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Magnetic tunnel junctions using Co/Ni multilayer electrodes with perpendicular magnetic anisotropy

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Magnetic and magneto-transport properties of amorphous Al₂O₃-based magnetic tunnel junctions (MTJ) having two Co/Ni multilayer electrodes exhibiting perpendicular magnetic anisotropy (PMA) are presented. An additional Co/Pt multilayer is required to maintain PMA in the top Co/Ni electrode. Slight stacking variations lead to dramatic magnetic changes due to dipolar interactions between the top and bottom electrodes. Tunnel magneto-resistance (TMR) of up to 8% at 300 K is measured for the MTJ with two PMA electrodes. The TMR value increases when the top PMA electrode is replaced by an in-plane magnetized Co layer. These observations can be attributed to significant intermixing in the top Co/Ni electrode. © 2015 AIP Publishing LLC.

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I. INTRODUCTION

Magnetic tunnel junctions (MTJs) having electrodes with perpendicular magnetic anisotropy (PMA) have attracted considerable interest because they are promising candidates for spin transfer torque magnetic random access memories (STT-MRAM).¹⁻³ Finding PMA materials that simultaneously show large tunnel magneto-resistance (TMR), low damping, low switching current density, and high thermal stability remains a challenge in implementing STT-MRAM. One of the most important systems investigated to date is the CoFeB/MgO/CoFeB stack where perpendicular anisotropy is created at the CoFeB/MgO and MgO/CoFeB interfaces.^{1,4} Appropriate choices of buffer and capping layers can enhance the interface PMA.^{5,6} The main limitation of using CoFeB is the thinness of the electrodes (<1.6 nm) which is necessary for PMA, as at this thickness thermal stability can be an issue.⁷ Rare-earth/transition metal ferrimagnet alloys,⁸ [Fe_{1-x}Co_x/Pt] multilayers (MLs),^{9,10} and L1₀ (Fe,Co)Pt alloys¹¹ have also been tested since they have large PMA. However, both these systems have large damping and low spin polarization.¹² Spin polarization can be improved by inserting CoFeB at the MgO barrier interface.¹³

Co/Ni MLs have attracted considerable attention for spin-transfer applications since they meet the requirements previously listed. In terms of anisotropy, the Co/Ni interface produces PMA as large as few MJ/m³.¹⁴ Changing the thickness of Co allows the magnitude of the PMA to be easily tuned. The saturation magnetization of Co/Ni MLs is approximately 700 kA/m, although the exact value will depend on the individual Co and Ni layer thicknesses. Gilbert damping of Co/Ni MLs mostly ranges between 0.01 and 0.02, depending on the composition.^{15,16} Finally, high spin polarization has been deduced as a result of spin transfer induced domain wall motion experiments.¹⁷ The importance of such a set of characteristics for achieving low critical current and sub-nanosecond switching time has been already demonstrated in metallic Co/Ni-based spin-valves nanopillars.^{3,18} However, no magneto-resistance or spin-transfer torque experiments

have yet been reported for MTJs using PMA Co/Ni electrodes and a tunnel barrier. The difficulties of growing a bcc MgO (100) barrier on top of fcc Co/Ni (111) stack, as well as Co/Ni on a MgO barrier is the limiting factor.¹⁹ Recently, You *et al.*¹⁹ succeeded in growing MgO-based MTJs using two PMA Co/Ni electrodes but only magnetometry measurements were provided.

In this letter, we report an investigation of magnetic and magnetotransport properties of two amorphous Al₂O₃-based magnetic tunnel junctions having one or two fcc (111) Co/Ni PMA electrodes. Magnetometry measurement reveals that subtle variations of magnetization or anisotropy in the top electrode can strongly affect its magnetic reversal properties due to dipolar coupling between electrodes. Magneto-transport measurements demonstrate up to 8% TMR at RT for a MTJ with two Co/Ni PMA electrodes. Here, TMR is defined as the normalized difference between parallel and anti-parallel alignment of the two electrodes magnetization. The TMR increases to 16% at 20 K. Replacing the top PMA soft electrode by an in-plane magnetized Co (15 nm) layer increases the TMR by a factor of two at room temperature. We discuss our results in terms of the structural features of the electrodes.

