Experimental report

29/04/2025

Proposal: **4-05-865**

Council: 4/2023

Title: Studying the universal properties of pyrochlore Heisenberg antiferromaget in the presence of correlated disorder

Research area: Physics

This proposal is a new proposal

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Samples: CsNiCrF6

Instrument	Requested days	Allocated days	From	To
IN5	5	5	22/09/2023	25/09/2023
			29/09/2023	01/10/2023
ORIENTEXPRESS	1	1	19/09/2023	20/09/2023

Abstract:

We are interested in the effects of correlated disorder in structure and magnetism, a realization of such is fluoride pyrochlore CsNiCrF6. In CsNiCrF6 ice rules control the distribution of Ni2+ and Cr3+ cations on the pyrochlore lattice, resulting a special form of correlated disorder. Heisenberg Na2+ and Cr3+ spins develop an antiferromagnetic Coulomb phase at low temperature evidenced by pinch points in the diffuse scattering. We also find features in dynamics and correlations that are identical with other canonical antiferromagnetic Coulomb phases such as the PHAFM; this proposes that such Coulomb phases would have universal features. In our experiments and numerical investigations we can compare a real example of a Coulomb phase (CsNiCrF6), a model (with Monte-Carlo and spin dynamics simulations), and a canonical Coulomb phase that we can also simulate, such that overall we can investigate the origin and extent of universal emergent dynamics in classical antiferromagnetic Coulomb phases. Here we propose an experiment to capture the complete magnetic dynamics spectrum in CsNiCrF6 in reciprocal space, this will enable us to converge our studies on this system.

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Studying the universal properties of pyrochlore Heisenberg antiferromaget in the presence of correlated disorder

The compound $CsNiCrF_6$ is one of the few established occurance of correlated disorder. It realises correlated chemical disorder on a pyrochlore structure formed jointly by Cr^{+3} and Ni^{+2} (to form a *charge ice*). Early studies showed the magnetism in $CsNiCrF_6$ to be similiar to that of pyrochlore Heisenberg AFM, showing no magnetic long-range order and diffuse magnetic scattering[1]. A more recent work suggested magnetic properties originate from the interactions (templated by the underlting chemical disorder) between $S_{\mathrm{Cr}^{3+}} = 3/2$ and $S_{\mathrm{Ni}^{2+}} = 1$[2]. The same work reports Magnetic excitations to have quasi-elastic Lorentzian (qel) lineshape, similar to pyrochlore Heisenberg AFM (PHAFM)[3], and low-energy magnetic dynamics to resemble the dynamics of PHAFM. This experiment aimed to investigate the magnetic excitations $CsNiCrF_6$ over a large volume of reciprocal space to identify the effects of correlated disorder on PHAFM, more particularly characterising the Q-dependence of the qel width.

Experiment detail: A single crystal sample of $CsNiCrF_6$ was aligned in the (h, h, l) plane on an Al finger. There was a slight misalignment on the sample which was corrected for generating the sqw files in the software Horace exploiting the large out-of plane coverage of IN5. The experiment was done with two settings of IN5, with low-energy ($\lambda = 4.35$ $\mathring{A}$) and high-energy ($\lambda = 2$ $\mathring{A}$), to capture the full spectrum of the excitations. The experiment was started with the low-energy settings at the base temperature of the cryostat (1.7 K). We collected data, rotating the sample about (-1,1,0) axis over 150° degrees (run numbers 246797 to 246948); scans were done with 1° step and for 10 minutes. Later, a smaller scan was done to improve the data quality close to the (1,1,1) position (range of 50°, run numbers 246949 to 246999).

In our experiment at the IN5 we observed a spurious feature at low momentum transfer extending in energy depicted in Figure 1. This feature shows in form of a sharp peak in energy cuts (shown by an arrow in Figure 2 panel (b)). To address this problem we collected a dataset with a similar settings, raising the temperature to the room temperature to effectively minimizing all coherent magnetic contribution (run numbers 247005-247130). Due to a problem with neutron production, the data collection at 300 K was forced to stop and resumed the week later. The second experiment was started similar to the first one, correcting the sample alignment in the software. The dataset at 300 K was completed (run number 248113-248188) finding the second dataset compatible with the one obtained in the first part of the beamtime. The dataset at 300 K was obtained combining the two datasets.

