Effects of annealing on the magnetic properties and microstructures of Ta/Mo/CoFeB/MgO/Ta films

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ABSTRACT: We report a study on the evolution of perpendicular magnetic anisotropy (PMA) in Ta/Mo/CoFeB/MgO/Ta junctions annealed at 350°C for different periods of time. The PMA quickly developed after a short annealing time followed by a slow approach to saturation. This evolution of PMA was accompanied by structural changes. The surface roughness (R_q) gradually increased with increasing annealing time to 30 min; further increases in annealing time resulted in a dramatic increase in R_q . The effects of film composition and chemical state on magnetic anisotropy in the as-deposited Ta/Mo/CoFeB/MgO/Ta films and annealed films were studied. The X-ray photoelectron spectroscopy results demonstrated that more Fe oxides were formed at the CoFeB/MgO interface after 1 min of annealing compared to in the as-deposited sample, which is critical to obtain the PMA. High-resolution transmission electron microscopy analysis revealed that the CoFeB and MgO layers are predominantly amorphous in sample as-deposited

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and after 1min annealing. However, the CoFeB and MgO layers are crystallized in sample after 30 and 60 min annealing, which is beneficial to get high PMA.

KEYWORDS: annealing, perpendicular magnetic anisotropy (PMA), X-ray photoelectron spectroscopy (XPS), High-resolution transmission electron microscopy (HRTEM), oxygen migration

1. INTRODUCTION

Magnetic tunnel junctions (MTJs) have been extensively studied owing to their wide range of applications in spintronic devices. Different materials have been used as ferromagnetic electrodes in MTJs; among these materials, CoFeB is the most commonly used in spintronic devices due to its high tunneling magnetoresistance ratio, fast speed, low damping constant, and low power consumption¹⁻⁶. In all types of MTJs based on MgO and CoFeB, thermal annealing after film deposition is essential to achieve a high tunnel magnetic resistance (TMR)⁷. Normally, CoFeB/MgO/CoFeB junctions are annealed for time periods ranging from tens of minutes to hours at temperatures between 340°C and 500°C^{8,9,10}. However, both TMR and perpendicular magnetic anisotropy (PMA) are subject to deterioration after long periods of annealing due to the diffusion of Ta, B, and other elements^{11,12}.

In MTJs, short annealing times show promise for achieving desirable characteristics. For example, high TMR was obtained by annealing at 380°C for shorter periods of time¹³. After a short period of thermal annealing, a large PMA was achieved in CoFeB along with a high thermal stability factor, while the Gilbert damping constant was simultaneously reduced to only 0.011¹⁴. Using extended x-ray-absorption fine structure (EXAFS) analysis, another work reported

the B depletion and crystallization of the CoFeB layers within a few seconds after the post-growth high-temperature annealing of a CoFeB/MgO/CoFeB film¹⁵.

PMA is critical to obtain a high TMR. Therefore, it is imperative to investigate the evolution of PMA in perpendicular MTJs. PMA is thought to originate from the hybridization of Fe (Co) and O orbitals at the CoFeB/MgO interface¹⁶, which is known to experience significant changes upon annealing¹⁷. Therefore, it is of great interest to investigate interfacial phenomena to determine the optimal annealing time for maximizing PMA and minimizing inter-diffusion. In our studies on Ta/Mo/CoFeB/MgO/Ta multilayers, we found that PMA developed after a short period of annealing and investigated the effects of the composition, interfacial chemical state, and crystallization on the magnetic properties of Ta/Mo/CoFeB/MgO/Ta films.

