Crossover between in-plane and perpendicular anisotropy in Ta/CoxFe100-x/MgO films as a function of Co composition

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Crossover between in-plane and perpendicular anisotropy in Ta/Co$_x$Fe$_{100-x}$/MgO films as a function of Co composition

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Interfacial magnetic anisotropy in Ta/Co$_x$Fe$_{100-x}$ (CoFe)/MgO films for alloy compositions
spanning pure Co to pure Fe has been studied in order to investigate the role of chemical
composition in the onset of perpendicular magnetic anisotropy at the CoFe/MgO interface. Out-of-
plane magnetization is not observed in Ta/Fe/MgO ($x = 0$) and Ta/Co/MgO ($x = 100$), for all
ranges of CoFe thickness (t), but a t-dependent crossover between in-plane and out-of-plane
anisotropy is found for $x = 20$, 50, and 80. Interestingly, effective magnetic anisotropy K_u as well
as interfacial anisotropy K_i are maximized for Co$_{50}$Fe$_{50}$ at a fixed $t = 0.8$ nm. The results suggest
that the degree of filling of valence bands in the CoFe adjacent to the interface, which determines
the relative population of the anisotropic d-bands, plays an important role in the interfacial
anisotropy brought on by CoFe-O hybridization at the metal/oxide interface. © *2013 American
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There has been intensive research interest in
Ta/CoFe(B)/MgO stacks with perpendicular magnetic ani-
sotropy (PMA), which are used in tunnel junctions with large
tunnel magnetoresistance for realizing magnetic random
access memory,[1] and which also exhibit structural inversion
asymmetry (SIA) associated with enhanced efficiency of
current-driven domain wall (DW) motion.[2] To realize high-
performance devices, it is crucial to optimize the PMA for
more efficient magnetic switching induced by current.[3] In
such films, the PMA is expected to originate from interfacial
anisotropy (K_i) at both the CoFe(B)/MgO and Ta/CoFe(B)
interfaces after CoFe(B) crystallization into the bcc
phase.[1,4,5] At the metal/oxide interface in particular, symme-
try breaking and CoFe-O hybridization are believed to split
the 3d bands of the CoFe, leading to strong PMA.[6] The
degree of oxidation of metal atoms at the interface[6–8] and the
composition ratio of them which determines the band filling[9]
are considered to be two critical factors to engineer K_i. For
the former, it has been shown that K_i is extremely sensitive
to the degree of oxidation at the Co/MOx interface in
Pt/Co(Fe)/MOx trilayers, where M is a metal such as Al,
Mg, Ta, etc.[6] The origin of the oxygen (O) induced PMA in
such films has been suggested to be related to charge-
transfer between Co or Fe and O atoms through the hybrid-
ization of O 2p orbitals with Co 3d and Fe 3d orbitals.[10–13]
However, for the latter, systematic studies on the composi-
tion dependence of the K_i in this system is lacking and more
understanding about the relation between composition and
K_i is needed. In this study, we experimentally investigate K_i
in Ta/Co$_x$Fe$_{100-x}$(CoFe)/MgO for alloy compositions span-
ning pure Co to pure Fe to determine how the composition
of CoFe affects K_i in this system.

Magnetic films with the stack structure Si (100)/50 nm
SiO$_2$/5.0 nm Ta/(0.5–1.2) nm Co$_x$Fe$_{100-x}$ (CoFe)/1.8 nm MgO/
5.0 nm Ta for alloy compositions $x = 0$, 20, 50, 80, and 100
were prepared using rf sputtering for MgO under 3 mTorr Ar
and dc sputtering for metallic layers under 2 mTorr Ar.

In order to characterize magnetic properties, in-plane
and out-of-plane magnetization curves were measured using
a vibrating sample magnetometer (VSM). We extracted the
magnetic anisotropy energy (K_u) and anisotropy field (H_k) of
each structure by examining the saturation magnetization
(M_s) and saturation field (H_{sat}) in hard-axis curve. The in-
plane and out-of-plane coercive fields (H_c) were obtained
from hysteresis loops measured by the magneto-optic-Kerr-
effect (MOKE) with the field sweep rate of 1.0 Hz in the lon-
gitudinal and polar configuration, respectively.

