Experimental report

02/02/2016

Proposal:	**4-04-469**	**Council:**	10/2014
Title:	Magnetic Excitation in Mn2Ni Single Chain Magnets		
Research area:	Physics		

This proposal is a new proposal

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Samples: [Mn2(saltmen)2Ni(pao)2(py)2](ClO4)2 (abbreviated Mn2Ni)

Instrument	Requested days	Allocated days	From	To
IN6	2	2	03/08/2015	05/08/2015

Abstract:

Single-chain magnets are an interesting, novel class of molecular nanomagnetic materials. In order to understand the relaxation of the magnetization in these materials, it is essential to understand the lowest energy excitations. These were predicted to be either excitations localized on one of the units composing the chain, or collective excitations delocalized over the chain (spin waves). We have studied the single chain magnet abbreviated as Mn2Ni by by means of broadband high-frequency electron paramagnetic resonance. We observe a zero-field transition at ca. 331 GHz (1.37 meV) and a further, weaker transition. For a local excitation, one would expect the ground state transition at 5D, and excited state transitions at 3D and D, where D is the second rank axial zero-field splitting. However, no excited state transitions were observed. Inelastic neutron scattering is the ideal technique to resolve this issue, leading to an improved understanding of the relaxation of magnetization in Mn2Ni and in single-chain magnets in general. Excitation energies up to 1.4 meV are involved.

Experimental report for “Magnetic excitation in Mn_2Ni single chain magnet”

Single-chain magnets (SCM) are one-dimensional coordination polymers which exhibit a slow relaxation of the magnetization which is only of molecular origin. The have a (usualy) ferromagnetic intra-chain coupling and a negative single-ion zero-field splitting. We investigated here the SCM $[Mn_2(\text{saltmen})_2Ni(\text{pao})_2(\text{py})_2](ClO_4)_2$ (abbreviated Mn_2Ni), where saltmen^{2-} = *N*,*N*'-(1,1,2,2-tetramethylethylene) bis(salicylideneiminate), pao^- = pyridine-2-aldoximate and py = pyridine. The parameters of the spin system have already been determined by EPR spectroscopy, and are $J_{MnMn} = 0.224\,\text{meV}$, $J_{MnNi} = -3.20\,\text{meV}$ and $D_{Mn} = -0.439\,\text{meV}$. The aim was to find out whether the magnetic excitations in this system are localized excitations (spin cluster), where the tilted spin is localized on an atom in the chain, or collective excitations (spin wave), where the tilted spin is delocalized over the whole chain. Because of the wave-like structure of the latter, a Q dependence should be visible with inelastic neutron scattering. From EPR spectroscopy, spin wave behaviour was predicted, but we wanted to deepen this analysis and improve the degree of evidence.

A 900 mg non-deuterated, polycrystalline sample has been stored in aluminium foil, which has been put into a aluminium can. The measurements were performed with an ILL orange cryostat at a temperature of $T = 1.5\,\text{K}$ and an incident wavelength of $\lambda = 5.12\,\text{Å}$. For the treatment of the data, bad detectors have been removed and the spectra were normalized to vanadium.

The excitation was observed at an energy of of $E = 1.40\,\text{meV}$ and does not show a Q dependence on the first sight (see figure 1). Unfortunately, the intensity of the signal was not as high as This is due to the fact that we only had access to a polycrystalline sample because of synthetic difficulties. For a single-crystal, one would expect a Q dependence of $E = 2JS(1 - \cos(ka))$, where J is the coupling constant, S the spin quantum number, k the wave vector of the spin wave, and a the spatial distance between two spins. In a polycrystalline sample, the scattering vector can be decomposed into a vector parallel to the chain and a part perpendicular to the chain, $\vec{q} = \vec{q}_{||} + \vec{q}_{\perp}$, which leads to different wavelengths of the oscillation are expected due to different angles between the scattering vector and the direction of the chain. All these oscillations superimpose each other, which leads to a complex pattern, where the intensity of the excitation is dependent on the momentum transfer. This behaviour is indeed reproduced, as shown in figure 2, where the parameters from EPR spectra and the distance of the crystal structure have been used.

