Experimental report

05/03/2021

Proposal: 4-05-828 **Council:** 10/2020

Title: The natural mineral szenicsite: a frustrated antiferromagnetic chain with competing NN and next NNN interactions

Research area: Physics

This proposal is a resubmission of 4-04-502

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Experimental team: Ursula Bengaard HANSEN

Local contacts: Bjorn FAK
Mechthild ENDERLE
Ursula Bengaard HANSEN

Samples: Cu3(MoO4)(OH)4

Instrument	Requested days	Allocated days	From	To
IN20 Flatcone	7	0		
PANTHER	3	3	04/02/2021	08/02/2021

Abstract:

A simple frustrated magnetic model system is the spin-1/2 linear chain with competing nearest neighbour (NN) and next-nearest neighbour (NNN) interactions. Despite having been extensively studied in theory, no real material with antiferromagnetic NN- and NNN interactions and a non-zero excitation gap have ever been observed. Here we propose to study the natural mineral szenicsite (Cu3(MoO4)(OH)4) that has recently been proposed as a model candidate material. In the proposed interaction scheme, the magnetic Cu2+ ions form triple chains along the c-axis. The two outer chains are predicted to be decoupled from the central chain due to frustrated interchain couplings, they can be described as weakly 1D coupled antiferromagnetic dimers. The central chain is predicted to be a quasi-isolated uniform Heisenberg chain with antiferromagnetic NN and NNN interactions in a ratio close to the exactly solvable Majumdar-Ghosh model. We here propose to study the nature of the magnetic excitations emerging from the two types of 1D chains in szenicsite by means of inelastic neutron scattering.

The natural mineral szenicsite: a frustrated antiferromagnetic chain with competing NN and next NNN interactions

- Proposal number: 4-05-828
- dates: 04/02/2021-07/02/2021, 4 days
- Local contact: Björn Fåk
- sample: Szenicsite (Cu3MoO4(OH)4) polycrystalline sample, m = 7.55 g
- mounted on an aluminium pin

The natural mineral szenicsite (Cu3(MoO4)(OH)4) is very rare and large single crystals has only been found in one location, a small area of approximately 1 m3 in the Jardinera No. 1 mine in Chile's Atacama province [1]. This mineral has recently been proposed as a model candidate for the spin-1/2 the spin-1/2 linear chain with competing nearest neighbours (NN) and next nearest neighbour (NNN) interactions [2], which is one of the simplest model system in low dimensional magnetism. When the NNN coupling J_2 is antiferromagnetic the chain is frustrated regardless of the sign of the NN coupling J_1. Despite being conceptually simple, the model has a very interesting phase diagram governed by the ratio between the two exchange couplings $\alpha = J_2/J_1$.

However, in the case of szenicsite, the picture is more complicated than the simple 1D-model. The crystal structure of szenicsite is orthorhombic (Pnnm) with lattice constants a = 12.559 Å, b = 8.518 Å and c = 6.072 Å [1,5]. Within the unit cell, there are three different sites of the magnetic spin-1/2 Cu2+ ions and as a result the Cu2+ ions form triple chains along the c-axis. The CuO4 plaquettes of the Cu2- and Cu3-sites share common edges and constitute, flat ribbon, the central chain. The two outer chains are made up from the corner sharing plaquettes of the Cu1-ions. Each triple-chain subsystem is predicted to be decoupled from the next triple-chain subsystem [2]. A microscopic magnetic model of szenicsite has been derived using density functional theory [2]. In Fig. 1 the structure of the Cu-chains are shown together with a schematic drawing of the proposed exchange coupling scheme. In Ref. [2], it is argued that the frustrated interchain exchange couplings (J_a & J_b) essentially decouple the central chain from the two side chains. The two side chains can then again be understood as coupled 1D AFM dimers, where the coupling between the dimers is predicted to be almost negligible, $J'_D/J_D = 0.06$ [4]. The middle chain is hence an effectively isolated uniform AFM Heisenberg chain with NN and NNN interactions, the predicted ratio between the two couplings indeed puts szenicsite close to the Majumdar-Ghosh point: $J_2/J_1 = 0.45$ [3].

