Magnetic properties of a Pt/Co_2FeAl/MgO structure with perpendicular magnetic anisotropy*

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Microstructures and magnetic properties of Ta/Pt/Co_2FeAl (CFA)/MgO multilayers are studied to understand perpendicular magnetic anisotropy (PMA) of half-metallic full-Heusler alloy films. PMA is realized in a 2.5-nm CFA film with B2-ordered structure observed by a high resolution transmission electron microscope. It is demonstrated that a high quality interface between the ferromagnetic layer and oxide layer is not essential for PMA. The conversions between in-plane anisotropy and PMA are investigated to study the dependence of magnetic moment on temperature. At the intersection points, the decreasing slope of the saturation magnetization (M_s) changes because of the conversions. The dependence of M_s on the annealing temperature and MgO thickness is also studied.

Keywords: half-metallic, perpendicular magnetic anisotropy

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1. Introduction

As a key technique, magnetic tunnel junctions (MTJs) have speeded up the development of spintronics devices such as magnetic read-head sensors and magnetic random access memories (MRAM) due to the tunneling magnetoresistance (TMR) effect. A typical MTJ structure contains two ferromagnetic electrodes separated by a nonmagnetic barrier, and possesses a large TMR ratio with the two magnetic layer magnetization alignments parallel and antiparallel to each other. The TMR was observed first by Moodera *et al.*[1] and Miyazaki and Tezuka[2] in MTJs with an AlO_x barrier at room temperature. Mathon and Umerski[3] predicted theoretically that an extremely large TMR ratio of over 1000% can be obtained in epitaxial Fe/MgO/Fe MTJs due to coherent tunneling. Many groups experimentally realized the prediction in MTJs with MgO barriers.[4−6] According to Jullière's model, the infinite TMR could be realized if the spin polarization (SP) of the tunneling current is 100%.[7] Therefore, another promising way to improve the TMR ratio is to employ high spin polarized ferroelectrodes such as half-metallic ferromagnets (HMFs) in MTJs. Recently, many experimental and theoretical studies on HMFs have been reported.[8−11]

As a special class among HMFs, the full-Heusler alloy with a formula of X_2YZ (X and Y are transition metals and Z is a main group element) has been successfully employed in MTJs to obtain large TMR values.[8,9,12,13] However, all the ferroelectrodes in these devices are of in-plane magnetization. As a result, it is hard to secure good thermal stability when devices are patterned into the nanoscale. Realizing perpendicular magnetic anisotropic HMFs is a promising way to solve this problem and preserve a high SP simultaneously. Co-based full-Heusler alloys are ideal candidates because of their high values of Curie temperature (T_c) and SP at room temperature.[14−16] Perpendicular magnetic anisotropy (PMA) was observed in Pt/Co_2FeAl (CFA) multilayers and Cr (or Pt)/CFA/MgO/Pt structures.[17,18] However, the critical thickness for a CFA layer to maintain PMA is lower than 1.0 nm so that the current may not be polarized completely when it has passed through the device, thereby causing a reduced SP. Another

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problem is that it is difficult to realize and maintain half-metallic B2 or $L2_1$ structures in such thin CFA films. In our previous work, PMA was observed in a full-Heusler alloy film such as a Tb-CFA co-sputtered film,[19] Ni/CFA multilayer, and Pt/CFA/MgO trilayer.[20] In the Pt/CFA/MgO trilayer, we successfully achieved PMA in a CFA film as thick as 2.5 nm. In the present paper, we conduct a detailed study on the microstructure and magnetic property of the Pt/CFA/MgO structure. The cross-sectional high resolution transmission electron microscopic (HRTEM) images show that there exists a CFA layer with a thickness of 2.5 nm in the B2-ordered structure. We also investigate the effects of conversions between in-plane magnetic anisotropy (IMA) and PMA on the temperature dependence of magnetic moment and the variation of saturation magnetization (M_s) with annealing temperature (t_{an}) and the thickness of MgO layer (t_{MgO}).

