Experimental report

06/05/2022

Proposal: **5-32-915**

Council: 10/2020

Title: Uncovering correlated paramagnetism in mixed-metal 2D thiocyanates

Research area: Physics

This proposal is a new proposal

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Samples: Co_xNi_{1-x}(NCS)2

Instrument	Requested days	Allocated days	From	To
D7	4	4	22/05/2021	26/05/2021

Abstract:

Two-dimensional magnets offer a platform for exploring both fundamental questions in condensed matter physics and a potential route to new information technology integrating magnetic and electronic functionality. Recently, it has become clear that mixed-metal magnets can have properties distinct from either end member, suggesting that these materials could possess novel physics in their own right.

In this proposal we will investigate the transition between Co(NCS)2, a stripe antiferromagnet, and Ni(NCS)2, which has ferromagnetic interactions within the layer but orders antiferromagnetically, by studying mixed-metal compounds (Co,Ni)(NCS)2. Our bulk characterisation indicates that although they appear homogenous, there are significant non-linearities in their magnetic behaviour, indicative of short-range magnetic correlations. The unique capabilities of D7 will allow us to measure diffuse scattering signatures of both structural and magnetic correlations, and inderstanding how these correlations emerge will not only allow us to understand the behaviour of this interesting class of materials but shed light on the interaction between disorder and magnetism in layered magnets.

Uncovering correlated paramagnetism in mixed-metal 2D thiocyanates

Data was collected for the three mixed-metal thiocyanate frameworks $Co_xNi_{1-x}(NCS)_3$ x = 0.25, 0.5, 0.75 on the polarised diffuse scattering D7 diffractometer. The incident neutron wavelength was λ = 4.8707 Å, giving a reciprocal space range of $0.15 \leq Q \leq 2.5$ Å^{-1}. The FullProf program was used to refine the ordered magnetic phase at 1.5 K. The magnetic diffuse scattering data were analysed using the programs SPINVERT, SPINCORREL and SCATTY.

$Co_{0.25}Ni_{0.75}(NCS)_2$ was measured at four temperature points: 1.5, 25, 50 and 75 K. The magnetic Bragg reflections which appear at 1.5 K can be indexed to a propagation vector of (0, 0, ½) and a magnetic space group of C_c2_1/c. The magnetic diffuse scattering shows ferromagnetic correlations to an interatomic spacing (**r**) of 10 Å, decreasing in magnitude as **r** increases.

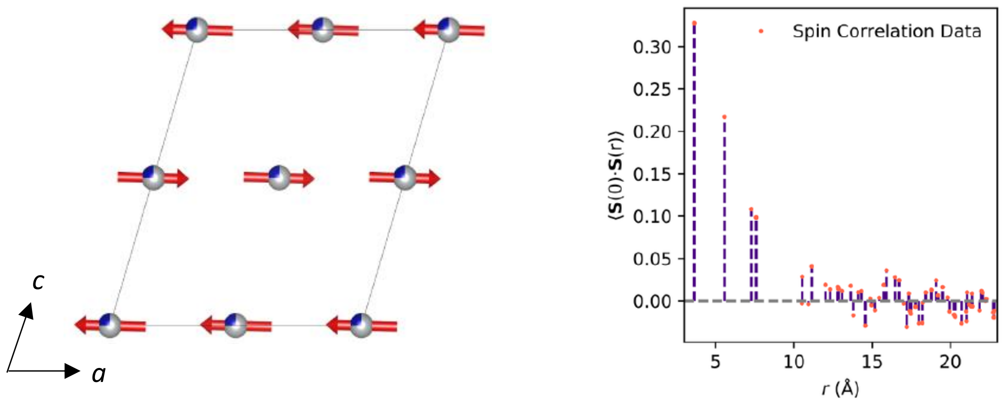

Figure 1. Left: Ordered magnetic structure of $Co_{0.25}Ni_{0.75}(NCS)_2$; right: spin-spin correlation function values for increasing atomic distances at 50 K.

