1 **Enhanced post-annealing stability of perpendicular Ta/CoFeB/Mg/MgO**
2 **multilayers by inhibiting Ta diffusion**

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8 **Abstracts:** To reveal the underlying mechanism of Mg influence on the enhanced
9 post-annealing stability of perpendicular Ta/CoFeB/Mg/MgO multilayers, the X-ray
10 photoelectron spectroscopy analysis **has** been performed. It is found that a certain
11 amount of Mg interlayer at the CoFeB/MgO interface could prevent the Ta oxidation,
12 and consequently lower the diffusion motivation of Ta from the bottom layer to the
13 CoFeB/MgO interfaces to some extent in the annealing process. The prevention of Ta
14 diffusion realizes the effective hybridization of Fe and O at the CoFeB/MgO interface
15 and maintains interfacial magnetic anisotropy ($K_{\text{CoFeB/MgO}}$). As a result, the
16 perpendicular magnetic anisotropy at high annealing temperatures is maintained.

17 **Keywords:** CoFeB/MgO, perpendicular magnetic anisotropy, interfacial anisotropy
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1. Introduction

The spin-transfer-torque magnetic random access memory (STT-MRAM) has attracted considerable attention in the development of nonvolatile memories with low power assumption and high speed performance [1]. Compared with the in-plane magnetic anisotropic (IMA) materials, the magnetic materials with large perpendicular magnetic anisotropy (PMA) can fulfill the thermal stability in nano-meter range. In addition, the magnetic tunnel junctions based on thin films with PMA can reduce the critical current density for STT switching [2]. As a result, perpendicular magnetic tunnel junctions (P-MTJs) have been considered as a candidate for future magnetic random access memory elements. In 2010, Ikeda et al. reported observing PMA in CoFeB layer sandwiched by MgO and Ta layers, which has potential applications in STT devices [3]. The recent discovery of ultrathin CoFeB/MgO/CoFeB tunnel junctions with perpendicular magnetic anisotropy, which satisfies the criteria of high tunnel magnetoresistance, high anisotropy constant and low switching current, has aroused great concern as a promising candidate for spintronics materials [4]. Traditionally, the annealing treatment at 350 °C–400 °C is crucial for the formation of the crystalline structure of magnetic electrodes and high TMR and **for matching** with the complementary metal oxide semiconductor (CMOS) processing temperature [5,6]. However, the PMA will degenerate when the annealing temperature is beyond 350 °C [3]. In previous studies, it was reported that the Mg interlayer introduced at the CoFeB/MgO interface in in-plane MTJ Ta/CoFeB/MgO/CoFeB/Ta can influence the magnetic and TMR properties [7-9]. For perpendicular MTJ Ta/CoFeB/MgO/CoFeB/Ta, Q. L. Ma reported **the top electrode MgO/CoFeB/Ta can still exhibit PMA** at the annealing temperature as high as 400 °C by introducing Mg layer at the CoFeB/MgO interface [10]. Other explorations also have been conducted in order to improve the annealing stability of PMA for CoFeB/MgO system [11-13]. Although great progresses in improving the post-annealing stability of PMA have been made, the underlying mechanism of

1 realizing the thermally stable PMA still remains elusive. In our work, the multilayers
2 of Ta/CoFeB/Mg(t_{Mg})/MgO/Pt with t_{Mg} **varying from** 0 nm to 1.2 nm were prepared. It
3 is found that when a certain thickness of Mg layer is introduced, strong PMA is
4 maintained after annealing at temperature as high as 400 °C, indicating
5 high-post-annealing stability. That is significantly contrast to the sample without Mg
6 interlayer, which turns to IMA when the annealing temperature exceeds 350 °C. X-ray
7 photoelectron spectroscopy (XPS) measurement is investigated to study the chemical
8 states of different elements at CoFeB/MgO interfaces for the samples with and
9 without the Mg interlayer. From the XPS results, the existence of certain thickness of
10 Mg can prevent the **Ta atoms diffusing to** the CoFeB/MgO interface to some extent,
11 which results in the maintained PMA at relatively high annealing temperature.

