ISIS Neutron Training Course – Single Crystal Spectroscopy Practical

Introduction

The material we will study is copper germanate, $CuGeO_3$. This material is quite close to being an ideal 1-dimensional spin chain, with a strong exchange interaction (J) between spins in the chains but rather weak interactions between chains. $CuGeO_3$ exhibits two interesting phenomena – spinons, and a spin-Peierls transition.

You can understand what spinons are by looking at figure 1 below. In the ground state, at time=zero, all of the Cu^{2+} spins (which have S=1/2) in the chain are antiferromagnetically aligned. A neutron comes in, and flips over one of the spins by magnetic interaction, and loses some energy in the process (this is what we will be measuring later), at time=t1. The system is no longer in its ground state, but something interesting unfolds as we allow time to pass. The overall change in energy is zero if we then flip more spins in such a way that we separate the chain into 3 domains, in each of which the preferred antiferromagnetic alignment is sustained. The excitations of such a system are therefore the domain walls, which can propagate along the chain. The S=1/2 entity at the domain wall is, in this simplified picture, the spinon. (Note that spinons are fermions, whereas magnons have S=1 and are bosons).
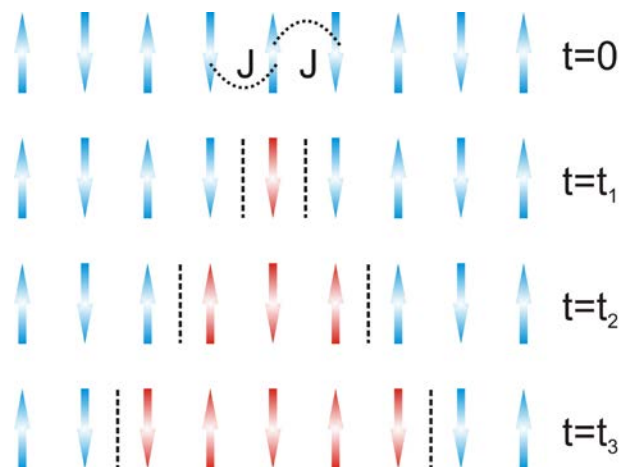

Figure 1 – Evolution of a spin-flip excitation in a 1D Heisenberg S=1/2 antiferromagnetic chain.

The other phenomenon that we are hoping to see evidence of is a spin-Peierls transition, shown in figure 2. This is a transition where the one-dimensional lattice distorts due to magneto-elastic coupling, as doing so puts the system in a lower energy ground state. In the case of $CuGeO_3$ this distortion takes the form of pairs of spins moving slightly closer together – this is known as weak dimerization. The effect is that instead of each spin interacting by exchange with equal strength J to both its nearest neighbours, as is shown at t=0 in figure 1, the interaction between dimerized spins is a little bit stronger (J_1) and between spins in different dimers it is a bit weaker (J_2).
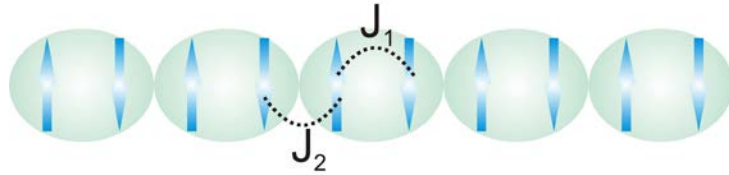
Figure 2 – Dimerization of the spins in the chain due to a spin-Peierls transition.

The physics of spinons is actually pretty complicated. However the prediction for what you expect to measure ends up being pretty simple. There is a continuum of allowed energies and momenta/wavevectors for the spinons, and the lower and upper boundaries are given by the following equations:

$$E_{lower} = \pi \mid J \, sin(qa) \mid \text{..............(1)}$$

$$E_{upper} = \pi \mid 2J \, sin(qa/2) \mid \text{........(2)}$$

Here J is the exchange between nearest neighbour Cu^{2+} ion, a is the spacing between atoms in the chain, and q is the wavevector of the excitation. (Note that we are ignoring the more complicated dimerized spin case for now).

Exercise 1: Make a sketch of the boundaries of the spinon continuum given by equations (1) and (2), so you know what we are expecting to see in our measurement.

Mounting the Sample

We will mount the sample in the closed-cycle refrigerator (CCR) on MERLIN, on the end of a stick which can be rotated inside the instrument. This is the standard setup on MERLIN. Our first consideration is what to use to shield the sample:

Q1: What governs our choice of shielding material?

Q2: What should we use – cadmium or B_4C?

