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ABSTRACT

The CoFeB thickness, $t$ dependence of the effective first- and second-order magnetic anisotropy, $K_1^{\text{eff}}$ and $K_2$, for MgO/(Co$_{1-x}$Fe$_x$)$_{80}$B$_{20}$/Ta films ($x=0.3-1.0$) is investigated. As Co$_{40}$Fe$_{40}$B$_{20}$ thickness decreases, $K_1^{\text{eff}}$ increases and shows a perpendicular magnetic anisotropy for $t=1.2$ nm. On the other hand, in-plane magnetic anisotropy is observed for $t \geq 1.4$ nm. Also, a 1.3-nm-thick CoFeB sample demonstrates an easy-cone behavior, which suggests that the magnitude of $K_1^{\text{eff}}$ and $K_2$ becomes comparable. By plotting the product of $K_2$ and $t-d$ as a function of $t-d$, where $d$ is a magnetic dead layer thickness, linear dependences with negative y-axis intercepts are displayed for all ranges of $x$. The extracted interfacial $K_2$, $K_i^{(2)}$ are varied depending on the compositions in the range of $-0.024$ to $-0.042$ erg/cm$^2$ for $x=100\%$ and 30, 50%, respectively. A magnetic phase diagram summarizing the results of $K_1^{\text{eff}}-2\pi M_s^2$ and $K_2$ suggests that the ratio of $K_2$ against $K_1^{\text{eff}}-2\pi M_s^2$ is varied depending on the compositions. These results give us a guideline to achieve the desired magnetic properties of CoFeB for spintronic applications.

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I. INTRODUCTION

Magnetic tunnel junctions (MTJs) using CoFeB/MgO/CoFeB are a building block for spintronic devices due to their large tunnel magnetoresistance (TMR) ratios up to 604% at room temperature, which enables MTJs to be used in practical applications such as magnetoresistive random access memory (MRAM) and magnetic field sensor devices. A conventional method to control these devices’ performances is mainly the use of the first-order uniaxial magnetic anisotropy, $K_1$ of the CoFeB thin film in such MTJs. Since an interfacial magnetic anisotropy is mostly generated on the interface of Fe/MgO by orbital hybridization, a manipulation of the $K_1$ in CoFeB has been established by means of thickness, compositions, and voltage controls and contribute to the development of low-power-consumption MRAM and high-sensitive magnetic sensor devices. On the other hand, recent studies have proposed the use of the second-order uniaxial magnetic anisotropy, $K_2$ which remarkably benefits spintronic devices. For MRAM applications, if a free layer of MTJ satisfies the conditions of $K_1-2\pi M_s^2<0$ and $K_2>-1/2(K_1-2\pi M_s^2)$, an easy-cone state can be observed due to the magnetization canting and notably, this state can decrease the switching current density for a writing scheme using
Spin-transfer torque. Moreover, for magnetic sensor devices, due to industrial development, electric vehicles receive much attention and linear magnetic field sensors are desired for current monitoring systems. An improved linear conductance output of MTJ has been demonstrated by reducing the $K^2$ of the free layer, and complete linear conductance has been predicted when $K^2=0$. For these purposes, as both large and small $K^2$ are significantly profitable for spintronic devices, a controlling method of $K^2$ should be established. Referring to the $K^2$ study for other ferromagnets, an evident linear relationship between $K^2$ against $1/t$ has been observed in a CoFe/Ni multilayer, where $t$ is the thickness of the CoFe/Ni layer and the magnitude of interfacial $K^2$ was found to be one order smaller than that of $K_1$. Additionally, a sizable voltage controlled magnetic anisotropy (VCMA) effect has been found in the $K^2$ of epitaxial Cr/Fe/MgO films. These results may ensure the existence of an interfacial contribution in $K^2$ as is the case with well-defined $K_1$. Similarly, the $K^2$ of Ta/CoFeB/MgO films have been investigated. Although it is implied that there may be an interfacial effect even for $K^2$ because it follows the $1/t$ trend, the data lacks a quantitative evaluation of the interfacial contribution. Hence, in this paper, we studied the thickness dependence of $K^2$ for MgO/CoFeB/Ta films with different CoFeB compositions and report the composition dependence of interfacial magnetic anisotropy of $K^2$.

