Experimental report

07/01/2022

Proposal: **CRG-2757**

Council: 4/2020

Title: Time-resolved study of the multiferroic domain inversion in Ni3V2O8

Research area: Physics

This proposal is a new proposal

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Samples: Ni3V2O8
NaFeGe2O6

Instrument	Requested days	Allocated days	From	To
IN12	5	4	25/02/2021	01/03/2021

Abstract:

Ni3V2O8 exhibits a sequence of magnetic phase transitions, including a multiferroic phase. Our time-resolved neutron scattering experiment revealed an Arrhenius like relaxation behaviour of multiferroic domains in Ni3V2O8, when switching the polarity of an applied electric-field. It was possible to follow these relaxation times over ≈4 decades in time as a function of temperature at fixed electric field. In order to study also the electric-field amplitude dependence of relaxation times in Ni3V2O8, and in order to analyse lower tempeatures we propose to continue the experiments on IN12.

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The Kagomé-staircase system $Ni_3V_2O_8$ exhibits a sequence of magnetic phase transitions, including a multiferroic phase. Our previous time-resolved neutron scattering experiment on IN12 (CRG-2290) revealed a simple Arrhenius like relaxation behaviour of multiferroic domains as a function of temperature. Due to finite beamtime and the time-consuming handling of the time-resolved high-voltage setup, it was not possible at that time to also record the electric field dependence of multiferroic domain inversion. Meanwhile, our setup and the experimental procedure has been improved, which allows the investigation of temperature and electric-field dependent relaxation behaviour in much shorter time. Respective time-resolved experiments on $TbMnO_3$, $NaFeGe_2O_6$, $(NH_4)_2[FeCl_5(H_2O)]$, CuO (all conducted at IN12) revealed a simple combined Arrhenius-Merz law that describes the multiferroic domain inversion as a function of temperature and also electric field [1,2, reports 5-42-499 and CRG-2573]. To verify this combined law to be also valid in multiferroic $Ni_3V_2O_8$, we proposed to supplement the previous time-resolved and temperature-dependent data sets of relaxation times in $Ni_3V_2O_8$ by electric-field dependent measurements on IN12.

The orthorhombic system (*Cmca*) undergoes a sequence of magnetic phase transitions including a multiferroic one at low temperatures [3-9]. First, at T=9.1K an incommensurate (k_{inc}=(0.27 0 0)) spin-density wave (SDW) with moments pointing along a direction evolves. Subsequently, the magnetic arrangement transforms into a spiral structure at T_{MF}=6.3K, when simultaneously a ferroelectric polarization develops. The ferroelectric polarization is directed along b direction and its sign is coupled to the chiral spin structure via the inverse Dzyaloshinskii-Morya interaction (DMI), which enforces the controllability of the spin-spiral handedness by external electric fields. Below 4K, the structure becomes commensurate and no multiferroic behaviour can be observed in this phase.

To study the multiferroic domain inversion, longitudinal polarization analysis was utilized as the spin-flip channels $I_{x\bar{x}} = -i(M_\perp \times M_\perp^*)$ and $I_{\bar{x}x} = i(M_\perp \times M_\perp^*)$ along x-direction (the conventional coordinate system with Q||x was chosen) sense the chirality of the magnetic structure. The so-called chiral ratio $r_\chi = -i(M_\perp \times M_\perp^*)/|M_\perp|^2$ thus describes the domain population of a particular handedness. It must be noted that the maximum values $r_\chi \pm 1$ can only be achieved for ideal scattering geometries, which enforce a scattering vector perpendicular to the rotation plane of spins. Further, also elliptical distortions reduce the chiral ratio. To provide an almost ideal sample setup for the time-resolved investigation of the multiferroic domain inversion in $Ni_3V_2O_8$, the sample was mounted within the scattering plane (1 0 0)/(0 1 1) to access the magnetic reflection (1.27 1 1). The sample was furthermore cramped between aluminium plates, which were tightened together by insulating Polytetrafluoroethylene (PTFE) screws. These plates were connected to our time-resolved high-voltage setup, which supplied +/-4kV and allows for field inversions within several μs.

The main goal of the allocated beamtime was to record as many switching curves as possible over a broad temperature and electric field range. Figure 1 a) and b) display exemplary two recorded switching curves together with a fit of the relaxation process (red line). It can be seen that the relaxation processes in respectively both directions are symmetric, which entails that the system does not exhibit a preference for a particular domain type.

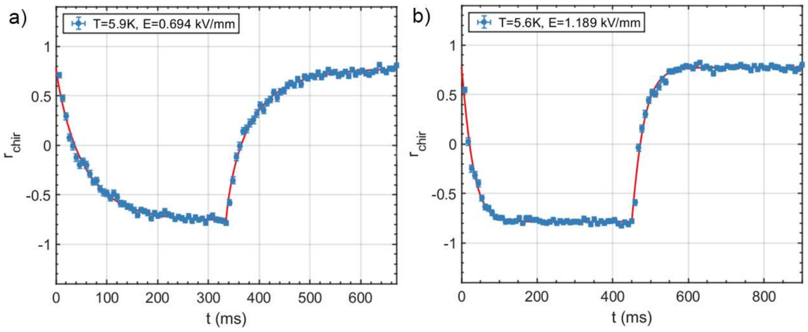

Figure 1: Two exemplary switching curves for different fields and temperatures. The blue points refer to the measured data and the red solid line belongs to the fit.

