-range with lower bound less than 0.5 $Å^{-1}$ but with D7 still possessing relatively large flux to compensate for polarisation analysis and to achieve a dynamic range below 3 meV (since our measurements on IN4C provided evidence of E < 3 meV magnetism), a wavelength of 4.8 Å (E_i = 3.32 meV) was chosen.

Preliminary Data: The sample was cooled whilst measuring since a.c. magnetic susceptometry (figure **a**) implied a very small superconducting volume (< 20%). The flipping ratio F.R. did not exhibit any significant decrease that would accompany a transition into the superconducting regime. Consequently, all measurements below T_c did not require a ZFC. Although not evident in the magnetic cross section, if one averaged the magnetic cross section over the |**Q**|-range probed (excluding |**Q**| values near the lower/upper measurement limit), the magnetic cross section does exhibit a temperature dependence (figure **b**) that is strikingly similar to that of the cuprates [3], a family of T_c's where magnetic correlations are indeed suspected to play a key role in the superconductivity mechanism. Furthermore, the stark increase in the average magnetic cross section below T_c is most distinctive near Bragg peaks. As is shown in figure **b**, there is no clear experimental indication of pseudo-gap transition proposed at T ~ 80 K [4] but there is a clear increase (within experimental error) in the average magnetic cross section below T_c.

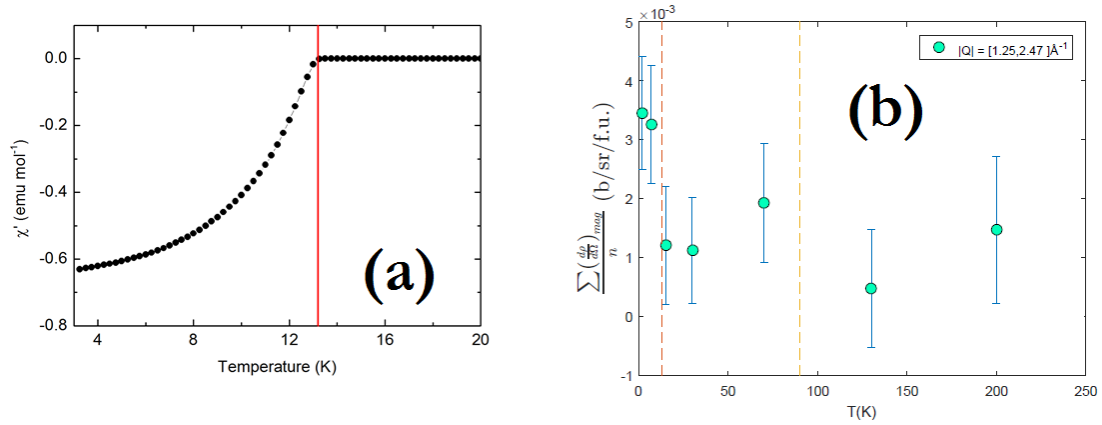

Figure: (a) In-phase component of the ZFC a.c. susceptibility confirming a superconducting transition at 13.2(1) K indicated by the red vertical line. (b) The average magnetic cross section (integrated over |**Q**| = [1.25,2.47] $Å^{-1}$) as a function of temperature measured on D7 (λ = 4.8 Å). The increase in the magnetic cross section below the superconducting transition (red line) is indicative of enhanced magnetic fluctuations/excitations accompanying the superconducting state. The green line indicates the approximate temperature where the pseudo-gap phase has been predicted to appear [4] but a transition we failed to experimentally measure.

Future Studies: Our IN4C study was correct that magnetic excitations may lie below 3 meV. Since there is no energy discrimination on D7, the magnetic scattering is in fact integrated over all energies up to E_i = 3.32 meV. Although the data from D7 is extremely encouraging, the exact location in S(|**Q**|,E) space is still relatively unknown. The data from D7 confirms that the magnetic excitations are prevalent at low energies

(< 3 meV) and low |**Q**| (< 2.5 $Å^{-1}$). The data from D7 confirms that the next step in pursuing magnetism in the superconductivity of $LiTi_2O_4$ is to use a high flux/high resolution cold time-of-flight (TOF) spectrometer such as IN5 in order to access the appropriate regions of S(|**Q**|,E) space with adequate resolution and comparable flux to D7 to compensate for the low moment associated with a S = ½ system. A cold TOF experiment is recommended before performing a cold triple-axis (TAX) experiment because although there is substantial proof that the magnetic intensity are increasing most starkly around the Bragg peaks, this has yet to be confirmed in other experiments, mostly due to phonon contamination. Once the TOF data has been obtained, a dispersion relation may be observed but most importantly, the particular energies and |**Q**| required to be probed in the already limited S(|**Q**|,E) region of interest due to D7 and IN4C, can be retrieved and any fine structure can then be probed by a cold TAX with an advantage of an increase of approximately two orders of magnitude of flux.

Conclusion: Our work on D7 has provided substantial evidence for the role of magnetic fluctuations in the superconductivity mechanism of the S = ½ spinel oxide superconductor $LiTi_2O_4$, agreeing with the conclusion of another high profile study [4].

References: [1] Christianson *et al.* Nat. Lett. **456**, 7224 (2008). [2] Johnston *et al.* Mater. Res. Bull. **8**,777 (1973). [3] Mangin-Thro *et al.* Nat. Comm. **6**, 8705 (2015). [4] Jin *et al.* Nat. Comm **6**, 7183 (2015).