Figure 1 shows out-of-plane (Fig. 1(a)) and in-plane
hysteresis loops (Fig. 1(b)) measured for Ta/0.8 nm CoFe/
MgO films with different Co compositions ($x = 0$, 20, 50, 80,
and 100), respectively. The Ta/0.8 nm CoFe/MgO films
exhibit PMA only for $x = 20$ and 50 as shown in Fig. 1(a),
whereas for all other compositions, the out-of-plane rema-
nent magnetization vanished and in-plane magnetic anisot-
ropy (IMA) was observed for this CoFe thickness. With
$x = 0$ (pure Fe case), out-of-plane anisotropy was not
observed although the out-of-plane saturation field was low,
suggesting that an interfacial contribution to PMA may
exist[10–12] but it is insufficient to overcome the demagnetiz-
ing field. Increasing the Co content in the alloy enhances
PMA for intermediate x, but, in the Co-rich case ($x = 80$) and
the pure Co case ($x = 100$), the out-of-plane saturation field
is several kOe, indicative of strong in-plane anisotropy.
Hence, although strong PMA is well-known in Pt/Co/MOx
films,[6] in the present case of a Ta underlayer, Co-O bonding
alone is insufficient to generate out of plane magnetization.
It is generally observed that PMA in thin Co films is

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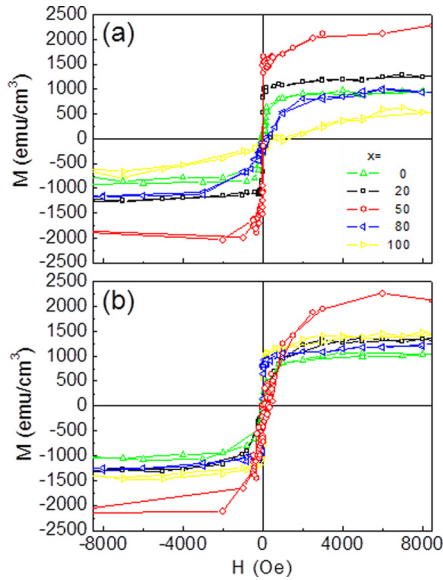gbeach@mit.edu.

FIG. 1. (a) Out-of-plane and (b) in-plane magnetization curves for 0.8 nm CoFe/MgO films with several Co compositions ($x = 0$, 20, 50, 80, and 100).

FIG. 2. Dependences of magnetic properties of 0.8 nm CoFe/MgO films on Co composition ($x = 0$, 20, 50, 80, and 100). (a) Coercive field (H_c) and saturation magnetization (M_s). Inset: Detailed Kerr hysteresis loop corresponding to Ta/0.8 nm $Co_{50}Fe_{50}$/MgO. (b) Magnetic anisotropy field (H_k) and anisotropy energy (K_u). The dashed lines separate OP and IP magnetized regimes with respect to the Co composition.

associated with strong fcc(111) texture, which can be achieved in Co grown on Pt, but not on Ta, suggesting that both microstructure and interfacial bonding chemistry are important in establishing PMA in these films.

The magnetic anisotropy energy, K_u, is given by $K_u = M_s H_k / 2$ with $H_k = H_{sat} + 4\pi M_s$, where M_s is the saturation magnetization, H_k is the magnetic anisotropy field, and H_{sat} is the hard-axis saturation field. The dependence of K_u, H_k, M_s, and coercive field H_c on alloy composition for the Ta/0.8 nm CoFe/MgO films is shown in Fig. 2. Note that for the H_k and K_u data in Fig. 2(b), negative values correspond to in-plane magnetized (IP) regimes, while positive values correspond to out-of-plane magnetized (OP) regimes. Both H_c and M_s increase up to $x = 50$ and go through their maximum values of 14 Oe and 1889 emu/cm^3 at $x = 50\%$, respectively, in Fig. 2(a). They then decrease as the Co composition increases beyond 50%. It is notable that the coercivity in all of these films is less than 15 Oe, which would be beneficial for achieving a low critical current density for current-driven DW motion. Similarly, the H_k and K_u have their maximum values of 25 kOe and 2.4×10^{-7} erg/cm^3 at $x = 50\%$, respectively, in Fig. 2(b). These data indicate that the alloy composition plays an important role in generating interfacial PMA in the CoFe/MgO system.

