Microstructural evolution of perpendicular magnetization films with an ultra-thin Co$_2$FeAl/MgAl$_2$O$_4$(001) structure

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A B S T R A C T
We investigated the structure of 1-nm-thick Co$_2$FeAl (CFA)/MgAl$_2$O$_4$ (MAO) epitaxial heterojunctions in order to understand the origin of perpendicular magnetic anisotropy (PMA). The CFA/MAO stacks were fabricated by plasma oxidation of Mg-Al grown on the CFA layer and subsequent post-annealing process. Before post-annealing (as-oxidized), the stack showed in-plane magnetization with poor crystallinity of the MAO layer. After post-annealing, the stack showed PMA accompanied by the improved crystallinity in the MAO layer, leading to the formation of a lattice-matched CFA/MAO(001) epitaxial interface. These behaviors indicate that the crystallization of the MAO layer is strongly promoted by atomic templating effect from a lattice-matched underlayer. In addition, we also found that this MAO crystallization effectively enhanced PMA at the interface. At the same time, significant Al atomic diffusion from CFA to MAO was confirmed. This Al diffusion was observed in the as-oxidized stack, and this was further promoted by the post-annealing. Therefore, the strong PMA at the CFA/MAO interfaces is attributed to (i) the reduced in-plane magnetocrystalline anisotropy due to the lattice-matching and (ii) the promoted hybridization between Fe and O orbitals due to the Al re-distribution near the interface. These results include important findings for constructing high performance perpendicularly-magnetized magnetic tunnel junctions for non-volatile magnetic memory applications.

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1. Introduction

Successful engineering of magnetic tunnel junctions (MTJ) consisting of a ferromagnetic metal (FM)/non-magnetic tunnel barrier (barrier)/FM trilayer has laid the foundation for the practical application of spintronic devices such as read heads of a hard disk drive and non-volatile magnetic random access memory (MRAM) [1–3]. For these applications, CoFeB based FM layers and an MgO barrier are commonly used for MTJs since large tunnel magnetoresistance (TMR) ratios are observed in their combination, i.e., CoFeB/MgO/CoFeB. The large TMR ratios originate from the spin-dependent coherent tunneling mechanism through the $\Delta_1$ bands of bcc CoFeB(001) and MgO(001). However, it is known that a large number of misfit dislocations are introduced at their interfaces due to the existence of 3–4% of lattice mismatch, making it difficult to improve the performance of current MgO-based MTJs further. Recently, MgAl$_2$O$_4$ (MAO) spinel has been attracting attention as a new barrier for MTJs since MAO shows good lattice-matching with various FM materials, e.g., lattice mismatches with Fe(001) $\sim$ –0.2%, Co$_5$Fe$_5$S$_9$(001) $\sim$ 0.3%, and Co$_2$FeAl (CFA)(001) $\sim$ –0.2% [4–7]. MAO-based MTJs have shown large TMR ratios exceeding 300% at room temperature (RT) [8,9], indicating TMR enhancement by the coherent tunneling effect, similar to MgO-based MTJs. It was found that the good lattice-matching with an MAO barrier significantly suppresses the bias voltage dependence of TMR ratios [5,10,11], suggesting the importance of creating dislocation-free barrier interfaces for good spin-dependent tunneling.

An MAO barrier can be produced on an epitaxial film by post-oxidation of an Mg-Al based layer such as an Mg/Al bilayer [4,5] or an MgAl alloy layer [6,8–10,12]. Also, achievement of a high-quality and flat MAO(001) barrier by direct sputtering of a sintered MgAl$_2$O$_4$ target was recently reported [13]. These results indicate that various preparation methods are available for creating an epitaxial MAO barrier. However, obtaining a (001)-oriented crystalline MAO thin film is relatively difficult; an epitaxial template layer may be necessary for forming an epitaxial MAO layer. Especially, a crystalline MAO layer cannot be directly grown on an amorphous layer such as CoFeB [14], which is in contrast to MgO layers [15]. This difference may be related to differences in chemical
Characteristics between MgO and MAO; e.g., MAO has a greater covalent nature and a higher chemical stability than MgO [16]. Therefore, it is essential to understand the crystallization mechanism of MAO in order to improve the TMR properties of MAO-based MTJs.

In addition to a large TMR ratio, miniaturization of MTJs down to below a few tens of nm is required for practical application to high-density MRAMs and magnetic logics. In order to attain high thermal stability to ensure long data retention in such a small MTJ cell, it has become increasingly important to obtain a perpendicularly magnetized MTJ (p-MTJ) [17–23]. It is known that perpendicular magnetic anisotropy (PMA) appears in an ultrathin FM layer adjacent to an oxide layer, such as Co/AlOx [24–26], CoFeB/MgO [17,19,27], or Fe/MgO [26,28–34], which is a suitable form for obtaining p-MTJ structures with large PMA energies and TMR ratios. This interfacial PMA at 3d-transition FM/oxide interfaces is attributed to the anisotropic orbital magnetic moments of FM induced by the spin-orbit interaction at the interface [26,29]. Therefore, the PMA characteristics are significantly affected by the interfacial electronic states, which are mainly determined by the atomic arrangement near the interface [26,29–31]. This indicates that controlling the interfacial oxidation state and reducing the dislocation/defect density near the oxide interface are indispensable for improving the interfacial PMA energy. Heusler alloy type CFA/MgO(001) interfaces were also reported to exhibit high interfacial PMA energies, e.g., 1.0 erg/cm² [35], 0.65 erg/cm² [36], 0.77–0.95 erg/cm² [37], and 2.2 erg/cm² [38]. However, high-resolution transmission electron microscopy (TEM) analysis of the CFA/MgO interfaces revealed significant Al diffusion from the CFA layer into the MgO layer and the resulting loss of the Heusler-like local crystal structure of the CFA layer near the MgO interface [39]. Nevertheless, it was suggested that this Al redistribution played an assisting role in enhancing PMA, thus demonstrating its importance in controlling the local interfacial structure and chemistry.