We found the spurion to be present at 300 K, at the same energy and with a similar lineshape. We then fitted the spurion peak at high temperature with a Voigt function and used its central position and width in the fitting funtion consisting of the qel, elastic line and the spurion to fit the low temperature scans (at same Q position) and extracting the qel parameters (Figure 2 panel (a)).

The last part of the experiment we used the $\lambda = 2$ $\mathring{A}$ incident neutron energy settings to collect the high-energy magnetic excitaions. At the sample temperature 1.7 K we collected a dataset over 100° degree sample rotation (run numbers 248000 to 248101).

The data obtained in this experiment contributed to the results in a submitted manuscipt title "Algebraic loop liquid in the pyrochlore CsNiCrF6".

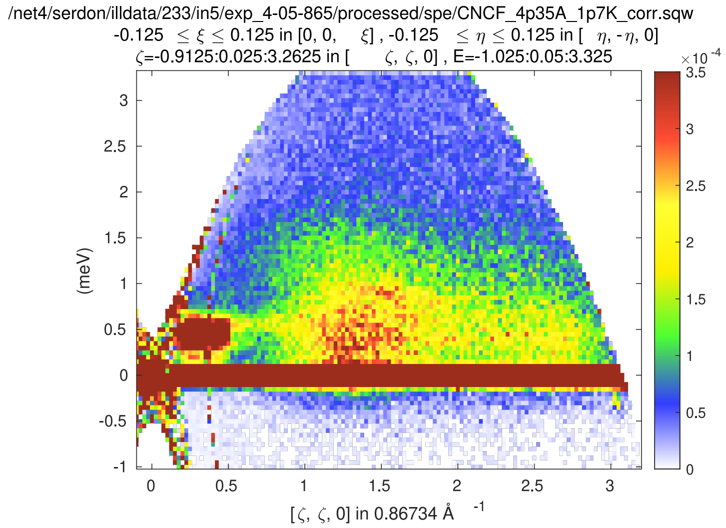

Figure 1: $\mathbf{Q} - E$ slice of the spectrum at 1,7 K showing the spurion at very low-Q (at $\mathbf{Q} = 0$) and dispersing magnetic contribution.

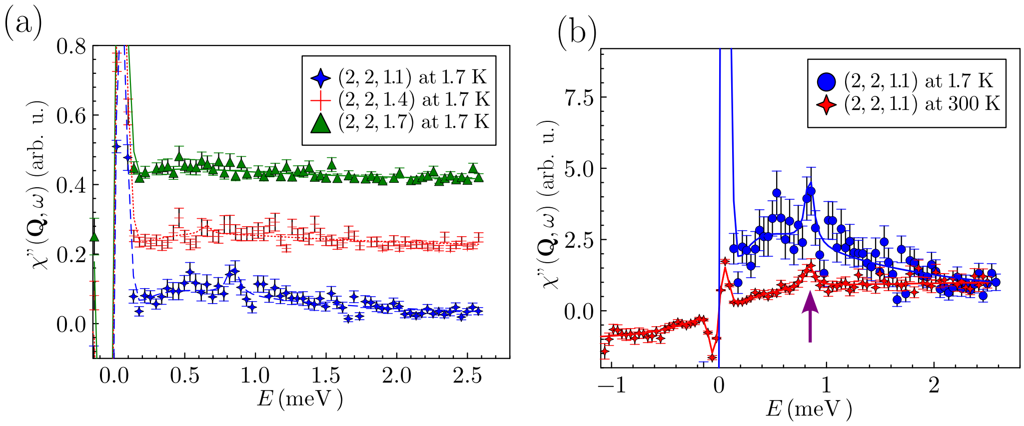

Figure 2: Fits to the dynamical susceptibility and spurious feature at low energies. a) different fits of the low-temperature data showing good fit of the magnetic signal and the spurion. b) Comparison of the base-temperature and room temperature with their fits showing the presence of the spurion at same energy.

References:

[1]: Harris M. J. *et al*, Phys. Rev. Lett. (1994).
[2]:Fennell T. *et al*, Nat. Phys. (2019).
[3]:Conlon P., Chalker J., Phys. Rev. Lett. (2009).