Experimental report

15/09/2023

Proposal: **4-05-849**

Council: 10/2022

Title: Magnetic excitations in a quantum spin ladder (C5H9NH3)2CuBr4

Research area: Materials

This proposal is a new proposal

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Samples: (C5H9NH3)2CuBr4

Instrument	Requested days	Allocated days	From	To
IN5	8	8	05/06/2023	13/06/2023

Abstract:

Quantum spin ladders bridge the gap between one and two dimensions and have complex excitation spectra, which depend on the relative strengths of the leg and rung exchange interactions. While the realization of strong-rung ladders is fairly common, only one strong-leg ladder has been studied in detail so far. Here, we propose to use neutron spectroscopy to unveil the magnetic excitations in the previously overlooked quantum spin ladder, (C5H9NH3)2CuBr4, and confirm whether it is a strong-leg system as suggested by bulk measurements.

Magnetic excitations in a quantum spin ladder $(C_5H_9NH_3)_2CuBr_4$.

Introduction

Quantum spin ladders describe coupled chains of spins with a two distinct exchange interactions: an inter-chain interaction, J_{leg}, and an intra-chain interaction, J_{rung}. The relationship between these two interaction terms can drastically change the excitation landscape, making quantum spin ladders an exciting candidate for material based quantum simulators as the tuning of only two exchange parameters can represent a plethora of many body Hamiltonians.

To this end, the so called "strong-leg" (SL) regime is preferable as quantum fluctuations are more prominent. These systems are much rarer than their counterpart of the "strong-rung" (SR) regime, with only one SL material having been studied previously: $(C_7H_{10}N)_2CuBr_4$, other wise known as DIMPY. It is therefore necessary for new SL materials to be studied to identify the universal features. It is especially important that these materials can have tuneable exchange parameters.

$(C_5H_9NH_3)_2CuBr_4$ (referred to as CPA from now on) is a proposed SL material. CPA's orthorhombic structure with ladders running along the b direction (**Fig.1c**), with the interactions between copper spins proceeding through halogen bonds in series for the leg direction but in parallel for the rung direction, make it an ideal system to tune the exchange parameters with uniaxial pressure. From our bulk susceptibility measurements, we extracted exchange parameters of J_{leg}~1 meV and J_{rung}~0.5 meV (**Fig.1b**). Additionally, previous inelastic neutron scattering (INS) measurements show a clear dispersion we could fit to the SL model.

Here, we aimed to resolve the low-energy section of the magnetic dispersion as we saw a clear "two peak" feature at the gap in previous INS measurements. We also possibly saw such behaviour throughout the dispersion but was very difficult to resolve and so wanted to investigate this as well. Additionally, capitalising on the higher sample mass and higher statistics available we aimed to resolve the whole excitation spectrum in detail and extract the key exchange parameters with higher accuracy but also interactions in other directions which we had not accounted for.

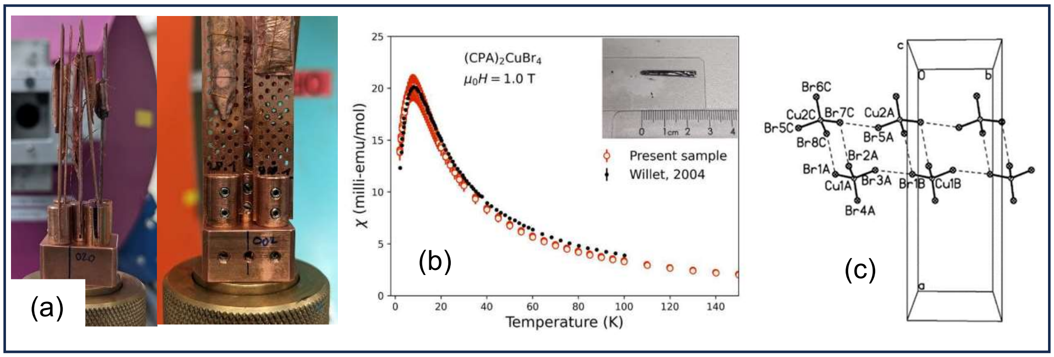

Figure 1: (a) the crystals aligned for this experiment. (b) magnetic susceptibility data on our samples showing SL behaviour. (c) the crystal structure showing the ladders living on the *b* axis.

Experimental Procedure

5 fully deuterated single crystals were coaligned using the ORION instrument at the Paul Scherrer Institute. The total mass was 968.7 mg. The LT1 dilution refrigerator was used to reach a base temperature of 50 mK. The same environment was used for later temperature scans of 100 mK, 200 mK, 300 mK, 500 mK, 1K, 3 K, 5 K and 10 K. Different incident neutron wavelengths were used to investigate each area of interest in the dispersion; 6 Å (low energy), 5.5 Å (up to top of dispersion) and 4 Å (far above top of the dispersion) were used. 4 Å was used to count the background.

