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X-ray analysis of the magnetic influence of oxygen in Pt/Co/ AlO_x trilayers

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X-ray spectroscopy measurements have been performed on a series of Pt/Co/ AlO_x trilayers to investigate the role of Co oxidation in the perpendicular magnetic anisotropy at the Co/ AlO_x interface. It is observed that varying the degree of oxidation modifies the magnetic properties of the Co layer, inducing a magnetic anisotropy crossover from in plane to out of plane. The microscopic structural properties are analyzed via x-ray photoelectron spectroscopy measurements. It is shown that increasing the oxidation time enhances the amount of interfacial oxide, which may be at the origin of perpendicular magnetic anisotropy.

Perpendicular magnetic anisotropy (PMA) in Co/Pd, Co/Pt and Co/Au multilayers¹ has attracted much interest in the scientific community questioning the fundamental origins of such PMA. A number of theoretical^{2–5} and experimental^{6,7} studies have revealed the seminal role of hybridization of the atomic orbitals at the interface and its resulting influence on the interfacial PMA. These hybridizations modify the 3d orbitals energies of Co, inducing crystal field effects which, in combination with spin-orbit coupling, eventually favor out-of-plane interfacial magnetic anisotropy.⁵

Although PMA was first observed in ferromagnetic/noble metals multilayers, it has also been demonstrated in Pt/Co/ AlO_x trilayers.^{8–10} The origin of PMA can be found in the hybridization of O 2p orbitals with Co 3d orbitals.¹¹ Tsymbal and Belashchenko¹¹ demonstrated the fundamental influence of resonant states on the electronic structure in F/I (F is a ferromagnet and I an insulating oxide) interface. Oleinik *et al.*¹² described the role of interfacial oxygen in the enhancement of the magnetic moment of cobalt. The charge transfer which occurs between Co and O enhances the asymmetry of the valence band of Co, increasing the interfacial spin polarization. This charge transfer reduces the energy of Co d orbitals pointing toward O, creating a splitting between in-plane ($d_{xy}, d_{x^2-y^2}$) and out-of-plane (d_{xz}, d_{yz}, d_{z^2}) d orbitals (parameter Δ in Ref. 5). Consequently, one can expect a strong enhancement of PMA induced by Co–O bondings.

In this article, we investigate the role of interfacial oxidation in the anisotropy crossover of a thin Co layer in Pt/Co/ AlO_x sandwiches. The macroscopic magnetic characteristics are analyzed in parallel with the microscopic structural properties of the samples obtained via x-ray photoelectron spectroscopy (XPS)

Pt (3 nm)/Co (0.6 nm)/Al (1.6 nm) trilayers were deposited on a thermally oxidized silicon wafer by conventional dc magnetron sputtering with a base pressure of 5

$\times 10^{-8}$ mbar. The samples were oxidized by using oxygen rf plasma with a partial pressure of 3×10^{-3} mbar and a power of 10 W. The oxidation time t was varied from 15 to 60 s. These samples were highly stable after oxidation so that no capping layer had to be deposited on the top of AlO_x .

The macroscopic magnetic properties of these samples were studied by extraordinary Hall effect (EHE) in a standard four-point probe Hall geometry^{8,13} which allows us to probe the out-of-plane component M_z of the magnetization. The field variation $M_z(H)$ must be interpreted considering the effective magnetic anisotropy energy (MAE) of the Co layer. This effective MAE is the combination of three anisotropy sources: interfacial anisotropies (Pt/Co and Co/ AlO_x) and bulk anisotropy including shape, magnetocrystalline, and stress contributions. Figure 1 shows the hysteresis loops obtained by room temperature EHE for the trilayer sample when the field is applied perpendicular to the plane of the layers, after oxidation with different exposure times.

