Experimental Report

15/01/2014

Proposal: **5-42-355** **Council:** 10/2012

Title: Toroidal moment in the spin-glass system NixMn1-xTiO3 (x=0.42)

This proposal is a new proposal

Researh Area: Physics

Main proposer: **HOLBEIN Simon**

Experimental Team: HOLBEIN Simon
BAUM Max

Local Contact: STEFFENS Paul

Samples: NixMn1-xTiO3 (x=0.42)

Instrument	Req. Days	All. Days	From	To
IN14 CPA	6	7	07/06/2013	13/06/2013
			26/06/2013	01/07/2013
IN3	0	4		

Abstract:

The discovery of spin-induced ferroelectricity in frustrated transition-metal-oxides attracted much interest due to their exciting physical mechanism and due to their potential applications. In most materials studied the magnetoelectric properties arecoupled with the so-called spin chirality via the inverse Dzyaloshinskii-Moriya interaction. Another multi-spin variable that couples directly with magnetoelectric properties is the toroidal moment which is given by the cross product between positions of the magnetic ions and their spins. So far the coupling between the multi-spin variables vector chirality and toroidal moment is studied in systems that exhibit long-range magnetic structure. However, very recently Yamaguchi et al. showed that long-range order actually is not needed. They find an antisymmetric finite magnetoelectric effect in a material that does not exhibit a long-range magnetic ordering. In Ni0.42Mn0.58TiO3 only a spin-glass ordering is observed that nevertheless seems to induce a linear magnetoelectric effect. The magnetic ordering in this material is however little characterized and we therefore wish to study it with neutron scattering methods.

Toroidal moment in the spin-glass system $Ni_xMn_{1-x}TiO_3$ (x=0.42)

The discovery of spin-induced ferroelectricity in frustrated transition-metal-oxides attracted much interest due to their exciting physical mechanism and their potential applications [1]. So far the coupling between the multi-spin variables vector chirality and toroidal moment is studied in systems that exhibit long-range magnetic structure. However, in $Ni_{0.42}Mn_{0.58}TiO_3$ only a spin-glass ordering is observed that nevertheless seems to induce a linear magnetoelectric effect [2]. The magnetic ordering in this material is little characterized and we therefore wish to study it with neutron scattering methods.

The strategy of this experiment was twofold. First we want to pole the crystal using crossed magnetic and electric fields to investigate an evidence of the induced toroidal moment as proposed by reference [2]. Using spherical neutron polarimetry with the CryoPAD option of IN14 a field-induced reorientation of magnetic moments can be detected. In the second part we want to characterize the magnetic correlation lengths in zero field and to perform inelastic scans to search for a spin gap.

The experiment was carried out using a single crystal of $Ni_{0.42}Mn_{0.58}TiO_3$, which has been grown at Cologne University. They show an excellent crystal quality and the temperature dependence of the magnetization perfectly agrees with that reported in reference [2]. For the elastic part of the experiment we used a flat shaped crystal which was hold in between two aluminum plates in order to apply an homogenous electric field along the [110] direction. For the inelastic part a large crystal (30x6x6 mm^3) was mounted on an aluminum holder.

Throughout the experiment, we worked at k_i=1.55 $Å^{-1}$ and k_i=1.2 $Å^{-1}$, with CryoPAD option and Be-filter on k_f. The flipping ratio on a magnetic Bragg peak was almost 34 which corresponds to a polarization of the neutron beam of approx. 97 %. To pole the crystal in the first part of the experiment, an electric field of at least 500 V/mm was applied along the direction [110] upon cooling below the spin glass transition. For the application of a magnetic field, the cryostat was taken out of CryoPAD and mounted in an external electromagnet. Magnetic fields of 1 T were applied to the directions [120], [100] and [001].

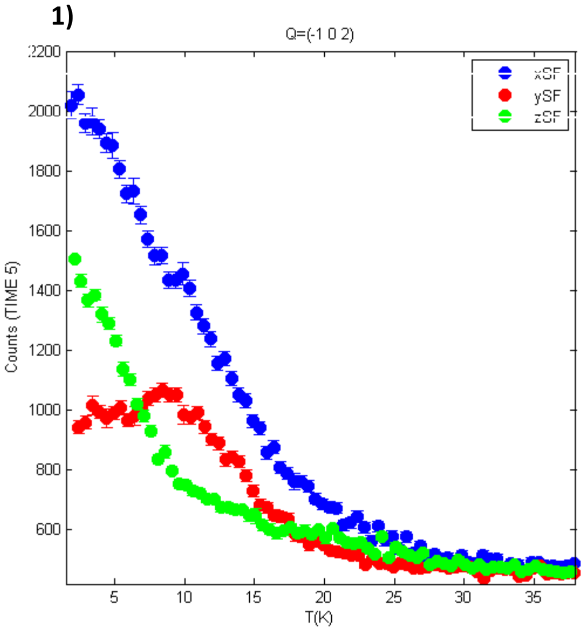
At low temperatures, we found diffuse magnetic signals at positions corresponding to both magnetic structures of the mother compounds MnTiO3 (AFM1) and NiTiO3 (AFM3) [3]. Polarisation analysis of the Bragg peak intensities supports this assumption: On AFM1-like positions the magnetic moment lies in the ab-plane whereas on AFM3-like positions the moment shows a component parallel to the z-direction. The magnetic reflections could be well described with a Lorentz function suggesting a cluster glass type ordering. In the spin-glass phase the frozen fluctuations of both magnetic structures, AFM1 and AFM3, coexist. Fig. 1 and Fig. 2 show the different temperature dependence of the spin flip intensities at the position of AFM1 and AMF3-like Bragg point.

