Strong perpendicular magnetic anisotropy in $Co_2FeAl_{0.5}Si_{0.5}$ film sandwiched by MgO layers*

Wang Sheng(王 圣), Li Xiao-Qi(李晓其), Bai Li-Juan(白丽娟), Xu Xiao-Guang (徐晓光)†, Miao Jun(苗 君), and Jiang Yong(姜 勇)

State Key Laboratory for Advanced Metals and Materials, School of Materials Science and Engineering, University of Science and Technology Beijing, Beijing 100083, *China*

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$Co_2FeAl_{0.5}Si_{0.5}$ (CFAS)-based multilayers sandwiched by MgO layers have been deposited and annealed at different temperatures. Perpendicular magnetic anisotropy (PMA) with the magnetic anisotropy energy density $K_u \approx 2.5\times10^6$ erg/cm^3 (1 erg $= 10^{-7}$ J) and the coercivity $H_c = 363$ Oe (1 Oe $= 79.9775$ A $\cdot$ m^{-1}) has been achieved in the Si/SiO_2/MgO (1.5 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm) film annealed at 300 °C. The strong PMA is mainly due to the top MgO layer. The structure can be used as top magnetic electrodes in half-metallic perpendicular magnetic tunnel junctions.

Keywords: half-metallic ferromagnets, MgO layers, perpendicular magnetic anisotropy

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1. Introduction

Spin polarization of ferromagnetic electrodes is a key factor in the performance of spintronic devices such as magnetic tunnel junctions (MTJs) and spin valves (SPVs). For traditional transition metallic ferromagnets, the spin polarization is about 40%–50%. However, half-metallic ferromagnets (HMFs) possess a theoretical spin polarization of 100%, since their minority spin states are zero at Fermi energy.[1] The application of HMFs in MTJs or SPVs could greatly enhance the tunneling magnetoresistance (TMR) or the giant magnetoresistance (GMR).[2,3] Among HMFs, $Co_2FeAl_{0.5}Si_{0.5}$ (CFAS) has a larger spin polarization than other Co-based full-Heusler alloys, e.g., Co_2FeAl and Co_2FeSi,[4] and exhibits the weakest temperature dependence among all known HMFs. Therefore, CFAS is a promising ferromagnetic (FM) material for spintronic devices.[5]

Perpendicular magnetic anisotropy (PMA) has been a hot field in recent years because FM electrodes with PMA could bring faster and smaller MTJ-based devices than those with in-plane magnetic anisotropy.[6] However, no traditional PMA film can satisfy all three requirements of high thermal stability at reduced dimension, low critical current for spin-torque-induced switching, and high magnetoresistance.[7,8] Therefore, it is important to realize PMA in half-metallic full-Heusler alloy thin films due to their high spin polarization and low Gilbert damping constant.[9] Recently, we have studied strong PMA in Pt/Co_2FeAl/MgO trilayers with a thick Co_2FeAl layer of about 2.5 nm.[10,11] However, this structure can only serve as the bottom electrodes in MTJs or SPVs. It is more difficult to realize PMA in full-Heusler alloy-based top electrodes on MgO, even though a weak PMA with poor squareness was obtained in the MgO/Fe/Co_2FeAl/Pt multilayer after 350-°C annealing for 1 h with a 5000 Oe perpendicular magnetic field applied.[12] Moreover, the Co_2FeAl thickness was only 0.8 nm in MgO/Fe/Co_2FeAl/Pt multilayer,[12] which cannot guarantee the half metallic B_2 or $L2_1$ phases after 350-°C annealing.

According to the previous studies,[10–12] the PMA comes from the interface between the Co-based Heusler alloy and MgO layers. Moreover, CFAS films have a good lattice match with MgO layers. Therefore, we choose to study CFAS-based multilayers with a CFAS thickness of 2.5 nm sandwiched between two MgO layers, in which a clear PMA has been observed after 300-°C annealing with no external magnetic field. The structure could be applied as the top electrodes in perpendicular MTJs or SPVs.

2. Experiments

Thin films with a stack structure of MgO (1.5 nm, 2 nm)/CFAS (2.5 nm)/MgO (t_{MgO})/Pt (5 nm) were deposited on Si/SiO_2 substrates by direct-current (DC) magnetron sputtering except for the MgO layer, which was deposited by radio-frequency (RF) magnetron sputtering using an MgO target. The thickness of the top MgO layer t_{MgO} varied from 0.6 nm to 1.0 nm. The cap layer of Pt was required to prevent oxidation from outside. The base pressure of the sputtering system was better than 1.0×10^{-5} Pa. The annealing process

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†Corresponding author. E-mail: xgxu@ustb.edu.cn

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was carried out in a vacuum chamber under 1.0×10^{-4} Pa at different temperatures for 30 min with no magnetic field. The magnetic hysteresis loops were measured by alternating gradient magnetometer (AGM) at room temperature.