2. EXPERIMENTAL SECTION

Ta/Mo/CoFeB/MgO/Ta films were deposited on thermally oxidized Si/SiO₂ substrates via magnetron sputtering with a base pressure of 3.0×10^{-8} Torr and an Ar gas (99.99%) sputtering pressure of 3 mTorr. The substrates were rotated at 40 rpm during deposition to insure uniform film thickness. After deposition, the samples were annealed in a vacuum furnace under a pressure of 4×10^{-7} Torr. The magnetic properties were measured using a vibrating sample magnetometer (VSM), and the anomalous Hall effect (AHE) resistance was determined using a standard four-point technique with a test current of 1 mA at room temperature. The magnetic properties of the system were evaluated based on the AHE using a standard four-point technique, which has been widely used to characterize perpendicular ferromagnetic films^{18,19}. The elemental chemical states were investigated by X-ray photoelectron spectroscopy (XPS; ESCALAB 250Xi). The chamber vacuum before XPS analysis was better than 3×10^{-9} Torr. All binding energies were adjusted using the C 1s line at 284.6 eV as a reference to eliminate the effect of surface

charging. The roughness was measured by white light interferometry (WLI). The vertical uncertainty depends mainly on the roughness of the measured surface. For smooth surfaces, the accuracy of the measurement is limited by the accuracy of the positioning stage. The lateral positions of the height values depend on the corresponding object point that is imaged by the pixel matrix. These lateral coordinates, together with the corresponding vertical coordinates, describe the surface topography of the object. Recent developments in both instrumentation and in measurement software for WLI have increased the vertical (i.e. height) resolution of these instruments to give a capability of better than 0.01 nm, which makes it a practical tool for measuring surface roughness in many applications. The microstructures were characterized by high-resolution transmission electron microscopy (HRTEM; Titan Themis 200). Films with structures of Ta (5 nm)/Mo(1 nm)/CoFeB(0.9 nm)/MgO(2 nm)/Ta(2 nm) were deposited and annealed at 350°C for different periods of time (0.5, 1, 30, 60, and 90 min). The nominal layer thicknesses of Mo must be less than 1 nm because of the inter-mixture of Ta/Mo and Mo/CoFeB layers.

3. RESULTS AND DISCUSSION

Fig.1 shows the AHE response of the as-deposited samples and the samples after annealing for 1, 30, 60, and 90 min. The as-deposited sample (black curve) exhibited in-plane magnetic anisotropy. In contrast, the samples annealed at 350°C for 1, 30, 60, and 90 min exhibited an out-of-plane magnetic anisotropy. The hysteresis loops of the samples measured along the in-plane and out-of-plane directions are shown in Fig. 2. Figs. 2a–f show the perpendicular and in-plane M–H curves of the as-deposited sample and the samples annealed at 350°C for 0.5, 1, 30, 60, and

90 min, respectively. The as-deposited sample exhibited in-plane magnetic anisotropy (IMA). However, after annealing at 350°C for 0.5 min, the easy axis transitioned from the in-plane direction to the out-of-plane direction. After annealing at 350°C for 30 min, the out-of-plane magnetic hysteresis loops exhibited high remanence and good squareness, indicating that the easily magnetized axis was perpendicular to the plane of the film. The sample remained perpendicularly magnetized after annealing for longer periods of time. Annealing at a moderate temperature can rearrange elements at interfaces, resulting in the removal of disorder generated during sputtering growth, which is advantageous to obtain PMA.

To investigate the effect of annealing time on magnetic anisotropy in the Ta/Mo/CoFeB/MgO/Ta films, we determined the effective magnetic anisotropy energy (K_{eff}) and the coercivity (H_c) for each sample. The effective magnetic anisotropy energy (K_{eff}) was determined for each sample by using the following expression^{20,21,22}:

$$K_{eff} = H_k \times M_s / 2$$

where M_s is the saturation magnetization and H_k is the effective perpendicular magnetic anisotropy field along the hard axis.