12 **2. Experimental**

13 All samples were deposited on thermally oxidized Si substrates by magnetron
14 sputtering system at room temperature. The sample structure is substrate/Ta
15 (3)/CoFeB (1.1)/Mg (t_{Mg})/MgO(2)/Pt(3), where all the thicknesses are the nominal
16 thickness in nm and t_{Mg} **is the introduced Mg thickness** varying from 0 nm to 1.2 nm.
17 The nominal composition of CoFeB is Co:Fe:B=40:40:20. The base pressure of the
18 sputtering system was better than 1.5×10^{-7} Torr and a working argon pressure was 2
19 mTorr. The Ta, CoFeB, Pt and Mg were deposited by DC sputtering with powers of
20 100 W, 100 W, 100 W and 10 W, respectively. The MgO was deposited by RF
21 sputtering with a power of 120 W. The deposition rates for Ta, CoFeB, Pt, Mg and
22 MgO were 1 Å/s, 0.45 Å/s, 1.875 Å/s, 0.150 Å/s and 0.057 Å/s, respectively, which
23 were double-checked by using X-ray reflectivity and light interference profiler. The
24 nominal thickness of each layer was controlled by the deposition time through a
25 computer. The annealing treatment was carried out in a vacuum furnace (vacuum
26 better than 3×10^{-7} Torr) for 30 min without any external magnetic field. The magnetic
27 hysteresis loops and the interfacial chemical states were investigated by using
28 vibrating sample magnetometer (VSM) and X-ray photoelectron spectrometer (XPS),

1 respectively. All experimental measurements were performed at room temperature.

2 **3. Results and discussion**

3 As is well known, K_{eff} is the effective magnetic anisotropy energy which is used
4 to characterize the orientation of magnetization in magnetic thin film. Negative K_{eff}
5 corresponds to IMA while positive K_{eff} corresponds to PMA [14]. In our experiment,
6 K_{eff} is determined from the integrated difference between out-of-plane and in-plane
7 magnetization vs. magnetic-field curves, which can be written as:

$$8 \quad K_{\text{eff}} = K_v + \frac{K_{\text{interface}}}{t} - 2\pi M_s^2 \quad (1)$$

9 where K_v is the volume magnetic anisotropy which can be neglected, $K_{\text{interface}}$ is the
10 interfacial magnetic anisotropy, t is the thickness of CoFeB and M_s is the saturation
11 magnetization of CoFeB. Fig. 1(a) shows the effect of annealing temperature (T_a) on
12 the effective magnetic anisotropy energy (K_{eff}) for Ta(3)/CoFeB(1.1)/Mg(t_{Mg})/MgO(2)
13 /Pt(3) multilayers with t_{Mg} =0 nm, 0.6 nm and 1.2 nm. Fig. 1(b) shows the variation of
14 K_{eff} with different Mg interlayer thickness (t_{Mg}) at annealing temperature T_a =375 °C.
15 And Fig. 1(c) and 1(d) show the representative out-of-plane and in-plane magnetic
16 hysteresis loops for the samples with t_{Mg} =0.6 nm and t_{Mg} =0 nm after 375°C annealing
17 treatment marked as points c and d in Fig. 1(a). As can be seen from Fig. 1(a), K_{eff} for
18 all the as-deposited multilayers is negative, indicating that the as-deposited
19 multilayers are IMA, neglecting the existence of Mg. With T_a increasing, K_{eff} becomes
20 positive for the multilayers with t_{Mg} =0 nm and 0.6 nm, indicating that the annealed
21 multilayers obtain PMA. When the annealing temperature reaches 350 °C, the value of
22 K_{eff} for the film with t_{Mg} =0 nm turns to negative. However, the K_{eff} for the film with
23 t_{Mg} =0.6 nm reaches its maximum positive value. As the further increase of T_a , K_{eff} for
24 the film with t_{Mg} =0.6 nm decreases but still keeps positive, indicating the PMA. For
25 the film with t_{Mg} =1.2 nm, K_{eff} keeps negative over the entire range of T_a , which may
26 be due to the destruction of effective hybridization between Fe in CoFeB and O in
27 MgO owing to the introduction of relatively thick Mg interlayer. From Fig. 1(b), at
28 T_a =375 °C, when t_{Mg} =0 and 0.2 nm, the multilayers show IMA with negative K_{eff} .