To distinguish the volume from interfacial contributions to PMA in the Ta/CoFe/MgO films, the thickness (t)-dependence of magnetic anisotropy, $K_u = K_v - 2\pi M_s^2 + K_i/t$ was measured, where K_v and K_i are the volume anisotropy and interfacial anisotropy, respectively. Figure 3 shows $K_u * t$ versus t for $x = 0$, 20, 50, 80, and 100. Out of plane magnetization is absent in Ta/Fe/MgO and Ta/Co/MgO, for all t, but a t-dependent crossover between in-plane and out-of-plane anisotropy is found for $x = 20$, 50, and 80. In particular, even though PMA has been theoretically predicted at the Fe/MgO interface[10,11] due to the hybridization of Fe 3d orbitals and O 2p orbitals, we do not observe PMA in Ta/Fe/MgO even
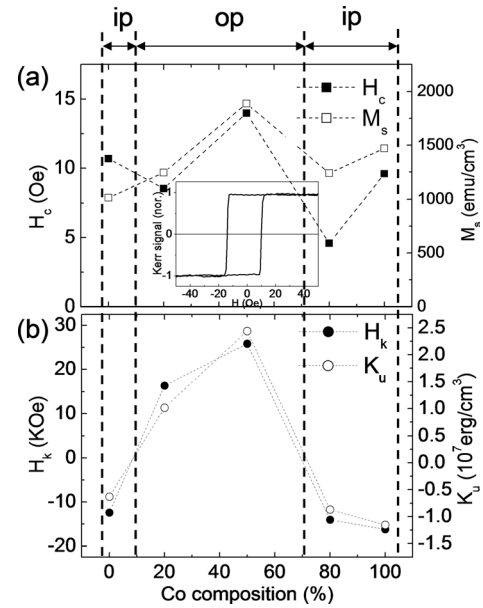
down to $t = 0.5$ nm. We believe the reason is that the effect of underlayer on the out-of-plane anisotropy at Fe/MgO interface is not considered in the theoretical result.[10,11] From the intercepts by extrapolation, K_i for the intermediate Co-Fe compositions is roughly estimated to be 6.4 erg/cm^2 for $x = 20$, 8.2 erg/cm^2 for $x = 50$, and 3.2 erg/cm^2 for $x = 80$, respectively. The volume contribution is comparable to the demagnetization field $-2\pi M_s^2$ implying that K_v is insignificant. This suggests that the PMA of the films is primarily due to an interfacial contribution.

To better elucidate the origin of the crossover between IMA and PMA in the films with respect to the Co composition, we consider two possible mechanisms that are expected to exhibit strong compositional variation: orbital moment anisotropy and magnetostriction. Symmetry breaking at an interface[6,14,15] lifts the degeneracy of the 3d orbitals, leading to an energy splitting between those oriented in the plane ($3d_{xy}$, $3d_{x^2-y^2}$) and those with an out-of-plane character ($3d_{yz}$, $3d_{zx}$, and $3d_{z^2}$). From a crystalline field ascribed to this energy difference, the magnetic anisotropy energy K_u^e can be given by[14,15]

$$K_u^e = -\frac{G}{H}\frac{\xi}{4\mu_B}(m_{orb}^{op} - m_{orb}^{ip}), \qquad (1)$$

where the ratio G/H is a constant related to the band structure, ξ is the spin-orbit coupling parameter, μ_B is the Bohr magneton, and $m_{orb}^{op\,(ip)}$ is the out-of-plane (in-plane) orbital moment. Taking the values for the constants[14] in Eq. (1) to be $G \sim 10^{-3}$ eV, $H \sim 0.05$ eV, $\xi \sim 0.05$ eV, the composition dependence of K_u^e may be estimated using the dependence of $m_{orb}^{op\,(ip)}$ on the number of valence electrons as reported in Ref. 14. Fig. 4(a) shows that as the number of valence
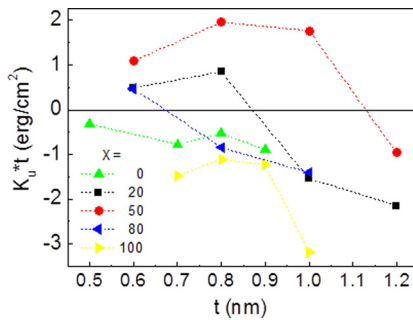
FIG. 3. Magnetic anisotropy energy (K_u) times the CoFe thickness (t) versus t for CoFe/MgO films with several Co compositions ($x = 0$, 20, 50, 80, and 100). The films with $K_u*t > 0$ are out-of-plane magnetized and those with $K_u*t < 0$ are in-plane magnetized.

