

Composition Dependence of Perpendicular Magnetic Anisotropy in Ta/Co_xFe_{80-x}B₂₀/MgO/Ta (x=0, 10, 60) Multilayers

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The perpendicular magnetic anisotropy of sputtered CoFeB thin films covered by MgO was investigated by vibrating sample magnetometry. Three different Co_xFe_{80-x}B₂₀ alloys were studied. Under out-of plane magnetic field, the saturation field was found to increase with increasing the Co content. The magnetization and interface anisotropy energy were obtained for all samples. Both showed a marked dependence on the MgO overlayer thickness. In addition, their variations were found to be non-monotonous as a function of the Co concentration.

Keywords : perpendicular magnetic anisotropy, CoFeB/MgO, interface anisotropy, CoFe alloy

1. Introduction

Magnetic tunnel junctions (MTJs) with a crystalline MgO barrier show a large magnetoresistance and offer new possibilities for developing high performance memories and logic circuits. MTJs possessing ferromagnetic layer with a large perpendicular magnetic anisotropy (PMA) are expected to provide interesting features such as high thermal stability and low switching energy consumption [1]. Ultrathin CoFeB films in contact with MgO show a large PMA, which originates from the interface anisotropy, and have been recently used in MTJs [2]. Several studies tried to optimize the PMA and to clarify its origin by studying its dependence on the thickness of the magnetic layer [2-4], on the buffer layer [4], the oxidation condition at the interface [5-7] and the annealing temperature. We have also reported the MgO overlayer thickness dependence of the PMA, and found that the PMA strongly depends on the MgO overlayer thickness [8]. However, the variation of the PMA with the composition of CoFeB alloys [9] has not been fully investigated experimentally, and such a study may help us to understand the mechanism of PMA in CoFeB thin films.

In the present paper, we estimate the effective magnetic anisotropy energy and the interface anisotropy for three selected compositions of CoFeB. Because the MgO overlayer might be different when grown on different underlayers, the MgO overlayer thickness dependence of PMA was also studied for a precise comparison of those alloys.

2. Experiment

Samples with a structure Ta(5)/CoFeB(1.3)/MgO(0.7-6.0)/Ta(5) (nominal thicknesses in nanometers) were fabricated on thermally oxidized Si substrates using a Canon-ANELVA C-7100 ultra high vacuum magnetron sputtering. Figure 1 (a) depicts the stacking structures of the samples. The thickness of CoFeB was fixed at 1.3 nm for all samples. The selected compositions are Fe₈₀B₂₀, Co₁₀Fe₇₀B₂₀ and Co₆₀Fe₂₀B₂₀. After growth, the films were annealed at 300 °C for 1 h in ultra high vacuum. The magnetization (*M-H*) curves of the samples were measured by vibrating sample magnetometer (VSM) with perpendicular and in-plane magnetic fields.

3. Result and Discussion

Figure 1(b) shows the magnetization curves measured with out-of-plane magnetic field of the three targets, with 1.0 nm-thick MgO overlayers. The smallest saturation field

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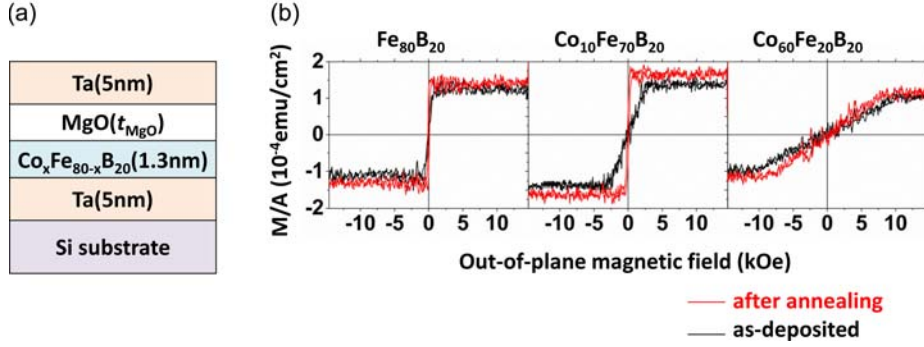


Fig 1. (Color online) (a) Schematic view of sample stacking structures. (b) Magnetization curves of samples with 1.0 nm MgO overlayer for the three alloys (black curves: as-deposited, red curves: annealed at 300 °C). The saturation field increases with increasing the Co concentration.

