Experimental report

12/11/2018

Proposal: 4-05-704 **Council:** 4/2018

Title: Magnetic dimer excitations in azurite

Research area: Physics

This proposal is a new proposal

Main proposer: Ursula Bengaard HANSEN

Experimental team: Ursula Bengaard HANSEN

Local contacts: Mechthild ENDERLE

Samples: Cu3(CO3)2(OH)2

Instrument	Requested days	Allocated days	From	To
THALES	7	0		
IN12	7	0		
IN20	14	7	04/09/2018	11/09/2018

Abstract:

The natural mineral azurite (Cu3(CO3)2(OH)2) has been proposed as a candidate model of the distorted diamond chain. Magnetisation measurements have revealed a characteristic 1/3 plateau, that is the signature of a spin fluid phase within this model. However, the ratio and sign of the exchange couplings, describing the magnetic nature of azurite, are still under much debate. Previous inelastic neutron scattering experiments have shown a complex excitation spectrum where there are signatures of both dimer and monomer spins, that could not be described using a simple 1d Heisenberg antiferromagnetic chain model. Here we propose to study the excitation spectrum of azurite in more detail using both polarised and unpolarised neutrons in hope of answering some of the open questions regarding the spin dynamics in this system.

Experimental report: Magnetic dimer excitations in Azurite

no. 4-05-704

Introduction

The natural mineral Azurite ($Cu_3(CO_3)_2(OH)_2$) is a proposed model system for the distorted diamond chain model. Here the spins-1/2 are arranged in a diamond shaped units, that are assembled into a chain system, as depicted in Fig. 1. The exchange couplings consist of a dimer bond, J_2, connecting the two magnetic sites across the diamond, and two exchange couplings J_1 and J_3 connecting the monomer sites along the chain. This system displays dimerisation, ferrimagnetism and a spin-fluid phase depending on the ratio between the exchange couplings J_1 , J_2, and J_3 [1]. One of the characteristics of the distorted diamond chain with $J_2 >> J_1, J_3$ is a characteristic plateau in the magnetisation curve at 1/3 of the saturation magnetisation. The origin of this plateau can be explained by the formation of a singlet state by the two dimer sites connected by J_2, while the spin on the third monomer site will be polarised along the field. Since this configuration is based on a singlet state, it is completely of quantum nature, without any classical analog. However, despite several studies the size and relative importance of the exchange couplings in Azurite are still under debate [2-4].

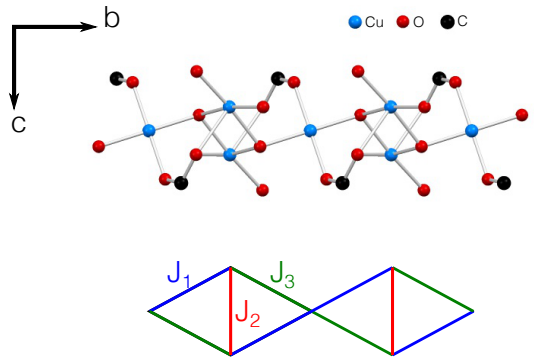

Fig. 1: The structure of azurite and the proposed diamond chain model.

Experimental setup

The IN20 instrument was used without any polarisation having k_f fixed to 2.662 $Å^{-1}$. The magnetic field at the sample position was controlled using the 15 T cryomagnet. The large single crystal of azurite (mass = 3 g) was mounted in the (H K H) scattering plane on an aluminium sample holder.

Results

In this experiment we have studied the magnetic dimer excitations of Azurite both in zero field and at 9 T, in the plateau phase. Examples of the inelastic neutron scattering data at 9 T are shown in Fig. 2. The data show a subtle modulation of the lowest lying triplet branch which hints to a more complicated exchange coupling scheme than the one proposed in Fig. 1. The excitation spectrum of between 0 and 2 meV in the (H K H) plane was not within the suitable range of the IN20 instrument and would be interesting to study using a cold triple axis instrument.

In the following table an overview of the different Q- and energy ranges covered in this experiment are summarised.