II. EXPERIMENTAL METHODS

The samples were prepared on thermally oxidized silicon substrates, where the oxide layer thickness was 400 nm, using magnetron sputtering with a base pressure of 5×10^{-8} mbar. The deposition was performed at room temperature. Co/Ni and Co/Pt MLs, as well as the Ta and Pt layers, were grown by dc-magnetron sputtering. Three MTJ samples were produced which all have (i) the same bottom PMA electrode Ta(5)/Pt(10)/Co(0.6)/[Ni(0.6)/Co(0.3)]*3 (thicknesses in nm) and (ii) the same Al₂O₃ (2.5 nm) barrier obtained through the deposition of 1.5 nm Al layer following by an oxidation in a Ar+O₂ plasma. The three samples then had different top electrodes deposited; sample A had a PMA top

electrode consisting of [Co(0.2)/Ni(0.6)*3/Pt(1)]/[Co(0.6)/Pt(1)]*3, Sample B had a PMA top electrode consisting of [Co(0.3)/Ni(0.6)*3/Pt(2)]/[Co(0.6)/Pt(1)]*3 and Sample C top electrode consists in an in-plane magnetized Co(15) single layer (again all thicknesses in nm). UV lithography was used to pattern samples B and C into MTJ devices with junctions size from $10 \times 10 \mu\text{m}^2$ up to $50 \times 50 \mu\text{m}^2$ having $1 \text{ G}\Omega \cdot \mu\text{m}^2$ RA product. Magnetic characterization was performed at 300 K using an Alternative Gradient Field Magnetometer (AGFM). The transport properties were measured using a Physical Properties Measurement System (PPMS) cryostat over a temperature range from 20 to 300 K.

III. RESULT AND DISCUSSION

Fig. 1 shows normalized magnetization curve measured on samples A and B, using an AGFM. In the case of sample B, as the field is applied perpendicularly to the layers, we observe loops with full remanent magnetization and two successive jumps at reverse fields of 130 Oe and 270 Oe, respectively. The first magnetization jump has a larger magnitude than the second one. This indicates that the top electrode with the largest total moment [Co(0.2)/Ni(0.6)*3/Pt(2)]/[Co(0.6)/Pt(1)]*3, is softer than the bottom Pt(10)/Co(0.6)/[Ni(0.6)/Co(0.3)]*3.

This result is counter-intuitive since Co/Pt ML is expected to have a much larger PMA than Co/Ni ML.²⁰ However, the well established layer by layer growth of the bottom Co/Ni ML on a smooth (111) textured Pt buffer²¹ has to be compared with the island-like growth process of the top ML on a Al_2O_3 oxide barrier.²² Moreover, the top ML may not be well (111) textured on the amorphous barrier, and it has been found that (100) and (110) grain significantly reduce PMA in Co/Ni ML.²³ The tail of the first magnetization jump is typical of the dipolar interactions (so-called demagnetization field) in PMA film thicker than few nanometers²⁴ but that a fully anti-parallel state is reached before the second step. Sample A shows a different behavior, a slight decrease of the Pt interlayer thickness in the top layer as compared with sample B leads to a drastic change in the normalized magnetization versus field loop with the disappearance of the anti-parallel plateau (Fig. 1). The fact that top and bottom layers reverse together is due to dipolar

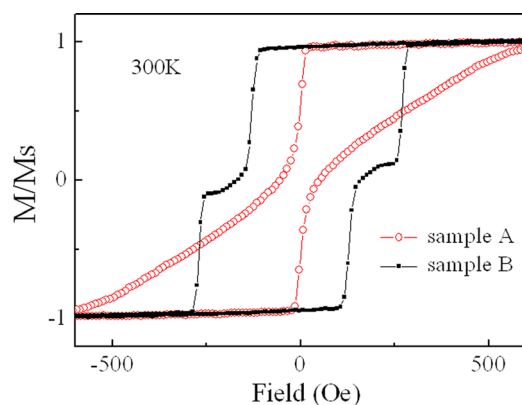


FIG. 1. Room temperature normalized magnetization vs field measurements of $\text{Co(0.6)/[Ni(0.6)/Co(0.3)]*3/AlOx(2.5)/[Co(0.2)/Ni(0.6)*3/Pt(y)/[Co(0.6)/Pt(1)]*3}$ MTJ under out-of-plane applied magnetic field with $y = 1$ (red open circle, sample A) and 2 nm (black solid square, sample B).

interactions which effectively couples the two layers.^{25,26} As a consequence, one has to carefully tune the electrodes not only to insure PMA but also limit the inter-layer dipolar coupling.

