Probing one-dimensional fluctuations in a spin-1/2 antiferromagnetic chain using MACS

Introduction:

Recent developments in neutron instrumentation have allowed the study of fundamentally new phenomena in quantum mechanics. One example, which is the subject of this case study, is the excitation spectrum of the spin-1/2 chain. For a ground state antiferromagnet, the low-energy fluctuations are characterized by propagating spin-waves (or magnons) and these can be derived using first order perturbation theory in a similar way to the calculation of lattice excitations (or phonons). Such theories involve the use of second quantization and assume a spin ground state and proceed by calculating transverse spin fluctuations.

While these techniques and theories have been largely successful in describing the low-energy excitations in a variety of materials, they do not take into account quantum effects which become important in investigating the fluctuations of low spin magnets. The corrections owing to the value of the spin, is elegantly demonstrated by the following Laurent series for the correction to the spin-wave stiffness for a two-dimensional Heisenberg antiferromagnetic derived by Igarashi [1].

$$Z_p = 1 - \frac{0.235}{2S} - \frac{0.041 \pm 0.003}{(2S)^2} + \dots$$

It can be seen from inspection that larger the spin-values (S), the less important the corrections. However, for small spin-values and in particular the case of S=1/2, the corrections become large and therefore a careful treatment of the spin dynamics must take into account quantum mechanics.

The example of a spin-1/2 antiferromagnetic chain is an extreme case as it involves both low spin and also low-dimensionality. The ground state does not exhibit long-range order, as was first found Bethe [2], and differs significantly from the classical Neel type order found in a classical model. A classic antiferromagnetic chain can be readily solved using perturbation theory and the analysis produces the low-energy dispersion to have the following form.

$$E = 2J|\sin(Q)|$$

This relation has been confirmed in a number of linear-chain antiferomagnets with the classical S=5/2 system (TMMC) being a prototypical example [3].

For S=1/2, the solution is quite different. Des Cloizeaux and Pearson showed that the lowest-lying excited states are given by the following formula [4].

$$E = \pi J|\sin(Q)|$$

In this case study, we will evaluate the validity of this expression and characterise the dynamics of a spin-1/2 chain (CuPzN).

S=1/2 one-dimensional chain (CuPzN):

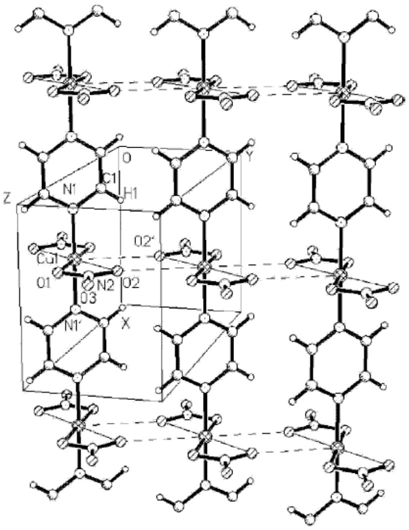

Figure 1*: Crystal structure of CuPzN showing the Cu2+ ions (hatched spheres) are linked through pyrazine rings to form one-dimensional chains. The chain axis (a) is vertical on the page, with the b axis nearly horizontal.*

CuPzN, $Cu(C_4H_4N_2)(NO_3)_2$, is orthorhombic with space group *Pmna.* The magnetic copper ions form chains along the [100] direction, with one copper unit cell along he chain [5]. The Cu ions along the chain are coupled magnetically through the pyrazine molecules, as illustrated in Fig. 1.

The crystals studied here, were grown by slow evaporation of aqueous solutions of Cu(II) nitrate and the corresponding pyrazine. Large single crystals formed after several months with faster evaporation resulting in smaller needle like crystals. The same used here has total mass of 73 mg and consists of two crystal coaligned with a mosaic of less than 0.5 degrees.

Given that the pyrazine provides the means of the superexchange (and the coupling between S=1/2 Copper ions), the magnetic dynamics are expected to be highly one-dimensional. The chains are held together by strong covalent bonds, where the bonding between chains is due to much weaker Van der Waals forces.

MACS Cold triple-axis spectrometer:

To investigate the spin fluctuations in this one-dimensional system, the MACS cold triple-axis spectrometer was used. MACS consist of 20 spectroscopic detectors which are used in conjunction with a double bounce graphite analyzer. Further details on MACS will be provided during the presentation and tutorial. For this experiment, a fixed final energy of 5meV was used and the sample was cooled using an orange cryostat.

The sample was aligned in the HK0 scattering plane. To map out the excitations, the chain axis was placed perpendicular to the incident beam so that all detectors sampled a unique value along the **a** direction. This is a unique method that can be used in low-dimensional systems and allows the use of multi-detector systems to be of great value in the study of these systems. Further examples of how this has been used effectively for the study of other low-dimensional magnets (such as the cuprate superconductors) will be discussed during the tutorial.

Des Cloizeaux and Pearson (dCP) relation:

During the first part of this tutorial, verify the dCP relation for CuPzN from the data extracted. Perform constant-Q cuts through the data and extract a value for the super exchange constant from the relation provided above.

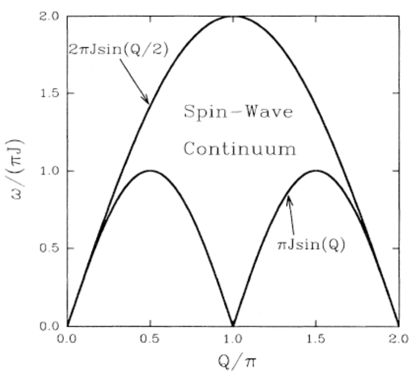

Figure 2: *A schematic of the spin-wave dispersion for a linear chain.*

Take a close look at the lineshapes from a constant-Q scan through the data. Are the peak widths symmetric and resolution limited? Do they represent well defined propagating spin-wave modes?

A careful investigation of the dispersion illustrates that while the overall dispersion seems to follow the dCP relation, there is a distinct high-enery tail to the lineshapes. A constant-Q slice illustrates the presence of a continuum lineshape which as first observed in the one-dimensional chain $KCuF_3$. A schematic of the excitation spectrum is presented in Figure 2.

Spinons:

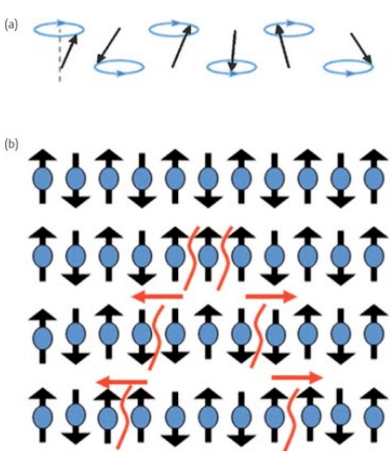

Figure 3: *Description of spinons and compared to linear transverse spin-wave excitations.*

Verify the dispersion above (and illustrated in Fig. 2) through the use of constant-Q slices. The high-energy continuum represents the fundamental excitation of S=1/2 chain known as a spinon. These excitations can also be interpreted as domain walls or solitons of the antiferromagnetic background.

The experimental signature of this excitation is significant spectral weight observed over a broad range in momentum and energy transfer. Because the excitations are broad in energy and momentum, they are typically quite difficult to measure and characterize and were first measured in the linear chain $KCuF_3$ using the MARI chopper time-of-flight spectrometer at ISIS [6]. The presence of a multi detector array and large incident flux, such as available on MACS, allows these types of excitations to be readily measured and characterized.

References:

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