Experimental Report

Proposal: **5-31-2275** **Council:** 10/2012

Title: Magnetostructural coupling and spin dynamics in the "breathing" pyrochlore antiferromagnets LiInCr4O8 and LiGaCr4O8

This proposal is a new proposal

Researh Area: Physics

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Samples: LiInCr4O8
LiGaCr4O8

Instrument	Req. Days	All. Days	From	To
D2B	3	2	09/07/2013	11/07/2013
IN4	3	3	01/07/2013	04/07/2013

Abstract:

The chromium spinels of general formula ACr2O4 exemplify the pyrochlore antiferromagnet with strong spin-lattice coupling. We have prepared the A-site ordered compounds LiInCr4O8 and LiGaCr4O8, which realize the so-called "breathing" pyrochlore lattice; a pyrochlore lattice where the Cr-Cr bond lengths alternate on adjacent tetrahedra. Both of these materials are found to order with an accompanying change in the cell parameters at TN~14 K, but at higher T exhibit rather different behavior. The magnetic susceptibility of the In compound may be described by isolated tetrahedra, while the Ga compound is well fitted by by classical Monte Carlo simulations. This difference may be traced back to the difference in the amount of alternation in bond lengths between small and large tetrahedra. We propose here an investigation of the nuclear/magnetic structure and dynamics of both at low T using the D2B diffractometer and the IN4 spectrometer. In particular, we are interested in the low temperature magnetic structures, changes in nuclear structure across TN, and the possible presence of a "resonant" mode from the tetrahedra. We thus ask 6 days of beamtime: 3 days on D2B and 3 days on IN4.

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Magnetostructural coupling and spin dynamics in the "breathing" pyrochlore antiferromagnets $LiInCr_4O_8$ and $LiGaCr_4O_8$

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The chromium spinel family ACr_2X_4 (A=Zn, Mg, Cd..., X=O, S, Se...) manifests a number of novel magnetic phenomena, including strong magneto-elastic coupling, multiferroicity, localized resonances, and spin liquid physics. We have recently produced two new materials in this family, $LiInCr_4O_8$ and $LiGaCr_4O_8$, where the A site is populated by two different cations in an ordered fashion. The effect of this on the B-site is to distort the pyrochlore lattice such that small and large tetrahedra alternate in a pattern we call the "breathing" pyrochlore lattice. By changing the A site cations, it should thus be possible to interpolate between isolated tetrahedra and the regular pyrochlore lattice. In the case of the present materials, the distortion found to be smaller for A'=Ga than for A'=In. We performed a combined neutron diffraction and inelastic time-of-flight spectroscopy study on $LiInCr_4O_8$ and $LiGaCr_4O_8$ in order to:

- Determine the nature of the phase transitions observed in C_p, and the low temperature nuclear and magnetic structures. To achieve this, we used the D2B diffractometer at $\lambda = 1.59$ Å. The best compromise between resolution and flux was found with $10'$ of collimation.

- Elucidate the excitation spectra in the same temperature range. This was done on the IN4 spectrometer using wavelengths of 1.6 and 2.2 Å.

Preliminary refinements [Fig. 1] of the 20 K structures of both materials show the buildup of considerable strain prior to transitions observed at $T_1^{\mathrm{In}} = 17.8$ K and $T_1^{\mathrm{Ga}} \sim 17$ K. In the A'=In case, the transition is found to be purely structural in origin. Magnetic order eventually sets in at $T_1^{\mathrm{In}} = 12.9$ K, with at least two **k**-vectors. No evidence of unit cell multiplication was observed, but the statistics are rather poor. For A' =Ga, T_1^{Ga} represents a first-order transition into a regime where partial magnetic and partial structural order coexist. The new structural phase is tetragonal in symmetry, and its phase fraction increases until T_2^{Ga} is reached, beyond which which additional magnetic peaks appear and the structural order is frozen.

The excitation spectra of the materials at $T > 20$ K are consistent with their relative degrees of breathing. For the more highly distorted A' =In compound, a gapped mode centered at 6.2 meV is observed in $S(Q, E)$ at 50 K. This is found to soften considerably in advance of the structural transition, revealing a novel coupling between magnetism and structure in this material. For A' =Ga, the spectrum is quasi-elastic until T_1^{Ga} is reached, beyond which a resonant feature at ~ 6 meV appears. T. A similar resonant mode is observed in A' =In, first appearing at 14.5 K$< T_1^{\mathrm{In}}$, and becoming strongly enhanced below T_1^{In}. The behaviour below the lower transition is analogous to $ZrCr_2O_4$ for both materials.

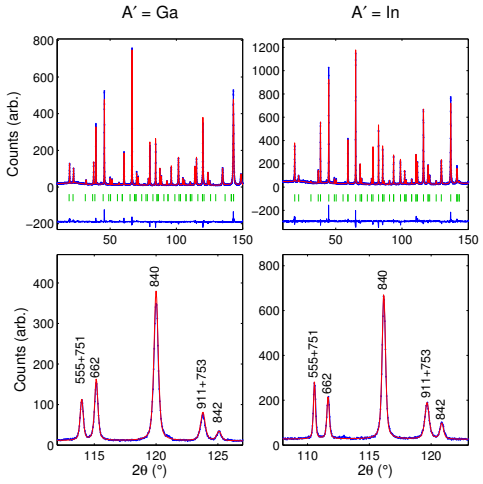

Figure 1: (a-b) Rietvield refinements of neutron diffraction data from D2B, ILL, taken at $\lambda = 1.594$ Å. The experimental data points are shown in blue, with the Rietveld refinement in red, and the difference also in blue. The green tick marks indicate positions of allowed nuclear peaks in the cubic $F\bar{4}3m$ space group. (c-d) View of a narrow portion at high angle indicating the systematic broadening of the $(hk0)$ type Bragg peaks. The instrumental resolution is indicated by the solid black line.

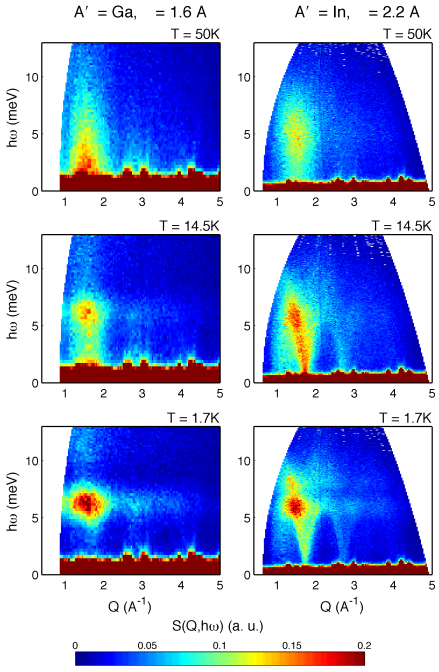

Figure 2: Temperature dependences of $S(Q, E)$ for both systems.