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High tunneling magnetoresistance ratio in perpendicular magnetic tunnel junctions using Fe-based Heusler alloys

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Heusler alloys Fe2Cr1−xCoxSi (FCCS) with different Co compositions x have been predicted to have high spin polarization. High perpendicular magnetic anisotropy (PMA) has been observed in ultra-thin FCCS films with magnetic anisotropy energy density up to 2.3 × 106 erg/cm3. The perpendicular magnetic tunnel junctions (p-MTJs) using FCCS films with different Co compositions x as the bottom electrode have been fabricated and the post-annealing effects have been investigated in details. An attractive tunneling magnetoresistance ratio as high as 51.3% is achieved for p-MTJs using Fe2CrSi (FCS) as the bottom electrode. The thermal stability Δ can be as high as 70 for 40 nm dimension devices using FCS, which is high enough to endure a retention time of over 10 years. Therefore, Heusler alloy FCS is a promising PMA candidate for p-MTJ application. © 2015 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4937917]

I. INTRODUCTION

MgO-based magnetic tunnel junctions (MTJs) have attracted great interest due to large tunneling magnetoresistance (TMR) effect1,2 and potential applications in magnetoresistive random access memory (MRAM). MTJs with perpendicular magnetization of ferromagnetic electrodes have attracted more attention recently, as they require lower spin transfer torque (STT) switching current density due to the absence of the demagnetization term. In addition, they show higher thermal stability owing to their larger anisotropy energy as compared to the established in-plane magnetized materials.3–5

The conventional PMA materials explored so far include rare-earth/transition metal alloys,6,7 Co(Pd, Pt, Ni) multilayers,8,9 and L10-ordered (Co, Fe)Pt alloys.10,11 However, these materials suffer from some limitations including insufficient chemical and/or thermal stability and low spin polarization. In order to achieve good device performance, such as low switching current and high TMR, PMA materials with high spin polarization, low damping constant, and good lattice matching with MgO are desired. Recently, interfacial perpendicular anisotropy induced by MgO interface was shown as an efficient way to realize PMA for conventional magnetic storage materials, such as CoFeB.12 However, the damping constant of CoFeB increases sharply as its thickness decreases (less than 2 nm),12 which will lead to the increase in the intrinsic critical switching current density. This constitutes a disadvantage for STT switching.

Recently, full Heusler alloys have attracted much attention as PMA materials, such as Co2FeAl13,14 and Co2FeAl0.5Si0.5.15 Only Co2FeAl Heusler alloy has been reported to give a TMR ratio of 91% in perpendicular magnetic tunnel junctions (p-MTJs).13 So far, there are no reports on the PMA in Fe-based Heusler alloys. One of which is Fe2Cr1−xCoxSi (FCCS), which has been predicted to have high spin polarization.16 The Co composition x was known to tune the Fermi level close to the center of minority band gap in order to achieve better thermal stability and high spin polarization. We have successfully utilized them in in-plane MTJs17 and we have also obtained PMA in ultra-thin FCCS films.18 In this study, we have fabricated investigated the magnetic and electrical properties of p-MTJs with ultra-thin FCCS films.

