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# Time scales of bias voltage effects in Fe/MgO-based magnetic tunnel junctions with voltage-dependent perpendicular anisotropy

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## ABSTRACT

Interplay between voltage-induced magnetic anisotropy transition and voltage-induced atomic diffusion is studied in epitaxial V/Fe (0.7nm)/MgO/Fe(5nm)/Co/Au magnetic tunnel junction where thin Fe soft electrode has in-plane or out-of-plane anisotropy depending on the sign of the bias voltage. We investigate the origin of the slow resistance variation occurring when switching bias voltage in opposite polarity. We demonstrate that the time to reach resistance stability after voltage switching is reduced when increasing the voltage amplitude or the temperature. A single energy barrier of about 0.2 eV height is deduced from temperature dependence. Finally, we demonstrate that the resistance change is not correlated to a change in soft electrode anisotropy. This conclusion contrasts with observations recently reported on analogous systems.

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## 1. Introduction

Epitaxial magnetic tunnel junctions (MTJs) composed of two ferromagnetic electrodes separated by a single-crystal MgO(001) barrier remain a model system for studying the physics of spin-dependent tunneling during the last 15 years. Tunnel magnetoresistance (TMR) effect is observed in epitaxial MgO-based MTJs because of specific spin filtering, which does not exist in Al-O-based MTJs [1,2]. MTJs are currently used in many spintronic applications: magnetic sensors, magnetic read head in hard disk drive, spin transfer magnetic random access memories (STT-MRAM). CoFeB is often chosen as electrodes for MgO-based MTJs in these applications. First, it provides TMR as high as 1100% at 5 K [3]. Secondly, CoFeB/MgO interface favors perpendicular magnetic anisotropy which is preferred since it improves the writing process as well as the thermal stability of the MRAM-cell [3]. Third, it has been demonstrated that CoFeB/MgO interface anisotropy can be controlled by applying a voltage through the MgO. The ability to dynamically control the characteristics of this interface with an electric field opens a new way to control the writing process in MRAM that would substantially reduce the energy cost and writing time [4-6]. Therefore understanding and controlling the effect of the voltage at MgO interface is currently of great interest.

In ferromagnetic metal / oxide bilayers, such as Co / AlOx [1] and CoFe / MgO [3,7,8], high perpendicular magnetic anisotropy (PMA) originates from the hybridization between Fe and Co with oxygen atoms. Most of the works on voltage control of PMA has focused on the charge accumulation at the metal / oxide interface when applying a voltage on the MgO tunnel barrier in MTJ [4-6]. Charge accumulation of the Fermi level is expected to shift the Fermi level energy. This effect is reversible, linear and symmetric in voltage. PMA changes with voltage is usually characterized by  $\beta = \Delta K \cdot t_{\text{MgO}} / \Delta V$  where  $\Delta K$  is the PMA variation,  $t_{\text{MgO}}$  is the MgO layer thickness and  $\Delta V$  is the voltage. With charge accumulation, typical values for  $\beta$  are around 50 to 100  $\text{fJ} \cdot \text{V}^{-1} \cdot \text{m}^{-1}$  [4-6]. Some other experimental studies have highlighted either irreversible phenomena or asymmetric variations of PMA with voltages [9,10]. In these experiments, values of  $\beta$  are much larger than observed previously and theoretically predicted [9,10]. It has been demonstrated very recently that ion displacements at the interface must be responsible for these observations [11-13]. In Ref. 13, using electron microscopy, high resolution transmission (HRTEM) and spectroscopy of electron energy loss (EELS) on a cross section of a bilayer Co / GdO<sub>x</sub>, the authors show a migration from the front of oxygen upon application of a voltage. The same conclusions were obtained by X-ray dichroism at Co/GdO<sub>x</sub> and FeCo/MgO interfaces [11,12]. Oxygen ions migration in oxides is

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already operating as a resistive switching mechanism in the phase change devices and memristors [12,14].

