# Experimental report

07/09/2022

**Proposal:** **4-05-708** **Council:** 4/2018

**Title:** Field-dependent spin dynamics in spin-1/2 Heisenberg antiferromagnetic chain

**Research area:** Physics

**This proposal is a new proposal**


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**Samples:** $CuSO_4.5D_2O$

| Instrument | Requested days | Allocated days | From | To |
|---|---|---|---|---|
| IN5 | 7 | 7 | 16/01/2020 | 23/01/2020 |
| IN3 | 3 | 3 | 10/01/2020 | 13/01/2020 |


**Abstract:**

We wish to study the evolution of the spinon continuum of the Heisenberg antiferromagnetic spin 1/2 chain $CuSO_4.5D_2O$ as a function of the magnetic field below and up to saturation. Theory predicts the presence of new string-type quasi-particles in addition to "modified" spinon continua. We plan to compare our results quantitatively to theory. The Zeeman-transition of a second, isolated Cu-site, is present in all spectra at finite field and requires good resolution to isolate the chain spectra. A supplementary measurement at 10T will lift all excitations above the range of interest and allow a "perfect" background subtraction. IN5 equipped with the 10T magnet and dilution insert is ideally suited for this purpose, the experiment can not be performed on IN6 (form and width of the resolution) nor on a triple axis (resolution not sufficient). We will need two positions with the 10T magnet per field (quarter-, half-, 3/4-, full saturation, and 10T), and therefore ask for 7d of IN5 equipped with 10T and dilution fridge.

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## MOTIVATION

$CuSO_4 \cdot 5D_2O$ belongs to the model example of the 1D AFM quantum Heisenberg system. Its crystal structure is triclinic and was firstly published in [1]. By means of inelastic neutron scattering, M. Mourigal et al. [2] have demonstrated excellent one-dimensionality of $CuSO_4 \cdot 5D_2O$. There are two copper $Cu^{2+}$ ions in the unit cell, which constitute two different magnetic subsystems: $Cu_1$ ions at (0,0,0) form one dimensional chain along a-direction with $J$ = 0.252(17) meV, and $Cu_2$ ions at (0.5,0.5,0) ferromagnetically coupled with $J_{22}$ = 0.012(18) meV. The interaction between two different sites, $Cu_1$ and $Cu_2$, is found to be $J_{12}$ = 0.020(22) meV, which is less than $0.1J$.

In [2], authors have studied spin dynamics of 1D AFM chain in zero and above saturation fields. In fully polarized state H = 5 T, where the excitation spectrum is correctly described by linear spin-wave theory, they could extract exchange parameters as well as the values of g-tensors for each copper site. In zero field, they have concluded that two- and four-spinon excitations need to be considered to match 98(8)% of the spectral weight, where 30% relate to four-spinon states.

In intermediate magnetic field, one has to deal with exotic excitations such as psinons, antipsinons and Bethe strings [3]. To have a full overview of chain dynamics in the applied magnetic field, we have performed an inelastic neutron experiment on time-of-flight spectrometer IN5.

## EXPERIMENTAL DETAILS

$CuSO_4 \cdot 5D_2O$ sample was glued to a Cu holder, thus ensuring a good thermalization and inserted into a vertical cryomagnet reaching up to 10T together with a dilution fridge. An experiment was performed at T = 100 mK above the Néel temperature $T_N$ [4] and at selected field values including H = 0T, 1.5T, 3T, 3.3T, 3.5T, and 9.5T. Incident neutron wavelength was = 6 Å with chopper speed 12000 rpm, thus providing energy resolution of 0.048 meV FWHM at zero energy transfer. A sample was aligned with two reciprocal axes (1 0 0) and (0 1 1) in the scattering plane. The choice of this scattering plane is due to that $q_{1D}$ = (1 0.3877 0.2953) axis is very close to the scattering plane (2.8° out).

## RAW DATA

After data reduction performed in MANTID and using Horace for data visualization, we obtained S(Q,w) maps for different field values, which are shown in Fig.1. The magnetic signal is clearly distinguishable in the energy region of $0.1 < E < 1$ meV. A flat dispersion-less mode is observed due to the second $Cu^{2+}$ ion at different energies depending on the strength of the magnetic field. A large elastic signal at zero energy transfer is seen mainly due to incoherent scattering from the sample. An additional field-independent scattering can be found at the top $0.75 < E < 1.6$ meV and at the lower part $E < 0.75$ meV in low-$h$ region $h < 0.3$ rlu of each spectrum. We could observe a similar scattering in the spectrum of an empty magnet, thus we conclude that these features are not from the sample and related to the sample environment and will be treated as a background.

## PRELIMINARY RESULTS

Data preparation consisted of the following steps. From each data-set, one had to subtract 1) background scattering, which is related to the sample environment; 2) incoherent scattering, and 3) multiple scattering. In addition, one had to correct data with the $Cu^{2+}$ magnetic form-factor. Fig.2 shows S(Q,w) maps after all applied corrections together with theoretical calculations provided by M.Kohno. As one can see, experimental data and theory are in a good qualitative agreement apart from the flat dispersion-less mode due to the second copper ion. All features are well captured during the experiment including very weak ones in higher energy region associated with Bethe strings.

## ONGOING ANALYSIS

To be able to compare spin dynamics of 1D AFM chain with theory on the quantitative level, it would be nice if we could subtract paramagnetic mode from the experimental data-sets. For that, we might need more corrections to apply including non-trivial ones, such as transmission of the magnet. This work is currently in progress.

