Experimental report

16/11/2016

Proposal: 5-31-2480 **Council:** 4/2016

Title: Magnetic structure of geometrically frustrated SrGd2O4

Research area: Physics

This proposal is a new proposal

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Samples: SrGd2O4

Instrument	Requested days	Allocated days	From	To
D20	3	4	29/08/2016	02/09/2016
			15/09/2016	16/09/2016
D2B	3	0		

Abstract:

We propose to investigate the low temperature magnetic structure of the geometrically frustrated SrGd2O4 compound by powder neutron diffraction using a sample with non-absorbing Gd isotope. SrGd2O4 belongs to the SrLn2O4 (Ln = Lanthanide) crystal family. In this compound, magnetic ions are linked through a network of triangles and hexagons suitable for the establishment of magnetic frustration. Heat capacity measurements have revealed the presence of two magnetic transitions at TN1 = 2.72K and TN2 =0.47K. In field susceptibility measurement have also show the field sensibility as well as the anisotropy of the magnetic structure of this compound. Due to the spin only magnetic moment of Gd, SrGd2O4 gives a unique opportunity to study magnetism in the SrLn2O4 family member, whose properties are determined mostly by the exchange interactions, rather then by the single ion physics.

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D20 PND magnetic structure investigation of $SrLn_2O_4$, Ln=Gd and Nd.
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From this measurement, we report that $SrNd_2O_4$ stabilizes a long range ordered magnetic structure on one of the two Nd^{3+} sites in the vicinity of 2.2 K. $SrGd_2O_4$ stabilizes a magnetic structure on both Gd^{3+} sites at 2.73 K and undergoes a second magnetic transition at 0.57 K.

The PND experiment has been divided into two parts in order to study both expected transitions for the two compounds using a standard cryostat and a dilution insert. For the low temperatures measurement, a Cu sample container was used in order to maximize the sample thermalisation. The transitions occurring at higher temperatures were then studied within a vanadium container in high resolution mode. The $SrGd_2O_4$ powder contains the Gd^{160} isotope (98%), reducing significantly the absorption of the sample. Both materials were studied with a 2.41 Å wavelength.

$SrGd_2O_4$: The following diagram presents the main results of the measurement performed on the $SrGd_2O_4$ polycrystalline sample, see fig.1. The magnetic intensities (10 K background subtracted) measured at 100 mK, 1.6 K and 4 K i.e. below, between and above the two transition temperatures are shown. At 4 K, the result displays a clear magnetic diffuse scattering revealing the appearance of short ranged magnetic ordering in the system. Then at 1.6 K, well defined magnetic peaks have emerged indicating the stabilization of a long-ranged magnetic order (transition occurring at 2.73 K). The presence of both diffuse and localized magnetic scattering at 1.6 K reveals that both long and short-ranged magnetic phases coexist at this temperature. At 100 mK, the diffuse scattering reported at higher temperature is negligible and extra magnetic peaks have appeared (e.g. 2θ equal 24 and 38°) revealing the establishment of a second long-ranged magnetic order. A precise temperature study of the evolution of the magnetic scattering was also performed revealing clearly the two magnetic transition occurring in the system at 2.73 and 0.57 K.

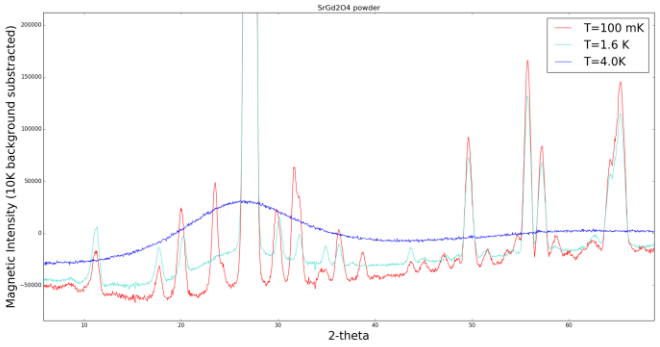

FIGURE 1: TEMPERATURE EVOLUTION OF THE MAGNETIC SCATTERING OF $SrGd_2O_4$.

Magnetic structure determination: From a Fullprof treatment of the magnetic data collected at 1.6 K (in both Cu and V sample container), we have retrieved the magnetic structure of $SrGd_2O_4$ stabilized at T_{N1}. Impurity reflections present at 22, 29 and 32 degrees have been disregarded and the model fits perfectly the data (Chi square: 2.419), see Fig. 2a. The magnetic structure of $SrGd_2O_4$ (stabilized at 2.73 K) is commensurate with the lattice and presents a (k=0) propagation vector, see Fig.4. On both Gd sites, the magnetic structure displays ferromagnetic order along the chains i.e. along the b-axis. The magnetic

moments are pointing also along b. The chains are then coupled antiferromagnetically, with the closest neighboring chain of the same Gd type forming magnetic triangular ladders. The chains are in addition coupled antiferromagnetically with the nearest chains of the other Gd type. Finally, the magnitude of the ordered magnetic moments was calculated to be different on the two sites with values of 2.1 and 3.0 μ_B (it is not possible to attribute a value to a site in particular since both configurations are accepted by our theoretical model).