2. Experiment

A sample with a structure of Si/SiO_2 (500)/Ta (3)/Pt (20, 40)/CFA (2.0, 2.5)/MgO (t_{MgO}) (thicknesses in nanometers) (as shown in the inset of Fig. 1) was deposited by magnetron sputtering on a thermally oxidized Si(100) substrate. The base pressure before sputtering was lower than 9.0×10^{-6} Pa. The annealing process was carried out in a vacuum chamber at a pressure of better than 2×10^{-4} Pa for 0.5 h in the absence of an external magnetic field. For comparison, a sample with a stack of Ta(3)/Pt(5)/CFA(2)/Pt(4) without MgO was prepared under the same conditions. The magnetic properties were studied by alternating the gradient magnetometer (AGM) and vibrating the sample magnetometry (VSM). The surface roughness and cross sectional microstructure were characterized by atomic force microscopy (AFM) and HRTEM, respectively.

3. Results and discussion

Figure 1 shows the hysteresis loops for the Ta (3)/Pt (20)/CFA (2.0)/MgO (0.5) structure annealed at 250 °C. It is obvious that the CFA film is of PMA with a small perpendicular saturation field (H_s). The PMAs of our samples and other similar structures can be achieved only after annealing.[18,20,21] Since the CFA film is a soft magnetic material,[22] the magnetic moments are difficult to magnetize perpendicularly, thereby leading to a low remanence ratio.

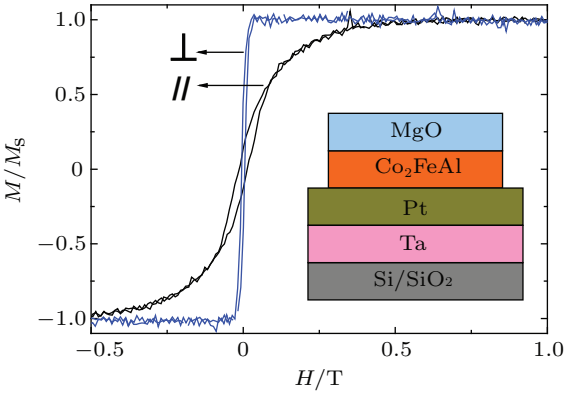


Fig. 1. (colour online) M–H loops for the Ta (3)/Pt (20)/CFA (2.0)/MgO (0.5) structure annealed at 250 °C, and the inset shows a schematic plot of the structure.

In order to understand the interface information, HRTEM is employed to study the cross-sectional structure of the Ta (3)/Pt (40)/CFA (2.5)/MgO (0.5) sample annealed at 350 °C. The interfaces of Ta/Pt and Pt/CFA can be seen clearly from Fig. 2(a), but the CFA/MgO interface cannot be distinguished. Since the sample still shows PMA after annealing at 350 °C, a high-quality interface between the ferromagnetic and oxide layers is not essential to the inducing of PMA. The thick Pt layer shows a columnar structure with a large roughness of about 0.81 nm characterized by AFM measurement. The large roughness together with the diffusion during the annealing process may lead to the unclear CFA/MgO interface. However, the thickness of the CFA layer is 2.5 nm and twice larger than that in Ref. [18]. From the high magnification image shown in Fig. 2(b), lattice fringes of Pt can be seen clearly. The lattice spacings of $d \approx 0.22$ nm and 0.20 nm correspond to the Pt(111) and Pt(200) planes, respectively. In the CFA film, a B2-ordered structure is evidenced by the lattice spacing of $d_{CFA(200)} \approx 0.28$ nm.

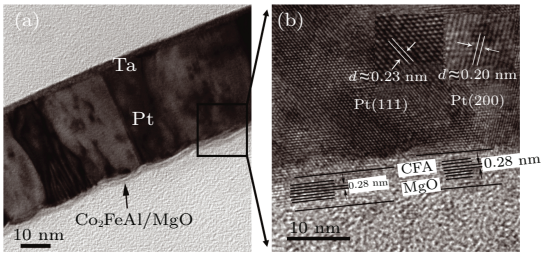


Fig. 2. Cross-sectional HRTEM images of the Ta (3)/Pt (40)/CFA (2.5)/MgO (0.5) structure annealed at 350 °C. Panels (a) and (b) are low and high magnification images, respectively.