(H_s), was obtained with Fe₈₀B₂₀. The saturation field increases as a function of the Co concentration in the alloys. After annealing, H_s drastically decreases, indicating the enhancement of PMA. The annealed sample of Fe₈₀B₂₀ alloy has a spontaneous perpendicular magnetic easy axis with a full remanence.

The MgO overlayer thickness dependence was investigated for the three alloys. Figure 2 shows the values of (a) magnetizations per area (M/A), and (b) the product of PMA energy and thickness of magnetic layers ($K_{\text{eff}}t$) as a function of the MgO overlayer thickness. The PMA energy was estimated from the area enclosed between the magnetization loops measured with in-plane and out-of-plane magnetic fields. Positive (negative) $K_{\text{eff}}t$ indicates an out-of-plane (in-plane) easy axis of magnetization. In this figure, a clear reduction of the magnetization is observed in samples with thin MgO layers. These observations may be related to intermixing with the Ta underlayer (for all samples) and with the Ta capping layer (for samples with thin MgO). The presence of a magnetic dead layer due to Ta incorporation has already been reported [10]. In the whole range of MgO overlayer thicknesses, and for both as-deposited and annealed samples, the magnetization of Fe₈₀B₂₀ alloy was smaller than the one of Co₁₀Fe₇₀B₂₀ and larger than the one of Co₆₀Fe₂₀B₂₀ alloy, as expected from the Slater-Pauling behaviour [11]. The PMA energy depends more strongly on the MgO overlayer thickness, as shown in Fig. 2(b). In general, larger values of $K_{\text{eff}}t$ were obtained with thinner MgO. The perpendicular magnetic easy axis was observed in annealed Fe₈₀B₂₀ with thin MgO (0.7 nm to 1.5 nm).

The magnetic anisotropy energy density writes: $K_{\text{eff}} = K_B - 2\pi M_s^2 + K_I/t$ [12], with K_B the bulk crystalline anisotropy, $2\pi M_s^2$ the shape energy, K_I the interface anisotropy energy, and t the nominal thickness of CoFeB. Assuming a negligible K_B , we can estimate the interface

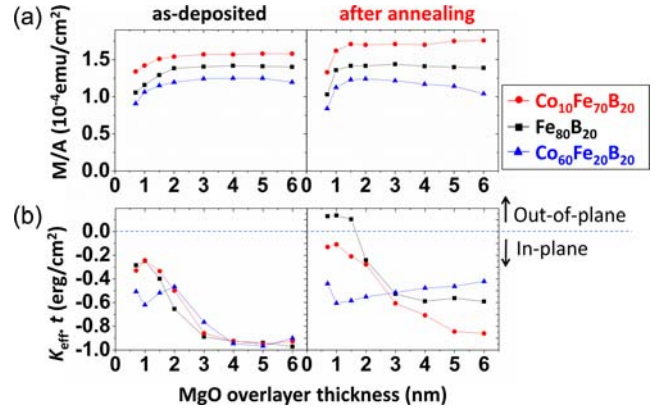


Fig 2. (Color online) MgO overlayer thickness dependence of as-deposited and annealed samples. (a) Magnetization per unit area (M/A). The M/A value decreases for samples with thin small MgO layers. (b) Product of the effective magnetic anisotropy energy (K_{eff}) and the thickness of the CoFeB layer (t).

anisotropy energy K_I by the following relation: $K_I = K_{\text{eff}}t + 2\pi M_s^2 t$. The MgO thickness dependence of K_I is plotted in Fig. 3. There is a peak in the interface anisotropy for all alloys. For Fe rich alloys (Fe₈₀B₂₀ and Co₁₀Fe₇₀B₂₀), the largest K_I was found in both as-deposited and annealed samples with a thin MgO overlayer (1.0 nm-1.5 nm). For the Co rich alloy, the peak appears at a MgO thickness of 2.0 nm and disappears upon annealing. This different behaviour may be related to a bcc-fcc phase transition which occurs in Co rich alloys (70-75% of Co) [11, 13].