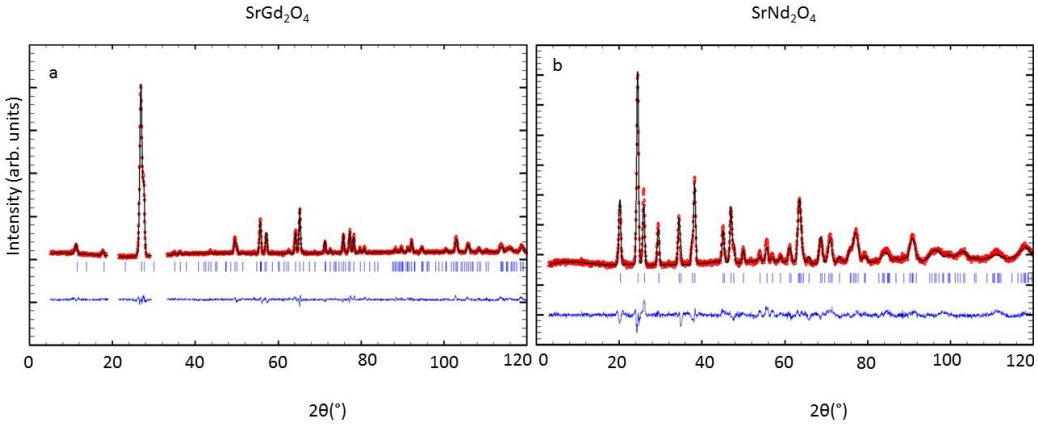

Figure 2: Fullprof treatment of the magnetic intensity measured on, a) $SrGd_2O_4$ (impurity reflections are disregarded to perform this treatment) and on b) $SrNd_2O_4$ at 1.6 K. Data and fit are represented in black and red, respectively.

$SrNd_2O_4$: The diagram presented in Fig. 3 shows the evolution of the magnetic scattering of $SrNd_2O_4$ within the temperature range of 10 to 1.6 K. The appearance of a weak diffuse scattering located around 2θ=20° at 5 K is a clear indication that magnetic correlations present in the system start to express themselves. We then report a significant increase of this diffuse scattering while cooling down to 2.5 K suggesting that the establishment of a short ranged magnetic order is progressing in the system. At 1.9 K, well defined magnetic Bragg peaks have replaced the previous diffuse scattering revealing the presence of a long-ranged magnetic order. The data collected at 1.6 K look fairly similar to the previous 1.9 K data despite a slight increase of the Bragg peaks magnitude. Thus we expect that the magnetic structure stabilized at 1.9 K remains stable at 1.6 K. Data collected using the dilution insert have confirmed that no other magnetic transitions do occur in the system down to the lowest temperatures.

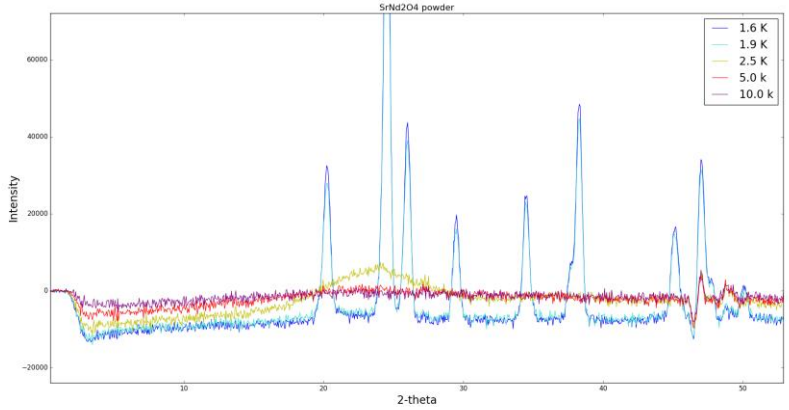

FIGURE 3: TEMPERATURE EVOLUTION OF THE MAGNETIC SCATTERING OF $SrNd_2O_4$

Magnetic structure determination: From our PND data, the magnetic structure of $SrNd_2O_4$ has been determined with the Fullprof program, see Fig. 2b (fit) and Fig. 4. Only the magnetic ions sitting on one of the two Nd sites contribute to the establishment of the magnetic structure, (it is not possible to determine if the magnetic order is present either on site 1 or on site 2). The magnetic structure is commensurate with the lattice and presents a (k=0,0.5,0.5) propagation vector. The magnetic order consists of antiferromagnetic chains running along the crystallographic b-axis. Neighboring chains present the same motif but shifted by b/2. Magnetic moments are pointing within the ac-plane. This structure is then coupled antiferromagnetically along c with the nearest ladder of the same type.