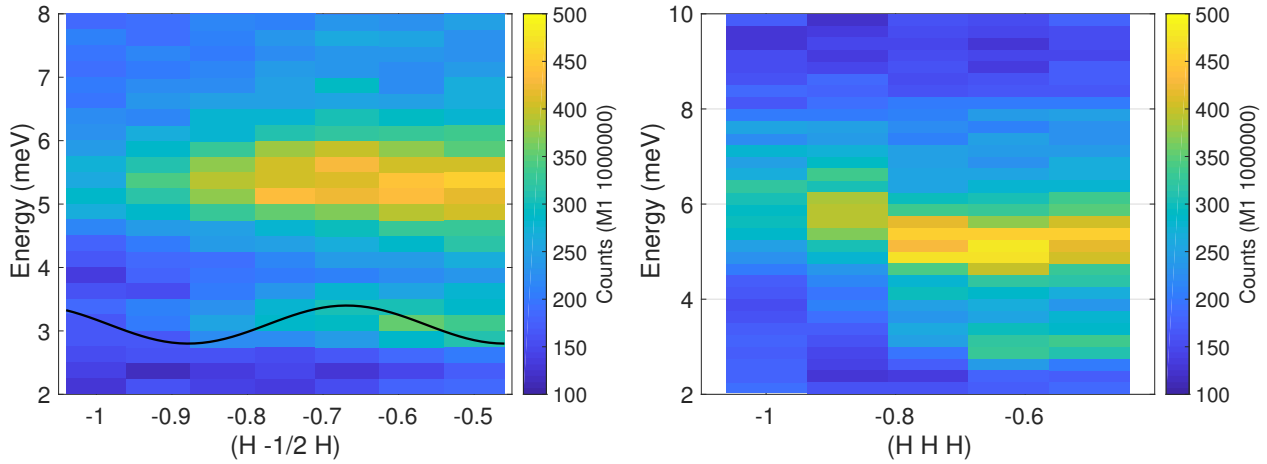

Fig. 2: Inelastic neutron-scattering data showing the dispersion along (H -1/2 H) and (H H H). Measurements were taken at 2 K and 9 T using the thermal triple axis instrument IN20. The black line is a guide to the eye.

Q	Energy range	Counting time	H	T
(-1 0 -1), (-1 -0.5 -1), (-0.75 -0.5 -0.75), (-0.5 -0.5 -0.5), (-0.75 -0.75 -0.75), (-0.5 -0.75 -0.5), (-0.25 -0.75 -0.25), (-0.5 -1 -0.5), (0 -0.75 0), (0 -1 0), (1 -0.5 1), (0.75 -0.5 0.75), (0.5 -0.5 0.5), (0.75 -0.75 0.75), (0.5 -0.75 0.5), (0.25 -0.75 0.25), (0.5 -1 0.5),	2 to 10 meV in 0.25 steps	~2 min pr point.	0 T	1.8 K
(-1 0 -1), (-1 -0.5 -1), (-0.75 -0.5 -0.75), (-0.5 -0.5 -0.5), (-0.5 -0.75 -0.5), (-0.5 -1 -0.5), (1 -0.5 1), (0.5 -0.5 0.5), (0.5 -1 0.5)	2 to 10 meV in 0.25 steps	~2 min pr point.	9 T	1.8 K
(0 K 0) K = -1, -1.1, -1.2, -1.3, -1.4, -1.5, -1.6, -1.7, -1.8, -2.0	2 to 12 meV in 0.25 steps	~6 min pr point	9 T	1.8 K

[1] K. Okamoto *et al*., J. Phys. Condens. Matter 11, 10485 (1999), K. Okamoto *et al*., J. Phys. Condens. Matter 15, 5979 (2003). [2] H. Kikuchi *et al*., Phys. Rev. Lett. 94, 227201 (2005).
[3] K. C. Rule *et al*., Phys. Rev. Lett. 100, 117202 (2008). [4] J. Kang *et al*., J. Phys. Condens. Matter 21, 392201 (2009).