In order to study the temperature dependence of magnetic moment, a M–T curve of the as-prepared sample is characterized by VS under a 0.25-T perpendicular field and presented in Fig. 3. Since the as-prepared sample is of IMA with a large perpendicular H_s (about 0.6 T),[20] the magnetic moment is not saturated while the temperature is low, with the magnetic easy axis kept unchanged from in-plane to out-of-plane. From the curve, it can be seen that the Curie temperature T_c of the sample is not reached yet even though the temperature has arrived at 500 °C and the value reported is extremely high (about 900 °C).[23]

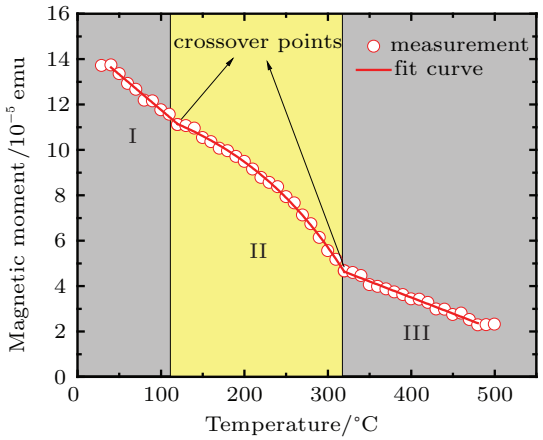


Fig. 3. (colour online) An M–T curve measured under a perpendicular field of 0.25 T for the Ta(3)/Pt(20)/CFA(2.0)/MgO (0.5) structure.

It is interesting that there are two crossovers for the slope of the curve, which is unexpected. The evolution can be observed in the fit curve, which is different from the scenario of other PMA systems, owing to the special mechanism of the PMA origination.[24,25] The curve can be divided into three parts according to the changes of the slope. The magnetic moment decreases linearly with temperature increasing in parts I and III and reduces exponentially in part II. The two crossover points are at about 110 °C and 310 °C, which are close to the experimental PMA critical temperatures of 150 °C and 350 °C, respectively. It is reasonable to consider that the two intersecting points in the curve are consistent with both critical conversions between IMA and PMA, and the difference of about 40 °C can be attributed to the different measuring temperatures. Therefore, the sample has PMA and is saturated at 110 °C. In part II, the magnetic moment decreases slowly at first, and then sharply. The slow decrease of magnetic moment may be induced by the optimized average surface roughness (R_a) and the improved CFA ordering structure in the heating process.[26,27] As the temperature further increases, the accelerated diffusion and over oxidation at the interface leads to a sharp decrease of the magnetic moment. The promotion of oxidation results in an enhancement of hybridization between Co (Fe) 3d and O 2p, which is confirmed by H_c increasing with t_{an} going up to 350 °C.[20]

Figure 4 shows the M_s of Ta(3)/Pt(20)/CFA(2.0)-/MgO(0.5) structure as a function of t_{an} For the sample with ultrathin CFA ($t_{CFA} < 0.5$ nm), a very weak magnetic signal is observed, indicating the over oxidation of the CFA film. Therefore, it is reasonable that the M_s of the as-prepared sample (about 815 emu/cm^3) is lower than that of the pure CFA. The effective thickness of the CFA layer is estimated to be about 1.6 nm compared with that of the Ta(3)/Pt(5)/CFA(2)/Pt(4) structure. The Ta/Pt/CFA/Pt structure is of IMA even after annealing, demonstrating that the main PMA origination of the Ta/Pt/CFA/MgO structure is the hybridization between Co (Fe) 3d and O 2p orbitals, instead of the Pt/CFA interface. The M_s reduces after annealing and increases gradually from 150 °C to 400 °C except for the point of 350 °C. Two reasons may correspond to the M_s development with t_{an}. The effective magnetic layer decreases due to the diffusion between the CFA and Pt layers and oxidation in the annealing process. On the other hand, superparamagnetic islands may form in the discontinuous and discrete regions due to the rough Pt layer and the thin CFA layer.[24] As the flatness of the sample is improved with t_{an} increasing, some of the superparamagnetic islands disappear gradually, which may play a more important role when the increase of M_s with t_{an} varying from 150 °C to 250 °C is taken into account. In addition, it was reported that the chemical order and local stoichiometry in the CFA film can be improved by increasing t_{an}, which may also lead to a little increase of M_s.[27] The sudden dropping of M_s at 350 °C might be related to the over oxidation and serious diffusion. The variation of M_s as a function of t_{MgO} is also studied and shown in Fig. 5. The M_s decreases monotonically with the increase of MgO thickness. The variation of M_s indicates that the thicker MgO can lead to a more serious oxidation. In this way, the degree of oxidation as well as the strength of the PMA can be accurately regulated by carefully controlling the MgO thickness and annealing conditions.