Discussion:

$SrGd_2O_4$: So far only the magnetic structure stabilized at 2.73 K has been solved however from the data it seems clear that the system undergoes a second magnetic transition at 0.57 K. The magnetic structure stabilized at 2.73 K shows magnetic long range order established on both Gd sites making $SrGd_2O_4$ a special case in the $SrLn_2O_4$ family (only $SrYb_2O_4$ was previously reported to present long range order on both sites). Also, the antiferromagnetic coupling between first neighbor ladders reveals the existence of magnetic interactions between the two different Ln sites, this feature is exclusive to $SrGd_2O_4$. The analysis of the magnetic structure stabilized below 0.57 K is currently in progress.

$SrNd_2O_4$: This compound is the only known member of the $SrLn_2O_4$ family showing long-range magnetic order with a non-zero propagation vector and antiferromagnetic ordering along the 1-D chains (along the leg of the ladders). In fact, Nd^{3+} is bearing a magnetic moment significantly smaller than Er^{3+}, Ho^{3+}, Dy^{3+}, Yb^{3+}, Gd^{3+} and it seems in this particular case that the local anisotropy of the magnetic sites plays a leading role in the establishment of the magnetic structure. In addition, the antiferromagnetic ladder coupling along the c-axis seems to reveal the presence of extended interaction between neighboring ladders.

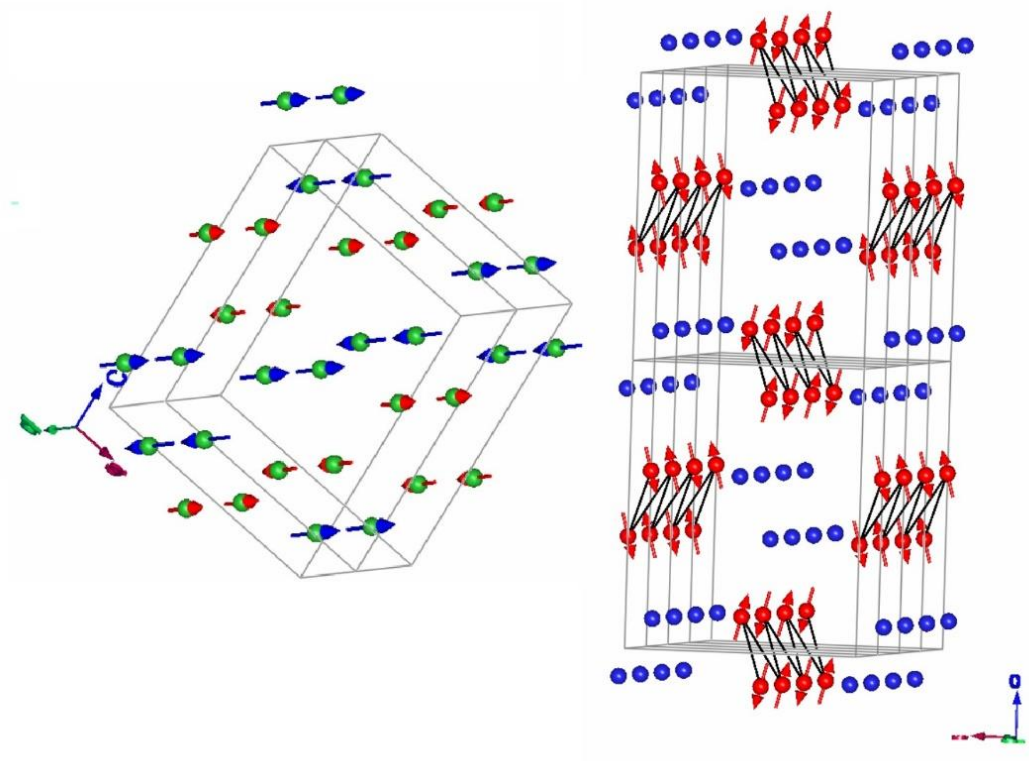

Figure 4: Left, magnetic structure of $SrGd_2O_4$ stabilized at 2.73 K. Right, magnetic structure of $SrNd_2O_4$ stabilized at approximatively 2.2 K. The crystallographic axes a, b and c are represented in red, green and blue respectively.