Results

In the 6 Å scans, the two-gap feature is clearly identified with two distinct minima in the dispersion at *k* = 0.5. This confirms the feature we observed in previous INS measurements. When moving to the higher energy part of the dispersion using 5.5 Å, we see two very clear modes that were not visible in the previous experiments, with a possible tertiary mode at very top of the dispersion. This is backed up by cuts through the gap showing a third less intense peak. In between the distinct minima of the dispersion at integer point in *k* it is unclear whether the spectrum is dispersive or whether the mode has a minima. In the 4 Å measurements, even more features are seen in the higher energy spectrum. These are presently tough to identify but could be arising from additional exchange interactions. When cutting through the *h-l* plane we see a clear one-dimensional dispersion. This confirms that our dispersion is contained along the *b* direction, but we have additional exchange interactions/parameters that we have not accounted for yet.

The temperature dependence of the excitation spectrum shows that the magnetic excitations are present even up to 1 K with only a small decrease in intensity. At 3 K they are also present but the modes begin to overlap as they become more disperse. At 5 and 10 K, the excitations are almost completely disappeared.

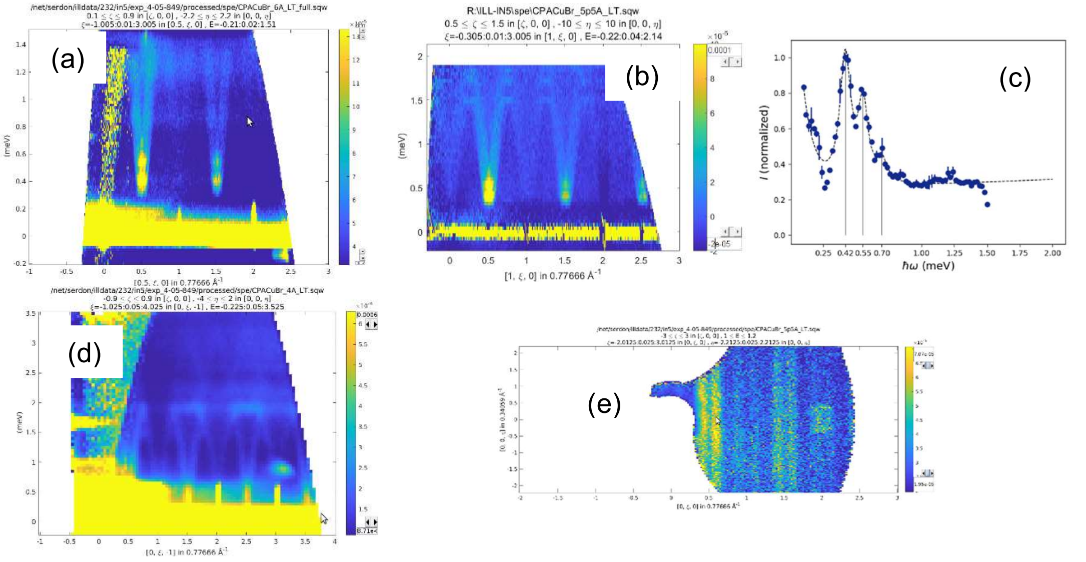

Figure 2: (a) 4 Å and (b) 5.5 Å and (d) 6Å measurements. (c) the three peaks seen when cutting across the gap. (e) the one dimensional dispersions in the *h-l* plane

Conclusions and Future Prospects

In total, we have observed the magnetic excitation spectrum of CPA in great detail. We have identified three modes within the dispersion, suggesting that this system does not behave in the traditional SL regime but there are additional exchange parameters that are not being accounted for in our current thinking. We have also observed that the excitations are clearly one dimensional, confirming out understanding that they are contained to live along the *b* axis. The temperature dependence also shows no drastic changes to the excitation spectrum, only a decrease in intensity where the spectrum dies at around 5 K.

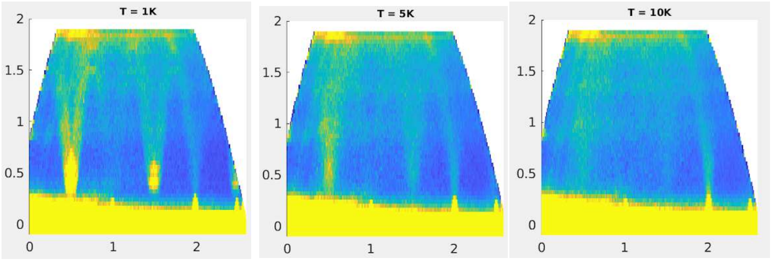

Figure 3: the temperature dependence of the dispersion up to 10 K.

It is clear further analysis is needed to understand the fundamental physics underlying the excitation spectrum in this material. A more careful background subtraction, where one removes the phononic features from the dispersion, could help in revealing the three modes in greater detail. It is also clear that a better understanding of this material from a theoretical point of view is needed. Simulations and modelling to match this dispersion will also be attempted. Further studies of this material under field and pressure to see how the modes change would also give us a clearer understanding of the exchange interactions in the material.