Temperature dependence of the exchange bias properties in polycrystalline $BiFeO_3/Ni_{80}Fe_{20}$.

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Multiferroics (MF) based nanostructures combining ferroelectricity (FE) and antiferromagnetism (AF) are attracting a lot of interest in the scientific community. Indeed, a controlled magnetoelectric coupling in a single nanostructure will bring a direct way to control the magnetic state of the MF material. Therefore, MF materials are good candidates for use in nanodevices, like magnetic memories or high frequency devices [1–3]. However, most of MF materials possess a Néel temperature well below 300 K, reducing the number of candidates available for room temperature applications. Still, a good candidate for exchange coupling at room temperature is BiFeO₃ (BFO). An interesting use of MF would be in MF/Ferromagnet exchange coupled bilayers as the net magnetization could then be controlled through the coupling by the use of an electric field. Such coupling named exchange bias was discovered by Meiklejohn and Bean [4]. It results in a characteristic horizontal shift of the hysteresis cycle (bias), and can be interpreted as unidirectional anisotropy.

Previous studies carried out on epitaxial or bulk BFO have demonstrated the key role of the crystalline ordering (eg. the strain) for driving some of the multiferroic properties (eg. the presence of a magnetic cycloid) [5, 6]. Furthermore, it is well known that the unidirectional anisotropy depends on structural disorder at the interface, which includes crystalline disorder. Therefore, it is of interest to study the exchange bias properties not only in epitaxial systems but also in polycrystalline ones. Following this, we have recently demonstrated that an exchange coupling may be induced in polycrystalline BiFeO₃/Ni₈₀Fe₂₀ (Py/BFO) bilayers [7, 8]. At 300 K, the interfacial exchange coupling energy in the polycrystalline BFO/Py bilayers is similar to those reported in previously studied epitaxial BFO/F bilayers [9–11]. However, a temperature dependence study could reveal differences between epitaxial and polycrystalline systems.

In this study, we report a temperature dependent analysis of the exchange bias properties in BFO/Py bilayers as a function of the BFO thickness. The temperature dependence of the exchange bias field (H_e) and the coercive field (H_c) are first presented. To understand exchange bias magnetization reversal and the magnetic anisotropies, we will then present an azimuthal study of H_e and H_c at 300 K and 77 K. Finally, results of a controlled field cooling protocol applied on all samples will be discussed to understand anisotropy energy distribution. The bilayers were grown by radio-frequency sputter deposition, with the following structure: Si/Pt(14 nm)/BiFeO₃(t_{BFO})/Ni₈₀Fe₂₀(10 nm)/Pt(10 nm), with t_{BFO} among 0 nm, 29 nm and 177 nm. To induce uniaxial anisotropy, a 300 Oe field H_{dep} was applied during the growth. The XRD analysis confirmed a single polycrystalline structure for the BFO layer.

To investigate the reversal mechanism, we measured the azimuthal magnetic behavior using a vectorial vibrating sample magnetometer (VVSM) at room temperature. Azimuthal evolutions of H_e and H_c show a complex pattern (fig.a), revealing the presence of a biquadratic anisotropy. A misalignment between the deposition field and the angular position of the maximum value of H_c is also observed for $t_{BFO}=0\,\mathrm{nm}$ and 29 nm, and vanishes for the thickest sample. It indicates that a transfer of anisotropy through exchange coupling with reversible spins is present between the layers.

As the anisotropy energy may depend on the temperature, we employed a MPMS SQUID (Quantum Design) with sample temperatures between $10\,\mathrm{K}$ and $390\,\mathrm{K}$. After a field cooling along the deposition field, H_e and H_c were deduced from hysteresis cycles at increasing T up to room temperature (fig.b). The results show a coercive field decreasing exponentially with T increasing. The evolution of the exchange bias is more complex, as it is not monotonic for all bilayers. This behavior differs from usual F/AF exchange bias systems where a monotonic enhancement of both coercive and exchange field with decreasing temperature is observed. This unusual comportment was previously observed in epitaxial [12] and polycrystalline [13] BFO/F bilayers and its origin is not clear.

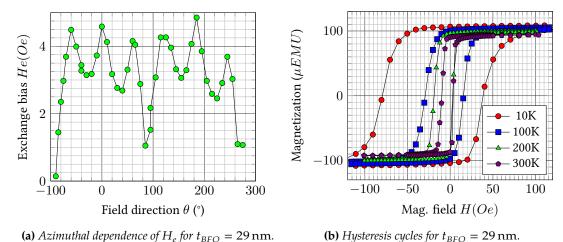
In order to understand the bias evolution with temperature and probe the anisotropies directions at

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low temperature, we measured magnetic hysteresis cycles at 77 K depending on the azimuthal position of the sample compared to the deposition field direction. For all samples, the azimuthal symmetry axes are not temperature dependent but a change of shape is noticeable, revealing a modification of the anisotropies' configurations, involving different ratio and/or misalignment between anisotropy axes. Such modifications do not result in a non-monotonic trend of H_e in most F/AF systems. A theoretical study will be presented to understand this non-monotonic trend.

Also, a controlled temperature sweep protocol was performed, giving access to the so called blocking temperature distribution, interpreted for polycrystals to AF domain distribution. We show that this distribution is similar to epitaxial systems, despite structural differences. We will discuss that considering frustration or domain structure within BFO.



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