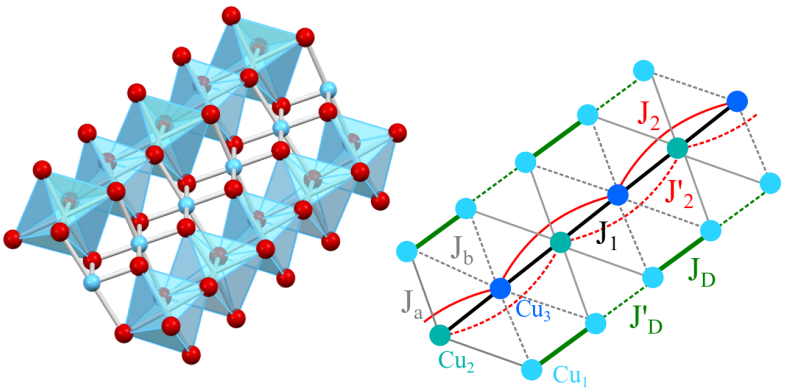


Fig. 1: Crystal structure of the triple chains and a schematic drawing of the exchange couplings in the microscopic magnetic model of szenicsite.

Both the DFT calculations and susceptibility measurements support that there should be two different energy scales for the two types of magnetic excitations: ~6 meV for the AFM Heisenberg chain with NN and NNN interactions and ~16 meV for the magnetic dimer excitations.

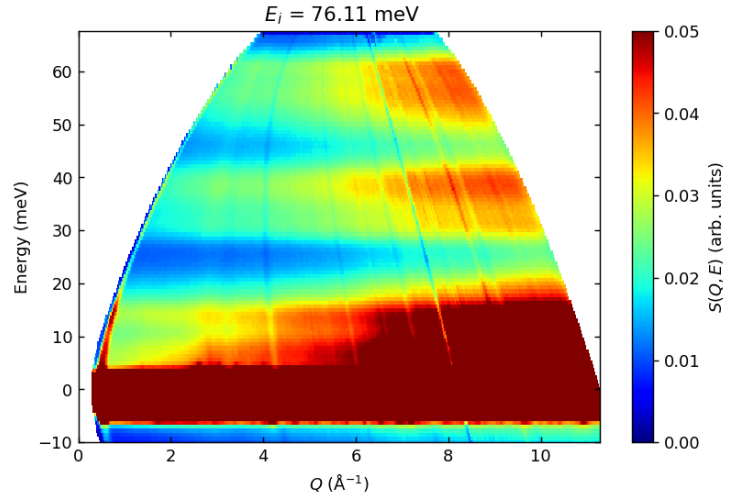
The goal of the PANTHER experiment was to get an overview of the different magnetic excitation energies. It is clear that the energy scale central NN- and NNN AF-chain might be too low to be accessible within the PANTHER energy range. The polycrystalline was mounted on an aluminium pin and was rotated during the experiment using an A3-rotation stage in order to emulate a powder spectrum.

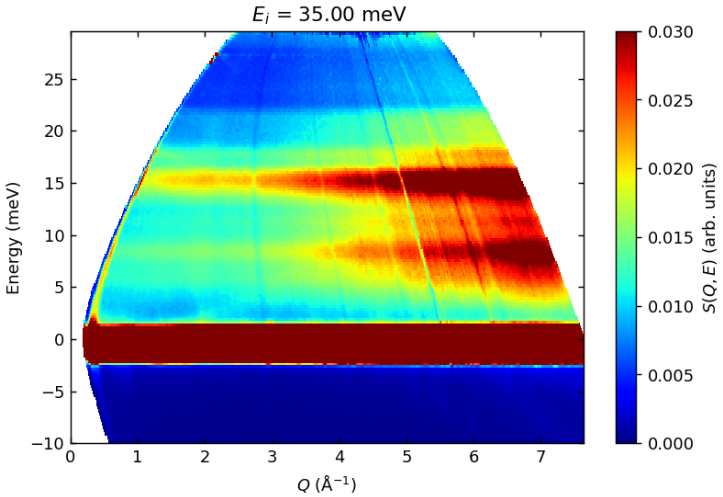
The table below contains on overview of the different experimental conditions including the counting time for each setting and in the figures below is an overview of the measured spectra.