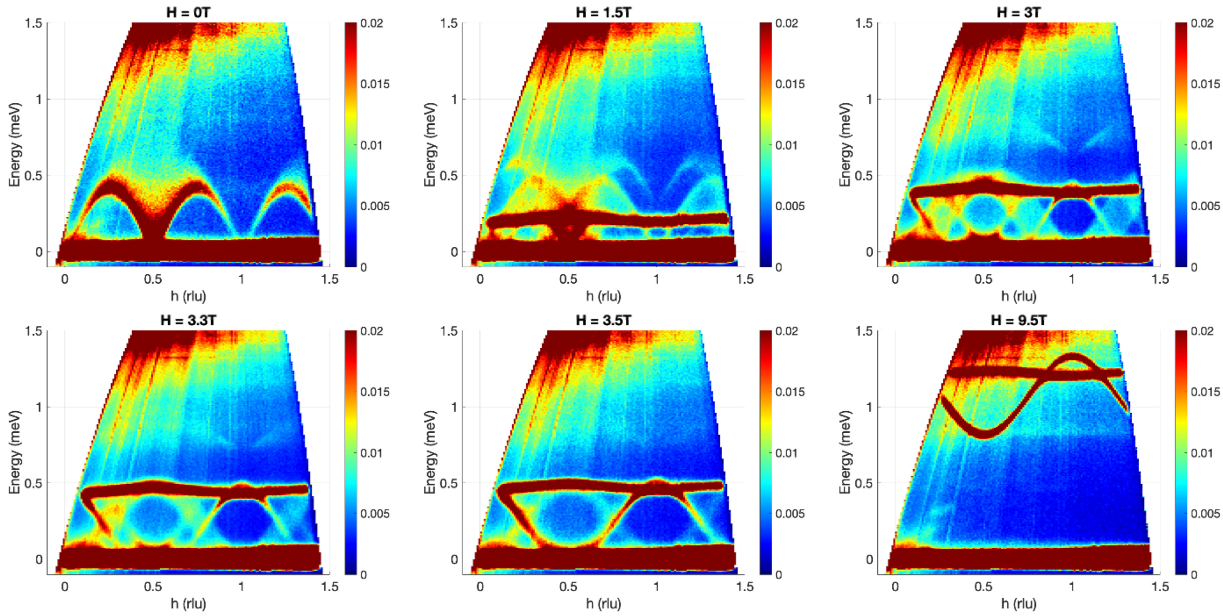


Fig.1. Measured dynamic structure factor S(Q,w) along chain direction in vertical magnetic fields H =0, 1.5, 3, 3.3, 3.5, 9.5T.

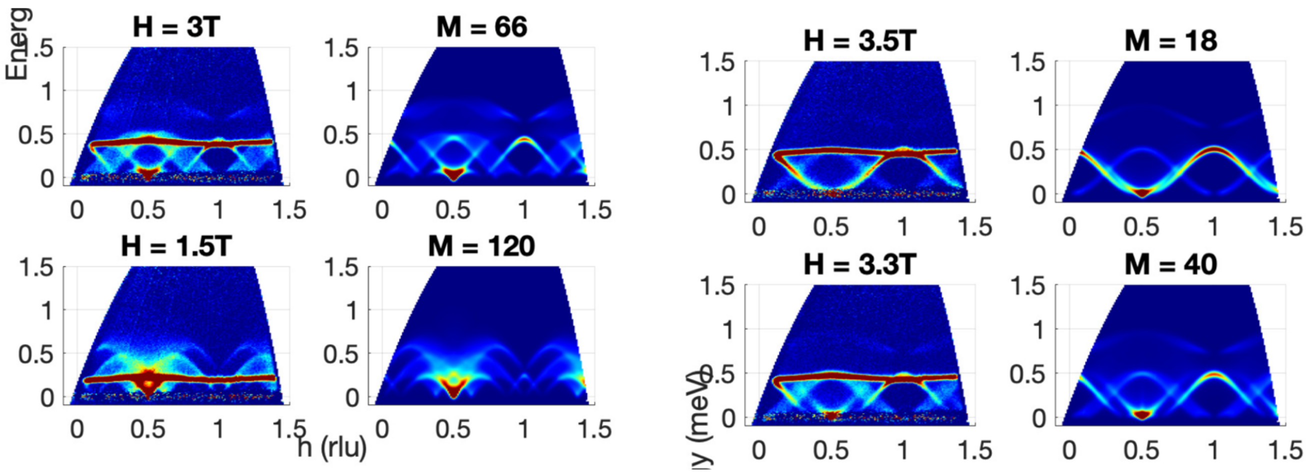


Fig.2. Experimental dynamical structure factors (left column) and corresponding Kohno's theoretical calculations (right column).

## REFERENCES

[1] C. A. Beevers and H. Lipson. 'The crystal structure of copper sulphate pentahydrate, $CuSO_4.5H_2O$'. In: *Proc. R. Soc. Lond. A* 146 (1934), p. 570.
[2] M. Mourigal et al. 'Fractional spinon excitations in the quantum Heisenberg antiferromagnetic chain'. In: *Nat. Phys.* 9 (2013), p. 435.
[3] M. Kohno. 'Dynamically Dominant Excitations of String Solutions in the Spin-1/2 Antiferromagnetic Heisenberg Chain in a Magnetic Field'. In: *Phys. Rev. Lett.* 102 (2009), p. 037203.
[4] M. W. Van Tol and N. J. Poulis. 'A high-field phase transition in the linear chain compound $CuSO_4 \cdot 5H_2O$'. In: *Phys.* 69 (1973), p. 341.