II. EXPERIMENTAL METHOD

All the films with the stacking of Ta(3)/MgO(2)/(Co$_{1-x}$Fe$_x$)$_{80}$B$_{20}$($t$)/Ta(5) (thickness in nm) were prepared on Si/SiO$_2$ substrates at room temperature where $x$ and $t$ are the Fe concentration of CoFe from 30 to 100% and the nominal thickness of CoFeB determined by the sputtering rates, respectively. All metallic and oxide layers were deposited by DC and RF sputtering, respectively, with a base pressure less than $1.0\times10^{-6}$ Pa. After the post annealing of the samples at 300°C without an external magnetic field in a vacuum furnace, all measurements were carried out at room temperature as follows. The magnetization curves were measured by using a superconducting quantum interference device (SQUID) magnetometer to evaluate the saturation magnetization. Angular dependent ferromagnetic resonance (FMR) was performed to obtain the magnetic parameters such as the effective and second-order magnetic anisotropy field, $H_{k}^{\text{eff}}$ and $H_{k}^{2}$, respectively, where a 9.4-GHz microwave frequency was applied and resonance spectra were lock-in detected. The surface morphology was also scanned by using atomic force microscopy (AFM).

III. RESULTS AND DISCUSSION

Figure 1(a) is a plot of areal magnetic moment density, $m$ as a function of the nominal thickness, $t$ for each composition. As can be seen, $m$ increases linearly against $t$ because of the relationship of $m=M_s(t-t_d)$, where $M_s$ and $t_d$ are the saturation magnetization and magnetic dead layer thickness, respectively. The slope and $x$-axis-intercept for this line correspond to $M_s$ and $t_d$, respectively, and their composition dependancies are summarized in Figs. 1 (b) and (c). In Fig. 1(b), $M_s$ roughly follows the Slater-Pauling curve, where we expect that such a large magnetization comparable to the bulk value might be a result of the crystallization by boron.
diffusion during the annealing process. For a MgO/CoFeB/Ta structure, boron moves to the Ta capping layer and CoFeB crystallizes with bcc structure by using a MgO template.\(^{15}\) Figure 1(c) shows that the magnetic dead layer thickness is in the range of 0.2 to 0.4 nm for all compositions, which agrees well with the previous work of MgO/CoFeB/Ta stacks\(^ {20,22}\) where the dead layer is believed to be produced by Ta migration during the sputtering process.\(^ {23}\) Thus, the magnetic properties of our thin films are in good agreements with previous studies.

Figure 2 shows the typical angular dependence of resonance field, \(H_{\text{res}}\) for \(x=50\%\) measured by the FMR technique. The magnetic field angle, \(\theta_H\) is measured from the film normal. As the CoFeB thickness decreases, the \(H_{\text{res}}\) of 0 deg. decreases and that of 90 deg. increases, in which we can expect that the magnetic anisotropy changes gradually depending on the CoFeB thickness. Since the angle of the smallest or largest \(H_{\text{res}}\) corresponds to the easy or hard axis of the ferromagnet, respectively, a clear behavior of perpendicular magnetic anisotropy (PMA) is seen for \(t=1.2\) nm. On the other hand, that of in-plane magnetic anisotropy is seen for \(t\geq1.4\) nm due to interfacial anisotropy. The inset of Fig. 2 shows an enlarged view for the 1.3-nm-thick CoFeB on a wider scale from −60 to 180 deg., where we can see the two- and four-fold symmetry with the comparable amplitude, which means that the effect of \(K_2\) is not negligible. Also, the minimum of \(H_{\text{res}}\) does not exist at 0 and 90 deg. but at ca. 47 deg., suggesting that it exhibits an easy-cone behavior.\(^ {10,24}\) Thus, we confirmed the presence of a second-order magnetic anisotropy in our film. The lines of Fig. 2 are the fitting to the experiment using the same method of our previous study.\(^ {14,25}\) As a result of the fitting, the magnetic parameters of \(H_{\text{K}1\text{eff}}\) (=2\(K_1/M_s\) + 4\(K_2/M_s\) - 4\(\pi M_s\)) and \(H_{K2}\) (=4\(K_2/M_s\)) are obtained, for example, a 1.2-nm-thick Co\(_{40}\)Fe\(_{40}\)B\(_{20}\) sample shows the best-fitting parameters of \(H_{\text{K}1\text{eff}}=1759\) Oe and \(H_{\text{K}2}=473\) Oe.