Fig.3 summarizes the dependences of H_c and K_{eff} on annealing time for each sample. As shown in Fig. 3a, H_c increased with annealing time and reached a maximum at 90 min of annealing. The H_c values for the samples annealed for 1 and 90 min were 24 and 34.5 Oe, respectively. Fig. 3b shows that K_{eff} increased dramatically with increasing annealing time and reached a maximum at an annealing time of 30 min; upon further increases in annealing time, decreased gradually. Although the samples annealed for 0.5 and 1 min exhibited PMA, their values were only 1.8 or 2.4 Merg/cm³, respectively. However, compared to the sample annealed

at 1 min, the value nearly doubled when annealing time was increased beyond 30 min. To be integrated into complementary metal-oxide-semiconductor transistor (CMOS) logic circuits, MTJs must exhibit high annealing stability at temperatures ranging from 350°C–400°C³. In this study, the values of Ta(5 nm)/Mo(1 nm)/CoFeB(0.9 nm)/MgO(2 nm)/Ta(2 nm) ranged from 1.8 to 5.0 Merg/cm³ over the tested range of annealing times, while H_c changed from 24.0 to 34.5 Oe. These values satisfy the needs of industry in terms of annealing temperature along with the magnetic random access memory (MRAM) requirements in terms of high intensity.

In order to clarify the effects of annealing time on PMA, we investigated the properties of the MgO barrier layer and the CoFeB/MgO interface using WLI, XPS, and HRTEM. We first examined the surface morphologies of the samples annealed for different times using WLI. Fig. 4 shows the dependence of the root-mean square of surface roughness (R_q) on annealing time based on the WLI measurements. R_q increased gradually with annealing time increased to 30 min. Upon further increases in annealing time, R_q increased dramatically. As shown in Fig.4, the relationship between surface roughness and the annealing time were non-monotonic.

XPS is one of the most powerful tools for the investigation of chemical bonding and oxidation states. We carefully etched the as-deposited sample and the sample annealed for 1 min using a time step between 0 and 75 s. Based on the Ta, Mg, Co, Fe, and Mo XPS spectra (not shown), we determined that the chemical information of the CoFeB/MgO interface and the bottom Ta layer could be revealed after Ar ion etching for 15 and 60 s, respectively.

Fig.5 shows Ta 4*f*, Fe 3*p*, and Mo 3*d* XPS spectra for samples. Figs. 5a, b, and c show the Ta 4*f* XPS spectra as-deposited (black color) and after 1 min annealing (red color) after etching for 45, 60, and 75 s, respectively. Metal thin films vacuum deposited on SiO₂ substrates made

from thermally oxidized Si wafers may react with SiO₂. The change in Gibbs free energy of this reaction is negative (i.e., the reaction is thermodynamically favorable). In this study, the atoms that were sputtered off from the target and arrived at the substrate were provided with a kinetic energy of approximately several to tens of eV. Therefore, the reaction is dynamically possible. When a Ta thin film was vacuum deposited on an oxidized silicon wafer, Ta₂O₅ was formed at the Ta/SiO₂ interface²³. The reaction resulted in an intermixing layer of Ta/Ta₂O₅ at the Ta/SiO₂ interface. The XPS spectrum of Ta exhibited a strong spin-orbit splitting with the metallic Ta and Ta-oxide peaks. Based on the XPS handbook²⁴, peak at 21.50 eV is a metallic Ta 4*f*_{7/2} peak, peak at 23.10 eV is its 4*f*_{5/2} peak, peak at 26.20 eV is a 4*f*_{7/2} peak of Ta⁵⁺, and peak at 28.10 eV is its 4*f*_{5/2} peak. In the spectrum of the film etched for 45 s (Fig. 5a), only small Ta₂O₅ signals could be distinguished, and strong Ta peaks were present. Increasing the etching time greatly enhanced the Ta⁵⁺ peak and weakened the Ta peak. At an etching time of 75 s, the intensity of the Ta⁵⁺ peak was much stronger after etching for 75 s compared to after etching for 45 s. Ta₂O₅ was formed due to the oxidation of Ta adjacent to the Si/SiO₂ substrate on the bottom of the sample by SiO₂. However, the intensity of the Ta peak after 1 min of annealing (red color) was stronger than that for the as-deposited sample (black color; Figs. 5b and c). This indicates that some of the Ta₂O₅ was restored to Ta and O (Ta₂O₅ → 2Ta + 5O) after a short period of annealing, which is in agreement with other results²⁵. Therefore, the annealing treatment led to a decrease in the amount of Ta₂O₅ and an increase in the amount of Ta at the Ta/SiO₂ interface of the Ta/Mo/CoFeB/MgO/Ta film.