Here, we investigate resistance change with time in V/Fe/MgO/Fe epitaxial MTJs where voltage control of PMA has been demonstrated. After changing MTJ bias-voltage from zero to + (-) bias, we observed a continuous increase (decrease) of the resistance over a few minutes to a few hours at room temperature, depending on the bias value. The kinetic of the resistance change is shown to be activated both by electric field (voltage) and temperature. Contrary to the already mentioned previous reports, for our system the resistance evolution in time is not correlated to an anisotropy evolution.

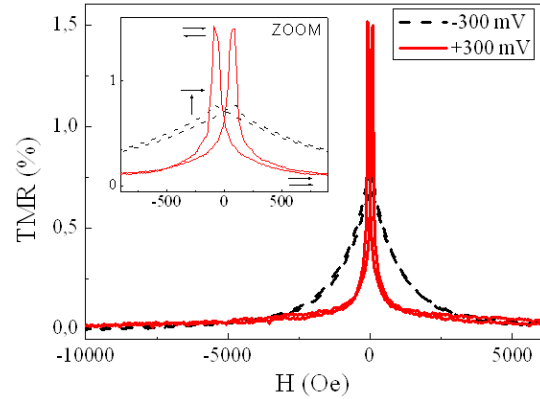
## 2. Experiments

Single-crystal V(5nm)/ Fe(0.7 nm) / MgO(1.2 nm) Fe(5 nm) / Co(5 nm) / Au(5 nm) MTJ was grown by molecular beam epitaxy (MBE), with a base pressure lower than  $10^{-10}$  mbar, on MgO (001) single-crystal substrate as described in details in our previous report [7,10]. Vanadium was deposited at room temperature (RT) and post-annealed at 600 °C. Fe was added at RT and annealed at a lower temperature of 400 °C to smooth the Fe surface, but avoid Fe/V intermixing observed in other samples annealed above 600°C. The MgO barrier and hard magnetic Fe/Co top layer were then deposited at RT and not annealed. The epitaxial relationship, growth mode, number of deposited monolayers (ML), and surface flatness were controlled in-situ using reflection high energy electron diffraction. UV lithography was used to pattern MTJ devices with a junction size of  $30 \times 30 \mu\text{m}^2$ . Then we measured resistance and magnetoresistance in a commercial cryostat Physical Property Measurement System coupled to a home-made electrical transport rod and connecting chip, on which the MTJs are wire-bonded with 2 probes geometry.

## 3. Results and discussions

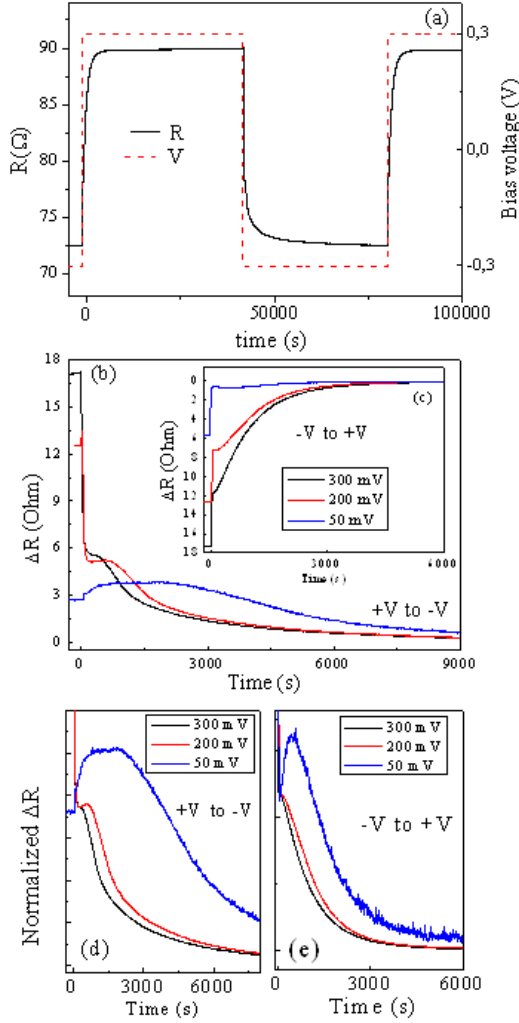
In Fig.1, we present the magnetoresistance versus in-plane field amplitude for two different MTJ bias voltages. The voltage sign is defined such that it is positive when the lower Fe electrode is the positive electrode and the top Fe/Co electrode is the negative electrode. The magnetoresistance curve at +300 mV is typical of MTJs with two in-plane anisotropy electrodes. At high field both magnetizations are aligned, i.e. parallel to each other, along the in-plane field and the resistance is minimum and equal to  $R_P$ . As the field reverses, the magnetization of the softer electrode (the bottom Fe layer) reverses with the field, the resistance increases up to  $R_{AP}$ . Then as the negative magnetic field amplitude increases the hard Fe/Co top electrode magnetization reverses and the resistance returns to  $R_P$ . TMR is defined as  $[R(H)-$