1 With the Mg thickness increasing, the samples exhibit PMA at $t_{\text{Mg}}=0.4$ nm, 0.6nm, 0.8
2 nm. But once t_{Mg} exceeds 0.8 nm, the PMA turns to IMA again. It can be concluded
3 that appropriate thickness of Mg interlayer plays a crucial role in the thermal stability
4 of PMA for CoFeB/MgO system. Fig. 1(c) and 1(d) show the representative
5 out-of-plane and in-plane magnetic hysteresis loops for the films with $t_{\text{Mg}}=0.6$ nm and
6 $t_{\text{Mg}}=0$ nm after 375 °C annealing treatment. For the film with $t_{\text{Mg}}=0.6$ nm, the
7 out-of-plane magnetic hysteresis loop with high remanence ratio and good squareness
8 demonstrates a well-developed PMA. On the contrary, for the film with $t_{\text{Mg}}=0$ nm, the
9 easy magnetic axis is in the film plane. It can't be neglected that the saturation
10 magnetization is 1303 emu/cm³ and 1033 emu/cm³ for the sample with $t_{\text{Mg}}=0.6$ nm
11 and 0 nm, respectively.

12 Traditionally, it is suggested that the PMA mechanism of CoFeB/MgO system is
13 mainly arising from the CoFeB/MgO interfacial anisotropy. Consequently, the
14 chemical states of different elements at the CoFeB/MgO interface may influence the
15 property of annealing stability for PMA. XPS measurement is considered as an
16 effective tool to study the chemical states of different elements at the interfaces
17 [15,16]. The representative samples for XPS measurements are
18 Ta(3)/CoFeB(1.1)/MgO(2)/Pt(3) (denoted as S₁) and
19 Ta(3)/CoFeB(1.1)/Mg(0.6)/MgO(2)/Pt(2.4) (denoted as S₂) and annealed at 375 °C. S₂
20 is prepared with the capping Pt layer of 2.4 nm in order to make the CoFeB/MgO
21 interfaces of S₁ and S₂ locate at the same depth. The base pressure of the XPS system
22 was better than 5×10⁻⁸ Pa. The source of the X-ray was Al K_α with the energy of
23 1486.6 eV. The energy analyzer was operated at constant pass energy of 30 eV. The
24 beam energy of Ar ion gun was 2 keV with a working pressure of 2×10⁻⁵ Pa. The XPS
25 detectable depth d can be calculated by the equation $d=3\lambda\sin\alpha$ [17], where λ is the
26 inelastic mean-free paths (IMFPs) for photoelectrons and α is the take-off angle for
27 photoelectrons with respect to the films plane. The IMFPs can be calculated by using
28 the table compiled by Tanuma *et al* [18]. For Al K_α source, the IMFPs for Co 2p in
29 metallic Co, Fe 2p in metallic Fe, Ta 4f in metallic Ta and Mg 1s in metallic Mg are
30 1.18 nm, 1.36 nm, 1.89 nm and 8.1 nm, respectively. The IMFPs for their oxides are