Figs. 5d and 5e show the computer-fitted curves of the Fe 3*p* and Mg 2*p* high-resolution XPS spectra of the as-deposited sample and the sample annealed for 1 min after 15 s of etching (which corresponds to the MgO/CoFeB interface), respectively. According to the XPS

handbook²¹, the peaks located at ~50.0 eV, 52.7 eV, and 54.7 eV correspond to the Mg 2*p*, metallic Fe 3*p*, and Fe oxide 3*p* states, respectively, indicating that the interfacial Fe atoms were partially oxidized. However, the ratio of Fe oxide to metallic Fe calculated from the areas of the fitted curves was 0.18:1 for the as-deposited sample and 0.26:1 for the sample annealed for 1 min. This indicates that more Fe oxide was formed in the ferromagnetic layers of the annealed sample. The Mg 2*p* peak also shifted to higher binding energies after annealing. The analytical error in quantitative analysis of XPS is usually less than 10%. PMA in CoFeB/MgO structures originates from the hybridization of Fe (Co) and O orbitals at the interface of CoFeB/MgO; thus, the formation of moderate Fe oxide is beneficial for the generation of PMA.

Fig. 5f shows the Mo 3*d* XPS spectrum of the Ta/Mo/CoFeB/MgO/Ta film etched for 15 s after 1 min annealing. The corresponding Mo 3*d* photoelectron spectrum showed only metallic Mo components, without any detectable trace of oxide peaks, along with a strong spin-orbit splitting with the metallic Mo 3*d*_{5/2} Peak 1 at 227.2 eV and Mo 3*d*_{3/2} Peak 2 at 230.5 eV.

The microstructure was characterized by HRTEM for samples as-deposited and after 1, 30, and 60 min annealing. HRTEM image of the sample annealed for 1 and 60 min annealing are shown in Fig. 6a and 6b, respectively. The bottom and top Ta is definitely amorphous for samples after 1 min annealing. The CoFeB and MgO layers are also predominantly amorphous, since the lattice fringes are not visible in the CoFeB and MgO layers, indicating these layers are amorphous. The interface between CoFeB layer and MgO layer is ambiguous in sample after 1 min annealing (Fig. 6a). It is same as that of the sample as-deposited. However, the CoFeB and MgO layers are crystallized in sample after 60 min annealing. The fast Fourier transforms (FFT) of the square areas “C,” “D,” and “E” are shown in Figs. 6c, 6d, and 6e, where in the spots can be identified as the fcc (001) plane of MgO, bcc (001) plane of CoFeB, and bcc (110) plane of

Mo. These results demonstrate that the Mo inserted layer of the bcc structure had a (110) texture, whereas the CoFeB and MgO layers exhibited bcc (001) and fcc (001) textured, respectively. The MgO barrier interface became sharper and smoother after 60 min annealing. It's same as that of the sample after 30 min annealing. Fig. 6f shows the line profile of energy-dispersive X-ray spectroscopy (EDX) for the sample used for TEM observation. The peaks of the composition elements were detected from the top area in the following order; Ta, Co, Mo, Fe, B, O and Mg, respectively. The line profile showed that B and Mo likely diffuse during thermal annealing. This result indicates an inter-diffusion event at the CoFeB/MgO interface. However, this interdiffusion event seems to not affect the crystalline structures in our samples.