$R_P]/R_P$ . As the voltage bias is turned to -300mV, a different behavior is observed. As the in-plane field is reduced from 10kOe, the resistance continuously increases from  $R_P$  to reach a resistance of about  $R_{AP}/2$  around zero field. This is typical for the MTJ where the hard electrode has in-plane anisotropy and the soft electrode has out-of-plane anisotropy. The continuous increase of TMR is related to the rotation of the soft layer magnetization from in-plane to out-of-plane when the magnetic field is reduced.



**Fig.1. TMR as a function in-plane field amplitude, measured for bias voltage equal to +300 mV (black dashed line) and -300 mV (red solid line) at room temperatures.**

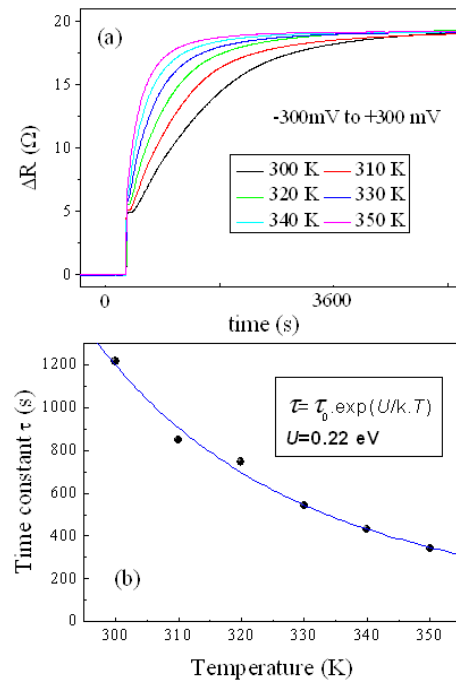
As discussed in Ref. [10] and Ref. [15], the Fe layer in the V/Fe/MgO bottom electrode undergoes four main contributions to magnetic anisotropy. The first one comes from the demagnetization fields that tend to align the magnetization in-plane. The second one comes from the Fe bulk cubic anisotropy. It is very small as compared with demagnetization field component. The third one is the V/Fe interface that we found to be zero [15]. The last one is the interface anisotropy at Fe/MgO interface [7]. The latest provides PMA as described in the introduction. So the main competition is between the demagnetization field which provide in-plane anisotropy and the Fe/MgO interface anisotropy which provides PMA. As changing the bias voltage, the Fe/MgO interface is modified and the PMA is changed so that at +300 mV the V/Fe/MgO electrode has in-plane anisotropy whereas at -300 mV it has out-of-plane anisotropy [10].



**Fig.2.** Resistance as a function of time during bias voltage switching procedure applying the magnetic field in-plane (4 kOe) at room temperature (a) and comparison of variation of the resistance at switching from +300 mV to -300mV and opposite (b), (c). (d) and (e) show normalized resistance as a function of time for “-” and “+” voltages at switching voltage from +V to -V and from -V to +V, respectively.