II. EXPERIMENT

The MTJ structures of MgO (100)/Cr (40 nm)/Fe2Cr1−xCoxSi (1 nm)/Mg (0.3 nm)/MgO (0.7 nm)/CoFe (0.1 nm)/CoFeB (1.5 nm)/Ta (5 nm)/Ru (5 nm) were fabricated using a sputtering system, where x is the nominal composition determined by the relative deposition rate of each target and set as 0, 0.3, 0.5, 0.7, 0.9, and 1. The FCCS films with different x were prepared by co-sputtering technique using Fe2CrSi and Fe2CoSi sputtering targets. Prior to the deposition, the MgO substrate was preheated at 600 °C for 1 h. The Cr buffer layer was in-situ annealed at Tia = 700 °C for 30 min after deposition at room temperature in order to achieve a very flat surface. Subsequently, the FCCS film was deposited on Cr at room temperature, followed by an in-situ annealing at Tia = 400 °C to promote ordered B2 or L21 structure. The MgO layer was grown by RF sputtering using MgO target at room temperature. The interface between the electrode and the MgO barrier plays a critical role in TMR effect. In order to prevent oxidation of FCCS film by MgO layer, a thin Mg layer of 0.3 nm was inserted before MgO deposition. The CoFe layer inserted between MgO barrier and CoFeB layer is to improve the interface. The post-annealing effect was investigated by ex-situ post-annealing with temperature ranging from Tpa = 320 °C to 400 °C in high vacuum in the presence of an out-of-plane magnetic field of 1 T for 1 h. An alternating gradient magnetometer (AGM) was used to...
characterize the magnetic properties of the MTJ structures. While the TMR ratio and Resistance-Area product (RA) of the p-MTJs were measured by Capres Current-In-Plane-Tunneling (CIPT) technique at room temperature.

III. RESULTS AND DISCUSSIONS

Among all the p-MTJs using FCCS films with Co composition $x = 0, 0.3, 0.5, 0.7, 0.9$, and 1, separate switching between the two ferromagnetic layers are only observed for p-TMJs using FCS (as shown in Fig. 1) and $\text{Fe}_2\text{Cr}_x\text{Co}_{0.3}\text{Si}$ after post-annealing. Figure 1 shows that a sharp magnetization reversal is observed at $340^\circ \text{C}$, indicating a better crystalline structure of MTJ structure at appropriate $T_{pa}$. The top and bottom electrodes switch simultaneously at $T_{pa} = 400^\circ \text{C}$ which may be due to diffusion of Cr atoms from the Cr buffer layer at high $T_{pa}$ which could destroy crystalline structure of bottom FCCS films. Similar observations were obtained for Co compositions ($x = 0.5, 0.7, 0.9$, and 1), which could be attributed to a lower PMA in FCCS films. According to our previous study, FCS has the strongest PMA and becomes poorer as $x$ increases, since Fe-O bonding is predominant and generates PMA in FCCS films. As a result, the poorer PMA in FCCS films with higher $x$ is not robust enough for p-MTJ applications.

Figure 2 depicts the RA product and TMR ratio as a function of $T_{pa}$ for p-MTJs using (a) FCS and (b) $\text{Fe}_2\text{Cr}_x\text{Co}_{0.3}\text{Si}$ as the bottom electrode, respectively. The RA value reduces with $T_{pa}$, which could be due to a better interface between MgO barrier and electrodes. The highest TMR ratio for p-MTJs using FCS is 51.3%, while the highest TMR ratio for p-MTJs using $\text{Fe}_2\text{Cr}_x\text{Co}_{0.3}\text{Si}$ is 35.3% as shown in Fig. 2(b). Thus, a higher TMR ratio is achieved for p-MTJs using FCS as compared to that of $\text{Fe}_2\text{Cr}_x\text{Co}_{0.3}\text{Si}$, which could be attributed to the stronger and more robust PMA in FCS film.

Magnetic anisotropy energy density ($K_U$) is estimated by $K_U = M_S H_k/2$, where $H_k$ is the perpendicular anisotropy field. The calculated $K_U$ are $2.8 \times 10^6 \text{erg/cm}^3$ for FCS, and $2.3 \times 10^6 \text{erg/cm}^3$ for $\text{Fe}_2\text{Cr}_x\text{Co}_{0.3}\text{Si}$, as shown in Fig. 3. The $K_U$ decreases as Co composition ($x$) increases. From these $K_U$ values, we calculated the thermal stability $\Delta U$ using equation $(K_U V)/k_B T$ for FCCS with different Co compositions for a 40 nm dimension device, where $V$ is the volume ($V=\text{area} \times \text{thickness} = 40 \text{nm} \times 40 \text{nm} \times 0.8 \text{nm}$), $k_B$ is the Boltzmann constant ($k_B = 1.38 \times 10^{-23} \text{JK}^{-1}$), and $T$ is the temperature ($T = 300 \text{K}$). The $\Delta U$ is as high as 70 for FCS, which is comparable to that of CoFeB in Ref. 22. It is high enough to endure a retention time of over ten years.

