Experimental report

24/05/2021

Proposal:	**5-42-486**	**Council:**	10/2018
Title:	Microscopic origin of the electricfield induced magnetization reversal in hexaferrites		
Research area:	Physics		

This proposal is a new proposal

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Experimental team:	Lingjia SHEN Emma CAMPILLO
Local contacts:	Robert CUBITT

Samples: Ba2-xSrxMg2Fe12O22 (x = 1.6)

Instrument	Requested days	Allocated days	From	To
D33	4	4	07/09/2019	11/09/2019
D22	4	0		

Abstract:

Magnetization switching by electric field holds great potential in realizing low energy dissipation magnetic random access memories and standby-power-free integrated circuits that rely on nonvolatile information encoded in the direction of magnetization. This effect, which has been realized in metals, semiconductors and multiferroic insulators, is interesting not only because of its technological importance, but also because it allows us to uncover novel properties of matter that are otherwise inaccessibl. This proposal aims to unveil the microscopic origin of the electric field induced magnetization sign switch (reversal) effect in hexaferrites.

Experimental Report

Instrument: D33

Experiment Number: 5-42-486

Title: Microscopic origin of the electric field induced magnetization reversal in hexaferrites

Experiment Date: 07/09/2019 – 11/09/2019

Experimental Team: Lingjia Shen, Emma Campillo, Elizabeth Blackburn (Lund University)

Local Contact: Robert Cubitt

Introduction

Magnetization switching by electric field holds great potential in realizing low energy dissipation magnetic random access memories and standby-power-free integrated circuits that rely on non-volatile information encoded in the direction of magnetization. This effect, which has been realized in metals, semiconductors and multiferroic insulators, is interesting not only because of its technological importance, but also because it allows us to uncover novel properties of matter that are otherwise inaccessible.

Magnetoelectric (ME) multiferroics are materials that combine coupled electric and magnetic dipoles. In some ferrites with hexagonal crystal structures, termed hexaferrites, it has been found that application of a magnetic / electric field can induce a change in the ferroelectric / magnetic properties [1-2], corresponding respectively the ME and converse ME effects. This cross control lays the foundation for fabricating multiferroics-based devices. However, the microscopic mechanism that governs the converse ME effects is still in debate. For example, electric field control of bulk magnetization has been realized in $Ba_{2-x}Sr_xMg_2Fe_{12}O_{22}$ (BSMFO, x = 1.6) recently (Fig. 1) [2]. This effect can be explained by a change in the spin helicity [3] or magnetic domain dynamics [4]. The experiment below has been performed to test these two scenarios. Specifically, the spin helicity can be obtained by imposing polarization analysis [5], while the magnetic domain dynamics can be measured by traditional small-angle neutron scattering (SANS) at very small momentum transfers [6].

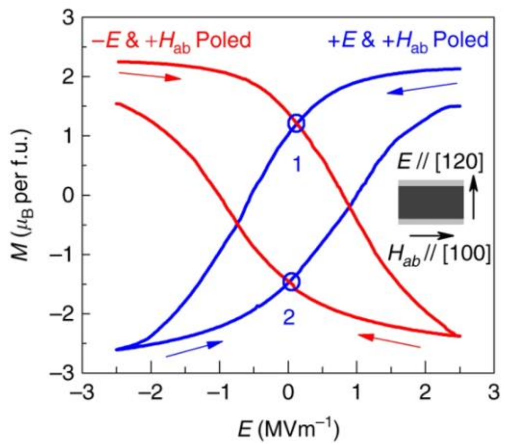

Figure 1: Electric field control of magnetization in BSFMO (x = 1.6) at 10 K [2]. The magnetic / electric field poling protocol is discussed in the Results section.

