Experimental Report

15/01/2014

Proposal: **5-32-778** **Council:** 10/2012

Title: Investigation of the nature of magnetic short-range ordering in the bilayered compounds Ca2.5-xLaxSr0.5GaMn2O8 (x = 0, 0.05, and 0.1)

This proposal is a new proposal

Researh Area: Physics

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Local Contact: NILSEN Goran
RITTER Clemens

Samples: brownmillerite type bilayered compounds Ca2.5-xLaxSr0.5GaMn2O8 (x = 0, 0.05, and 0.1)
Ca2.5-xLaxSr0.5GaMn2O8 (x = 0, 0.05, and 0.1)

Instrument	Req. Days	All. Days	From	To
D7	7	7	29/04/2013	03/05/2013
			25/07/2013	29/07/2013

Abstract:

The naturally occurring layered quasi-2D compound Ca2.5Sr0.5GaMn2O8 with mixed-valence of Mn3+ and Mn4+ (Mn3+: Mn4+:: 1:1) has attracted a considerable attention from the low-dimensional magnetic ordering point of view. We propose to investigate the nature and dimensionality of the short-range magnetic ordering both above and below the Neel temperature (over 140-170 K) in the electron doped naturally occurring brownmillerite type bilayered compounds,Ca2.5-xLaxSr0.5GaMn2O8 (x = 0.05, and 0.1). The evolution of the short-range correlation with increasing electron doping (x = 0, 0.05, and 0.1) will also be brought out. The diffuse scattering instrument D7 will be employed in its polarization analysis (elastic) spin-flip mode to filter out the weak magnetic diffuse scattering from the total scattering. The evolution of the short-range magnetic correlation length across the magnetic phase transition will be revealed. A magnetic phase diagram in the x-T plane will be drawn. The expected outcome will have significant impact to understand the physics of low dimensional magnetism.

Title of the Proposal: Investigation of the nature of magnetic short-range ordering in the bilayered compounds $Ca_{2.5-x}La_xSr_{0.5}GaMn_2O_8$ (x = 0, 0.05, and 0.1)

Experiment No. 5-32-778, Instrument: D7

Proposers: YUSUF Seikh Mohammad, BERA Anup Kumar, Local contacts: NILSEN Goran, RITTER Clemens

Magnetic ordering in a reduced dimension (D < 3), and its crossover to a higher dimension are of current interest and one of the fundamental issues in condensed matter physics. They can be mostly due to strongly anisotropic exchange interactions, or to the strongly frustrating topology of systems. The crossover of magnetic ordering can be driven by several external and internal parameters such as temperature, magnetic field, and pressure, substitutions, inhomogeneities, *etc*.

The naturally occurring layered compound $Ca_{2.5}Sr_{0.5}GaMn_2O_8$ with mixed-valence of Mn^{3+} and Mn^{4+} (Mn^{3+}: Mn^{4+}:: 1:1) has attracted a considerable attention from its low-dimensional magnetic ordering. The temperature dependent neutron diffraction study on $Ca_{2.5}Sr_{0.5}GaMn_2O_8$ confirms a dimensionality crossover of the magnetic ordering from two-dimensional (2D) to three-dimensional (3D) [1]. With lowering of temperature, a short-range 3D antiferromagnetic (AFM) ordering above 170 K, a 2D long-range AFM ordering over 160-170 K, and then a 3D long-range AFM ordering below T_N ~ 150 K [Fig. 1]. We have shown recently that the nature of short-range magnetic correlations above the T_N changes drastically with La-substitution with a possibility of dimensionality cross-over from 2D to 3D [2]. It was also shown that the magnetic properties of this type of mixed-valence layered compounds can be tuned, by electron doping in means of La^{3+} substitution at the Ca^{2+} site, to achieve new functional materials for possible spintronics applications [3].

For $Ca_{2.5-x}La_xSr_{0.5}GaMn_2O_8$ (x = 0, 0.05, and 0.1) compounds, a 3D long-range collinear antiferromagnetic (AFM) structure, as like the parent compound, was found below the Neel temperature T_N of the respective compound. With increasing electron doping, a decrease in T_N was also observed (T_N = 160, 150, and 142.5 K for compounds with x =0, 0.05 and 0.1, respectively) [2]. Well above T_N, 3D short-range AFM ordering was observed for all compounds. However, in the intermediate temperature range just above T_N, a strong effect of electron doping (La substitution) on the magnetic correlations has been observed [2]. In this temperature range an asymmetric Warren type peak profile, corresponding to the 2D magnetic correlations, was found for the parent compound. For the highest La-substituted compound (x = 0.1), the peak profile of the diffuse scattering is symmetric Lorentzian type, corresponding to the 3D short-range magnetic correlations [Fig. 2]. Interestingly, for the intermediate compound with x = 0.05, the peak profile of the diffuse scattering is possibly a mixed Warren and Lorentzian type [Fig. 2]. However, the true nature of the peak profile could not be confirmed from the unpolarized neutron diffraction data due to some limitations of the data quality e.g., (i) the weak intensity of the diffused scatterings, (ii) the limited resolution of the instrument, (iii) the presence of nuclear peaks [(020), (100), and (110)] in the same Q-range where the diffuse scatterings appear, and (iv) presence of two close magnetic Bragg peaks ((100) and (001) at Q = 1.17 and 1.2 $Å^{-1}$, respectively). The coexistence of both diffused scattering and magnetic Bragg peaks has also been observed for all compounds over a broad temperature range below respective T_N. All diffuse scatterings have been found centered on Q = 1.18 $Å^{-1}$, where the most intensed magnetic Bragg peaks [(100) and (001)] appear below T_N.