Table I summarizes TMR ratio in p-MTJs using Heusler alloys and CoFeB. Among these PMA materials, FCS has the strongest PMA and becomes poorer as $x$ increases, since Fe-O bonding is predominant and generates PMA in FCCS films. As a result, the poorer PMA in FCCS films with higher $x$ is not robust enough for p-MTJ applications.
smaller $M_S$, which allows for a low critical current for spin transfer torque switching in spin torque devices. We note that the PMA is observed in Co$_2$FeAl$_{0.5}$Co$_{0.5}$ film with larger thickness up to 4.8 nm. Although PMA is only observed for FCS with thickness up to 1.2 nm, the thermal stability $\Delta$ of FCS is as high as 70.

We have noticed that the highest TMR ratio achieved in the in-plane MTJs using FCCS with different Co compositions is 28% for Fe$_2$Cr$_{0.3}$Co$_{0.7}$Si. This suggests that Co doping has successfully tuned the Fermi level close to the center of minority bandgap to achieve better thermal stability and higher spin polarization. However, the highest TMR ratio in p-MTJs is obtained for FCS, i.e., $x = 0$. The PMA is originated from hybridization between Fe/Co 3$d$ and O 2$p$ orbitals, but it is dominated by the contribution of Fe atoms at the FCCS/MgO interface which is discussed in details in Ref. 18. This is consistent with the result reported for CoFeB in Ref. 24. As a result, PMA is strongest at $x = 0$. Therefore, there appears a trade-off between high spin polarization and strong PMA by adjusting the Co composition in FCCS. The thickness of FCCS in the in-plane MTJs is 30 nm, which is much larger than that of FCS in p-MTJs (1 nm). Since the $M_S$ values of ultra-thin FCCS films in p-MTJs are close to those of B2/L2$_1$-ordered epitaxial FCCS films in the in-plane MTJs, we infer that FCCS ultrathin films with PMA have high chemical ordering with good crystalline structure. In addition, the post-annealing at an appropriate $T_{pa}$ improves not only the structural properties of the upper electrode but also the interfacial structural properties, leading to an enhancement of the TMR ratio for both in-plane and perpendicular MTJs. The best post-annealing condition for in-plane MTJs is 350 °C, while that for p-MTJs is 380 °C. The TMR ratio drops dramatically at 400 °C for both cases. This is most likely due to interlayer diffusions. Hence, the best post-annealing conditions for both in-plane and perpendicular MTJs are similar (350 °C ≤ $T_{pa}$ ≤ 380 °C).

It is interesting to note that the TMR ratio is much larger in PMA than in the in-plane FCCS MTJs, which is in contrast to the CoFeB/MgO MTJs, where the PMA MTJ is generally of lower TMR ratio. Mechanism of the higher TMR in the PMA MTJs remains unclear. One possible origin is that the Fermi level at ultrathin FCS may shift, and fall into the minority band gap, resulting in high polarization of FCS film. Further confirmation needs to be done from the first principle calculations.

IV. CONCLUSION

The p-MTJs using Heusler alloys Fe$_2$Cr$_{1-x}$Co$_x$Si films with different Co compositions as the bottom electrode have been fabricated. The post-annealing effect is also studied in details. An attractive TMR ratio as high as 51.3% (35.3%) is achieved for p-MTJs using FCS (Fe$_2$Cr$_{0.7}$Co$_{0.3}$Si) as the bottom electrode. In addition, the thermal stability $\Delta$ of FCS can be as high as 70. These desirable properties render the Heusler alloy FCS is a promising PMA candidate for p-MTJ application.
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