1 about 0.1-0.2 nm larger than the values in metallic elements. In our experiment, the
 2 take-off angle α is set as 90° . And the chemical state information at CoFeB/MgO
 3 interface in Ta/CoFeB/MgO/Pt film can be detected after Ar ion etching 45 s with the
 4 etching rate of about 0.1 nm/s. To eliminate the electric charge effect, all the binding
 5 energies are calibrated by C 1s (284.60 eV). The XPS peak areas and peak
 6 decomposition are determined using Gaussian (80%)-Lorentzian (20%) curve fitting
 7 software provided by XPS system and peak areas are measured with a precision of 5%
 8 or better. Fig. 2(a)-(d) show the high-resolution XPS spectra of Co 2p_{3/2} and Fe 2p_{3/2}
 9 at the CoFeB/MgO interface for S₁ and S₂ annealed at 375 °C, respectively. The peaks
 10 1 and 1' locating at 777.8 eV represent metallic Co, showing no evidence of oxidation
 11 states. And the peaks 2 (2') and 3 (3') locating at 706.8 eV and 708.1 eV correspond
 12 to the metallic Fe and Fe oxides interacting with the O in MgO layer [19]. It is well
 13 known that the Co and Fe for the as-deposited multilayers are over-oxidized during
 14 the deposition of MgO layers, which results in the IMA [20]. After the annealing
 15 process, all the Co oxide and most of the Fe oxide are reduced to their metallic states
 16 and PMA is realized due to the appropriate hybridization of the Fe-3d and O-2p orbits
 17 at the ferromagnet (FM)/oxide interfaces. However, it is interesting that with the
 18 almost same oxidized states of Co and Fe, the multilayers with $t_{\text{Mg}}=0$ nm and $t_{\text{Mg}}=0.6$
 19 nm exhibit different magnetic anisotropy. The unknown reasons need to be explored
 20 further.

21 Other studies report that the deterioration of PMA for Ta/CoFeB/MgO stacks at
 22 relatively high annealing temperature (above 350 °C) is directly associated with the
 23 degradation of the interface anisotropy due to diffusion of thermally activated Ta
 24 atoms from the Ta seed layer into the CoFeB/MgO interface [21]. Whether or not the
 25 diffusion of Ta atoms is influenced by introducing Mg at the CoFeB/MgO interface
 26 needs to be explored. To analyze the Ta diffusion qualitatively, the Ta 4f spectra at
 27 various etching time for S₁ and S₂ are shown in Fig. 3(a) and 3(b). At etching time of
 28 0 s, no Ta peaks can be seen, demonstrating the Ta can't be detected in this depth. At
 29 etching time of 15 s, for S₁, peaks 3 and 4 locating at 25.5 eV and 27.4 eV represent
 30 the Ta oxide, but no Ta peaks is observed for S₂. It can be presumed that part of Ta

1 atoms in S_1 diffuse toward the CoFeB/MgO interface or MgO region during the
 2 annealing process. That is, the existence of Mg interlayer could inhibit the Ta
 3 diffusing up to the CoFeB/MgO interface. At etching time of 30 s and 45 s, for both
 4 S_1 and S_2 , peak 1 and peak 2 locating at 21.7 eV and 23.5 eV correspond to Ta $4f_{5/2}$
 5 and Ta $4f_{7/2}$ in metallic state, while peak 3 and peak 4 locating at the high bonding
 6 energy correspond to Ta $4f_{5/2}$ and Ta $4f_{7/2}$ in oxidized state. However, it can be seen
 7 that the oxidized peaks in S_1 is more obvious than S_2 , indicating more TaO_x exists.
 8 With further etching of 60 s, the Ta oxide disappears and only metallic Ta remains. To
 9 show the different percentage of TaO_x for S_1 and S_2 , the high resolution XPS spectra
 10 and fitting curves of Ta 4f for the as-deposited and annealed S_1 and S_2 at etching time
 11 of 45 s are shown in Fig. 4(a) and 4(b). Considering that the λ of Ta is about 1.89 nm,
 12 the Ta existing at CoFeB/MgO interface and at the bottom layer can be all detected at
 13 etching time of 45 s since XPS detectable depth $d=3\lambda$ is about 5.67 nm. It can be seen
 14 from the figures, in as-deposited state, peak 1 and peak 2 locating at 21.7 eV and 23.5
 15 eV for both S_1 and S_2 correspond to Ta $4f_{5/2}$ and Ta $4f_{7/2}$ indicating that Ta exists in
 16 metallic not oxidized state. In the annealed state, for S_1 , the original spectrum are well
 17 fitted by metallic Ta: Ta $4f_{7/2}$ (peak 1 at 21.7 eV) and Ta $4f_{5/2}$ (peak 2 at 23.5 eV) and
 18 an additional one that exists in the middle of the metallic and oxidized peaks referring
 19 to TaO_x for peak 3 and peak 4 locating at 25.5 eV and 27.4 eV [18]. By the fitting
 20 process, it can be concluded that 16% Ta is oxidized to TaO_x in Fig. 4(a). However,
 21 the annealed state of S_2 is different from that of S_1 . It is clearly seen the oxidized-Ta
 22 component (peaks 3 and 4 at 26.6 eV and 28.4 eV, respectively) decreases to about 6%
 23 in Fig. 4(b). How the Mg interlayer influences the Ta diffusion? To explore the
 24 possible reasons, the chemical states of Mg 1s for S_1 and S_2 in as-deposited and
 25 annealed states are studied by XPS shown in Fig. 4(c) and 4(d). Peak 1 in Fig. 4(c)
 26 locating at 1303.7 eV corresponds to Mg 1s in stoichiometric MgO (1303.8 eV). It is
 27 interesting that even though 0.6 nm Mg interlayer is introduced at CoFeB/MgO
 28 interface, it is oxidized Mg (peak 1 at 1303.4 eV) not metallic Mg existing for S_2 as
 29 shown in Fig. 4(d). The binding energy shifts to 1303.4 eV from 1303.8 eV for Mg 1s
 30 in stoichiometric MgO, suggesting that the introduced metallic Mg is oxidized during

