Experimental report

14/02/2017

Proposal: **4-02-473** **Council:** 4/2016

Title: An investigation on the role of spin fluctuations in the superconductivity of the spinel oxide $LiTi_2O_4$

Research area: Physics

This proposal is a new proposal

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Samples: $LiTi_2O_4$

Instrument	Requested days	Allocated days	From	To
IN5	6	0		
IN4	0	4	14/11/2016	18/11/2016

Abstract:

Among the over 300 known spinel compounds, only four exhibit superconducting properties with $LiTi_2O_4$ (LTO) being the only non-disordered 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) to the series $Li_{1+x}Ti_{2-x}O_4$. Disagreement between theory and laboratory measurements lead to the hypothesis that correlations play a significant role in the SC. This hypothesis was supported by extensive charge transport and 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 the IN5 with the aim to measure the T-dependence of the (|Q|,E) spectrum of isotopically pure 7LTO powder with the objective of confirming both (i) that LTO is not a conventional BCS superconductor and (ii) the crucial role played by fluctuations in the SC mechanism in LTO.

An investigation on the role of spin fluctuations in the superconductivity of the spinel oxide $LiTi_2O_4$ on IN4C: Experimental Report

Objective: The primary objective of the inelastic neutron scattering experiment on IN4C was to measure the non-elastic magnetic resonance mode associated with superconductivity in $LiTi_2O_4$. Theory proposes starkly different energy transfers for BCS and high T_c superconductors and consequently, once a magnetic resonance mode was measured, its location would be indicative if $LiTi_2O_4$ is indeed an unconventional superconductor – as is currently suspected after the work by Jin *et al.* [1] – or a BCS superconductor as was originally suspected after its discovery by Johnson *et al.* [2] almost 40 years ago.

Experimental Procedure: Since the desired magnetic resonance mode was predicted to be quite weak attributed to the small S = ½ of magnetic Ti^{3+}, minimal helium was used as an exchange gas. The $^{7}LiTi_2O_4$ powder sample was loaded into a 20 mm width x 35 mm aluminium can pre-loaded with a 16 mm central plunge to create an annulus that would maximise the amount of sample exposed to the beam.

Experimental Results: As is shown in Figure **a** below, the S(|Q|, E) spectrum of $LiTi_2O_4$ has two clear features: (1) intense Bragg peaks (confirming sample purity) and (2) strong acoustic and optical phonons emanating from the Bragg peaks. These excitations were confirmed to be phonons but (i) their dispersion, (ii) their temperature dependence and (iii) performing subtract with χ''(|**Q**|, E) at different temperatures. For the rest of the experiment, the wavelength was increased from 1.4 Å to 2.75 Å to 3.21 Å in order to probe dynamic ranges consistently closer to the elastic line with a corresponding increased access to lower |**Q**|. As shown in Figure **b**, there were no magnetic excitations in the predicted energy transfer (based on T_c) for either BCS or unconventional superconductivity whilst the phonons continued to be prevalent near the Bragg peaks.

There are a few explanations as to why we were not able to detect the magnetic resonance modes we desired: (1) there is a relatively high background at low energy transfers (compared to IN5 for example), (2) the flux was reduced significantly as we increased wavelength, (3) phonon contributions were extremely pronounced, especially in close proximity to the Bragg peaks that may smear out some possible magnetic excitations. In order to minimise the contribution of phonons, χ''(|**Q**|,E)/E was plotted as a function of energy transfer E as shown in Figure **c**. It can be shown that phonons will provide a constant flat background whilst magnetic excitations should appear above as a Lorentzian. As described in Figure **c**, there are indications of paramagnetism at high temperatures at low energy transfers (< 3 meV), albeit the dynamic range is limited due to the pollution by the elastic peak.

Conclusion & Future Work: With various temperatures and various incident energies, no clear experimental evidence for a magnetic resonance peak associated with superconductivity (either BCS or unconventional) was measured. Strong phonon contributions, coarse resolution, low flux with a strong background at low energy transfers prohibited any clear magnetic signal from being detected. Isolating most of the scattering signal from phonons, paramagnetic signal was detected at low energy transfers (< 3 meV) at high temperatures, although pollution from the elastic peak prohibited any determination of the exact energy transfer for this possible excitation. As this experiment was followed up by a very successful experiment on D7, exhibiting clear magnetic excitations below 3 meV – in agreement with IN4C – the natural extension of this work would be to use a cold high resolution/high flux time-of-flight spectrometer such as IN5 to isolate the signal alluded to in both IN4C and D7.

References: [1] Jin *et al.* Nat. Comm **6**, 7183 (2015). [2] Johnston *et al.* Mater. Res. Bull. **8**,777 (1973).

