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ABSTRACT
FeAlSi epitaxial films were fabricated on single crystalline MgO(100) substrates and their structural and magnetic properties were investigated to apply them to the free layer in magnetic tunnel junction (MTJ) based sensors. We found that the film composition must be precisely controlled, and the post-annealing temperature was varied to obtain a D0 3 -ordered structure and soft magnetic property. By adjustment of the film composition and optimizing annealing temperature, we succeeded in obtaining D0 3 -ordered FeAlSi thin films with a small surface roughness and low coercivity. The fabricated FeAlSi film is greatly useful as a free layer material in MTJ based sensor devices.

INTRODUCTION
The large tunnel magnetoresistance (TMR) effect in magnetic tunnel junctions (MTJs) with (001)-oriented MgO barriers was discovered, 1,2 and the MTJs are expected to be applied for highly sensitive magnetic field sensors such as bio-magnetic field sensors. For the sensor applications, MTJs are required to show high sensitivity and low noise. The sensitivity of sensor devices is defined as TMR/2Hk, 3 where Hk is the magnetic anisotropy field of the free layer. Therefore, the high TMR ratio and low magnetic anisotropy are required to achieve high sensitivity. For example, a high sensitivity of over 100%/Oe is required in 100×100 MTJ arrays to detect the bio-magnetic field such as human brain. 1 To get high sensitivity in MTJ sensor devices, various ferromagnetic materials such as Ni–Fe based alloys, 5 amorphous CoFeSiB, 6 and superparamagnetic CoFeB thin films 6 have been studied as free layer materials in MTJs. In our previous work, we observed a large sensitivity of 25%/Oe in an MTJ with a NiFe/Ru/CoFeB free layer 6 and 40%/Oe in an MTJ with a CoFeSiB/Ru/CoFeB free layer. 8 We have also demonstrated magnetoencephalography (MEG) measurements at room temperature by using such highly sensitive MTJs. 10 However, we need to improve the sensitivity of MTJ sensors to detect weaker bio-magnetic signals. One candidate to achieve both high TMR and small magnetic anisotropy is FeAlSi alloy. Fe 85 Al 5.4 Si 9.6 (in wt.%, named Sendust) alloy that has a D0 3 -ordered structure shows a very low magnetic anisotropy. 11 Its soft magnetic property is associated with the D0 3 -ordered structure at the optimum composition of Fe 85 Al 5.4 Si 9.6 (called Sendust center composition). The permeability of FeAlSi alloys is very sensitive to composition of the sample and sharply increases at the Sendust center composition. The permeability of FeAlSi alloys is very sensitive to composition of the sample and sharply increases at the Sendust center composition. In addition, a large TMR effect through the coherent tunneling is expected in MgO-based MTJs by using FeAlSi electrodes because of the structural and compositional similarity to bcc-Fe 1 12 and the good epitaxial relationship with the MgO tunneling...
FIG. 1. Annealing temperature dependence of the film composition for Sample A and B prepared by Fe$_{83.6}$Al$_{6.7}$Si$_{9.7}$ and Fe$_{81.6}$Al$_{7.7}$Si$_{10.7}$ sputtering targets.

barrier. Therefore, both low magnetic anisotropy and a high TMR ratio can be achieved by using D0$_3$-ordered FeAlSi film as the free layer in MTJs. Moreover, we can make the MTJ structure simpler than those of the previous MTJ sensor devices with NiFe/Ru/CoFeB or CoFeSiB/Ru/CoFeB synthetic free layers. This simplification of the MTJ structure will be a great benefit for the development of products.

However, although there are some reports on thick (μm-order) FeAlSi thin films for soft-magnetic property, thin films with nm-order thickness, which is needed for the MTJ devices, has never been investigated. In this study, we fabricated epitaxial FeAlSi films on single crystalline MgO(001) substrates by a magnetron sputtering technique. The post-annealing temperature and the film composition were varied to obtain FeAlSi thin films with a D0$_3$-ordered structure, (001)-orientation, smooth surface roughness, and soft magnetic properties to apply them to the free layer of MTJ-based sensor devices.