Magneto-resistance measurements performed on patterned sample B are shown in Fig. 2. A significant TMR was measured in the MTJ with two Co/Ni PMA electrodes. TMR values of 8% at 300 K and 16% at 20 K were measured for a 50 mV bias voltage. These values are smaller than the best reported TMRs (about 80%) for CoFeB/ Al_2O_3 -based MTJ.²⁷ Nevertheless, it is of the same order of magnitude as the previously reported for Al_2O_3 -based MTJs with PMA electrodes.^{9,10} Fig. 2(b) shows that the temperature dependence of TMR fits well with the $(1-\alpha T^{3/2})$ dependence usually reported and linked to the spin-polarization decrease and increase of the inelastic processes as the temperature increases.²⁸

The Brinkman model^{9,29} that describes the bias-voltage dependence of tunnel current can be employed to provide information on the barrier features. Room temperature $I(V)$ curve measured in parallel state is presented in Fig. 3(a) and compared with a fit to the Brinkman model using a 2.5 nm barrier width. A good match is obtained with a 1.18 eV zero bias barrier height and no barrier asymmetry. This barrier height value confirms the average quality of our Al_2O_3 layer. As barrier values of up to 3 eV can be achieved there is considerable scope for much larger TMR if we further improve our Al_2O_3 barrier layer.³⁰ Interestingly, no barrier asymmetry is needed in the Brinkman fit. This indicates that the bottom and top interfaces are similar. The same conclusion can be drawn from the voltage dependence of TMR (Fig. 3(b)).

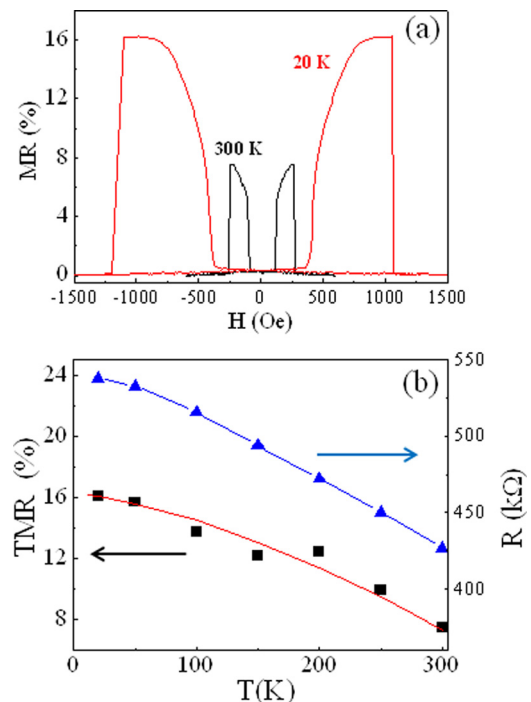


FIG. 2. (a) MR vs out-of-plane field for sample B with both PMA hard [Co/Ni] and soft [Co/Ni][Co/Pt] electrodes measured under 50 mV bias voltage, at the 300 K (black line) and 20 K (red line). (b) For the same sample, TMR (black points) and resistance (blue triangle) vs temperature. The red line corresponds to the theoretical $(1-\alpha T^{3/2})$ dependence of the TMR. The blue line is a guide for the eye.

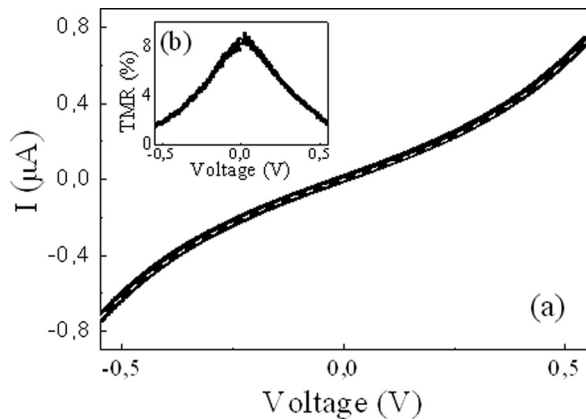


FIG. 3. (a) Experimental current vs bias voltage (black points) for sample B at 300 K compared with Brinkman fit (dashed white line). (b) TMR versus bias voltage measured on sample B at 300 K.

The decrease of TMR with increasing bias voltage mostly due to inelastic scattering by magnons, excitations and the shape of the electronic density of states,²⁸ is symmetric. It might have been anticipated that the TMR(V) curves would be asymmetric since the bottom and top electrode stacks are different, leading to a different density of states and thus spin polarization at the Fermi energy. The lack of TMR(V) asymmetry observed here suggests that diffusive processes strongly affect the magneto-transport properties. Such diffusive processes are well-known to be enhanced by interface roughness and structural defects in the layers. Consequently, increasing the crystalline quality of the layers should lead to larger TMR.