The origin of the sensitiveness of interface anisotropy with MgO overlayer is still unclear. With very thin MgO (less than 1 nm), we believe that some Ta diffuses from the capping layer through MgO and reduces K_I [8]. Indeed, 1 nm thick MgO is expected to be amorphous, which possibly favour the Ta diffusion. Above this thickness, crystallized MgO is formed, preventing Ta diffusion. The

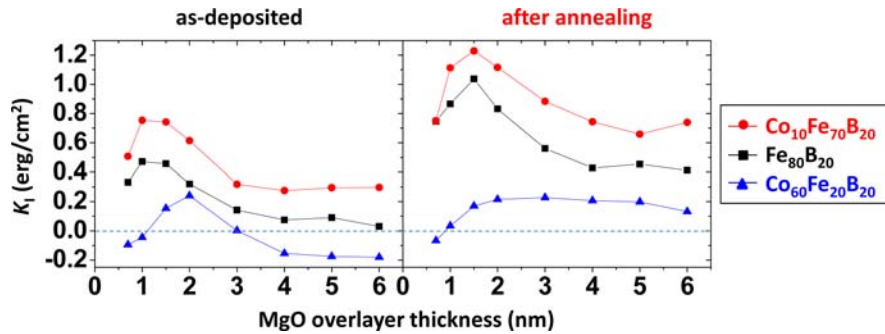


Fig 3. (Color online) MgO thickness dependence of the interface anisotropy energy (K_1) of as-deposited and annealed samples. There is a peak of interface anisotropy energy for each composition alloy.

MgO thickness dependence of K_I above 1 nm may be due to changes in the interfacial electronic structure when varying the MgO thickness [14, 15], changes in the interfacial oxidation [5-7] or stress-related effects. Further investigation is needed for clarifying the mechanism.

The above results show that the MgO overlayer thickness plays an important role for the interface anisotropy of CoFeB films. The magnetic properties are found to be very sensitive in the thin MgO overlayer region, as shown in Fig. 3. Note that, in this sensitive region, the influences of MgO overlayer are not the same for the different CoFeB alloys. Actually, for a given thickness, MgO layers can be different when grown on different underlayers. In contrast, the MgO thickness dependence is weaker in the thick MgO region. Therefore, in order to discuss the dependence of interface anisotropy on the alloys composition, we focus on samples with thick MgO (5.0 nm). The saturation magnetizations and the interface anisotropies of the three alloys are plotted in Fig. 4.

As mentioned above, the values of saturation magneti-

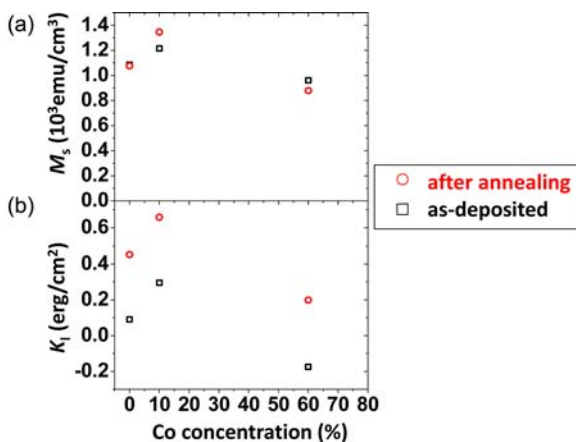


Fig 4. (Color online) Co concentration dependence of (a) saturation magnetization (M_s) and (b) interface anisotropy energy (K_1) of samples with 5.0 nm MgO overlayer.

zation are consistent with the well-known Slater-Pauling behaviour. Majority $3d$ states are gradually filled upon Co addition, which results in an increase of the magnetic moment. Further Co addition fills the minority $3d$ states, thereby reducing the magnetic moment. Interestingly, the interface anisotropy of CoFeB alloys shows a similar tendency. However, the anisotropy variations should originate from changes in the electronic structure at the interface rather than in the bulk. It is known that out-of-plane orbitals [p_z , d_{z^2} for the (001) surface] favour an in-plane easy axis [16]. According to a first-principle study, the hybridization of d_{z^2} orbitals with O $2p$ orbitals at the CoFe/MgO interface results in an enhanced interface anisotropy. This mechanism produces a larger anisotropy at the Fe/MgO interface than at the Co/MgO one [7]. In this picture, the detailed variation of anisotropy should be related to changes in the density of d_{z^2} states upon alloying, and could be indirectly related to the Slater-Pauling behaviour. Interface states may also have a strong influence [17].