The interfacial perpendicular anisotropy between oxide and ferromagnetic metal (Fe/MgO) was previously predicted by first-principles calculation and attributed to the hybridization of the Fe 3d and O 2p orbitals¹⁶. This means that the PMA for a ferromagnetic/oxide interface strongly depends on Fe3d-O2p orbital hybridization. In this study, the Ta₂O₅ in the Ta/Mo/CoFeB/MgO/Ta films was partially decomposed ($\text{Ta}_2\text{O}_5 \rightarrow 2\text{Ta} + 5\text{O}$), and oxygen was liberated after annealing at 350°C for 1 min. Thus, some of the O atoms released by TaO_x drifted toward the Mo and CoFeB layers via diffusion paths (grain boundaries). Subsequently, some of these oxygen atoms combined with Fe atoms in the CoFeB layer, leading to the formation of more Fe oxide (the Mo XPS result shows that Mo was not oxidized). This increased the content of Fe oxide at the CoFeB/Ta interface, promoting the formation of PMA. Another paper reported that interfacial oxygen migration induced the evolution of magnetic anisotropy in Pt/Co/MgO/Pt films, and annealing generated an obvious improvement in the Co/MgO interface and enhanced the effective Co-O orbital hybridization¹². This suggests that in the present study, the oxygen migrated from the Ta layer to the CoFeB/MgO interface, resulting in more effective Fe–O orbital

hybridization at the CoFeB/MgO interface, which is critical factor for the generation of PMA in Ta/Mo/CoFeB/MgO/Ta films after annealing. Therefore, oxygen migration plays a decisive role in the change from IMA to PMA in Ta/Mo/CoFeB/MgO/Ta films after annealing.

Magnetic anisotropy in films is closely related to interfacial structure, including the interfacial chemical state and interfacial oxygen migration. Mo plays a critical role in Ta–O–Fe bonding at the CoFeB/Ta interface; Mo causes oxygen to migrate from the Ta layer to the CoFeB layer in Ta/Mo/CoFeB/MgO/Ta films after annealing. Another study demonstrated that the insertion of an ultrathin Ru layer between the CoFeB and Ta layers in MgO/CoFeB/Ta/MgO films effectively induces PMA without annealing²⁶. In this study, oxygen migration at the CoFeB/Ta interface in Ta/Mo/CoFeB/MgO/Ta films was induced by Mo during a short period of annealing. The Mo-induced oxygen migration altered the interfacial electronic and chemical structures at the CoFeB/Ta interface, resulting in a Ta/Mo/CoFeB/MgO/Ta film with a high PMA.

4. CONCLUSION

We demonstrated the development of PMA in CoFeB/MgO structures during a short period of annealing in Ta/Mo/CoFeB/MgO/Ta films. Mo plays a critical role in Ta–O–Fe bonding at the CoFeB/Ta interface, causing oxygen to migrate from the Ta layer to the CoFeB layer after a short period of annealing. The XPS results show that the more formation of Fe oxide at the CoFeB/MgO interface is critical to obtain the PMA. The HRTEM results show that the bottom and top Ta, the CoFeB and MgO layers are predominantly amorphous in sample after 1min annealing. However, the CoFeB and MgO layers exhibited bcc (001) and fcc (001) textured after 30 and 60 min annealing, which is beneficial to get high PMA. Fe oxide at the CoFeB/MgO

interface plays decisive roles in the evolution of magnetic anisotropy from in-plane magnetic anisotropy to perpendicular magnetic anisotropy in Ta/Mo/CoFeB/MgO/Ta films.

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Figure captions

Fig.1 The AHE response of the as-deposited samples and the samples after annealing for 1, 30, 60, and 90 min

Fig.2 The perpendicular and in-plane M–H curves of the as-deposited sample(a) and the samples annealed at 350°C for 0.5(b), 1(c), 30(d), 60(d), and 90(e) min, respectively

Fig.3 The dependences of H_c and on annealing time for each sample

Fig.4 The dependence of R_q on the annealing time

Fig.5 The Ta 4f XPS spectra as-deposited (black color) and after 1 min annealing (red color) after etching for 45(a), 60(b), and 75(c) s; The computer-fitted curves of the Fe 3p and Mg 2p high-resolution XPS spectra of the as-deposited(d) sample and the sample annealed for 1 min(e) after 15 s of etching; The Mo 3d XPS spectrum(f) etched for 15 s after 1 min annealing

Fig.6 HRTEM image of the sample annealed for 1min (a) and 60 min (b)annealing; The line profile of EDX for the sample after 60 min annealing(f).