However, the analysis is not as clear as it might be. This is mainly due to the low intensity. The increase and decrease of the intensity over the momentum transfer might also arise from noise. Further measurements could improve this, especially if they are over a higher Q range.

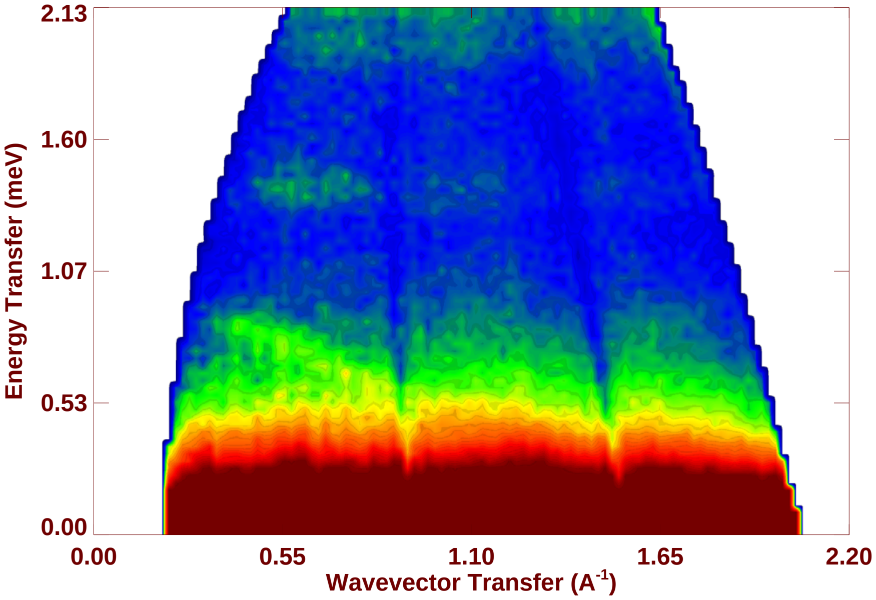

Figure 1: Q-E map of the INS spectrum of Mn2Ni at 1.5 K

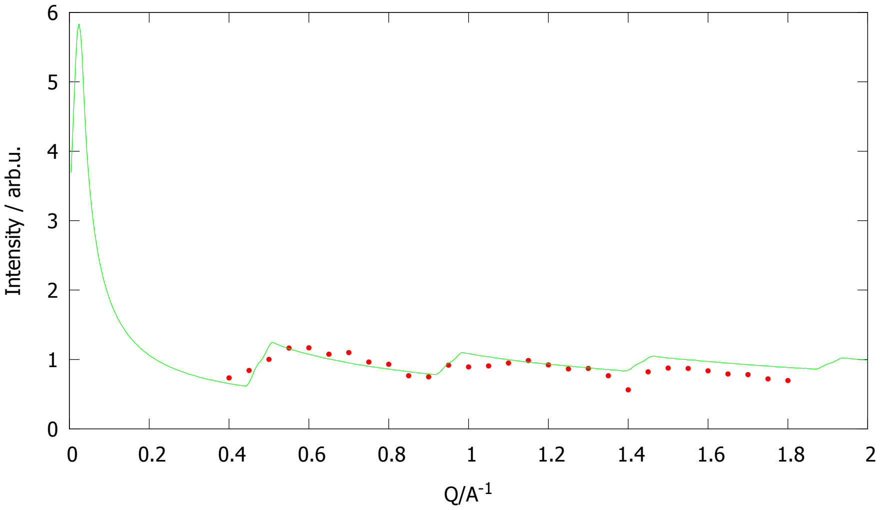

Figure 2: Q dependence of Mn2Ni at $E = (1.4 \pm 0.3)$ meV, experimental data (red dots) and the theoretical expected data (green line)