Sample Details and Instrumental Configurations

A single crystalline BSMFO (x = 1.6) with dimensions around 3*3*1 mm^3 was used; it was synthesized by Kun Zhai [2]. This material is reported to be hexagonal, with lattice parameters $a = b = 5.841$ Å and $c = 43.256$ Å at room temperature. The experiment was performed on the SANS instrument D33. The sample was mounted on the ILL electric field sample stick, which allows the application of an electric voltage up to 10 kV. It was mounted first on an aluminium rod using GE varnish, and then this was installed between ground plate and the high voltage plate, without direct contact. The sample was aligned so that the electric field was applied vertically, the magnetic field horizontal and parallel to the beam, and the [001] crystal axis was horizontal and perpendicular to the incident beam (Figure 2a). The sample stick was loaded in the Blue Charly cryomagnet for the final measurements. We also used the spin flipper to change the incoming neutron polarization. Two instrumental configurations were used. The low-Q configuration (0.005 Å^{-1} < Q < 0.029 Å^{-1}) was with the detector distance / collimation / wavelength values = 12.8 m /12.8 m / 5 Å; the high-Q configuration (0.025 Å^{-1} < Q < 0.29 Å^{-1}) was with the detector distance / collimation / wavelength values = 2.8 m /2.8 m / 5 Å.

Results

The electric field control of bulk magnetization in BSMFO (x = 1.6) has been realized using the following field poling protocols in sequence [2].

- The sample was first cooled to 10 K. A magnetic field of 5 T was then applied along the (100) direction. At 5 T, an electric field of ±2.5 MV / m was applied along the (120) direction (Fig. 1). The magnetic field was switched off. The magnetization was measured as a function of electric field with ±2.5 MV / m as the starting point.

Magnetic reflections

In Ref. 2, wide angle neutron diffraction on this compound is reported. At 1.5 K, an incommensurate longitudinal [IC-L, $\mathbf{k}_1$ = (0, 0, 0.75)] conical structure was found in zero-field. Between 0.0 T and 0.2 T, the magnetic structure is transformed into a two-fold transverse [TF-T, $\mathbf{k}_2$ = (0, 0, 1.5)] cone. After switching the magnetic field off from 0.5 T, the system remains in the TF-T conical state. Therefore, the converse ME effect in Fig. 1 has been claimed to originate from the TF-T conical spin state. We have studied the magnetic reflections in the small-angle regime.

We were able to observe magnetic reflections indexed by $\mathbf{k}_1$ and $\mathbf{k}_2$ (Fig. 2b-c); the $\mathbf{k}_2$ reflection survives after the magnetic field is removed [2]. However, we find that the $\mathbf{k}_2$ reflection is not sensitive to the incoming neutron polarization. Moreover, we find that the system undergoes another magnetic transition, which has not been reported in literature, around 4 T with $\mathbf{k}_3$ = (0, 0, 1.8) (Fig. 2d). The magnetic reflection modulated by the emergent $\mathbf{k}_3$ vector, on the other hand, is polarization sensitive. Since the converse ME effect was realized after the 5 T field poling, we believe that its underlying mechanism is related to this emergent structure.

We also explored the temperature dependences of these reflections at a selected number of magnetic fields. We observe a clear change in Bragg angle at a temperature around 150 K. We have started a limited investigation of this by X-ray diffraction, and our preliminary results indicate that there may be a small monoclinic distortion to the crystallographic structure.

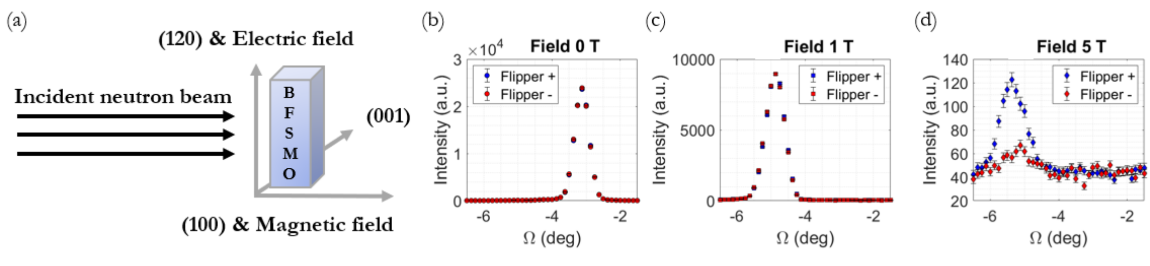

Figure 2: (a) Experimental geometry. (b-d) Rocking curves on the first magnetic Bragg peak of the IC-L cone, TF-T cone and emergent magnetic state with different incident neutron polarizations at 5 K.

