MAGNETOELECTRIC COUPLING IN $BiFeO_3$ SINGLE CRYSTALS AND THIN FILMS.

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$BiFeO_3$ is a multiferroic materials in which ferroelectric and anti-ferromagnetic orders coexist well above room temperature (T_N=643 K, T_C=1093 K), with a high polarization (over 100 µC/cm^2 [1]). We have shown at the LLB by neutron diffraction that these two order parameters interact and that the magnetization of the material can be modified by the application of an electric field. This opens the way towards the implementation of this material in spintronic devices in which magnetization could be controlled by a small electrical voltage rather than by currents or magnetic fields.

Although magnetoelectric materials have been known for decades, no *direct* proof of the coupling between the two co-existing physical effects had been reported in the literature. We have demonstrated this magneto-electric coupling by neutron diffraction on $BiFeO_3$ single crystals. One key to success was the exceptional quality of the high purity single crystals which are magnetically and electrically single domain.

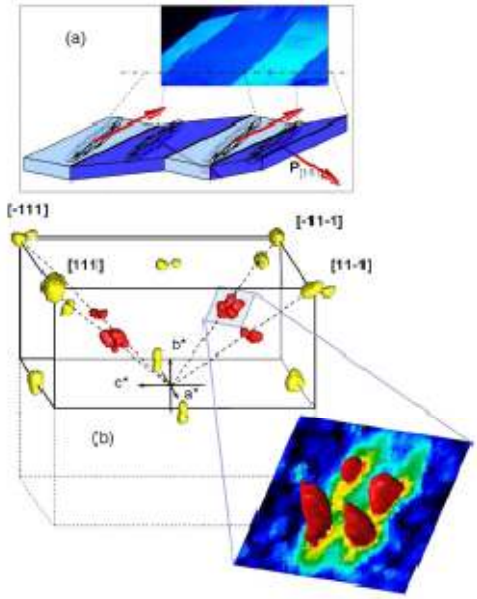

Fig. 1: Effect of an electric field on a BiFeO3 single crystal. (a) electric domains observed with polarized optical microscopy. (b) mapping of the neutron intensity in the reciprocal space. Two sets of splitting appear for the nuclear intensity (yellow spots): one because of the presence of two rhombohedral distortions along [111] and [1-11], the other because of the buckling of the crystal induced by twinning. The magnetic peaks (red spots) are further split because of the cycloids.

The first step of this work consisted in a verification of the magnetic structure of $BiFeO_3$ (several structures were consistent with powder diffraction data). Our single crystal results confirmed [2] that the spins of the Fe^{3+} ions lie in the (111) planes (G-type antiferromagnet) forming a circular cycloid with a long period of 62 nm.

In a second step, an electric field was applied on the crystal which was in a single ferroelectric domain state with the polarisation along [111]. The electric field was applied along the (001) and switched the sample into a multidomain ferroelectric state (Figure 1 (a)). Using neutron diffraction we showed that the propagation vector was the same as in the virgin state, but that the spins where then lying in two different planes. These can be accounted for by considering that 55% of the crystal volume has switched its polarization by 71°, and induced a tilt of the rotation plane of the Fe moments, thus inducing a spin flop of the antiferromagnetic sublattice (see Figure 2). These experiments evidence that although a macroscopic linear magnetoelectric effect is forbidden by symmetry, a rather strong coupling between M and P still exists at the atomic level via the Dzyaloshinskii-Moriya interaction.

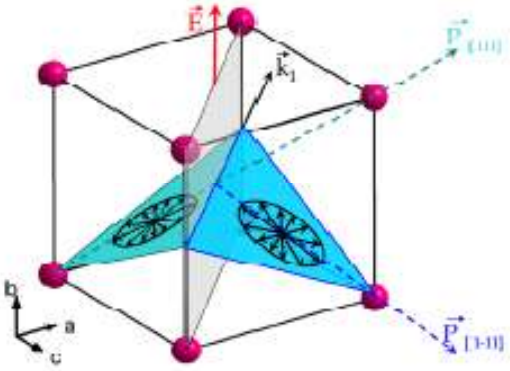

Fig. 2: Schematics of the planes of spin rotations and cycloids propagation vector for the two polarization domains separated by a domain wall (in light gray).

