Electric field-driven chemical reaction at the buried FeCo/MgO interface for potential use in low power spintronics devices

A challenging goal of spintronics is to manipulate electrically the magnetization direction in ferromagnets. In particular, this is a critical issue for the industrial development of magnetic random-access memories (MRAMs) composed of arrays of magnetic tunnel junctions (MTJs), the magnetization direction in each MTJ coding one bit of information. The usual way to switch the magnetization of a ferromagnetic layer is by using *current*-based methods: traditionally with the Oersted field generated by a current and recently with spin-transfer, by passing a large current through an MTJ. Interest on voltage-based methods is currently growing as these methods would require a much lower power consumption. We have recently shown that the perpendicular magnetic anisotropy (PMA) of ultrathin FeCo can be tuned with an electric field, leading to a reorientation of the easy magnetization axis from in-plane to out-of-plane under opposite voltages [1]. With this effect, precessional magnetization switching in MTJs could be induced by application of voltage pulses [2].

However, results that may seem inconsistent have been reported. The sign of the anisotropy change strongly depends on the ferromagnetic material and the nature of underlayers, and different time scales have been reported as well [1,3]. Actually, an electric field near a metal/insulator interface can have a variety of consequences on the properties of the metallic layer. First, capacitive charge accumulation modifies the density of states at the Fermi level and alters the electronic structure. Lattice distortions can also occur, as well as oxidoreduction and electromigration effects, with exchange of atoms between the metal and the insulator. When the metal is a ferromagnet, these effects may involve the electron spin, leading to magnetoelectric functionalities.

Investigation of these effects with spectroscopic techniques offers the opportunity to shed some light on the microscopic mechanisms occurring under voltage application. In particular, X-ray absorption near-edge spectroscopy (XANES) is particularly sensitive to the oxidation state, whereas X-ray magnetic circular dichroism (XMCD) is known to be a powerful tool for probing the atomic magnetic moment. In this paper, we report on the first XANES/XMCD measurements of an ultrathin FeCo film subjected to an electric field [4].

We studied multilayers with the following stacking: Au(50)/Fe_{0.9}Co_{0.1}(0.5)/MgO(1.8)/Cr(2)/Au(5) (asgrown sample) and Au(50)/Fe_{0.9}Co_{0.1}(0.5)/MgO(1.8)/ SiO₂(5)/Cr(2)/Au(5) (patterned sample), grown on MgO(001) by molecular beam epitaxy and SiO₂ sputtering (thicknesses are in nanometers). Pillars with a diameter of 200 μ m were patterned by electron beam lithography and ion etching. XANES measurements at the Fe $L_{2,3}$ edges were performed at the soft X-rays beamline **BL25SU**, with a beam diameter smaller than 200 μ m and partial fluorescence detection. For XMCD measurements, a 0.5 T magnetic field was applied at 30° from the normal of the sample surface.

Figures 1(a) and 1(b) show the Fe $L_{2,3}$ XANES and XMCD spectra of the as-grown and patterned samples (sketched in Fig. 1(d)). The spectra of the as-grown sample show line shapes similar to those of pure Fe. Microfabrication induces marked changes in these spectra. Namely, both L_2 and L_3 peaks present a shoulder on their high energy side (indicated by arrows in Fig. 1(a)), while the XMCD signal is markedly reduced. These features are characteristic of a nonmagnetic FeO oxide. This partial oxidization of Fe is actually due to an etching step necessary to obtain the MgO/SiO₂ bilayer. From the XMCD reduction, we estimated that 1.8±0.5 monolayers (MLs) at the FeCo/ MgO interface are oxidized. This corresponds to half of the grown 3.5 ML-thick FeCo film.

We measured the XANES/XMCD spectra of a pillar



Fig. 1. (a) XANES and (b) XMCD spectra at the Fe $L_{2,3}$ edge for the as-grown and patterned samples, indicating Fe oxidation after microfabrication. (c) XMCD loops of the two samples with a magnetic field oriented at 30° from the surface normal and with a reduction in perpendicular anisotropy due to oxidation. (d) Sample structures.



Fig. 2. (a) Fe $L_{2,3}$ XANES and (b) XMCD spectra of a pillar under a voltage of ± 2 V. Variations in XANES line shape and XMCD amplitude indicate oxidoreduction.

while setting an electric field across the MgO/SiO₂ bilayer. The FeCo electrode was grounded and a voltage was applied to the Cr/Au electrode. The voltage was set to +2 V, then to -2 V, three times repeatedly. The results presented in Fig. 2 were obtained by averaging the spectra acquired separately at +2 V and -2 V. Varying the voltage induces a change in the relative weight of the main peaks and oxide-related shoulders. This means that the oxidation (reduction) of Fe is favored by an outward (inward) electric field with respect to the FeCo electrode. The XMCD change under ±2 V, and therefore the change in the magnetic moment, reaches a large value of 25%, which corresponds to the oxidoreduction of approximately 0.26 ML of Fe at the interface. Interestingly, the reaction is found to be reversible. This can be confirmed in Fig. 3, which displays the XANES variations at the L_3 energy when cycling the voltage ten times between +2 V and -2 V. Averaging over all voltage cycles yields the voltage dependence given in Fig. 3(c).

From the polarity of the voltage leading to oxidation/reduction, it can be inferred that the driving force is the electrostatic force, and not the "wind force" due to momentum transfer from the flowing electrons. This is at odds with the previous observation of electromigration in MTJs, which was driven by the current (10⁶ A/cm²) and occurred through nanoconstrictions in the insulator [5]. In our case, the current density and electric field reach at most 20 A/cm² and 0.14 V/nm, respectively. Together with the reversibility of the reaction, these observations suggest that in our experiments, the oxidoreduction

originates from small changes in the positions of ions at the FeCo/MgO interface, uniformly over the surface.

The XMCD loops measured on the as-grown and patterned samples indicate that Fe oxidation, and therefore a negative applied voltage, reduces the PMA (Fig. 1(c)). This is opposite to the results reported in Ref. [1], in which a negative voltage induced an increase in PMA. Note that those studies dealt with as-grown films, showing no oxidization. This is an important result that sheds a new light on the voltage control of magnetism and clearly shows that several mechanisms (e.g., charge accumulation and oxidoreduction) can lead to magnetoelectric functionalities in solid-state devices. In future investigations, XANES/XMCD measurements will be performed to study the effect of charge accumulation on the magnetic anisotropy of these films.



Fig. 3. Time traces of (a) the applied voltage and (b) fluorescence yield for an excitation of 708 eV (Fe L_3) while cycling the voltage between +2 V and -2V. (c) Voltage dependence of the fluorescence yield after averaging ten voltage sweeps.

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