$Co_{0.5}Ni_{0.5}(NCS)_2$ was measured at the temperature points: 1.5, 20 and 30 K. The magnetic Bragg reflections which were present at 1.5 K can be indexed to a propagation vector of (0, 0, ½) and can be described by the magnetic space group C_c2_1/c. The magnetic diffuse scattering shows weak ferromagnetic correlations up to 6 Å. At **r** greater than this the magnitudes of the correlations are small.

$Co_{0.75}Ni_{0.25}(NCS)_2$ was measured at the temperature points: 1.5, 20 and 30 K. The magnetic Bragg reflections which appear at 1.5 K can be indexed to a propagation vector (1, 0, 0) and a magnetic space group P_A2_1/c. The magnetic diffuse scattering, has ferromagnetic correlation to its nearest neighbor, whilst the rest of the correlations between **r** = 5 and 10 Å show predominately antiferromagnetic correlations which are significantly weak.

The results from this experiment will form part of the thesis for a current ILL PhD student and will be published in a widely read journal.

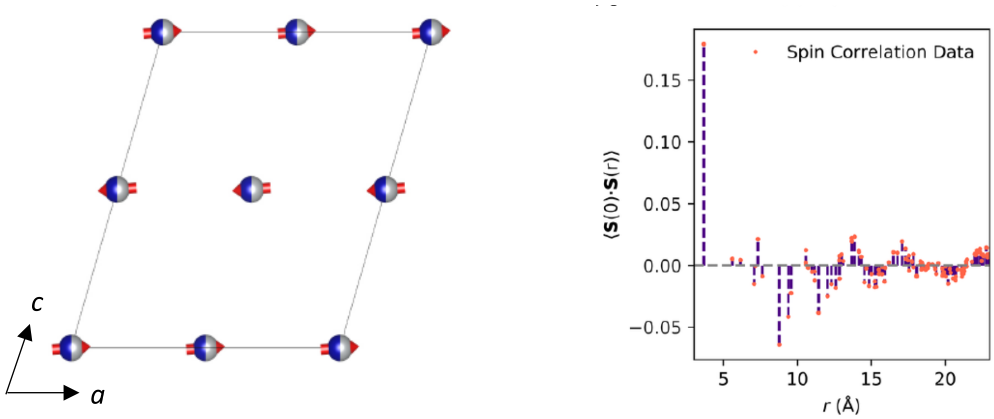

Figure 2. Left: Ordered magnetic structure of $Co_{0.5}Ni_{0.5}(NCS)_2$; right: spin-spin correlation function values for increasing atomic distances at 30 K.

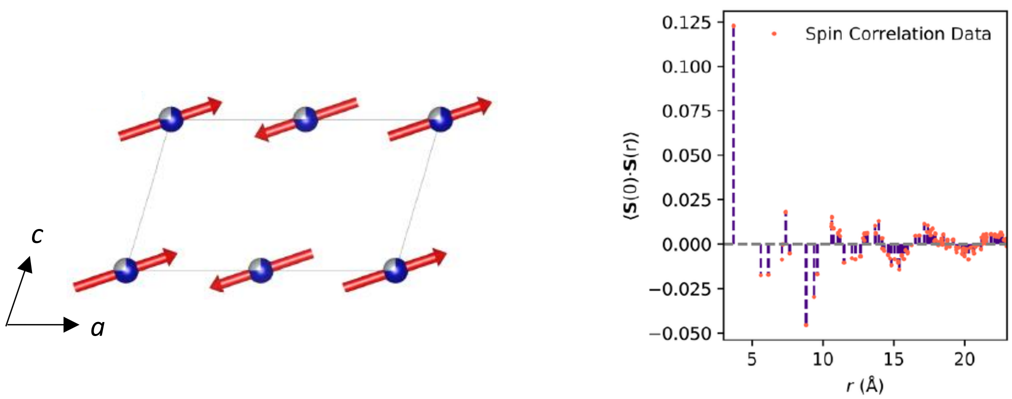

Figure 3. Left: Ordered magnetic structure of $Co_{0.75}Ni_{0.25}(NCS)_2$; right: spin-spin correlation function values for increasing atomic distances at 30 K.