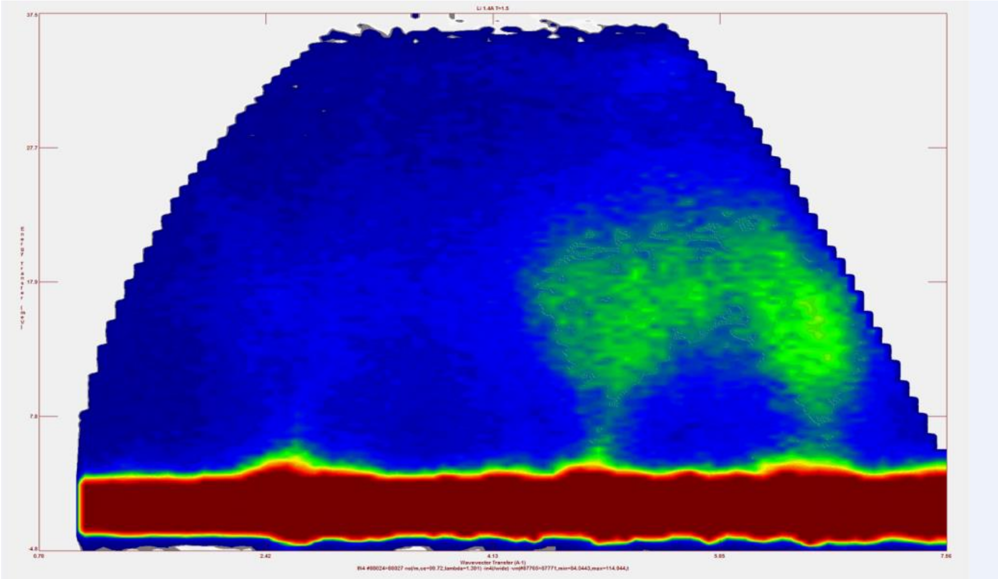

Figure a: Raw S(|**Q**|,E) spectrum of $^{7}LiTi_2O_4$ at 1.5 K (λ = 1.381 Å). The S(|**Q**|,E) spectrum is dominated by phonons emanating from the Bragg peaks.

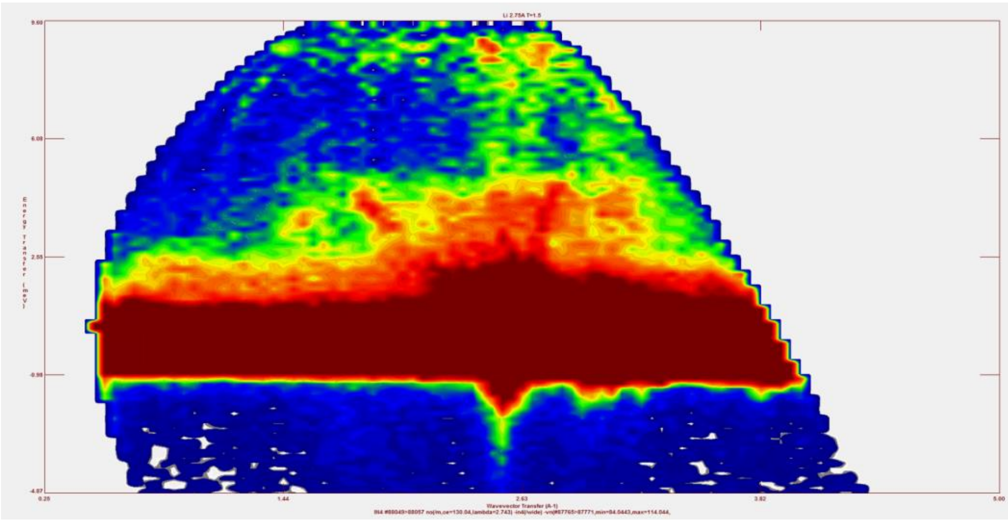

Figure b: Raw S(|**Q**|,E) spectrum of $^{7}LiTi_2O_4$ at 1.5 K (λ = 2.743 Å). The S(|**Q**|,E) spectrum is dominated by phonons emanating from the Bragg peaks.

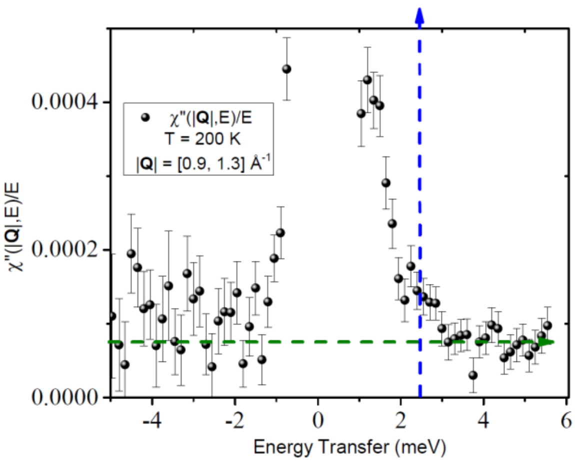

Figure c: Constant |**Q**|-integrated (|**Q**| = [0.9, 1.3] $Å^{-1}$) cut along E with λ = 3.21 Å at 200 K. The green line is a baseline representing contributions from phonons whilst the blue line represents the limits of the effects of elastic line pollution. There is a weak but detectable paramagnetic signal < 3 meV, suggestive that the magnetism is at much energy transfers than expected.