After applying a bias voltage, a certain delay time is required to obtain curves like the ones in Fig. 1. Indeed, after bias voltage is applied, a continuous MTJ resistance occurs over few minutes to few hours. In Fig. 2(a), we present how the MTJ resistance varies when switching bias voltage from -300mV to +300mV and +300mV to -300mV, under an applied field of 4 kOe, i.e. in the parallel state magnetic configuration of the MTJ. Resistance varies between 72.5 Ohm and 90 Ohm. In Fig. 2(b) and 2(c) we compare the curves obtained for three voltage amplitude  $V_{\max} = 300$  mV, 200mV and 50 mV. In both ascending and descending voltage switching, we notice that the change in resistance occurs in two phases: the first one is a large jump that seems to be immediate after voltage change and then a second part that is a slow variation. As the resistance is

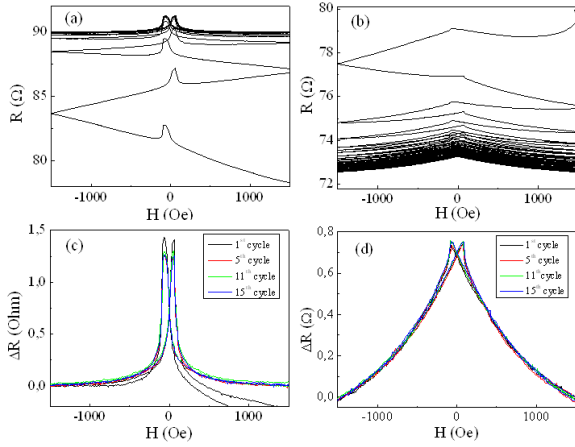
generally dependant on the voltage sign in MTJs (due to different top and down interfaces), resistance and the amplitude of the initial jump depend on the bias polarity. However the second phase in which a slow change of resistance is observed is not expected neither in one or the other voltage sign. Typical time transients (hours) are much larger than the RC time constant of the system or any time delay due to the acquisition. This slow resistance evolution as a function of time depends on the voltage amplitude. To perform a fair comparison, we normalized  $\Delta R$  for both  $-V_{\max}$  to  $+V_{\max}$  and  $+V_{\max}$  to  $-V_{\max}$  switchings (Fig. 2(a) and 2(b)). We observe the same behavior for the two switching. As  $V_{\max}$  increases from 50 mV to 300 mV, the time of requested for the resistance to stabilize decreases.



**Fig.3.** (a) Resistance as a function of time after bias voltage switching procedure from +300mV to -300mV applying the magnetic field in-plane (4 kOe) at the temperature range 300-350K. (b) Characteristic time  $\tau$  of resistance increasing as a function of temperature. The blue line corresponds to a fit considering that the resistance change is a thermally activated process requiring to overpass an unique energy barrier  $U$ .

To further investigate the origin of this slow variation of resistance with time, we performed voltage switching under 4kOe and at six temperatures ranging from 300 to 350 K. As the temperature increases, the time requested to achieve constant resistance decreases (Fig. 3(a)). So the observed effect is thermally activated. The time evolution of the resistance in the slow variation phase is well described by an exponential variation  $\Delta R(1-\exp(-t/\tau))$ . Fig. 3(b) represents the temperature dependence of the characteristic time  $\tau$ . This temperature dependence is nicely fitted by an exponential decay of the form  $\exp(-U/k.T)$  with  $U = 0.22$  eV. Therefore

the mechanism responsible for the slow resistance variation is thermally activated and is well described by a single energy barrier of 0.22 eV.



**Fig.4. Stabilization of resistance at voltage +300 mV (a) and -300 mV (b) (15 cycles). Comparison of few  $R(H)$  cycles with repetition of measurement 15 times at voltage 300 mV (a) and -300 mV (b). For comparison, the first, fifth, eleventh and fifteenth loops are plotted together after linear slope correction in (c) and (d) respectively.**