In the following we tried to pole the crystal by cooling it below the transition temperature in the presents of crossed magnetic and electric fields. As observed in reference [2], such an application of fields along [110] and [001] direction may lead to the development of a finite toroidal moment. So far a systematic dependence of the off-diagonal terms of the polarization matrices on the application could not be found. However, the interpretation of the data is hampered as the application of a magnetic field to the sample induced a weak ferromagnetic moment to the sample. Further detailed study of the collected polarization matrices will be necessary to clarify the impact of the crossed fields.

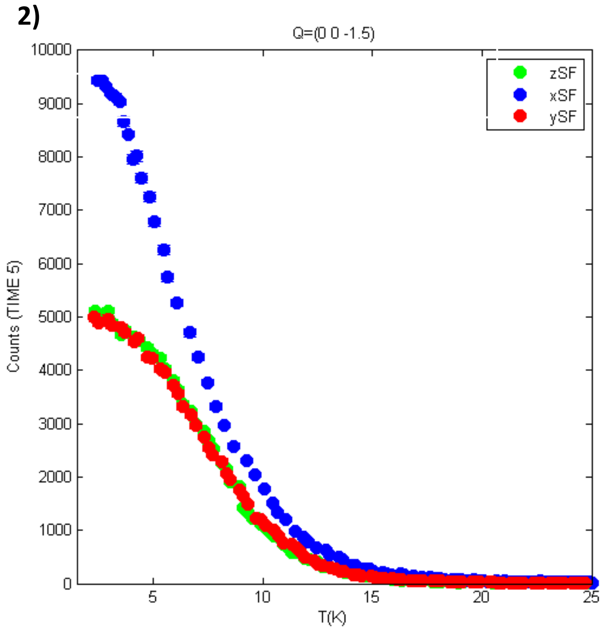
In the second part of the experiment we had a look at the excitation spectrum of this complex magnetic system. Using a large single crystal in the orientation 100/001, we were able to achieve high statistics even with the mounted spherical polarization analysis (Fig. 3). We found a rather broad spectrum at the magnetic zone center of the superposed AFM1 peak at Q=(-1 0 2). The polarization analysis verifies the origin of the inelastic signal to be purely magnetic, and by full xyz polarization analysis it appears isotropic. Perpendicular to the honeycomb plane, along the QL direction, we found a similar spectrum at several positions indicating the coupling of the planes to be rather weak.

[1] T. Kimura et al., Nature **426**, 55 (2003), **[2]** Y. Yamaguchi et al., Phys. Rev. Lett. **108**, 057203 (2012), **[3]** G. Shirane et al., J. of Phys. Soc. Japan 14, 1352 (1959)

1)



2)



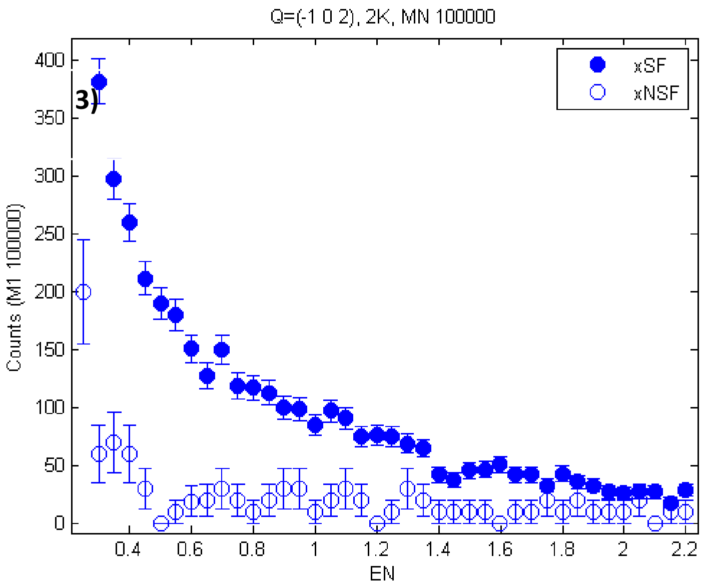


Fig. 1) Temperature dependence of the diagonal spin flip intensities at the AFM1-like reflection (-1 0 2).

Fig. 2) Temperature dependence of the diagonal spin flip intensities at the AFM3-like reflection (0 0 -1.5).

Fig. 3) Energy scan at AFM1 like reflection (-1 0 2) with polarisation analysis.