1 the sputtering process and compositional changed MgO_x ($x < 1$) is formed near the
 2 CoFeB/MgO interface. After the annealed process, peak 1' in Fig. 4(c) shifts to the
 3 low binding energy 1303.3 eV, indicating non-stoichiometric MgO_y ($y < 1$) is generated.
 4 However, peak 1' at 1303.3 eV in Fig. 4(d) doesn't change much compared with peak
 5 1 in the as-deposited state. From the result, it can be concluded that for S_1 part of O in
 6 MgO layer is transferred to oxidize Ta atoms, forming the MgO_y in the annealed
 7 process. For S_2 the binding energy of peak 1' and 1 are almost unchanged, suggesting
 8 that no O atoms or only smaller part of O in MgO is bonding with Ta. The results
 9 coincide with the different amount of Ta oxide in Fig. 4(a) and 4(b). As well known,
 10 the Ta oxide with a very high negative formation enthalpy (-2046 kJ/mol) is
 11 thermodynamically more stable than MgO (-602 kJ/mol) [7]. Once the Ta atoms
 12 diffuse to the CoFeB/MgO interface, the Ta oxides will be formed. For S_1 , during the
 13 annealing process, amount of Ta atoms diffuse to the CoFeB/MgO interface and are
 14 oxidized to TaO_x by stoichiometric MgO, as shown in Fig. 4 (a). And the diffusion
 15 motivation of bottom Ta to the CoFeB/MgO interface is believed to be the formation
 16 of Ta oxide. In contrast, for S_2 , the introduction of Mg interlayer makes the oxygen
 17 deficit MgO_x ($x < 1$) at the CoFeB/MgO interface and the percent of TaO_x is reduced
 18 due to the lower oxidation ability of non-stoichiometric MgO_x , as shown in Fig. 4 (b).
 19 Consequently, the Ta diffusion motivation is weakened at the same time. It's said that
 20 the diffusion of Ta into CoFeB will decrease saturation magnetization (M_s) [22],
 21 which is also consistent with the difference of M_s for Ta/CoFeB/MgO (1033 emu/cm³)
 22 and Ta/CoFeB/Mg(0.6)/MgO (1303 emu/cm³) as shown in Fig. 1(c) and 1(d). This
 23 indirectly confirms the blocking of Ta diffusion by introducing a certain amount of
 24 Mg interlayer. Traditionally, $K_{\text{interface}}$ is given by the sum of two components: $K_{\text{Ta/CoFeB}}$
 25 for the Ta/CoFeB interface and $K_{\text{CoFeB/MgO}}$ for the CoFeB/MgO interface. Previous
 26 work suggests it is the CoFeB/MgO interface that is primary responsible for the PMA.
 27 And the diffusion of Ta to the CoFeB/MgO interface will reduce $K_{\text{CoFeB/MgO}}$ and lead
 28 to PMA degradation. The introduction of a certain amount of Mg restrains the Ta
 29 diffusing toward the CoFeB/MgO interface, resulting in the effective hybridization of
 30 Fe and O and keeping interfacial perpendicular anisotropy $K_{\text{CoFeB/MgO}}$. As a result, the