Integration of multiferroics into spintronics devices requires growing thin films. The group of Agnès Barthelemy at the UMR CNRS/Thales managed to grow high quality single phase $BiFeO_3$ epitaxial films by pulsed laser deposition. The first issue was to

determine if the magnetic structure of thin films. Neutron diffraction has shown that the cycloidal order is lost in thin films [3] (see Figure 3a). This change in the magnetic order affects the magneto-electric coupling since it allows a linear effect

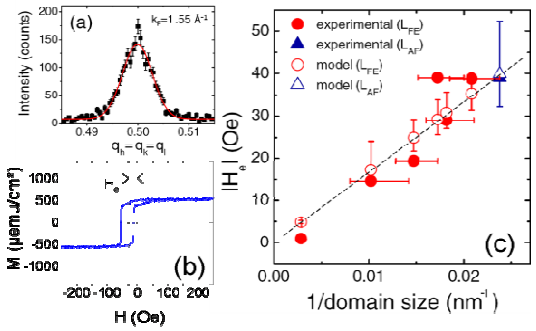

Fig. 3: (a) (1/2 1/2 1/2) magnetic Bragg peak observed with neutron diffraction on a 70 nm thick BiFeO3 (001) thin film. (b) Hysteresis loop of a CoFeB/BiFeO3(70nm) bilayer. The exchange field is noted He. (c) Exchange field as a function of the inverse of domain size. The data are in quantitative agrrement with the Malozemoff random field model.

The next step of the study has consisted in investigating the interaction of $BiFeO_3$ thin film coupled to a ferromagnetic layer (CoFeB in our case). In these heterostructures, both layers are coupled via the exchange bias mechanism [4] which was demonstrated by the shift of the hysteresis loops (Figure 3b). The Malozemoff model of the exchange bias states that the atomic scale disorder at the interface between ferro- and antiferromagnet results in domains within the antiferromagnetic layer. In this framework, the exchange field H_e is expected to be inversely proportional to the antiferromagnetic domain size (see Figure 3c). The anti-ferromagnetic domain size L_{AF} was extracted from neutron diffraction (measured on 4F1) and compared to the ferroelectric domains size L_{FE} measured by piezoresponse force microscopy. Both sizes have been shown to be identical which suggests that the magneto-electric coupling still exists in these thin film structures. The exchange field was also shown to be inversely proportional to the domain size as predicted.

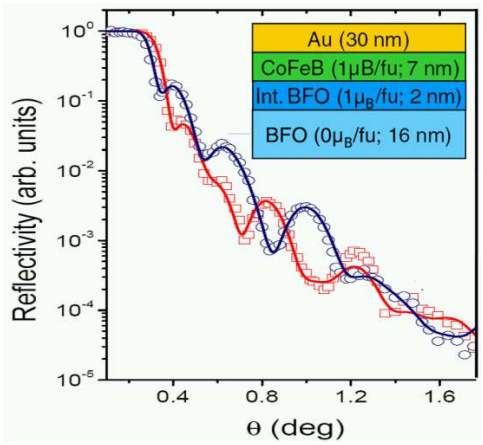

Fig.4: Polarized neutron reflectivity on a CoFeB/BifeO3 heterostructure showing the presence of an induced magnetization in the interfacial BFO layer).

Furthermore, the analysis of the data with Malozemoff's model suggests that a net magnetization should be observed in $BiFeO_3$ near the interface with CoFeB. To test this prediction, polarized neutron reflectometry was performed on CoFeB/$BiFeO_3$ bilayers on the PRISM spectrometer. The data (see Figure 4) are well reproduced by taking into account a 2 nm thick layer carrying a 1 μ_B per formula unit magnetic moment. This is far larger than the surface magnetic moment predicted by Malozemoff's model, and suggests that unpinned spins exist in $BiFeO_3$ near the interface along with the pinned moments predicted by the model. As the ferroelectric domain structure can be easily controlled by an electric field, these results open the route to the electrical manipulation of magnetization at room temperature, in $BiFeO_3$ -based exchange-bias heterostructures.

References:

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