On the basis of the values of \(H_{K1\text{eff}}\) and \(H_{K2}\) determined by FMR analysis, the effective and second-order magnetic anisotropy of \(K_{1\text{eff}}\) and \(K_2\) are evaluated by using \(H_{K1\text{eff}}m/(2(t-t_d))\) and \(H_{K2}m/(4(t-t_d))\), respectively. Figure 3 (a) shows the product of \(K_{1\text{eff}}\) and \(t-t_d\) as a function of \(t-t_d\) for each composition. (b), (c) Extracted \(K_{1\text{eff}}\) and \(K_{2}\) plotted against Fe concentration, respectively. (d) The product of \(K_2\) and \(t-t_d\) as a function of \(t-t_d\) for each composition. (a), (f) Extracted \(K_{1\text{eff}}\) and \(K_{2}\) plotted against Fe concentration, respectively. The points and lines for (a) and (d) are experimental and linear fitting, respectively.

![Figure 3](image-url)

**Figure 3.** (a) The product of \(K_{1\text{eff}}\) and \(t-t_d\) as a function of \(t-t_d\) for each composition. (b), (c) Extracted \(K_{1}\) and \(K_{2}\) plotted against Fe concentration, respectively. (d) The product of \(K_2\) and \(t-t_d\) as a function of \(t-t_d\) for each composition. (a), (f) Extracted \(K_{1\text{eff}}\) and \(K_{2}\) plotted against Fe concentration, respectively. The points and lines for (a) and (d) are experimental and linear fitting, respectively.
as a function of $t-t_d$, where $t-t_d$ is coordinated with the magnetically active thickness. To separate the interfacial and bulk contribution for $K_1^{eff}$, we fitted the experimental data by using Eq. (1)

$$K_1^{eff}(t-t_d) = (K_b^{(1)} - 2\pi M_s^2) (t-t_d) + K_i^{(1)}$$  \hspace{1cm} (1)

Here, $K_i^{(1)}$ and $K_b^{(1)}$ are the first-order interfacial magnetic anisotropy and first-order bulk magnetic anisotropy, respectively. Note that positive $K_i^{(1)}$ and $K_b^{(1)}$ represent PMA. We can see a good correspondence for the linear fitting to the experimental data from Fig. 3 (a), In Figs. 3 (b) and (c), the extracted $K_i^{(1)}$ and $K_b^{(1)}$ are plotted against $x$. The composition dependence of $K_i^{(1)}$ shows a gradual increase as $x$ increases to a maximum of 1.86 erg/cm$^2$ at $x=75\%$ and decreases to 1.56 erg/cm$^2$ at $x=100\%$, although $K_b^{(1)}$ is dominantly generated on the Fe/MgO interface rather than the Co/MgO interface from the first principle calculation. The surface roughness of $R_s$ is almost constant against the thickness with an average $R_s$ of 0.26 and 0.31 nm for $x=75\%$ and 100\%, respectively (not shown). Thus, the films of $x=75\%$ possess a flat surface than those of $x=100\%$, which might increase $K_i^{(1)}$ in $x=75\%$ compared with that in $x=100\%$. Figure 3 (c) shows that $K_b^{(1)}$ is in the order of 10$^6$ erg/cm$^2$, which is not negligible. We infer that such a large $K_b^{(1)}$ is not due to the magnetostriction effect since its coefficient is positive for almost all ranges of $x$, suggesting that an in-plane tensile strain from MgO may cause a negative $K_b^{(1)}$. On the other hand, from the view of thermochemistry, the formation energy of the Fe-Ta bond shows a largely negative value. Hence, this implies that the large $K_b^{(1)}$ for our sample may be a result of the bond orientation anisotropy. Next, we focus on the results of the second-order magnetic anisotropy. Figure 3 (d) shows the product of $K_i$ and $t-t_d$, where a linear relationship with a finite y-axis-intercept is clearly observed, which implies the presence of an interfacial effect even for $K_i$. As discussed above, the interfacial and bulk second-order magnetic anisotropy, $K_i^{(2)}$ and $K_b^{(2)}$, are extracted by using Eq. (2)

$$K_i(t-t_d) = K_b^{(2)} (t-t_d) + K_i^{(2)}$$  \hspace{1cm} (2)

