Experimental report

10/09/2022

Proposal: **EASY-1043**

Council: 4/2021

Title: Structure of Li4CuTeO6 at room temperature

Research area: Physics

This proposal is a new proposal

Main proposer: **Sylvain PETIT**

Experimental team:

Local contacts: Claire COLIN

Samples: Li4CuTeO6

Instrument	Requested days	Allocated days	From	To
D1B	3	3	10/10/2021	11/10/2021

Abstract:

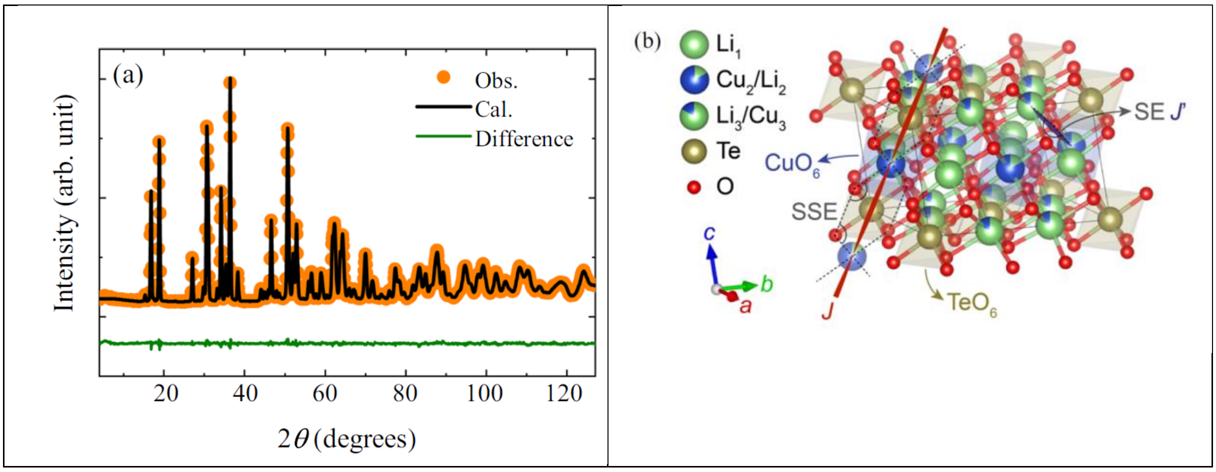
We would like to measure the diffractogram of Li4CuTeO6 at room temperature

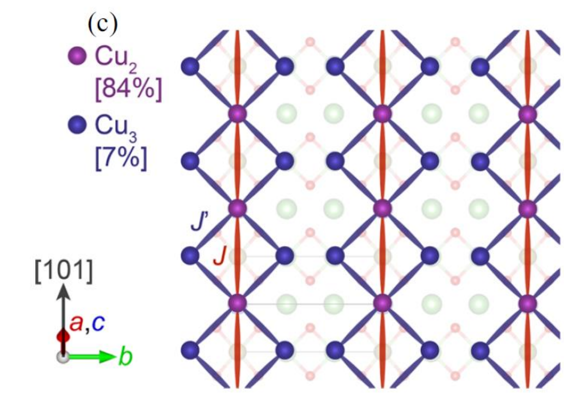
Exp report EASY-1043 at D1B

Results have been published in COMMUNICATIONS PHYSICS https://doi.org/10.1038/s42005-022-00879-2

Context: Geometrically frustrated magnets are defined as a subclass of magnetic materials where all pair-wise interactions cannot be satisfied simultaneously. They usually lack classical long-range order and host strong quantum fluctuations. Some of them, dubbed Quantum Qpin Liquids (QSL), even host fractionalized excitations, in contrast with spin-wave excitations found in conventional magnets [1]. Despite tremendous experimental efforts, however, proven experimental realizations remain rare. Indeed, this physics is extremely sensitive to perturbations such as next nearest-neighbor interactions, or magnetic anisotropy. In this paper, we pursue an alternative route, where disorder plays a prominent role. Indeed, disorder is unavoidable in real materials. However, provided quantum fluctuations are intrinsically strong, it can act as a new prism in revealing many interesting quantum phenomena [2-4]. Recently, it has been suggested that frustrated magnets, with quenched disorder in the form of material defects or a broad distribution of exchange interaction strengths, can exhibit a randomness-induced spin-liquid state [5]. In this work, we focus on the antiferromagnet Li_4CuTeO_6 (henceforth LCTO). Using neutron diffraction, magnetic measurements, specific heat, muon spectroscopy, density functional theory calculations and exact diagonalization, we establish that LCTO is a realization of this enigmatic novel ground state. In particular, neutron diffraction has proven to be of primary importance in understanding and unravelling the disorder issue in this material.

Rietveld refinement of data collected at room temperature on D1B (CRG@ILL) evidences a large anti-site disorder between Li^+ and Cu^{2+} in LCTO. Our analysis shows that the majority of Cu^{2+} ions (84%) at the 2d crystallographic site, form random-length spin chains, while a minority of Cu^{2+} ions (~7%) at defect 4g sites strongly couple to these spin chains, leading to significant frustration. This structure results in a model of randomly depleted 1D spin chains with exchange J, to which randomly occupied sites couple via an exchange J' (see Figure c for the definition of exchange coupling constants), hence introducing strong frustration. The large and negative value of the Curie–Weiss temperature reflects strong antiferromagnetic J and J' interactions. The data show that this compound neither undergoes a phase transition to long-range magnetic order nor spin-freezing down to at least 45 mK. Furthermore, specific heat and magnetization results reveal a data collapse behavior, which suggests the presence of a random-singlet state. Muon spin relaxation measurements corroborate a dynamic ground state, which is attributed to the presence of subdominant interchain interactions that couple the chains in a disordered network. Our results thus establish that LCTO hosts a randomness-induced spin-liquid-like state in a frustrated magnet





Crystal structure and spin model of LCTO. (a) Rietveld refinement of neutron-diffraction data at room temperature for the incident wavelength 1.28 Å. The solid circles represent the observed intensity (Obs.), whereas the black solid line is the calculated intensity (Cal.). (b) Visualization of one unit cell of LCTO where edge-sharing TeO_6 and CuO_6 octahedra connect Cu^{2+} ions on 2d (Cu2) sites with an exchange J through a Cu–O⋯O–Cu super-superexchange (SSE) bridge around Te^{6+}. Additionally, corner-sharing CuO6 octahedra connect Cu^{2+} ions on 2d (Cu2) and 4g (Cu3) sites with an exchange J' through a nearly-linear Cu–O–Cu superexchange (SE) bridge. (c) Resulting spin model of randomly depleted 1D spin chains of Cu2 sites with antiferromagnetic exchange J, to which randomly occupied Cu3 sites couple via an antiferromagnetic exchange J'.

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