## **Experimental report**

Proposal:	<b>5-54-171 Council:</b> 4/2014					
Title:	Magnetoelectric coupling in ferromagnetic/ferroelectric heterostructures probed by polarised neutron reflectomet					
Research are	a: Physics					
This proposal is	s a new proposal					
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Experimenta	l team: Manisha BISH	Г				
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Local contac	ts: Thomas SAER	BECK				
Samples: Fe	/BaTiO3					
Instrument		Requested days	Allocated days	From	То	
D17		5	5	05/11/2014	10/11/2014	

Abstract:

Multiferroics are materials with more than one ferroic order parameter, such as ferromagnetism and ferroelectricity. The magnetoelectric (ME) coupling between these ferroic order parameters makes these materials potential candidates for electronic devices. In composite multiferroics, ferromagnetic/ferroelectric interfaces play a crucial role in mediating the ME coupling. The aim of this work is to study the magnetic depth profile of Fe (ferromagnetic) thin films on BaTiO3 (BTO, ferroelectric) substrates under the influence of an electric field. The use of polarized neutron reflectometry will provide us the opportunity to obtain information on the in-plane depth-distribution of the Fe magnetization as a function of the applied electric field and to find the effective ME coupling distance away from the interface. We will also study the effect of an electric field on the magnetization reversal mechanism of the Fe/BTO system using magnetic field scans.

# Magnetoelectric coupling in ferromagnetic/ferroelectric heterostructures by polarised neutron reflectometry

### Abstract

Multiferroics are materials with two or more ferroic order parameters, such as ferromagnetism and ferroelectricity. The magnetoelectric (ME) coupling between these ferroic orders makes these materials a potential candidate for electronic devices. In composite multiferroics, ferromagnetic/ferroelectric interfaces play a crucial role in mediating the ME coupling [1]. The aim of this work is to study the magnetic spin structure of the Fe (ferromagnetic) thin films grown on  $BaTiO_3$  (ferroelectric) substrates under the influence of an electric field. The use of polarised neutrons provide information on the depth-distribution of the Fe magnetization as a function of the applied electric field. Thus allowing us to understand how the ME coupling is mediated and identifying the possibility of a magnetic dead layer forming at the interface.

#### Scientific background and aim

In ferromagnetic/ferroelectric (FM/FE) heterostructures, the ME coupling could be mediated by a number of mechanisms like strain mediated, charge driven and exchangebias coupling at the interface. [1, 2, 3, 4] It is clear that in such systems the coupling occurs via the interface, but so far there is very little knowledge about the structural or magnetic changes that occur *during* the application of an electric field. In our work, we study composite multiferroics consisting of Fe and FeCo thin films deposited on top of ferroelectric BaTiO<sub>3</sub> and LiNbO<sub>3</sub> single crystals (i.e., substrates). The macroscopic magnetic properties as a function of electric field were studied using longitudinal magnetooptical Kerr effect (MOKE), evidencing ME coupling in Fe/BaTiO<sub>3</sub> and FeCo/LiNbO<sub>3</sub> system. The maximum change in coercivity ( $\Delta H_c$ ) was observed to be ~ 86 % in Fe/BaTiO<sub>3</sub> heterostructures (see figure 1a) and ~ 57 % in FeCo/LiNbO<sub>3</sub> heterostructures (see figure 1b) on application of small electric fields, while higher fields result in small variations only. This suggests that the ME coupling deteriorates at high fields. We have already studied the effect of electric field on the Fe/BaTiO<sub>3</sub> interface using the isotope sensitive techniques of nuclear resonant scattering (NRS) of synchrotron radiation and Mössbauer spectroscopy. From these experiments, we could identify the structural, chemical and magnetic state of the interface upon growth of the Fe film on BaTiO<sub>3</sub> and unravel the evolution of the interface with the applied electric field. [5] In a recent NRS experiment, we have also identified the threshold electric field value for Fe/BaTiO<sub>3</sub> systems (200 kV/m) above which the ME coupling deteriorates. [6]