Figure 2a) displays all fitted relaxation times as a function of inverse temperature. Solid lines in this figure refer to a fit that follows the combined Arrhenius-Merz law with $\tau = \tau^* e^{\frac{A_0}{k_B E}\left(\frac{T_{\mathrm{MF}}-T}{T T_{\mathrm{MF}}}\right)}$. The parameter τ^* is the critical relaxation time, which describes the fastest possible domain inversion at the multiferroic transition temperature T_{MF}, and it was already speculated that this value is limited by the spin-wave velocity [1,2]. The second parameter A_0 is an activation constant and represents the pinning strength of multiferroic domains in the system. It is obvious from figure 2a) that these two parameters in the framework of the combined Arrhenius-Merz law are adequate to describe the temperature and electric-field dependence of multiferroic domain inversion. This is furthermore manifested by the appropriate scaling of the relaxation time with $1/E(1/T-1/T_{MF})$. (see Fig 2b)). It must be noted that for the sake of simplicity only one of both relaxation times is shown in Fig 2b). Nevertheless, the result holds also for the relaxation process in opposite switching direction. Unfortunately, the sample broke during the course of the conducted experiment, wherefore a thorough investigation of domain kinetics at even lower temperature and for a broader field range was prohibited. To exploit the remaining beamtime, we mounted the multiferroic system $NaFeGe_2O_6$ for the same kind of measurements. For this system and even for the same sample a comprehensive set of temperature and electric field dependent switching curves was already recorded on IN12 during a previous beamtime (CRG-2439). However, we utilized the last days of this current beamtime to concentrate on time-resolved measurements of multiferroic domain inversion at the lowest accessible temperature (T=1.5K).

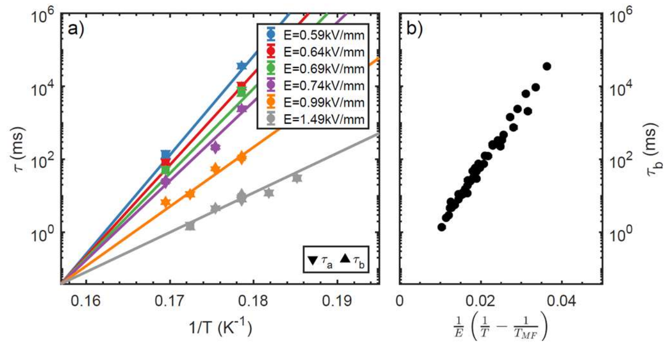

Figure 2: The left panels display the observed relaxation times, which are fitted by the combined Arrhenius-Merz-law. The right panels show the scaling of relaxation times with $1/E(1/T-1/T_{MF})$.

So far, the relaxation behaviour of multiferroic domains was seen to follow only a thermally activated and classical domain wall motion description. In contrast, for ferroelectric and magnetic domains, it was reported that also domain wall tunnelling can become relevant for domain inversion, when quantum fluctuations start to dominate at low temperatures [10-12]. The setup resembles the configuration from the previous experiment (see report CRG-2439).

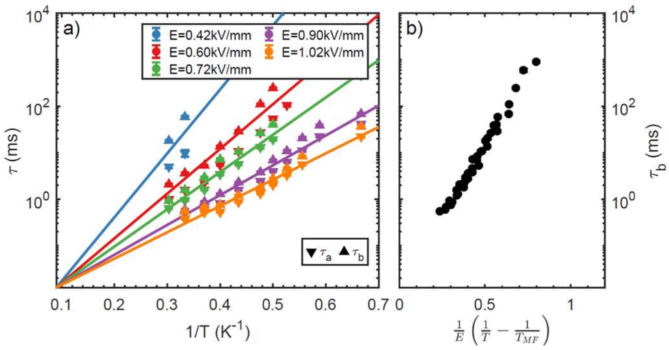

Figure 3: Both panels show the Arrhenius-Merz fit as well as the scaling behavior for $NaFeGe_2O_6$

Figure 3 show the recorded relaxation times (including the Arrhenius-Merz fit) and the respective scaling behaviour. It is obvious that also at the lowest temperature the multiferroic relaxation follows the combined Arrhenius-Merz law and no indication of quantum fluctuation driven domain wall tunnelling is observable. Nevertheless, quantum fluctuation can become relevant at even lower temperatures. This motivates further time-resolved experiments on multiferroic domain inversion, whereby respective investigations would necessitate a specialized sample environment e.g. He-3 refrigerators with appropriate and tested high-voltage adaptions.

We would like to acknowledge the allocation of this beamtime and especially we would like to thank Karin Schmalzl for the excellent on-site support. The low-temperature measurements on $NaFeGe_2O_6$ are published as an editor's suggestion in Physical Review B [2].

[1] J. Stein *et al.*, Phys. Rev. Lett. **127**, 097601 (2021), [2] S. Biesenkamp *et al.*, Phys. Rev. B **104**, 174405 (2021) [3] G. Lawes *et al.*, Phys. Rev. Lett. 93, 247201 (2004) [4] N.Qureshi *et al.*, Phys. Rev. B 88, 174412 (2013) [5] G. Lawes *et al.*, Phys. Rev. Lett. 95, 087205 (2005) [6] A. B. Harris *et al.*, Phys Rev. B 73, 184433 (2006) [7] A.B. Harris *et al.*, J. Phys.: Condens. Matter 20, 434202 (2008) [8] G. Lawes *et al.*, J. Phys.: Condens. Matter 20, 434205 (2008) [9] T. Yildrim *et al.*, J. Phys.: Condens. Matter 20, 434214 (2008) [10] J. Brooke *et al.*, Nature 413, 610 (2001) [11] O. G. Shpyrko *et al.*, Nature 447, 68 (2007) [12] F. Kagawa *et al.*, The Journal of Chemical Physics 7, 10675 (2016)