3. Results and discussion

Figure 1 shows the magnetization M against the magnetic field H loops (M–H loops) of the as-deposited sample of Si/SiO_2/MgO (1.5 nm)/CFAS (2.5 nm)/MgO (t_{MgO})/Pt (5 nm) with $t_{MgO} = 0.8$ nm. The sample shows a typical in-plane magnetization. Figure 2 shows the perpendicular M–H loops for the sample with $t_{MgO} = 0.8$ nm annealed at various temperatures. It can be seen that both the squareness ratio and the coercivity H_c increase as the annealing temperature increases up to 300 °C. However, further increasing the annealing temperature to 400 °C reduces saturation magnetization M_s and H_c of the multilayer. The evolutions of M_s and H_c can be understood by the competition between the crystallization of CFAS and the oxidation at the CFAS/MgO interfaces. During low temperature annealing, the CFAS begins crystallization, which causes the increase of M_s. At the same time, the oxidation at the CFAS/MgO interface induces the PMA of CFAS. As a result, the squareness ratio and M_s of M–H loops both increase with the increase of the annealing temperature at first. However, the CFAS is deeply oxidized after high temperature annealing at 400 °C, which results in the fading of PMA, as well as M_s.[13] Therefore, the PMA strongly depends on the annealing temperature, which is consistent with the previous results on Pt/Co_2FeAl/MgO trilayers.[11,12] Figure 3 shows the dependence of the magnetic anisotropy energy density K_u and M_s on the annealing temperature T_a, indicating that 300 °C is the optimum annealing temperature for CFAS-based multilayers to get a strong PMA.

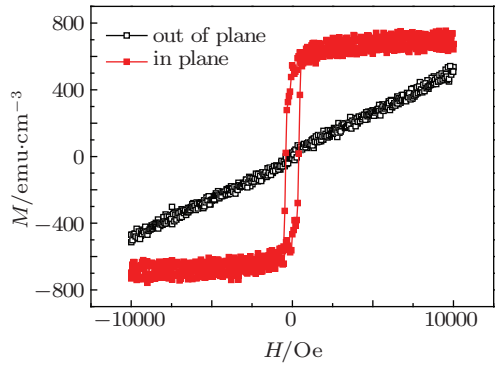


Fig. 1. (color online) In-plane and out-of-plane M–H loops for the as-deposited sample of MgO (1.5 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm).

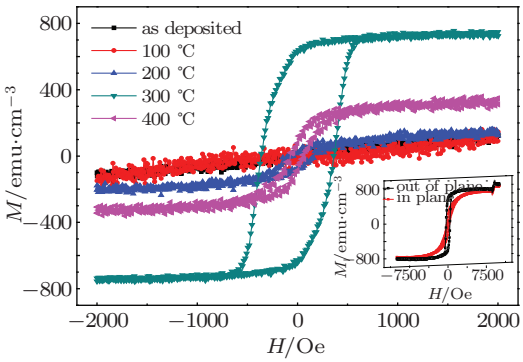


Fig. 2. (color online) The M–H loops measured under the perpendicular magnetic fields for the sample of MgO (1.5 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm) after annealing at different temperatures. The inset shows both in-plane and out-of-plane M–H loops for the sample annealed at 300 °C.

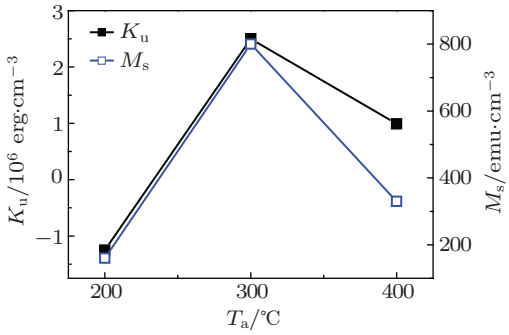


Fig. 3. (color online) Dependence of K_u and M_s on the annealing temperature for the sample of MgO (1.5 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm). The positive and negative K_u correspond to out-of-plane and in-plane magnetizations, respectively.

