Experimental Report

Proposal: **5-41-700** **Council:** 4/2012

Title: Magnetic Phase Transitions in Geometrically Frustrated SrGd2O4

This proposal is a new proposal

Researh Area: Physics

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Samples: SrGd2O4

Instrument	Req. Days	All. Days	From	To
D9	13	12	25/10/2012	06/11/2012

Abstract:

We propose to measure the development of low-temperature magnetism in SrGd2O4 using the hot neutron diffractometer D9 to minimise the absorption due to the Gadolinium ions. SrGd2O4 belongs to a family of compounds with the formula SrLn2O4 (Ln = Lanthanide). The triangular chains of Ln ions along the c-axis make the SrLn2O4 compounds suitable candidates to exhibit geometric frustration. From heat capacity measurements we have established that SrGd2O4 undergoes 2 low temperature phase transitions, TN1 = 2.72 K and then TN2 = 0.47 K. We have also seen the higher temperature (TN1) transition in single crystal susceptibility measurements, and observed a further transition upon the application of a magnetic field suggesting a rich H-T phase diagram for this material. The proposed D9 experiment would allow us to determine the nature of the magnetic phases in zero-field with possible future extensions to in-field measurements.

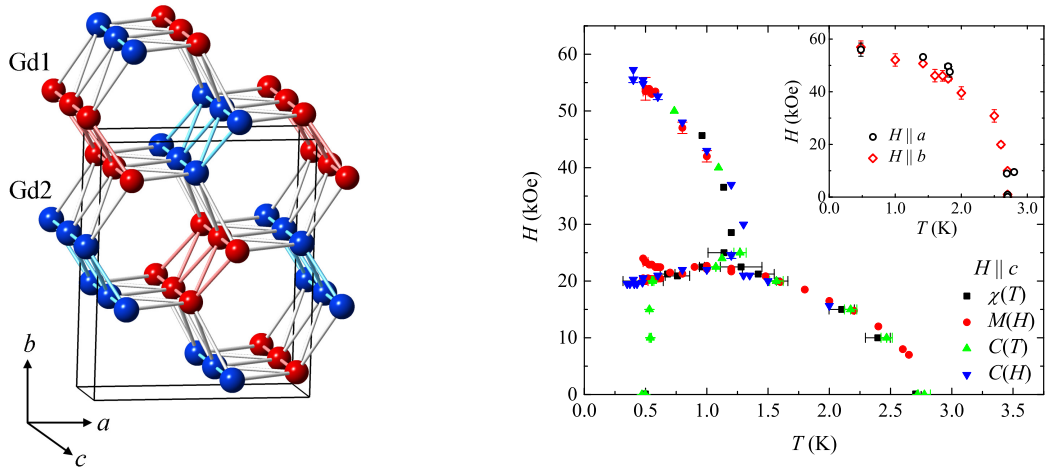

Fig. 1: (Left) Magnetic sublattice of $SrGd_2O_4$, with the two crystallographically inequivalent positions of the Gd^{3+} ions shown in different colours. When viewed along the c axis, honeycombs of the Gd^{3+} ions are visible. Zigzag ladders running along the c axis connect the honeycomb layers and give rise to geometric frustration. (Right) Magnetic H - T phase diagram for $SrGd_2O_4$ with the field applied along the c axis constructed from the susceptibility, magnetization and specific heat curves [2]. Inset: Field dependence of the upper critical temperature, T_{N1}, as observed in magnetization $M(H)$ and $M(T)$ measurements for $H \parallel a$ and $H \parallel b$.

$SrGd_2O_4$ is a geometrically frustrated magnet because the Gd^{3+} ions are arranged in a network of triangles and hexagons, and are coupled with antiferromagnetic exchange interactions. The crystal structure, space group *Pnam*, allows for the presence of two inequivalent sites of the rare earth ions in a unit cell [1], shown in Fig. 1. Magnetic susceptibility and heat capacity measurements on $SrGd_2O_4$ indicate that this compound undergoes two magnetic transitions, T_{N1} = 2.73 K and T_{N2} = 0.48 K, in zero-field [2]. The magnetic phase diagram [2] constructed from all the bulk property measurements on $SrGd_2O_4$ is shown in Fig. 1. The data illustrate that there is a large difference between the magnetic behaviour of $SrGd_2O_4$ and that of the other $SrLn_2O_4$ compounds [2–4], even though the positions of the magnetic ions and the strength of the exchange interactions are similar. This is not totally unexpected since spin-orbit coupling and crystal field anisotropies play a much more important role in establishing the low-temperature properties of other members of the $SrLn_2O_4$ series.