electrons increases up to 9.0 per atom, corresponding to Co, m_{orb}^{op} and K_u^e go to their maximum values. They then decrease rapidly as the number of valence electrons is increased beyond 9.0. The relation between m_{orb}^{op} and K_u^e arises from splitting of the bands giving rise to $m_{orb}^{op\,(ip)}$ moments, such that with increasing valence electron density, one band is preferentially filled up to a band structure-dependent limit, at which the orbital anisotropy takes a maximum. This enhancement of m_{orb}^{op} due to the anisotropic orbital configuration leads to the PMA. Beyond this limit, the PMA strength is reduced due to a decrease in the anisotropy of the orbital configuration.[16]

We also note that the bulk magnetostriction constant λ_s of CoFe alloys depends strongly on Co content and takes a maximum at $x = 0.5$. In Fig. 4(b), we plot K_i and λ_s, extracted from Ref. 16, versus Co composition. As expected, K_i and λ_s show the same trend and have maximum values of 8.2 erg/cm^2 and 68×10^{-6} at $x = 50$, respectively. It is therefore possible that the film stress yields a magnetostrictive anisotropy that can augment the interfacial anisotropy leading to a strong composition dependence of the net PMA in this system. Because λ_s also arises from magnetic anisotropy related to anisotropic band filling, it is likely that the composition dependence of m_{orb}^{op} is of fundamental importance in the crossover behaviors between IMA and PMA as a function of Co composition.

We experimentally observed the crossover behavior of the magnetic anisotropy in Ta/CoFe/MgO films for alloy compositions spanning pure Co to pure Fe and discussed how composition affects the magnetic properties of the films. Interestingly, PMA is absent for Ta/Fe/MgO and Ta/Co/MgO for all thicknesses examined here, although it is theoretically predicted to be generated at the Fe/MgO and Co/MgO interface due to hybridization between Fe (or Co) 3d orbitals and O 2p orbitals,[10,11] In the pure-Co case, strong PMA is well-known to occur in Pt/Co/MOx films[6] so its absence in the case of a Ta underlayer suggests that Co-O bonding alone is insufficient to guarantee out of plane magnetization. It is generally observed that PMA in thin Co films is associated with strong fcc(111) texture, which is usually achieved in Co grown on Pt, but not on Ta, suggesting that both microstructure and interfacial bonding chemistry are
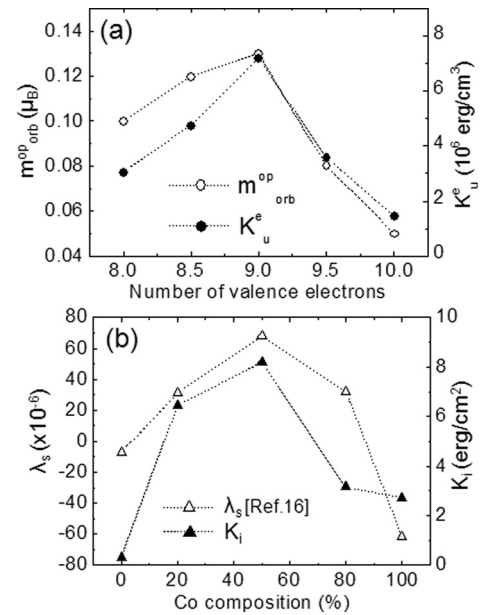

FIG. 4. (a) Dependences of out-of-plane orbital moment (m_{orb}^{op}) and estimated magnetic anisotropy energy (K_u) on the number of valence electrons. (b) Dependences of magnetostriction constant (λ_s) and interfacial anisotropy energy (K_i) on Co composition ($x = 0$, 20, 50, 80, and 100). λ_s values of CoFe alloys with several compositions ($x = 0$, 20, 50, 80, 100) have been culled from Ref. 16.

important in establishing PMA in these films. In the intermediate composition range, as the Co content increases, the degree of valence-band filling at the CoFe/MgO interfaces leads to distinct changes in the electronic structure of the valence band of the CoFe adjacent to the interface through hybridization between the 3d orbitals of Co and Fe the 2p orbitals of O. This composition dependent hybridization of 3d and 2p orbitals is expected to lead to a maximum of the PMA near $x = 50$, consistent with our experimental results, and is likely at the origin of the crossover behavior of the magnetic anisotropy as a function of composition.

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