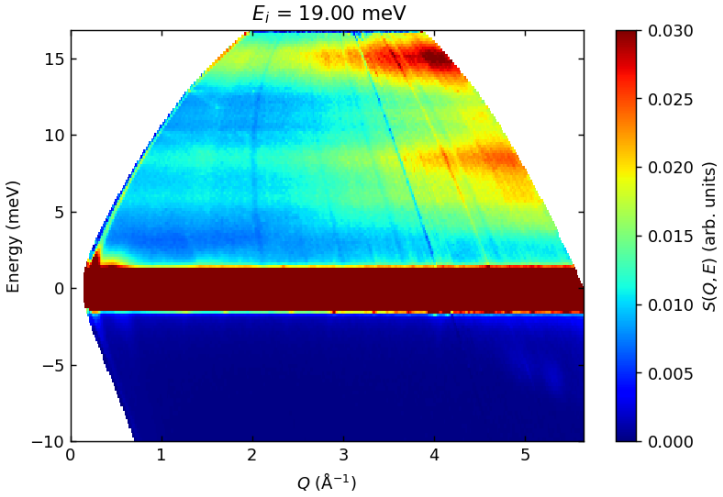
Ei (meV)	T (K)	A3 (°)	A3 step (°)	time total (mn)
12.5	2	60 : 240	30	620
12.5	54	60 : 240	30	420
12.5	110	60 : 240	30	480
19.0	2	60 : 240	30	528
19.0	110	60 : 240	30	420
35.0	2	60 : 240	15	865
35.0	110	60 : 240	30	600
76.1	2	60 : 240	30	420
76.1	110	60 : 240	30	420

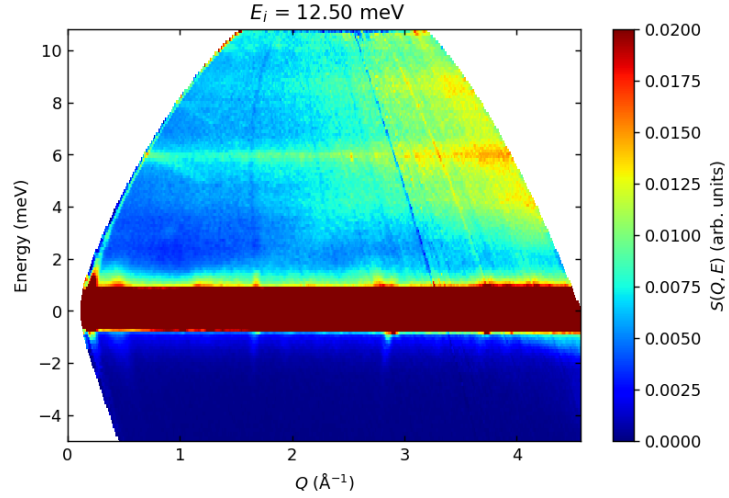
[1] P. C. Burns, Mineralogical Magazine, 62(04), 461–469, (1998) [2] S. Lebernegg, et al. Phys. Rev. B, 95(3), 5-8 (2017) [3] C. K. Majumdar and D. K. Ghosh, Jour. of Math. Physics, 10(8), 1388–1398 (1969) [4] K. Okamoto and K. Nomura, Phys. Lett. A, 169(6), 433–437 (1992) [5] J. Stolz and T. Armbruster, Neues Jahrb. Mineral. (Monatsh), 278 (1998).

T = 2 K

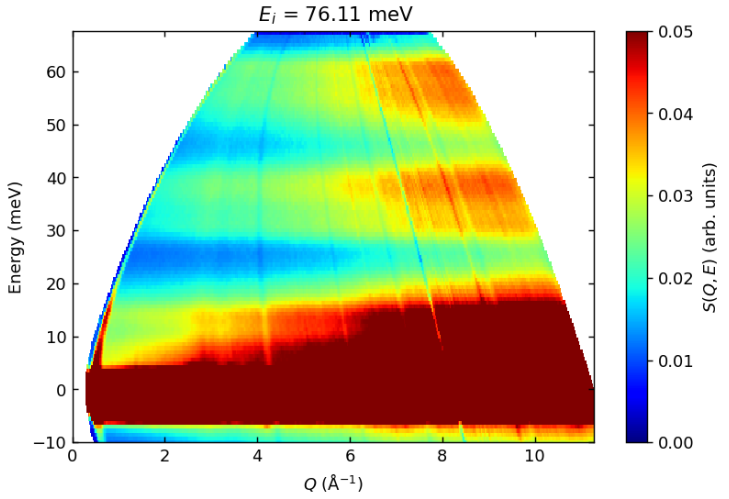

E_i = 76.11 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)