Figures 4(a) and 4(b) present the M–H loops of 300 °C annealed Si/SiO_2/MgO (1.5 nm)/CFAS (2.5 nm)/MgO (t_{MgO})/Pt (5 nm) multilayers with t_{MgO} of about 0.6 and 1.0 nm, respectively. It is obvious that the sample of $t_{MgO} = 0.6$ nm is in-plane magnetized, while the one with $t_{MgO} = 1.0$ nm shows a strong PMA. Comparing with the sample of $t_{MgO} = 0.8$ nm shown in the inset of Fig. 2, we find that a larger t_{MgO} (> 0.6 nm) favors PMA in our structure. It has been reported that the weak PMA in the MgO/Fe/Co_2FeAl/Pt multilayer is caused by the MgO/Co_2FeAl interfacial perpendicular anisotropy, and the very thin Fe layer (0.09 nm) is used only to promote this interfacial effect for more hybridization of the Fe-3d and O-2p electron orbitals.[12] Therefore, the PMA of the sandwiched structure comes from the effects of both bottom and top MgO interfaces, and the strong PMA of the multilayer with $t_{MgO} = 0.8$ nm can be attributed to the optimized oxidation degree at the CFAS/MgO interfaces. It has been suggested that the PMA in Co/Fe-based alloys is caused mainly by the hybridization between the Fe/Co-3d and O-2p orbitals.[13,14] Accordingly, the interface between CFAS and MgO layers is a key factor to affect the magnetic anisotropy of multilayers. In our samples, the sputtering rate of CFAS is nearly five times faster than that of MgO. After the deposition of the bottom MgO layer, the CFAS layer contacts the MgO insufficiently, which leads to a poor-quality interface between the bottom MgO layer and the CFAS layer. Due to the very low sputtering rate of the MgO, the top MgO layer contacts

the CFAS layer well. However, the thin top MgO layer of 0.6 nm might be discontinuous due to the island-like growth mechanism and the diffusion during high temperature annealing, which leads to a lower degree of oxidation between the CFAS layer and the top MgO layer. With the MgO thickness increasing from 0.6 nm to 1.0 nm, the area of interface between the CFAS layer and the top MgO layer increases, and the interface quality of CFAS/MgO is also improved. A better quality CFAS/MgO interface may enhance the hybridization between Fe/Co-3d and O-2p orbitals. Therefore, increasing the thickness of the top MgO layer greatly improves the PMA in MgO/CFAS/MgO/Pt multilayers. However, increasing the MgO layer thickness beyond 1.0 nm has no obvious effect on the PMA, since the MgO/CFAS interface is already continuous at $t_{MgO} = 1.0$ nm.

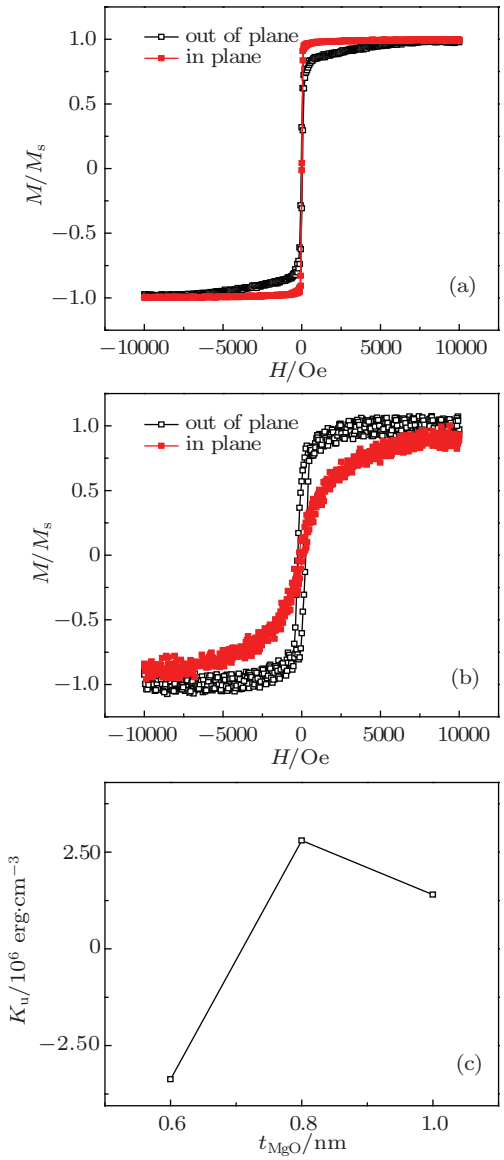


Fig. 4. (color online) In-plane and out-of-plane M–H loops for the multilayers of MgO (1.5 nm)/CFAS (2.5 nm)/MgO (t_{MgO})/Pt (5 nm) with (a) $t_{MgO} = 0.6$ nm and (b) $t_{MgO} = 1.0$ nm. (c) MgO thickness (t_{MgO}) dependence of K_u. All the multilayers were annealed at 300 °C.