From $t = 15$ s to $t = 25$ s, the EHE measurements show no hysteresis and zero remanence. Magnetization saturates at

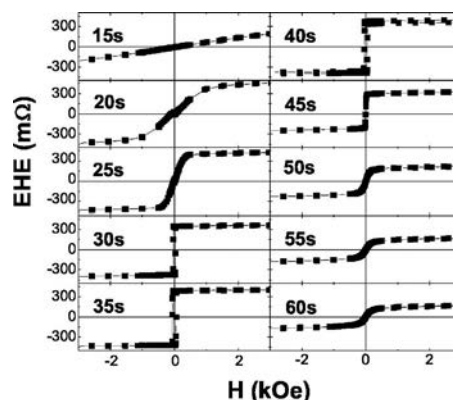


FIG. 1. Hall resistance as a function of the applied field for the Pt/Co/ AlO_x sandwiches oxidized from 15 to 60 s. The field is applied perpendicular to the plane of the trilayer.

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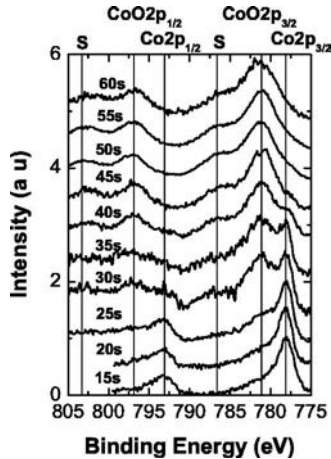


FIG. 2. X-ray photoelectron spectroscopy of Co $2p$ edges in Pt/Co/ AlO_x trilayers for various plasma oxidation times from 15 to 60 s.

high field ($|H| > 1$ kOe), where the perpendicular component of the Co magnetization M_z no longer depends on H and the residual slope comes from the ordinary Hall contribution.⁸ This demonstrates that the out-of-plane direction is a hard axis. EHE with in-plane field and superconducting quantum interference device (SQUID) measurements (not shown here) demonstrate that the magnetization forms an in-plane domain structure. One then needs to apply a large external field to saturate it in the out-of-plane direction. The saturation field is lower than $4\pi Ms$ (≈ 12 kOe) because of the contribution of Co/Pt PMA energy.¹ We also observe that the slope at zero field increases with the oxidation time.

From $t=30$ s to $t=45$ s, the samples show square hysteresis loops with sharp magnetization reversal and a nonzero coercivity. These features indicate that the out-of-plane direction becomes an easy axis, which is confirmed by EHE with in-plane field and SQUID measurements (not shown).

Finally, from $t=50$ s to $t=60$ s, the EHE measurements show no hysteresis and zero remanence. The amplitude of EHE signal decreases from $t=45$ s to $t=60$ s, which indicates that the effective thickness of the magnetic layer decreases. This is interpreted as due to the oxidation of the Co layer. The EHE slope at zero field decreases when increasing the oxidation time and the magnetization saturates at high external field. This last observation will be explained further.

The magnetic states deduced from Fig. 1 can be interpreted considering the magnetic anisotropy of the Co layer. Between $t=15$ s and $t=40$ s, the oxidation only modifies the Co/ AlO_x interface anisotropy. In contrast, from $t=45$ s to $t=60$ s, progressive oxidation of the Co layer must be taken into account.

To understand the microscopic origin of the anisotropy crossover associated with oxidation, we performed Co $2p$ XPS measurements. XPS measurements were carried out at the advanced photoelectric-effect experiments beamline of the Elettra synchrotron in Trieste (Italy) and performed setting the incident photon energy at $h\nu=1130$ eV, with an optimized incident angle of $\theta=60^\circ$. Figure 2 shows the spectra obtained for the ten samples. The spectrum of the $t=60$ s sample corresponds to pure CoO:¹⁴ two main peaks can be distinguished, corresponding to CoO $2p_{1/2}$ and CoO $2p_{3/2}$