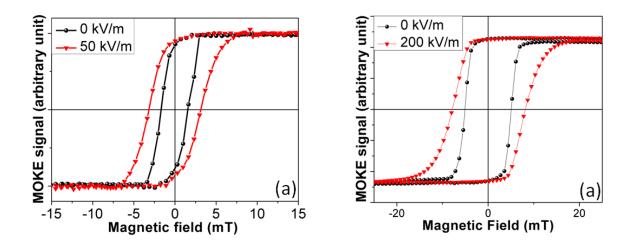


Fig. Hysteresis loops before and after applying electric field of (a) 50 kV/m on 5 nm Fe/BaTiO<sub>3</sub>(001) and (b) 200 kV/m on 5 nm Fe/LiNbO<sub>3</sub>(001)..

The primary goal of this experiment is to understand how the ME coupling is mediated in Fe/BaTiO<sub>3</sub> system by studying the effect of electric field (below the threshold value, where ME coupling is prominent) on the magnitude and orientation of the magnetic moments of the Fe film. We used polarised neutron reflectometry to study the electric field induced changes in the magnetization profile of the Fe film and resolve them as function of depth to identify the effective distance of ME coupling away from the interface. PNR is a suitable technique for our study, since it is depth-sensitive so allows to resolve the magnetization along the sample depth and is compatible with the in-situ application of electric and magnetic fields. PNR experiment was performed at the D17 diffractometer at ILL on samples with the following structure  $10 \text{ nm Al}/20 \text{ nm Fe}/\text{BaTiO}_3(001)$ and 10 nm Al/20 nm FeCo/LiNbO<sub>3</sub>(001). The samples were placed on a home-made holder designed for applying electric fields across the heterostructure. Polarized neutron reflectivity scans were measured first on the virgin samples with zero magnetic and electric fields, at saturation magnetic fields of 100 mT and at remanence. Further PNR spectra were measured at different applied electric fields from 0 kV/m to 200 kV/m in steps of 50 kV/m.

### **Results:**

Spectra recorded at 200 kV/m for Fe/BaTiO<sub>3</sub>(001) were significantly different in comparison to the initial state spectrum (zero electric field), shown in figure 2a. Spectra recorded on Fe/LiNbO<sub>3</sub>(0001) at 100 kV/m also showed slight differences in comparison to the initial state spectrum (shown in figure 2b).

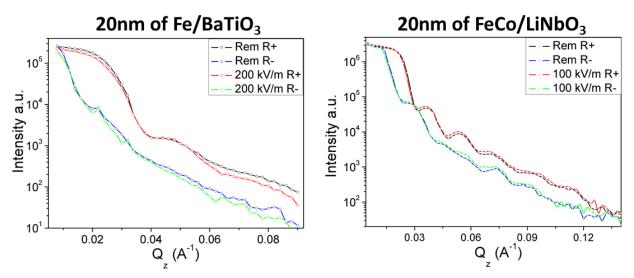


Figure 2 PNR spectra measured at remanence magnetic field after application of a magnetic field of 100 mT (Rem R+ and Rem R- for up and down neutron polarization respectively) on (a) 20 nm Fe/BaTiO<sub>3</sub> before and after application of 200 kV/m and on (b) 20 nm FeCo/LiNbO<sub>3</sub> before and after application of 100 kV/m.

However, it was observed that the Fe/BaTiO<sub>3</sub> sample had cracks on the surface after the experiment, possibly due to the temperature induced phase transition. BaTiO<sub>3</sub> is tetragonal above 285 K and it becomes orthorhombic at lower temperatures. Therefore, it is possible that the observed difference in the PNR spectra have not only contributions from the electric field induced changes in the magnetization of the heterostructure but also due to the developed cracks in the BaTiO<sub>3</sub> crystal. In order to study the influence of electric field on the magnetisation in our multiferroic heterostructures, further investigations are required. However, for this purpose we would need better experimental conditions during measurements like improved sample temperature control to avoid the structural phase transition of BaTiO<sub>3</sub> substrates and larger sample area for faster data acquisition. Use of the time of flight mode instead of the monochromatic mode would enable measurements at a single static angle providing more flexibility for setting the required sample environment, especially temperature control and electric field across the sample. The proposal will be resubmitted to continue the experiment.

#### **References:**

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