For this zero-field D9 experiment we used the four-circle He-flow cryostat, and the data were collected between $1.9 - 5.0$ K. The single crystal samples of $SrGd_2O_4$ were made using naturally abundant Gd, and a (short) 0.5121 Å wavelength was used to minimise absorption. The crystals were found to be of good quality, and we were able to follow the temperature dependence of several magnetic reflections that appear below $T_{\rm N1}$, with some examples shown in Fig. 2. We have found that *no* magnetic scattering appears between the nuclear peaks along three orthogonal directions in reciprocal space below 2.76 K, and no diffuse scattering is seen at temperatures far away from $T_{\rm N1}$ (in contrast to what has been observed for other $SrLn_2O_4$ compounds [4]).

Given that the magnetic order appearing below $T_{\rm N1}$ seems to be a **k** = 0 type structure, we have proceeded with the measurement of integer (hkl) reflections and collected two datasets at 5 and 1.9 K. Rietveld refinement using the higher temperature data has allowed us to confirm that the nuclear structure is identical to that previously reported from x-ray measurements [2]. FULLPROF refinement using the data collected at 1.9 K supports the initial conclusions that $SrGd_2O_4$ orders into a **k** = 0 magnetic structure below 2.76 K, however, the absorption corrections remain a very serious problem when trying to get an optimal fit to the data. Preliminary

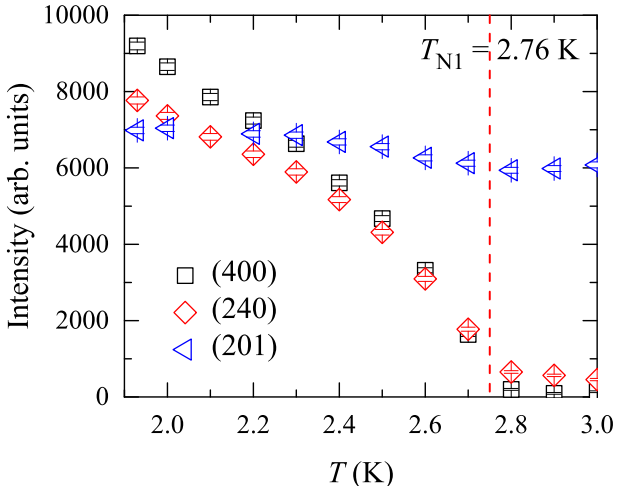

Fig. 2: Temperature dependence of the intensity of the (400), (240) and (201) peaks from single crystal $SrGd_2O_4$. The data shown are the raw intensity, where no absorption corrections have been applied. It appears that (400) is a purely magnetic reflection, whereas (240) has a small nuclear intensity above 2.8 K. For (201) the magnetic intensity sits on top of a large nuclear peak.

results suggest that only one of the Gd^{3+} sites carries a substantial magnetic moment of $4.5\mu_B$, while the other Gd^{3+} site remains disordered. We are in the process of further analysing the data collected at 1.9 K using different methods of taking the absorption into the account, however, in order to finalise the conclusions additional measurements on more favourably shaped samples might be required.

The dilution refrigerator part of the experiment, which was originally scheduled for 5 days, **did not take place** due to numerous technical problems with the instrument. The main mechanical problems (mainly related to the motor controllers and their links to the computers) were eventually diagnosed and rectified, but it took about three days to do this. By that time the procedure of cooling down to sub 1 K temperatures by using a dilution cryostat was deemed unreasonable, given that there will be no time left to explore a significant portion of reciprocal space. Thus it remains to be seen whether the two transitions observed in $SrGd_2O_4$ are due to the two Gd^{3+} sites ordering independently, or whether the lower temperature transition is effectively a rearrangement or modification of the **k** = 0 phase found at higher temperatures.

[1] H. Karunadasa, Q. Huang, B.G. Ueland, J.W. Lynn, P. Schiffer, K.A. Regan and R.J. Cava, Phys. Rev. B **71** 144414 (2005).
[2] O. Young, G. Balakrishnan, M.R. Lees and O.A. Petrenko, Phys. Rev. B **90** 094421 (2014).
[3] T.J. Hayes, O. Young, G. Balakrishnan and O.A. Petrenko, J. Phys. Soc. Japan **84** 024708 (2012).
[4] O.A. Petrenko, Low Temp. Phys. **40** 106 (2014).