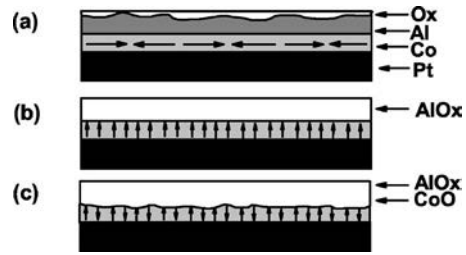


FIG. 3. Schematics of the influence of oxidation on the magnetic properties of Co layer in a Pt/Co/ AlO_x trilayer: (a) for small oxidation times, the magnetization lies in the plane forming domain walls; (b) for optimal oxidation times, the magnetization lies out of plane in a monodomain structure; (c) for long oxidation times, the magnetization forms out-of-plane stripe or labyrinth structure.

core levels (lying at 796.3 and 781.1 eV, respectively) and two satellite peaks (denoted by S and lying at 803.3 and 786.7 eV, respectively) which arise from the charge transfer between O $2p$ and Co $3d$. The main peaks are shifted toward larger binding energies with respect to Co metal peaks due to oxygen environment. The spectrum of the $t=15$ s sample is similar to that of pure cobalt,¹⁵ with Co $2p_{1/2}$ and Co $2p_{3/2}$ lying at binding energies of 729.5 and 778.1 eV, respectively.

The spectra clearly show that the Co/ AlO_x interface composition changes from pure Co to pure CoO during the oxidation process. Since XPS measurements are surface sensitive (the collected intensity is exponentially decreasing with a penetration depth of the order of 5–6 Å in Co), the spectra give information about the Co chemical composition averaged on the entire Co layer, with dominant contribution from the topmost Co monolayer (top Co–Al bondings, bottom and lateral Co–Co bondings). For samples oxidized from $t=15$ s to $t=25$ s (in-plane anisotropy), the spectra have mainly pure Co character, except at $t=25$ s, where a small contribution of CoO appears. For $t=30$ s to $t=40$ s, the spectra possess both Co and CoO characters, showing that most of the Co located at the Co/ AlO_x interface possesses Co–O bondings (Co–Al bondings are replaced by Co–O bondings), while the small contribution of pure Co in the XPS spectra arises from the remaining Co–Co bondings. Finally, from $t=45$ s to $t=60$ s, the Co character completely vanishes, indicating that the first Co monolayer (at least) is totally oxidized (Co–Al and Co–Co bondings are replaced by Co–O bondings).

We therefore conclude that the onset of PMA is related to the appearance of a significant density of interfacial Co–O bonds at the Co/ AlO_x interface. These results are summarized in Fig. 3. For small oxidation times [Fig. 3(a)], the Al layer is not fully oxidized and the magnetization of Co layer forms an in-plane domain structure. For intermediate (or optimal) oxidation times [Fig. 3(b)], O atoms have reached the Co/Al interface, so that most of the Co–Al bondings are replaced by Co–O bondings. In this case, the magnetic anisotropy of the Co/ AlO_x interface overcomes the other anisotropy sources and the magnetization of the Co layer lies out of plane in a single domain structure. The magnetic structure of the Co layers has been also confirmed both by

EHE with in-plane field and x-ray circular magnetic dichroism.¹⁶

Finally, for long oxidation times [Fig. 3(c)], the Co layer gets more and more oxidized. It is known that this oxidation preferentially takes place by oxygen diffusion through grain boundaries.¹⁷ This reduces the effective in-plane exchange stiffness and thereby lowers the domain-wall energy. As a result, the magnetization of the Co layer breaks up into up and down small domains forming labyrinth or stripe domain structures.¹⁸ This particular structure has been confirmed by both EHE with in-plane field and x-ray linear magnetic dichroism for over-oxidized samples.¹⁶

In conclusion, we have shown that interfacial oxidation of Co in Pt/Co/ AlO_x trilayers is at the origin of strong perpendicular magnetic anisotropy. Perpendicular anisotropy crossover induced by annealing in such structures has also been studied.¹⁶ *Ab initio* calculations, aiming at clarifying the role of Co–O hybridization in magnetization anisotropy energy, are presently underway.

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