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

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Interfacial magnetic anisotropy in Ta/Co_xFe_{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 Co₅₀Fe₅₀ 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 Institute of Physics. [<http://dx.doi.org/10.1063/1.4799779>]

There has been intensive research interest in Ta/CoFe(B)/MgO stacks with perpendicular magnetic anisotropy (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, symmetry 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 interface⁶⁻⁸ and the composition ratio of them which determines the band filling⁹ 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 hybridization of O 2p orbitals with Co 3d and Fe 3d orbitals.¹⁰⁻¹³ However, for the latter, systematic studies on the composition 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_xFe_{100-x}(CoFe)/MgO for alloy compositions spanning 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 SiO₂/5.0 nm Ta/(0.5–1.2) nm Co_xFe_{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 longitudinal 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 remanent magnetization vanished and in-plane magnetic anisotropy (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 exist¹⁰⁻¹² but it is insufficient to overcome the demagnetizing 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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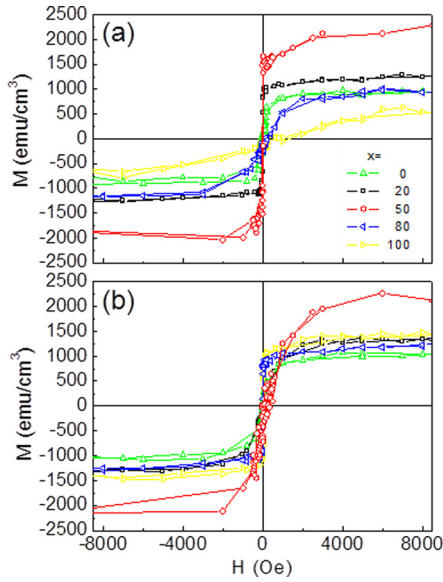


FIG. 1. (a) Out-of-plane and (b) in-plane magnetization curves for 0.8 nm CoFe/MgO films with several Co compositions ($x=0, 20, 50, 80,$ and 100).

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 $H_k = H_{\text{sat}} + 4\pi M_s$, 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 $2.4 \times 10^{-7} \text{ erg/cm}^3$ 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

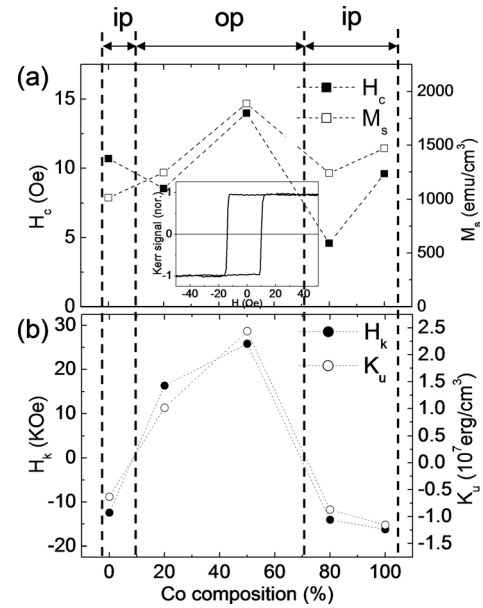


FIG. 2. Dependences of magnetic properties of 0.8 nm CoFe/MgO films on Co composition ($x=0, 20, 50, 80,$ and 100). (a) Coercive field (H_c) and saturation magnetization (M_s). Inset: Detailed Kerr hysteresis loop corresponding to Ta/0.8 nm Co₅₀Fe₅₀/MgO. (b) Magnetic anisotropy field (H_k) and anisotropy energy (K_u). The dashed lines separate OP and IP magnetized regimes with respect to the Co composition.

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^c can be given by^{14,15}

$$K_u^c = -\frac{G}{H} \frac{\zeta}{4\mu_B} (m_{\text{orb}}^{\text{op}} - m_{\text{orb}}^{\text{ip}}), \quad (1)$$

where the ratio G/H is a constant related to the band structure, ζ is the spin-orbit coupling parameter, μ_B is the Bohr magneton, and $m_{\text{orb}}^{\text{op(ip)}}$ is the out-of-plane (in-plane) orbital moment. Taking the values for the constants¹⁴ in Eq. (1) to be $G \sim 10^{-3} \text{ eV}$, $H \sim 0.05 \text{ eV}$, $\zeta \sim 0.05 \text{ eV}$, the composition dependence of K_u^c may be estimated using the dependence of $m_{\text{orb}}^{\text{op(ip)}}$ on the number of valence electrons as reported in Ref. 14. Fig. 4(a) shows that as the number of valence

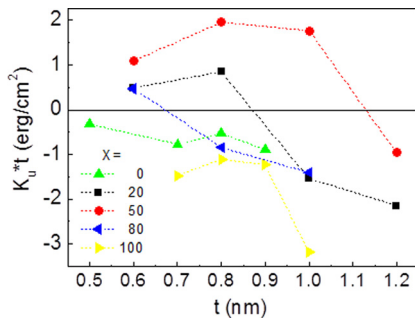


FIG. 3. Magnetic anisotropy energy (K_u) times the CoFe thickness (t) versus t for CoFe/MgO films with several Co compositions ($x=0, 20, 50, 80$, and 100). The films with $K_u^*t > 0$ are out-of-plane magnetized and those with $K_u^*t < 0$ are in-plane magnetized.

electrons increases up to 9.0 per atom, corresponding to Co, $m_{\text{orb}}^{\text{op}}$ 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 $m_{\text{orb}}^{\text{op}}$ and K_u^e arises from splitting of the bands giving rise to $m_{\text{orb}}^{\text{op(ip)}}$ 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 $m_{\text{orb}}^{\text{op}}$ 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 $m_{\text{orb}}^{\text{op}}$ 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 films⁶ 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

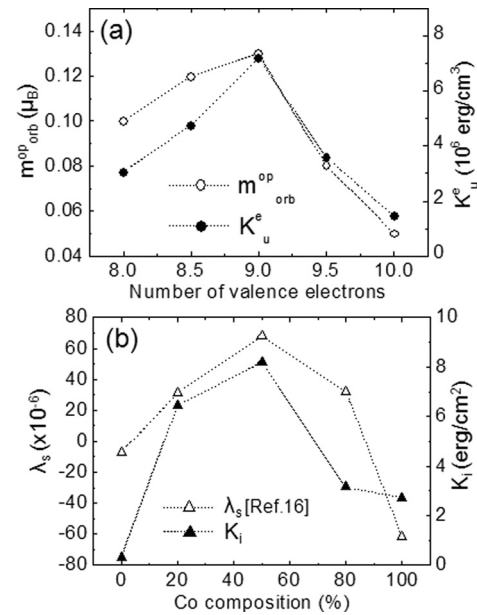


FIG. 4. (a) Dependences of out-of-plane orbital moment ($m_{\text{orb}}^{\text{op}}$) and estimated magnetic anisotropy energy (K_u) on the number of valence electrons. (b) Dependences of magnetostriction constant (λ_s) and interfacial anisotropy energy (K_i) on Co composition ($x=0, 20, 50, 80$, and 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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