Q3: What do we need to shield, and what should we avoid shielding?

Once you have answered these questions you will be ready to mount the sample, and load it onto the instrument. Take care to mark the part of the stick that is visible when in the CCR with an indication of the crystal's orientation.

Alignment

Before we do our measurement we need to know the orientation of the crystallographic axes with respect to the spectrometer. The lattice parameters of $CuGeO_3$ are given in the table below.

a (Å)	4.81
b (Å)	8.47
c (Å)	2.94
α (deg)	90
β (deg)	90
γ (deg)	90

Table 1 – lattice parameters of $CuGeO_3$

The rotation stage of the MERLIN CCR only has one degree of freedom, rotation about the vertical axis. This means that before the experiment started we had to choose a plane that would be horizontal. One of the axes in the plane is the crystallographic **c*** direction. To find the alignment quickly we will run MERLIN in white beam mode, with the Fermi chopper lifted out of the beam. This means that we will be able to see all of the Bragg reflections with accessible d-spacing in all of the detectors.

Exercise 2: To help understand what we will see, calculate the d-spacing of the Bragg reflections from the principal axes (i.e. (0,0,1), (0,0,2), etc), and also the following mixed reflections – (2,2,0), (2,0,2), (0,2,2).

With the help of your demonstrator you should now be able to identify the orientation of your sample with respect to the instrument.

Choosing a Configuration

In $CuGeO_3$ the 1-dimensional chain axis lies parallel to **c***. In figure 3 we show two possible orientations of the crystal – with the chain axis parallel to the incident neutron beam, and the chain axis perpendicular to the incident neutron beam. The bold blue lines indicate the intersection of the portion of reciprocal space measured by the detectors with the dispersion.

Exercise 3: Once you have determined what orientation you want, calculate the value of the rotation angle of the CCR that will give you this orientation.

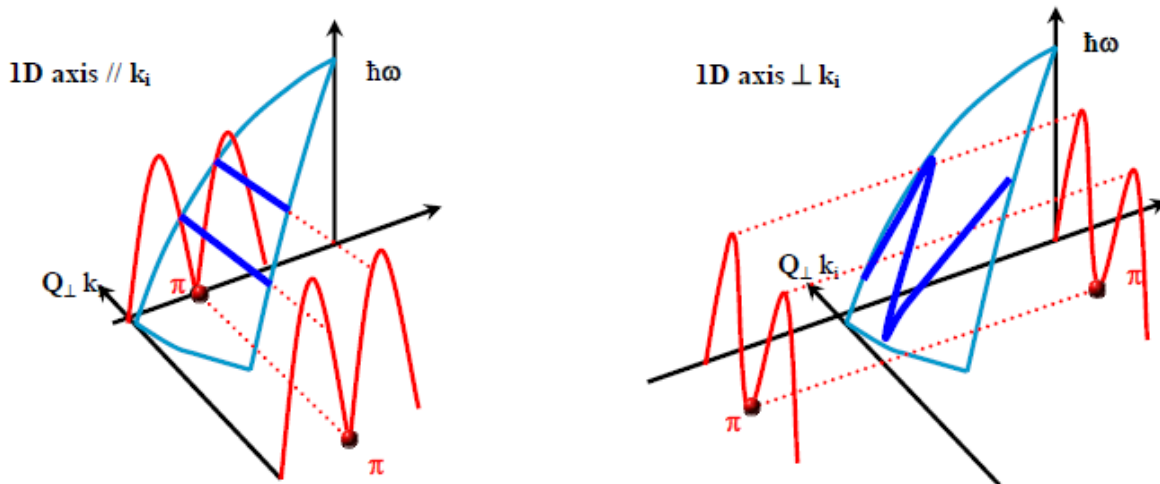*Q4: Which of these two orientations do you think would give the most informative measurement?*

Figure 3 – Generic dispersion of a 1D compound (red surface), projected onto the MERLIN detectors (blue segment) in the horizontal plane.

Choosing an Incident Energy and Chopper Frequency

We are now almost at the stage where we can do our measurement. After the Fermi chopper is back in place, we need to work out at what incident energy neutrons we would like to select with it, and at what frequency we want to spin it.

Q5: The chopper can only run at integer multiples of 50Hz. Why?

Exercise 4: Using the Pychop program, investigate what happens to the flux and energy resolution of the instrument when the chopper speed changes for a given incident energy.

We will use an incident neutron energy (E_i) of 50meV for this measurement.

Decide what chopper frequency we should use.