Lattice-mismatch between an ultra-thin FM layer and a barrier can cause large magnetocrystalline (bulk) anisotropy due to the induced lattice distortion of the FM layer. In the case of MgO barrier interfaces such as Fe(001)/MgO(001) (mismatch for bulk ~3.9%) [34] and CFA(001)/MgO(001) (~4.0%) [38], negative bulk anisotropy substantially reduces PMA. In contrast, a large interfacial PMA was observed in a lattice-matched CFA/MAO(001) epitaxial interface due to the reduction of the bulk anisotropy [40]. However, the interfacial crystal structure and elemental distribution near the CFA/MAO interface have yet to be clarified. Understanding the mechanism of the observed PMA as well as the epitaxial growth process of the MAO layer through monitoring the nano-scale structural evolution is indispensable to further improvement of PMA using an MAO interface.

In this article, we investigate ultrathin CFA (1 nm)/Mg-Al(-O) stacks using high resolution scanning transmission electron microscopy (STEM) imaging. First, we prepared three types of samples in order to evaluate how the multilayered structure and magnetic properties evolve during three different processing steps: a non-oxidized sample (CFA/MgAl), an as-oxidized sample (CFA/MAO), and an oxidized and post-annealed sample showing strong PMA (CFA/MAO/annealing [40]). Next, the elemental distributions of the as-oxidized and post-annealed samples were characterized using energy dispersive spectrometry (EDS) to clarify the possible PMA mechanism in the CFA/MAO stacks.

2. Experimental procedures

Epitaxial multilayers with CFA/MgAl or CFA/MAO structures were deposited on a Cr-buffered MgO(001) single crystalline substrate using magnetron sputtering at a base pressure less than 7 × 10⁻¹⁰ Pa. The stacking for the observed structures was Cr (40)/Co/FeAl (10)/Mg (0.2)/MgₙAlₘ (0.65)/no process or oxidation process)/Ru (2)/[non-annealed or post-annealed] (thickness in nm). Three stacks were prepared: (i) non-oxidized and non-annealed, (ii) as-oxidized (non-annealed), and (iii) oxidized and post-annealed. Insets of Fig. 1a–c shows their schematic structures. The oxidized barrier was formed by oxidizing the Mg/MgₙAlₘ bilayers through direct inductively coupled plasma (ICP) oxidation for 15 s using an Ar + O₂ mixture gas [10]. The fabricated sample (iii) was then ex-situ annealed at 275 °C (548 K) under a base pressure of 1 × 10⁻⁵ Pa to obtain perpendicular magnetization. Note here that the annealed samples were partly investigated in the previous work [40].

In-plane and out-of-plane magnetization hysteresis loops for the three samples were measured at RT using a vibrating sample magnetometer (VSM). Microstructural characterization was performed using an FEI Titan G2 80–200 Transmission Electron Microscope equipped with a spherical-aberration probe corrector. Cross-sectional TEM specimens were prepared by the lift-out technique using an FEI Helios Nanolab 650 Focused Ion Beam (FIB); protective coatings of Ni and Pt were applied during this process. STEM was used to evaluate the nanostructure of the layers; nanobeam electron diffraction (NBED) and EDS were utilized for crystallographic and chemical characterization, respectively. For all TEM analyses, the material was analyzed along the [100] zone axis of the MgO substrate.

3. Results

3.1. Magnetic properties

Fig. 1a–c shows the in-plane and out-of-plane magnetization curves for the non-oxidized, as-oxidized, and annealed samples, respectively. Perpendicular magnetization is observed only in the annealed sample [40]. The non-oxidized and the as-oxidized samples exhibit in-plane magnetization since the interfacial magnetic anisotropy, Kₐ, cannot overcome the shape anisotropy, which corresponds to 2πM² (for cgs units where M is the saturation magnetization). The Mₛ values and effective magnetic anisotropy, Kₑff, for each sample are plotted in Fig. 1d and e, respectively. Here, the positive Kₑff indicates perpendicular magnetization. Both Mₛ and Kₑff increase after both the oxidation and the post-annealing processes, indicating changes in chemical ordering and composition in the CFA layer and/or near the CFA/MAO interface after each process. It is seen that the Kₑff value is significantly enhanced by the post-annealing process. Also notably, the Mₛ value of the annealed sample reaches 1200 emu/cm³, which is greater than the value for bulk CFA (985 emu/cm³ [41]).

3.2. Microstructural evolution during fabrication

In a previous work, STEM imaging of an annealed CFA/MAO stack confirmed the crystallization of the MAO layer and formation of a lattice-matched CFA/MAO interface [40]. It was also reported that the interfacial PMA energy, Kₑ, at the CFA/MAO interface was maximized when optimal oxidation conditions and annealing temperature were selected. This indicates that the magnitude of the PMA energy is closely related to the interfacial states determined by such factors as crystallization of MAO, atomic diffusion, and degree of oxidation. Therefore, in this work, we monitored the microstructural evolution during different stages of processing to clarify the PMA mechanism.

Fig. 2a–c shows the annular dark field (ADF)-STEM images of the (a) non-oxidized, (b) as-oxidized, and (c) annealed stacks, respectively. In this observation, diffraction contrast was employed using a high camera length to reveal the atomic structures, which
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