Fig. 4 shows magneto-resistance measurements performed on sample C which differs from sample B as the top Co electrode has in-plane magnetized allowing an orthogonal configuration between the two electrodes at remanence. Interest in such an orthogonal magnetic geometry has grown in the past years because of its possible use in sensors,^{31,32} OST-MRAM,³³ and RF oscillators.^{34,35} In sample C, when the external magnetic field is large enough, the magnetizations

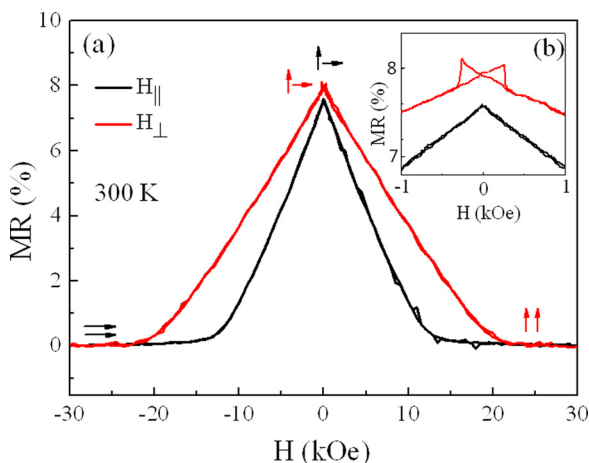


FIG. 4. (a) MR of sample C, i.e., Pt/Co(0.6)/[Ni(0.6)/Co(0.3)]*3/AlOx(2.5)/Co(15) MTJ measured applying the magnetic field in-plane (black line) and out-of-plane (red line) under 50 mV bias voltage at 300 K. (b) Corresponds to a zoom around zero field and highlights the reversal of PMA Co/Ni bottom electrode magnetization at -270 Oe.

of both electrodes are aligned along the field (applied either in-plane or out-of-plane). At zero applied field, the Co/Ni ML moment is perpendicular, whereas Co moment lies in-plane. Hysteresis occurs for the case when the applied field is out-of-plane as the bottom Co/Ni ML magnetization reverses. The coercive field is 270 Oe, identical to the value obtained from magnetometry measurements shown in Figs. 1 and 2 for sample B. In the case of an in-plane applied field, no hysteresis is observed for the Co layer. Note that the in-plane field curve provides a measure of the anisotropy field of the bottom Co/Ni ML which is approximately 12 kOe, in agreement with previous measurements.^{14,36} We note that the difference in resistance between the saturated state and the remanent state is about 8% at 300 K for this orthogonal magnetic configuration. This corresponds to a TMR (i.e., the normalized difference between parallel and anti-parallel alignment of the two electrodes magnetization) of 16% between the parallel and a hypothetical anti-parallel state. Hence, at room temperature sample C would in principal have two times higher TMR than sample B. This result is different to expectations since in recent spin-resolved photo-emission spectroscopy experiments, we observed that spin-polarization at the Fermi level for epitaxial [Co(x)/Ni(0.6)] ML with $0.1 \text{ nm} < x < 0.6 \text{ nm}$, is larger than for pure Co.³⁷ Since the bottom electrode and barrier quality are expected to be the same for both samples, it most probably indicates intermixing in the top Co/Ni ML that leads to lower than expected polarization and PMA, compared to a well layered stack. Indeed, the spin-polarization for a CoNi alloy is expected to be lower than for pure Co.³⁸

IV. CONCLUSION

In summary, our work demonstrates the potential of Al₂O₃-based magnetic tunnel junction (MTJ) with one or two perpendicular anisotropy (PMA) Co/Ni electrodes for future spin electronics device (MRAM, sensors, RF oscillators). Due to the island growth of Co/Ni on the Al₂O₃ barrier, the top Co/Ni electrode has to be covered by a Co/Pt stack in order to maintain the PMA in the top electrode. At 300 K, 8% TMR at 300 K was measured in the full PMA Pt/Co[Ni/Co]₃/Al₂O₃/[Co/Ni]₃/Pt/[Co/Pt]₃. Study of the tunnel barrier characteristics showed that our Al₂O₃ layer crystalline quality can be improved. Moreover, comparison with orthogonal anisotropy Pt/Co[Ni/Co]₃/Al₂O₃/Co MTJ indicated that intermixing must exist in the top Co/Ni electrode of the full PMA MTJ which lowers its polarization and PMA. Overall, this provides encouragement that it will be possible to achieve larger PMA and TMR values for Co/Ni-based magnetic tunnel junction with both PMA electrodes.

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