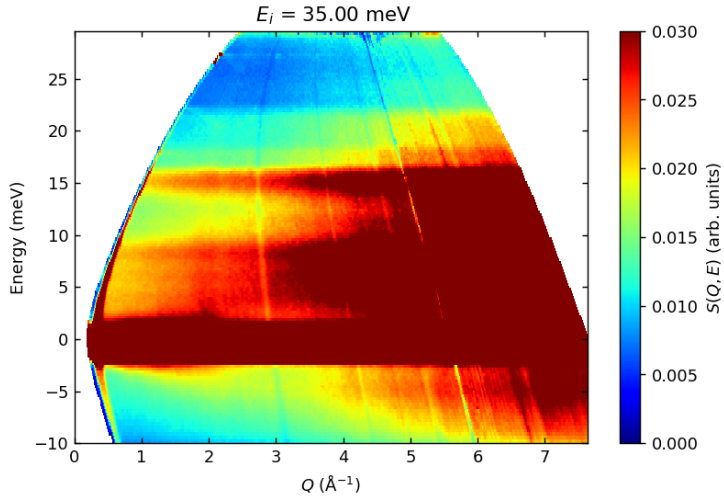

E_i = 35.00 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)

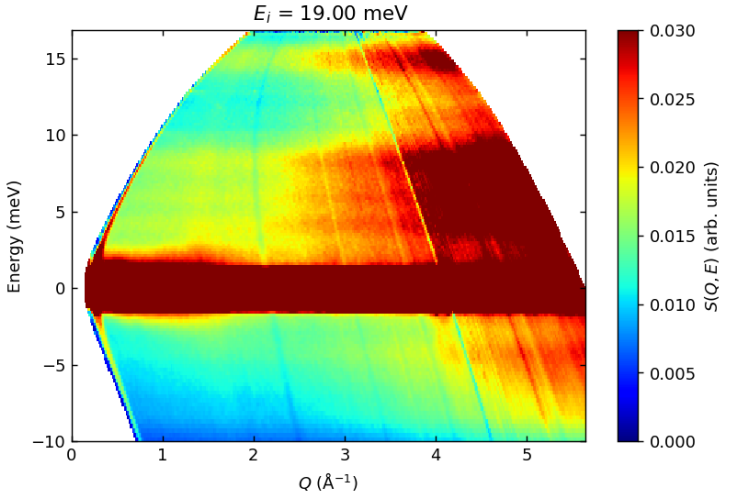

E_i = 19.00 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)

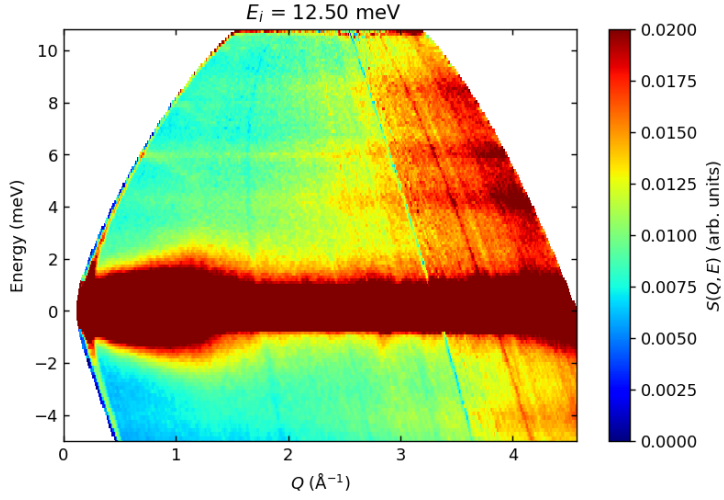

E_i = 12.50 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)

T = 110 K


E_i = 76.11 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)


E_i = 35.00 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)


E_i = 19.00 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)


E_i = 12.50 meV
Energy (meV)
Q (Å^{-1})
$S(Q,E)$ (arb. units)