Structural instability of FeCo ultrathin films grown on MgO(100)

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Abstract. Structures and their stability of FeCo ultrathin films grown on MgO(100) were investigated. Atomic force microscopy observations revealed dramatic structural changes from a continuous ultrathin layer to coarse islands by low temperature annealing. The results suggest that the thin layered structures of Fe based alloys as grown on MgO(100) are not so stable as expected from the relatively small lattice mismatch between Fe based alloys and MgO. This possibly explains annealing temperature dependence of tunnel magnetoresistance observed in a Fe/MgO/Fe(100) junction.

1. Introduction
Structures and their stability of thin films are of fundamental importance in science and technologies and are not fully understood in a wide range of materials although a great number of studies have been performed on thin film growth to date. Fe and Fe-based alloy thin films grown epitaxially on MgO(100) attract attention since MgO barrier magnetic tunnel junctions (MTJs) with Fe-based alloy electrodes play a significant role in current spintronic devices [1]. In this study, structures and their stability of Fe-based alloy “ultrathin” films grown on MgO(100) were investigated. The reason for choosing ultrathin films is that the effects of interfaces and/or epitaxial strain should appear clearly in thinner films. Annealing experiments were performed to see the stability (dynamic changes) of the film structures against (due to) epitaxial strain etc.

2. Experimental Procedure
Samples were prepared on a MgO(100) substrate by using an ultrahigh vacuum electron beam evaporator. The stacking structure is as follows: MgO(100) subs./MgO buffer 5nm/Fe 5nm/FeCo 50nm/MgO 2nm/FeCo 1.0 and 1.4nm. The Fe/FeCo bottom layers were annealed prior to the deposition of MgO and top FeCo layers. The evaporation sources used for MgO, Fe and FeCo layers were MgO (> 99.9at.%), Fe (> 99.95at.%) and Fe₅₀Co₅₀ (> 99.9at.%) blocks, respectively. In-situ reflection high-energy electron diffraction (RHEED) and ex-situ atomic force microscopy (AFM) were used for structural characterization. MTJs of MgO(100) subs./MgO buffer 10nm/Fe 60nm/MgO 2.1nm/Fe 5nm/IrMn 15 nm/Pt 7nm were also prepared to check the effect of post-annealing of tunnel magnetoresistance (TMR) in MTJs with a similar stacking structure. For the MTJs, top Fe/IrMn/Pt layers were deposited by sputtering.

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3. Results and discussion

Figure 1 shows RHEED patterns and AFM images for the sample with a 1.0 nm thick top FeCo layer. The streak RHEED pattern for the FeCo bottom layer annealed at 500°C shows that the layer is epitaxially grown and its surface is rather flat. The 2 nm thick MgO layer was also grown epitaxially. The RHEED pattern revealed that epitaxial strain is relaxed in the MgO lattice [2] and its crystallinity is poor, compared with MgO substrates. The RHEED patterns for 1.0 nm FeCo were spotty, indicating that island growth occurred on MgO(100). No significant changes appeared in the RHEED patterns through the post-annealing process in the temperature range from 200°C to 400°C. The weak streaks observed for the annealed states presumably come from the formation of facets of the FeCo islands. The AFM images for the FeCo islands at the annealed states correspond to the interpretation of the RHEED patterns. Considering the convolution effect of the AFM observations, the sizes of islands were roughly estimated to be 10 nm or smaller and 10 – 20 nm for 200°C- and 400°C-annealed states, respectively.

![RHEED patterns and AFM images](image-url)

Fig. 1. (a) RHEED patterns for each layer of a sample with 1.0 nm thick FeCo, including the results of an annealing process, AFM images for the FeCo layer annealed (b) at 200°C and (c) at 400°C.
Figure 2 shows a RHEED pattern and a AFM image for the sample with a 1.4 nm thick FeCo after annealing at 200°C for 10 min. Although the RHEED pattern is similar to that for the 1.0 nm thick FeCo annealed at 200°C, the result of the real space observation (Fig. 2(b)) is completely different. Namely, a number of large islands of a 100 nm scale appeared for the 1.4 nm thick FeCo after low temperature annealing. It has been confirmed by a previous study [3] and measurements of electrical resistivity and magnetization curves [4] that continuous layers of Fe and FeCo alloys are formed in the as-deposited state when the nominal thickness is larger than around 1.2 nm, while AFM is not useful to determine the critical thickness between continuous and discontinuous films due to the appearance of nanoparticle-like images even for continuous layers. Thus, the clear contrast in the AFM images (Figs. 1(c) and 2(b)) show that a dramatic structural change from a continuous layer to coarse islands occurred by low temperature annealing for the 1.4 nm thick FeCo layer. It is noted that a coarse-island structure of Fe on MgO, similar to that in Fig. 2(b), was also observed by transmission electron microscopy in a study on double tunnel junctions [5] and that the experiments using pure Fe instead of FeCo in this study showed almost the same results as the FeCo case.

The lattice mismatches between most of body-centered-cubic Fe-based alloys and MgO are relatively small (2-3 %), so that epitaxial growth of Fe-based alloy films on MgO(100) is easily achieved as is supported by many experiments. In this sense, it could be expected that thin layered structures of Fe-based alloys are stable on MgO(100) surfaces. However, the present study suggests that as-deposited Fe-based alloy layers grown on MgO(100) is not so stable as expected from the small lattice mismatches, probably due to strain and defects introduced in the growth process. In addition, the present results do not meet a simple idea that continuous layers are in general more stable than nanoparticles. Namely, in thin film growth processes, one may consider that the system becomes stable at the moment when the film structure changes from nanoparticles to a continuous layer with increasing the nominal thickness, but the present results presumably show that the one’s consideration is not in general correct.

The instability observed for the thin Fe-based alloy layers grown on MgO(100) could affect atomistic and/or layered structures even for thicker layers in actual spintronic devices. Concerning this idea, post-annealing temperature dependence of tunnel magnetoresistance was investigated for a Fe 60nm/MgO 2.1nm/Fe 5nm MTJ, in which no annealing was performed for the 5-nm Fe top electrode in the growth process. Fig. 3 shows magnetoresistance (MR) ratios and resistance-area products (RA) of the MTJ as a function of post-annealing temperature. At a relatively low temperature of 225°C, MR ratio takes a peak value, suggesting that low temperature annealing is sufficient to modify the MTJ...
structure for obtaining larger TMR. Further annealing above 225°C much affected the structure of the MTJ, resulting in degradation of TMR.

Considering the magnitude of lattice mismatch and/or the strength of chemical bonding to MgO, stability of Fe-based alloy thin layers should depend on the constitute elements, and therefore the results and discussion would be limited within simple Fe-based alloys such as Fe and FeCo. If the constitute elements are not only Fe and its neighbours, behaviour in annealing experiments is quantitatively different. Higher optimal annealing temperatures for TMR were reported in MTJs with Co2FeAl [6] and CoFeB [7,8].

4. Summary
Structural evolution of FeCo ultrathin films grown on MgO(100) were investigated by using RHEED and AFM. It was revealed that dramatic structural changes occurred by low temperature annealing, showing that the thin continuous layered structures of Fe based alloys are not so stable as expected from the small lattice mismatch. This possibly explains annealing temperature dependence of TMR in a Fe-based MTJ.

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