As shown in Fig. 4(b), the multilayer with $t_{MgO} = 1.0$ nm can maintain PMA even after 300-°C annealing. However, the H_c of the out-of-plane direction decreases compared with that of the multilayer with $t_{MgO} = 0.8$ nm. According to the lattice constant of MgO (0.43 nm), the layer may be incoherent when t_{MgO} is 0.8 nm. When t_{MgO} increases to 1.0 nm, the coherence of the MgO layer has been improved. This improvement may decrease the magnetic pinning sites at the interface, which reduces the value of perpendicular H_c. Moreover, the thicker MgO layer also contributes to more oxidation at the CFAS/MgO interface and leads to the decrease of H_c.

Figure 4(c) shows the values of K_u as a function of t_{MgO} for the samples of Si/SiO_2/MgO (1.5 nm)/CFAS (2.5 nm)/MgO (t_{MgO})/Pt (5 nm) after 300-°C annealing. The K_u is estimated by using the equation $K_u = M_s H_{an}/2$, where H_{an} refers to the anisotropy field and is estimated approximately by the saturation field in the hard magnetization direction. The positive and negative values of K_u correspond to out-of-plane and in-plane magnetizations, respectively. With the increase of t_{MgO}, K_u first increases and then decreases slightly. The maximum of K_u is as high as 2.5×10^6 erg/cm^3, and this large K_u can secure the good thermal stability of devices at a reduced dimension lower than 40 nm.

In order to further investigate the PMA of CFAS films, the dependence of $K_u t$ (multiplying K_u by t_{CFAS}) on the thickness of CFAS (t_{CFAS}) is studied at the annealing temperature of 300 °C. According to the curve of $K_u t$ versus t_{CFAS} shown in Fig. 5, the CFAS films show two anisotropic changes between in-plane and out-of-plane magnetizations when the thickness is around 2.5 nm. When the CFAS thickness is less than 2.2 nm, over-oxidation may lead to the in-plane anisotropy. A thickness greater than 2.8 nm also leads to in-plane anisotropy due to the intrinsic in-plane anisotropy of CFAS material.

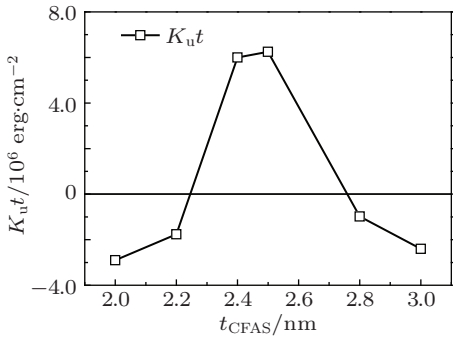


Fig. 5. Dependence of $K_u t$ on the CFAS layer thickness after 300-°C annealing.

The effect of the bottom MgO layer's thickness on the magnetic anisotropy has also been studied. We have fabricated a sample of Si/SiO_2/MgO (2 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm). Figure 6 shows its M–H loops of in-plane and out-of-plane magnetizations after 300-°C annealing. The

sample still exhibits a PMA property, which is a little weaker than that of the multilayer with a 1.5-nm bottom MgO layer. However, the thickness of 1.5 nm is thick enough for the MgO layer to serve as the barrier in MTJs, and a thicker MgO barrier may discourage the TMR effect for electron scattering. Therefore, the CFAS-based multilayers sandwiched by MgO layers is a potential candidate for top electrodes of perpendicular MTJs.

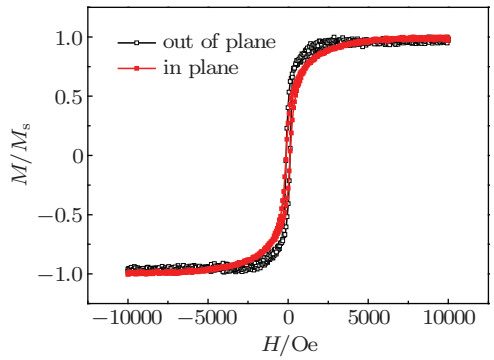


Fig. 6. (color online) In-plane and out-of-plane M–H loops for the sample of MgO (2 nm)/CFAS (2.5 nm)/MgO (0.8 nm)/Pt (5 nm) annealed at 300 °C.

4. Summary

In this paper, we study the magnetic properties of the MgO/CFAS/MgO multilayers. The strong PMA can be observed in this sandwich structure with a relatively thick CFAS layer of 2.5 nm. The annealed multilayers show good thermal stability, and the K_u is as high as 2.5×10^6 erg/cm^3. The strong PMA is induced by both top and bottom MgO/CFAS interfaces where the top one plays a more important role. Since the optimized thickness of the bottom MgO is 1.5 nm, the multilayer can be applied as a top electrode of a perpendicular MTJ.

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