4. Conclusion

We have studied the perpendicular magnetic anisotropy of Ta/Co_xFe_{80-x}B₂₀/MgO/Ta multilayers with three alloy compositions. A strong influence of the MgO overlayer thickness on the magnetic anisotropy was observed and shown to differ for the different CoFeB alloys. In general, the maximum interface anisotropy was obtained for MgO thicknesses between 1.0 nm to 2.0 nm. The magnetization and interface anisotropy showed similar tendencies as a function of the alloy composition (Co concentration), with a maximum in Co₁₀Fe₇₀B₂₀.

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References

- [1] T. Kishi, H. Yoda, T. Kai, T. Nagase, E. Kitagawa, M. Yoshikawa, K. Nishiyama, T. Daibou, M. Nagamine, M. Amano, S. Takahashi, M. Nakayama, N. Shimomura, H. Aikawa, S. Ikegawa, S. Yuasa, K. Yakushiji, H. Kubota, A. Fukushima, M. Oogane, T. Miyazaki, and K. Ando, IEEE International Electron Devices Meeting (IEDM) Technical Digest, 309 (2008).
- [2] S. Ikeda, K. Miura, H. Yamamoto, K. Mizunuma, H. D. Gan, M. Endo, S. Kanai, J. Hayakawa, F. Matsukura, and H. Ohno, *Nature Mater.* **9**, 721 (2010).
- [3] K. Lee, J. J. Sapan, S. H. Kang, and E. E. Fullerton, *J. Appl. Phys.* **109**, 123910 (2011).
- [4] D. C. Worledge, G. Hu, D. W. Abraham, J. Z. Sun, P. L. Trouilloud, J. Nowak, S. Brown, M. C. Gaidis, E. J. O'Sullivan, and R. P. Robertazzi, *Appl. Phys. Lett.* **98**, 022501 (2011).
- [5] L. E. Nistor, B. Todmacq, C. Ducruet, C. Portemont, I. L. Prejbeanu, and B. Dieny, *Magnetics*, IEEE Trans. Magn. **46**, 1412 (2010).
- [6] K. Nakamura, T. Akiyama, T. Ito, M. Weinert, and A. J. Freeman, *Phys. Rev. B* **81**, 220409(R) (2010).
- [7] H. X. Yang, M. Chshiev, B. Dieny, J. H. Lee, A. Manchon, and K. H. Shin, *Phys. Rev. B* **84**, 054401 (2011).
- [8] D. D. Lam, F. Bonell, S. Miwa, Y. Shiota, K. Yakushiji, H. Kubota, T. Nozaki, A. Fukushima, S. Yuasa, and Y. Suzuki, *J. Kor. Phys. Soc.* (to be published).
- [9] S. Yakata, H. Kubota, Y. Suzuki, K. Yakushiji, A. Fukushima, S. Yuasa, and K. Ando, *J. Appl. Phys.* **105**, 07D131 (2009).
- [10] S. Y. Jang, C. Y. You, S. H. Lim, and S. R. Lee, *J. Appl. Phys.* **109**, 013901 (2011).
- [11] B. D. Cullity and C. D. Graham, *Introduction to Magnetic Materials*, 2nd Ed. (2009) pp. 142-143.
- [12] M. T. Johnson, P. J. H. Bloemen, F. J. A. den Broeder, and J. J. de Vries, *Rep. Prog. Phys.* **59**, 1409 (1996).
- [13] Y. Ustinovshikov and B. Pushkarev, *J. Alloys Compd.* **424**, 145 (2006).
- [14] B. Ujfalussy, L. Szunyogh, P. Bruno, and P. Weiberger, *Phys. Rev. Lett.* **77**, 1805 (1996).
- [15] L. Zhong, M. Kim, and X. Wang, *J. Appl. Phys.* **79**, 5831 (1996).
- [16] D. Wang, R. Wu, and A. J. Freeman, *Phys. Rev. B* **74**, 14932 (1993).
- [17] F. Bonell, T. Hauet, S. Andrieu, F. Bertran, P. L. Fevre, L. Camels, A. Tejada, F. Montaigne, B. Warot-Fonrose, B. Belhadji, A. Nicolaou, and A. Taleb-Ibrahimi, *Phys. Rev. Lett.* **108**, 176602 (2012).