Experimental Report

15/01/2014

Proposal: **4-01-1300** **Council:** 10/2012

Title: Neutron Spin Echo investigation ofspin fluctuations in multiferroics

This proposal is continuation of: 4-02-422

Research Area: Physics

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Samples: MnWO4

Instrument	Req. Days	All. Days	From	To
IN11	12	7	02/07/2013	09/07/2013

Abstract:

We used recently high resolution Neutron Spin Echo (NSE) spectroscopy to investigate the critical dynamics at the antiferromagnetic transition and in the SDW phase down to the cycloidal transition. These experiments showed a noticeable broadening of the (0,q,1) magnetic Bragg peak around Tc and the diffuse scattering emerging out of the elastic Bragg peak is quasielastic at all temperatures above the cycloidal transition. In view of these results it becomes urgent to investigate, whether these puzzling features are special to TbMnO3 or intrinsic to mutliferroics. We therefore propose to examine the slow dynamics of MnWO4, which is another multiferroic oxide presenting a similar sequence of magnetic and ferroelectric transitions as TbMnO3.

Neutron Spin Echo investigation ofspin fluctuations in multiferroics

There is huge interest in the recently discovered families of multiferroic materials, in which the magnetic ordering directly causes ferroelectric polarization with strong coupling between the two phenomena. Multiferroics are promising in view of applications but the magnetoelectric coupling is also an interesting fundamental problem. The chiral magnetism in these multiferroics appears in two steps: upon cooling there is first a collinear order which transforms into a chiral order in a second transition which is accompanied with spontaneous ferroelectric polarization. One of the open issues concerns the dynamics of the multiferroic transition and within the multiferroic phase, in particular the dielectric response, which is coupled with the long-range and diffuse magnetic order.

At previous experiments (4-02-422 and 4-02-400) we used high resolution Neutron Spin Echo (NSE) spectroscopy to investigate the critical dynamics at the antiferromagnetic transition of $TbMnO_3$ and in the SDW phase down to the cycloidal transition. These experiments showed a noticeable broadening in energy of the (0,q,1) magnetic Bragg peak around T_C. The diffuse scattering emerging out of the elastic Bragg peak is quasielastic at all temperatures above the cycloidal transition. In addition triple-axis studies find a chiral diffuse signal persisting well above the long-range chiral phase.

In view of these results we performed a similar experiment on $MnWO_4$ [1-4]. This system is considered to be a prototypical multiferroic its electric polarization can be controlled by a magnetic field and its magnetic chirality can be controlled by an electric field [4]. At room temperature $MnWO_4$ crystallizes in the wolframite structure and can be described in the monoclinic space group P2/c. Manganese and tungsten ions are surrounded by edge-sharing O_6 octahedra, each forming a zigzag chain along the crystallographic c-direction. Chains of manganese and tungsten are arranged alternately in planes parallel to the a-axis. Upon cooling $MnWO_4$ undergoes three magnetic phase transitions in zero magnetic field seen by neutron diffraction. Below T_{AF3}~13.5K, an incommensurate (IC) SDW-structure with propagation vector $\mathbf{q}_{ic}$=(-0.214, 0.5, 0.457) is found, which upon further cooling transforms at T_{AF2}~12.7K into a chiral ordering with the same modulation vector. At T_{AF1}~7.6K a commensurate and collinear magnetic order is finally stabilized with propagation vector $\mathbf{q}_c$=(-0.25, 0.5, 0.5) [1,2]. In perfect accordance with the Dzyaloshinski Moriya scenario the ferroelectric polarization develops at the transition into the incommensurate non-collinear AF2 phase [3]; this transition is thus very similar to that in $TbMnO_3$ rendering $MnWO_4$ an ideal candidate to study the critical dynamics.
The IN11 experiment was done in paramagnetic NSE mode around $\mathbf{q}_{ic}$=(-0.214, 0.5, 0.457) the propagation vector of the SDW-structure. The NSE spectra were normalized by XYZ-polarisation analysis, where $\hat{z}$ was the vertical, $\hat{x}$ parallel to $\vec{Q}$ and $\hat{y}$ complementing the Cartesian set. As seen in Fig. 1a, the (spin flip) intensity at the position of the magnetic Bragg peak increases at T_{AF3} and an additional increase is found at T_{AF2}. However the two intensities shown, $I_{\hat{z}}^{sf}$ and $I_{\hat{x}}^{sf}$ for the neutron beam polarization either along the vertical $\hat{z}$ or along $\hat{x}$ are different and the increase at T_{AF2} is better seen on $I_{\hat{z}}^{sf}$. This effect is also illustrated by Fig. 1b, where the well defined minimum of the ratio $I_{\hat{z}}^{sf}/I_{\hat{x}}^{sf}$ indicates that the magnetic moments are along $\hat{z}$ between T_{AF2} and T_{AF3}, thus confirming previous results with Cryopad.
Fig. 2 shows the NSE spectra at the Bragg position, and the slowing down of the fluctuations at the onset of the magnetic Bragg peak at T_{AF3}. The resulting relaxation times are the blue points of fig.3. Below T_{AF3}, however, significant diffuse scattering persists around the Bragg peak, whose intensity gradually decreases. Due to the limited beam time we were not able to follow this temperature dependence accurately, but it seems that this diffuse scattering finally disappears at T_{AF2}. The surprising founding, which has strong similarities with the previous $TbMnO_3$ results, is that, besides the Bragg peak the diffuse scattering is unaffected by the magnetic transition at T_{AF3} whereas it clearly bears the signature of a q-dependent slowing down at T_{AF2}. Due to the limited

beam time we were not able to follow this feature down to T_{AF1} and also around the commensurate propagation vector $\mathbf{q}_c$=(-0.25, 0.5, 0.5).