One possible explanation for this slow change of resistance is that the MgO barrier does not behave like an ideal dielectric layer, due to defects in the MgO barrier, like vacancies or dislocations [16]. Thus, these defects may trap charges, and electromigration may occur to move these defects towards Fe/MgO interfaces as proposed in Ref. [17]. The presence of uncompensated charge in the barrier leads to electric poles that affect the tunnel transport and that can be slowly modified as the MTJ bias voltage switches polarity. So the electric polarization  $P$  increasing with time to some maximum value  $P_{max}$  progressively decreases the applied electric field thereby reducing the conductivity (increasing the energy barrier). This process may partly explain the long-lasting stabilization of resistance at the electric field polarity switching. Such trapping phenomenon may depend on the voltage polarity. Another origin of this phenomenon may come from ion migration inside the oxide barrier as reported in Fe/MgO [11,18], Co / GdO<sub>x</sub> [12,13] or other systems [14,19]. As described in Ref. [18], very small change such as 0.1 Å in the effective barrier thickness leads to few percents of tunnel barrier resistance change. Time scale from few minutes to few hours are typical for resistance variation induced by oxygen migration [12,18].

If strong oxygen migration occurs at the ferromagnetic electrode/barrier interface, interface PMA can be strongly affected [11,13]. In order to clarify the impact on the interface PMA of the phenomenon inducing the slow resistance change, we

performed fifteen successive resistance versus field loops immediately after changing the MTJ bias voltage polarity. In Fig. 4(a) and 4(b), the successive loops obtained after +300 to -300 mV and -300 to +300 mV respectively, are presented. The change of resistance is clearly observed with time. By correcting each loop with only linear slopes, we obtained Fig. 4(c) and 4(d). In both figures, we show the 1<sup>st</sup> loops curve, the 5<sup>th</sup>, the 11<sup>th</sup> and the 15<sup>th</sup>. In Fig. 4(c), one recognizes the TMR vs field loops described in Fig. 1 in the case of in-plane anisotropy soft electrode. In Fig. 4(d), one recognizes the TMR vs field loops described in Fig. 1 in the case of perpendicular anisotropy soft electrode. The 1<sup>st</sup>, 5<sup>th</sup>, 11<sup>th</sup> and 15<sup>th</sup> loops are mostly superposed so that we can conclude that no change of soft electrode anisotropy (i.e. no change of bottom Fe/MgO interface anisotropy) happens during the slow change of resistance after voltage switching. As interface PMA originate from Fe-O boundaries at Fe/MgO interface [8], one can conclude that changes in Fe-O bonds on the bottom electrode/barrier interface are not the origin of the slow resistance change after voltage switching. Thus, we can conclude that the resistance change originates from effect happening either at the top MgO/Fe interface, or inside the tunnel barrier volume as proposed in Ref. [17]. The asymmetric effect between the top and bottom interface is linked to the structural differences that have been already well established and related to the difference in growth conditions [20-23].

#### 4. Summary and Conclusions

As a summary, we have investigated epitaxial MgO sub./V/Fe(0.7 nm) / MgO(1.2 nm) Fe(5 nm)/Co/Au magnetic tunnel junction (MTJ) where the bias voltage can be used to tune the anisotropy of the bottom Fe electrode. When the bias voltage is positive, Fe(0.7nm) anisotropy lays in-plane. When the bias voltage is negative, Fe(0.7nm) anisotropy lays perpendicular to the plane. When we switch from positive to negative voltage (and vice-versa) with the same amplitude, the MTJ resistance changes in two steps. The first one consists in a sharp change. Due to the difference of top and bottom electrodes, the tunnel current direction probes different electronic states and the resistance is different. The second step corresponds to a much slower resistance variation over a few minutes to a few hours depending on the voltage amplitude and the temperature. As the voltage amplitude or the temperature increases the time to reach resistance stability is reduced. Experiments performed at various temperatures show that the phenomenon which creates the resistance change is a thermally activated process that can be related to a single energy barrier of about 0.2 eV height. Finally we demonstrated that the resistance change is not correlated to a change in soft electrode anisotropy so that Fe-O bonds at

bottom Fe/MgO interface are not at the origin of the resistance change. Subsequently, further investigation needs to be performed in order to address the role of the top interface and barrier volume.

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