Figures 3(e) and (f) are the summary of the extracted $K_i^{(2)}$ and $K_b^{(2)}$ as a function of $x$, respectively. In contrast to the positive $K_i^{(1)}$, the $K_i^{(2)}$ are negative for all ranges of $x$, suggesting that $K_2$ favors in-plane anisotropy on the interface. The magnitude of $K_i^{(2)}$ changes to almost double from $-0.024$ to $-0.042$ erg/cm$^2$ from $x=75\%$ to $x=30\%$–$50\%$. Although we believe that the origin of such composition dependence could involve the difference of atomic interfacial energy and/or lattice distortion, further investigations using the first principle calculation are required. Figure 3 (f) is the composition dependence of $K_b^{(2)}$, whose values are positive in the order of 10$^5$ erg/cm$^2$ in all ranges of $x$ and their magnitudes are smaller by roughly one order of magnitude compared with $K_i^{(1)}$. Also, the approximately opposite composition dependence of $K_b^{(1)}$ and $K_i^{(2)}$ can be seen from Fig. 3 (c) and Fig. 3 (f). Although the reason is not clear at present, we speculate that $K_b^{(2)}$ could be linked with the observed large $K_b^{(1)}$.

Figure 4 shows the magnetic phase diagram for MgO/CoFeB/Ta films with different compositions. We consider the magnetic anisotropy energy of $E$ under zero magnetic field as Eq. (3).

$$E = (K_1 - 2\pi M_s^2)\sin^2\theta + K_2\sin^4\theta$$  \hspace{1cm} (3)

By minimizing Eq. (3), the magnetic phase is categorized into four states such as in-plane, out-of-plane, easy-cone state, and metastable phase depending on the values of $K_1-2\pi M_s^2$ and $K_2^{eff}$. When $K_1-2\pi M_s^2 > 0$ and $K_2^{eff} > 1/2(\sqrt{K_1-2\pi M_s^2}/2\pi M_s^2)^2$ (green region), the magnetization direction is canted, and the minimum energy is shown at the angle of $\theta = \sin^{-1}(1 - 2\pi M_s^2/(K_1-2\pi M_s^2))^{1/2}$. On the other hand, when $K_1-2\pi M_s^2 < 0$ and $K_2^{eff} < 1/2(\sqrt{K_1-2\pi M_s^2}/2\pi M_s^2)$ (white region), the local minima appear at both $\theta=0$ and 90 deg., resulting in the metastable phase. Although the 1.3-nm-thick Co$_{90}$Fe$_{18}$B$_2$ film is approximately on the boundary of easy-cone state and out-of-plane, an apparent easy-cone behavior is demonstrated as discussed in Fig. 2. According to previous work, even if the condition of an easy-cone state is not satisfied accurately, an easy-cone behavior has been observed in the neighboring region, similarly to our case. The lines in Fig. 4 are the fitting using Eqs. (1) and (2) for each composition. Remarkably, the relationship of $K_1-2\pi M_s^2$ and $K_2$ varies depending on the compositions. For example, larger and smaller $K_2$ compared with $K_1-2\pi M_s^2$ are observed in $x=50\%$ and $x=30\%$, respectively. Previously, a clear easy-cone state has not been observed at room temperature in Co$_{90}$Fe$_{18}$B$_2$, which can be interpreted by our magnetic phase diagram showing a narrow region of easy-cone state for $x=75\%$. In this regard, $x=50\%$ is more advantageous for stabilizing an easy-cone state at room temperature for MRAM applications. On the other hand, a small $K_2$ of $x=30\%$ and 100\% is useful for magnetic sensor devices to achieve highly linear output. Thus, this magnetic phase diagram gives us a guideline...
for designing magnetic anisotropy for any device applications with CoFeB/MgO/CoFeB-MTJs.

IV. CONCLUSION

We studied the composition dependence of the first and second-order interfacial magnetic anisotropy for MgO/(Co1-xFe)x/Co2B/Fe3B/Ta films (x=0.3-1.0). On the basis of the FMR measurements, an easy-cone behavior is demonstrated for a 1.3-nm-thick CoFeBMt sample due to the comparable amplitude of $K_1^{(2)}$ and $K_2$ with a certain condition. The magnitude of extracted $K_1^{(2)}$ depends on the composition, exhibiting a maximum of $-0.024 \text{ erg/cm}^2$ for $x=100\%$ and a minimum of $-0.042 \text{ erg/cm}^2$ for $x=30$ and 50. Also, the magnetic phase diagram is created using experimentally-determined magnetic constants in which the ratio of $K_2$ against $K_1^{(2)}$ can be modified, for example, the ratio is large at $x=50\%$ and small at $x=30$ and 100%. These results will be beneficial for developing studies and interpretations of $K_2$ and enable device development.

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