According to our experimental results, the PMA of Ta/Pt/CFA/MgO structure with 0.5-nm MgO

shows a strong dependence on t_{an} and t_{CFA}. Figure 6 shows the PMA area with the free parameters of t_{an}, and t_{CFA} and PMA appears within the crossing space.

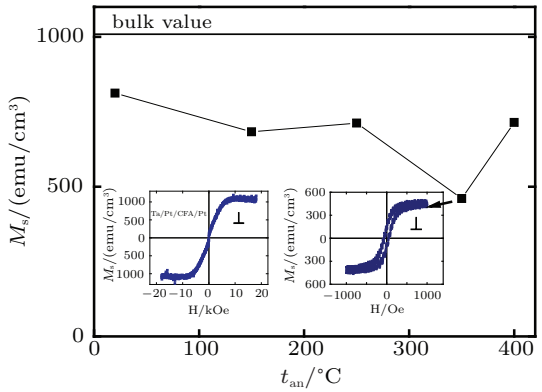


Fig. 4. (colour online) Dependence of M_s on t_{an} for the Ta(3)/Pt(20)/CFA(2.0)/MgO(0.5) structure. Insets show the perpendicular M–H loops for the as-deposited Pt(5)/CFA(2)/Pt(4) structure (left one) and the Ta(3)/Pt(20)/CFA(2.0)/MgO(0.5) structure after 350 °C annealing under a small field (right one), 1 Oe = 79.5775 A/m.

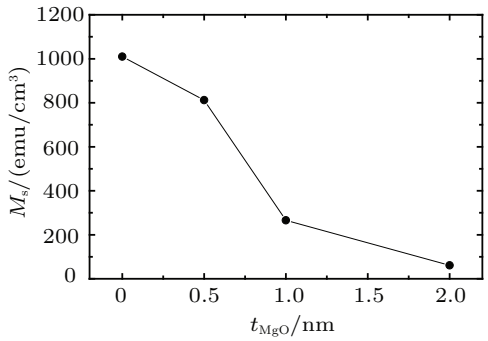


Fig. 5. M_s as a function of t_{MgO} for the as-deposited Pt(20)/CFA(2)/MgO (t_{MgO}) samples.

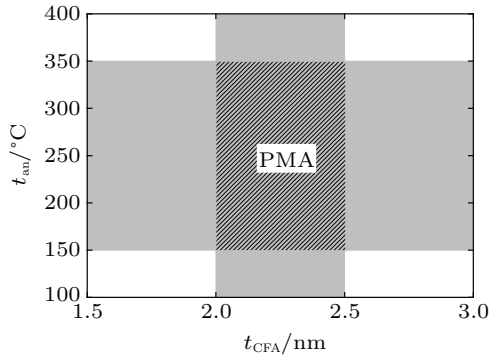


Fig. 6. Schematic diagram showing the region of PMA (cross space) for the Ta/Pt/CFA/MgO structure with 0.5-nm MgO.

4. Summary

In the present paper, we studied the microstructures and magnetic properties of the Pt/CFA/MgO multilayers. A B2-ordered structure was observed in a 2.5-nm CFA film by cross-sectional HRTEM. The variation of M_s depends on the conversion between IMA and PMA. The thickness of the MgO layer also has a strong effect on the magnetic property of CFA and the degree of oxidation at the CFA/MgO interface. The annealed Pt/CFA/MgO structure has potential applications in the high spin polarized SPVs and spin transfer torque in MgO-based MTJs.

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