With the aim to study the nature of the short-range magnetic ordering around T_N (both above and below), we have carried out polarized neutron diffraction measurements on x = 0, 0.05 and 0.1 compounds using D7 diffractometer. The diffraction patterns were recorded (i) at 150, 160, 165, and 175 K for x =0, (ii) at 140, 145, 148, 150, 152, 154, 156, 158, 168, 178, and 200 K for x =0.5, and (iii) at 125, 140, 145, 150, 160, 170, and 200 K for x =0.1 compound,

respectively. We have used an orange cryostat for low temperature measurements. The diffraction patterns were recorded by using an incident neutron beam with wavelength of λ = 4.8 Å with flipper ON and OFF states to get the pure magnetic scatterings.

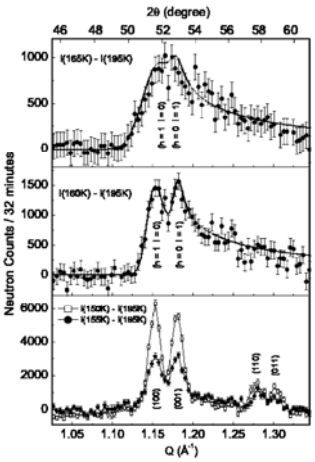

Fig. 1: The pure magnetic diffraction patterns for x =0 from G6.1 diffractometer [1].

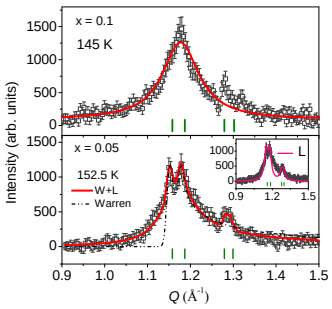

Fig. 2: The pure magnetic diffraction patterns for x = 0.05 and 0.1 compounds from G6.1 diffractometer [2].

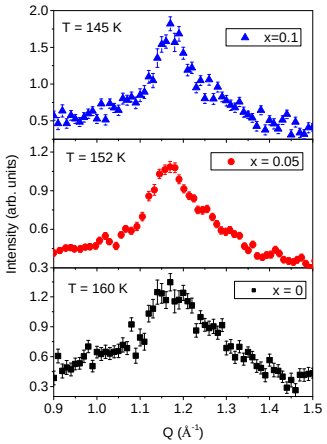

Fig. 3: The pure magnetic diffraction patterns [(magnetic spin flip + magnetic non spin flip)/2] from D7 diffractometer.

The pure magnetic scatterings at the interesting temperatures e.g., 160 K for x = 0, 152 K for x =0.05 and 145 K for x =0.1 compounds, respectively, are shown in Fig. 3. Only one broad magnetic peak has been found over the Q range from 0.9 Å^{-1} to 1.5 Å^{-1}. It is apparent from the peak profile that the Q-resolution of the D7 diffractometer is not sufficient enough to resolve the two magnetic peaks ((100) and (001) at Q = 1.17 and 1.2 Å^{-1}, respectively), hence, to study the nature of the magnetic ordering dimensionality (2D/3D/mixed 2D and 3D) from the direct fitting of the peak profiles by Warren function and/or Lorentzian function. In the case of a 3D ordering a symmetric Bragg peak occurs that can be represented by Lorentian function, (ii) in the case of 2D ordering, asymmetric peaks are observed that can be represented by Warren function, and (iii) in the case of 2D ordering with a short-range in the third dimension, it is expected to develop "corrugations" with increasing intensity in the vicinity of the incipient 3D Bragg peaks. The degree of asymmetry in the case of 2D depends on the 2D correlation length. Nevertheless, from the data from D7 we have separated out individual scattering contributions, such as magnetic non spin flip scattering, magnetic spin flip scattering, and magnetic average scattering [(magnetic spin flip + magnetic non spin flip)/2] which are useful for a Reversed Monte Carlo (RMC) analysis to derive spin-spin correlations. The RMC analyses on these data are on progress.

References:

1. S. M. Yusuf *et al.*, *Phys. Rev. B* **74**, 184409 (2006).
2. A. K. Bera *et al. J. Phys. Condens. Matter.* **23**, 426005 (2011).
3. A. K. Bera *et al.*, *J. Appl. Phys.* **107**, 013911 (2010).
4. H. Zhang *et al.*, *Phys. Rev. B* **45**, 10022 (1992).