1 PMA at relatively high annealing temperature is obtained.

2 **4. Conclusion**

3 During the high annealing process, the seed layer Ta diffuses to the CoFeB/MgO
4 interface, which is destructive for the PMA. However, the introduction of Mg at the
5 CoFeB/MgO interface lowers the motivation of Ta diffusion and results in the
6 effective hybridization of Fe and O at CoFeB/MgO interface. As a consequence, the
7 interfacial anisotropy $K_{\text{CoFeB/MgO}}$ is kept and PMA at relatively high annealing
8 temperature is maintained.

9 **Acknowledgments**

10 This work was supported by the National Science Foundation of China (Grant No.
11 51331002 and 51461014), by the Fundamental Research Funds for the Central
12 Universities FRF-SD-12-011A, by the Ph.D. Programs Foundation of Ministry of Education
13 (Grant No. 20120006130002), by the National key Scientific research projects of China
14 (Grant No. 2015CB921502).

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1 Figure captions:

2 Fig. 1. The effect of annealing temperature (T_a) on the effective magnetic anisotropy
3 energy (K_{eff}) for Ta(3)/CoFeB(1.1)/Mg(t_{Mg})/MgO(2) /Ta(2) multilayers with $t_{\text{Mg}}=0$,
4 0.6 nm and 1.2 nm (a), the variation of K_{eff} with different Mg interlayer thickness (t_{Mg})
5 at annealing temperature $T_a=375$ °C (b), and the representative out-of-plane and
6 in-plane magnetic hysteresis loops for $t_{\text{Mg}}=0.6$ nm (c) and $t_{\text{Mg}}=0$ nm (d) after 375C
7 annealing treatment marked as points c and d in Fig. 1 (a).

8 Fig. 2. The high-resolution XPS spectra and its fitting curves of Co 2p_{3/2} (a) and Fe
9 2p_{3/2} (c) at the CoFeB/MgO interface for S₁ and Co 2p_{3/2} (b) and Fe 2p_{3/2} (d) for S₂,
10 respectively.

11 Fig. 3. The XPS spectra of Ta 4f for S₁ (a) and S₂ (b) as a function of different etching
12 time.

13 Fig. 4. The high-resolution XPS spectra and its fitting curves of as-deposited and
14 annealed Ta 4f for S₁ (a) and S₂ (b) and Mg 1s for S₁ (c) and S₂ (d).

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Graphical Abstract

To reveal the underlying mechanism of Mg influence on the enhanced post-annealing stability of perpendicular Ta/CoFeB/Mg/MgO multilayers, the X-ray photoelectron spectroscopy analysis have been performed. It is found that a certain amount of Mg interlayer at the CoFeB/MgO interface could prevent the Ta oxidation to some extent due to the oxygen deficit MgO_x ($x < 1$), and consequently lower the diffusion motivation of Ta from the bottom layer to the CoFeB/MgO interfaces to some extent during the annealing process. The prevention of Ta diffusion realizes the effective hybridization of Fe and O at the CoFeB/MgO interface and maintains interfacial magnetic anisotropy ($K_{\text{CoFeB/MgO}}$). As a result, the perpendicular magnetic anisotropy at high annealing temperatures was maintained.