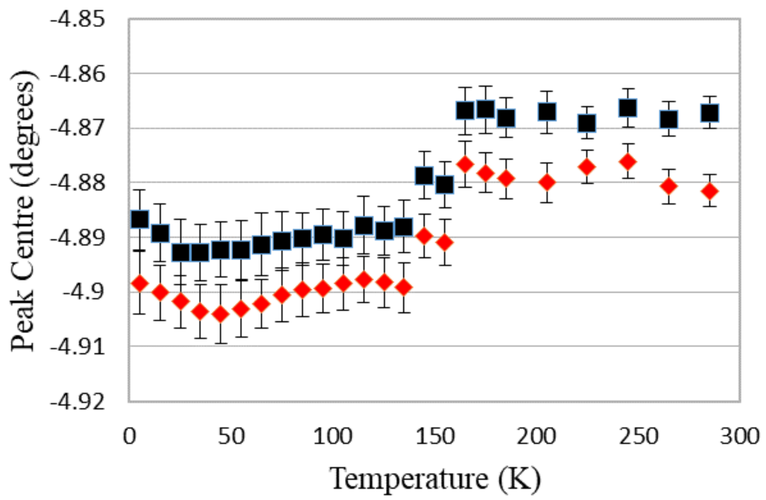

Figure 3: The temperature dependence of the peak centre of the $\mathbf{k}_1$ reflection. The black points were measured with the incoming neutrons polarized spin up, and the red points were measured with the incoming neutrons polarized spin down. The origin of the discrepancy in the observed values for the two spin configuration remains to be understood. These measurements were done after zero field cooling, and then applying 1 T on warming.

Flipping ratio measurements

We carried out half-polarized measurements, varying the polarization of the incident neutrons. It was not possible to analyse the polarisation of the scattered neutrons, as the Bragg angles of the $\mathbf{k}_2$ and $\mathbf{k}_3$ reflections exceeded the angular acceptance of the ^{3}He cell, and so this was not attempted. We investigated the electric field dependence of the $\mathbf{k}_2$ and $\mathbf{k}_3$ reflections using the field poling protocol described above. We were not able to establish a systematic change in either of the flipper$^+$ and flipper$^-$ channels, for two reasons. First of all, the $\mathbf{k}_3$ reflection is much weaker than the $\mathbf{k}_2$ reflection, and they are close in reciprocal space (Fig. 2c-d). Since the $\mathbf{k}_2$ reflection survives after the magnetic field is removed, it is very difficult to establish whether the $\mathbf{k}_3$ reflection also does so. Secondly, we encountered a leakage issue in the desired electric field range.

Low-Q small angle scattering

The analysis on the low-Q dataset, which can test the magnetic domain dynamics scenario, is still underway.

<u>Summary</u>

Our experimental observation does not support the conclusion made in Ref. 2, which suggested that the TF-T state is responsible for the converse ME effect in Fig. 1. We are planning to study this further, and are currently considering ways to deal with the leakage issue with electric field control.

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[2] K. Zhai et al., Nat. Commun. 8, 519 (2017)
[3] Y. Yamasaki et al., Phys. Rev. Lett. 98, 147204 (2007)
[4] V. Kocsis et al., Nat. Commun. 10, 1247 (2019)
[5] Y. Yamasaki et al., Phys. Rev. Lett. 98, 147204 (2007)
[6] A. Michels, J. Phys.: Condens. Matter 26, 383201 (2014)