



	Experiment title: Electric field induced interface oxidation in ferromagnetic/ferroelectric composites	Experiment number: MA-2023
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Report:

In a previous experiment (HE-3470), we studied the electric field driven evolution of the local magnetic state of the Fe/BaTiO₃ (BTO) (metal/ferroelectric oxide) interface using nuclear resonant scattering (NRS) at the ID22N beam line [1]. Based on the results of that experiment and Mössbauer spectroscopy done on similar samples in our lab, we presented a model for the metal/ferroelectric oxide heterostructure. During the growth of a metal on top of a FE oxide, electron transport occurs across the interface due to the work function difference between the metal and the oxide (with high dielectric constant), which leads to the formation of a built-in electric field (and an intermixing layer) at the interface. Depending on the direction of the built-in field, the direction of the externally applied electric field either promotes ion transport across the interface or opposes it until the external field over comes the built-in field (after applying an electric field above a critical value - ‘critical field’).

The aim of this experiment was to identify the values of ‘critical field’ for both Fe/BTO and Fe/LiNbO₃ (LNO) systems. All samples were prepared by molecular beam epitaxy and had the following layer sequence: 10 nm Al/4 nm ⁵⁶Fe/1 nm ⁵⁷Fe/LNO and 10 nm Al/4 nm ⁵⁶Fe/1 nm ⁵⁷Fe/BTO. LNO is particularly interesting for such a study, since it has a different work function for positively (6.2 eV) and negatively (4.6 eV) poled surfaces, pre-poling was expected to modify the interface oxidation behaviour (and hence modify the magnitude and sign of the ‘critical field’). We therefore also investigated the samples with positively and negatively poled LNO (Poling was done by applying an electric field of ± 20 MV/m on LNO single crystal substrates). The experiment was carried out using NRS with in-situ application

of an electric field at room temperature and atmospheric pressure at the ID18 beam line. The following samples were measured during the experiment : (1) Fe on virgin LNO, (2) Fe on positively poled LNO, (3) Fe on negatively poled LNO and, (4) Fe on virgin BTO. NRS time spectra were recorded for the as-grown samples and at each step after applying an electric field from 0 upto 1.2 MV/m in steps of 50 kV/m for the BTO sample and from 0 upto 1 MV/m in steps of 100 kV/m and further up to 10 MV/m in steps of 400 kV/m for LNO samples (convention- top Fe film as positive electrode and silver paint at the back of the ferroelectric substrate as negative electrode). The whole series of measurements was also repeated for the opposite polarity with a new set of samples. All measurements were taken at zero electric field. NRS time spectra for Fe/BTO samples are shown in fig. 1.

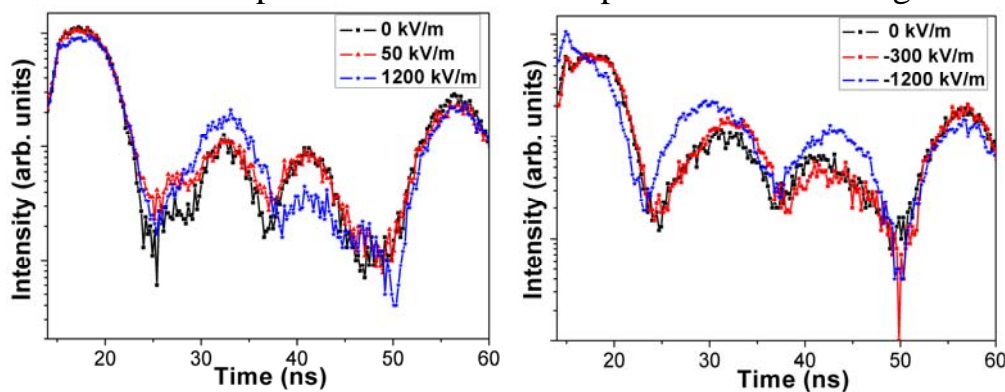


Fig 1. NRS time spectra for the sample with 10 nm Al/4 nm ^{56}Fe /1 nm ^{57}Fe /BTO (a) before and after applying a positive electric field (b) before and after applying a negative field.

Changes in the magnetic beat pattern were observed for the positive electric field at 50 kV/m but on applying a negative field, there was no change observed below -300 kV/m. Moreover, a positive field is observed to reduce the frequency of the magnetic beat pattern while a negative field increased the frequency (which is directly proportional to the hyperfine field of the magnetic site). In case of LNO samples, we could observe the effect of pre-poling on the magnitude of the ‘critical field’ at (and above) which the interface structure started evolving (for both positive and negative polarities). In case of positively poled LNO samples, the magnitude of the ‘critical field’ for positive electric field is much higher than that for the negative field (and vice versa for negatively poled LNO samples). These observations have been made by comparing the differences in the NRS time spectra and the data analysis is ongoing (in final phase). The complete analysis of the data will help us in identifying the values of the critical field for Fe/BTO and Fe/LNO systems and will provide us a detailed understanding of the ion transport mechanism across the interface as a function of an applied electric field and work function of the ferroelectric oxide. These results are expected to bring a better understanding of the metal/ferroelectric oxide interfaces in general and will be soon published in a followup paper.

References

1. S. Couet*, M. Bisht*, M. Trekels, M. Menghini, C. Petermann, M. J. Van Bael, J. P. Locquet, R. Rüffer, A. Vantomme and K. Temst, *Adv. Funct. Mater.* **24**, 71 (2014).