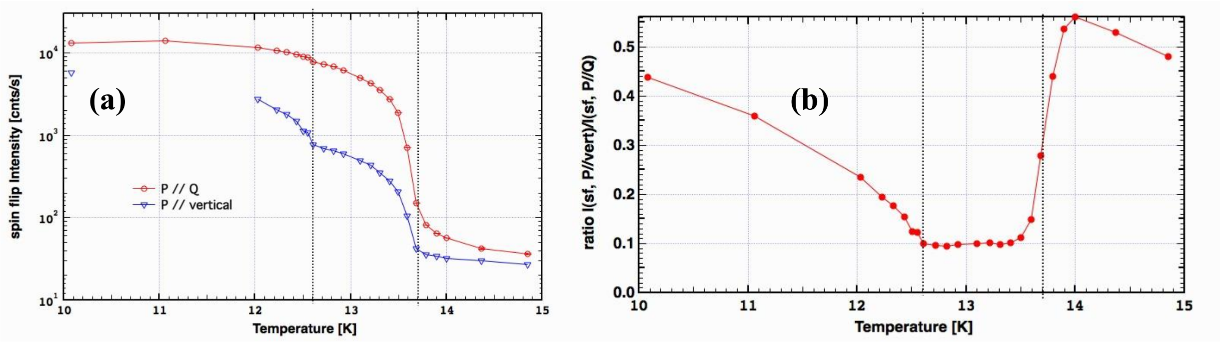

Fig. 1: (a) Normalised spin flip as function of temperature for the neutron beam polarization either along the vertical $\hat{z}$ or along $\vec{Q}$ ($\hat{x}$ direction of the xyz polarization analysis setup)
(b) ratio of the two intensities shown in (a) revealing thee preferred orientation along the vertical of the magnetic moments between the two transition temperatures T_{AF3} and T_{AF2} indicated by the dotted lines at both figures.

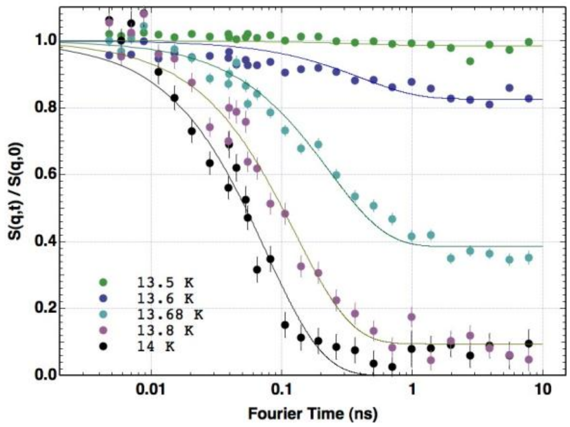

Fig. 2: NSE spectra at $\mathbf{q}_{ic}$= (-0.214, 0.5, 0.457). The spectra become elastic at the onset of the Bragg peak at T_{AF3}.

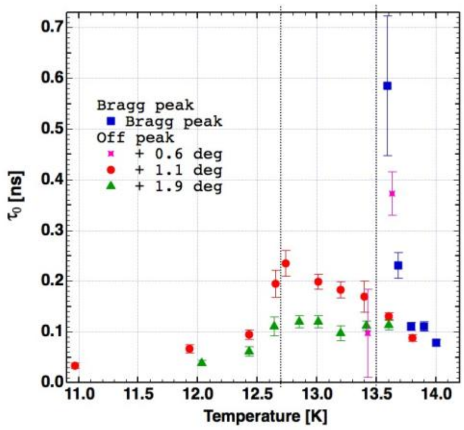

Fig. 3: Characteristic relaxation times obtained by the NSE echo spectra at the Bragg position and after sample rotation by 0.6, 1.1 and 1.9 deg.

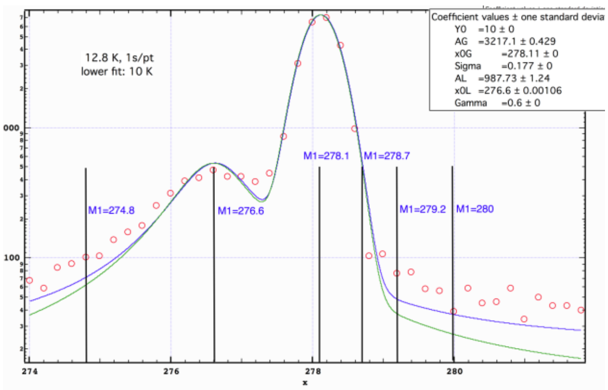

Fig. 4: Rocking scan of the magnetic Bragg peak indicating the positions of the NSE experiments.

[1] G. Lautenschläger et al., Phys. Rev. B. **48**, 6087 (1993) **[2]** H. Ehrenberg et al., J. Phys.: Cond. Matter **9**, 3189 (1997) **[3]** O. Heyer et al., J. Phys.: Cond. Matter **18**, L471 (2006)**,** A. Arkenbout et al., Phys. Rev. B **74**, 184431 (2006)**,** K. Taniguchi et al., Phys. Rev. Lett. **97**, 97203 (2006) **[4]** T. Finger et al., Phys. Rev. B. **81**, 54430 (2010).