EXPERIMENT

We fabricated MgO(20)/FeAlSi(30)/Ta(5) (in nm) films onto MgO(001) substrates for the free layer in MTJs to evaluate the epitaxial growth, D0$_3$-ordered structure, and soft magnetic properties of FeAlSi films. The films were deposited using magnetron sputtering ($P_{\text{base}} < 2 \times 10^{-6}$ Pa), and Ar pressure was 1.0 Pa for the MgO.

FIG. 2. XRD patterns (2θ-θ scan) for FeAlSi films with various annealing temperatures for (a) Sample-A and (b) Sample-B. φ scan profiles FeAlSi films annealed at various temperatures for (c) Sample-A and (d) Sample-B.
buffer layer and 0.1 Pa for the others. Two sputtering targets of Fe$_{83.6}$Al$_{6.7}$Si$_{9.7}$ and Fe$_{81.6}$Al$_{7.7}$Si$_{10.7}$ were used for FeAlSi film preparation to investigate the influence of film composition on the structural and magnetic properties. The composition for FeAlSi must be controlled to achieve the soft magnetic property as mentioned above. The prepared film composition resulted in Fe$_{86.7}$Si$_{8.7}$Al$_{4.7}$ (Sample-A) and Fe$_{83.8}$Si$_{9.2}$Al$_{7.2}$ (Sample-B) as deposition. Note that the composition of sputtered FeAlSi films is generally deviated from the target composition. The composition of the films was measured by the inductively coupled plasma (ICP) spectrometry for as deposited films. After being deposited, all films were annealed at $T_a = 300 - 500$ °C for 1 hour. The film composition measured with X-ray fluorescence (XRF) spectrometry slightly changed by the annealing process as shown in Fig. 1. Their crystal structure, surface roughness, and magnetic properties of FeAlSi films were measured using X-ray diffraction (XRD), an atomic force microscope (AFM), and a vibrating sample magnetometer (VSM), respectively.

**RESULTS AND DISCUSSION**

Figure 2(a) and (b) show the XRD patterns (2θ-θ scan) for prepared FeAlSi films annealed at various annealing temperatures. (004) peaks from A2 structure of FeAlSi were observed for both film compositions regardless of the annealing temperature. The result indicates that the prepared FeAlSi films on MgO substrates have (001)-orientation. In addition, (002) peaks from B2-ordered FeAlSi were clearly observed at $T_a \geq 400$ °C for Fe-rich Sample-A and $T_a \geq 300$ °C for Fe-poor Sample-B. We confirmed that the FeAlSi films possess a B2-ordered structure after the post-annealing process and the B2-ordering temperature depends on the film composition. Moreover, the intensity of (002) peaks increased as annealing temperature increased. The result means that B2-ordering in the films was improved by the increasing annealing temperature. Fig. 2(c) and (d) show the results of φ-scan measurements for FeAlSi films with various annealing temperature. Four-fold symmetric (111) peaks from D$_{03}$-ordered structure were observed at $T_a = 500$ °C (Sample-A) and $T_a \geq 400$ °C (Sample-B). This is the first successful fabrication of D$_{03}$-ordered FeAlSi epitaxial thin films with nm-order-thickness on MgO substrates, and we found that the ordering temperature for both D$_{03}$- and B2-structures depends on the film composition.

Figure 3(a) shows the annealing temperature dependence of average surface roughness $R_a$ measured by AFM for FeAlSi films. For Sample-A, the surface roughness was improved by the post-annealing at 300 - 500 °C for 1 hour. The film composition measured with X-ray fluorescence (XRF) spectrometry slightly changed by the annealing process as shown in Fig. 1. Their crystal structure, surface roughness, and magnetic properties of FeAlSi films were measured using X-ray diffraction (XRD), an atomic force microscope (AFM), and a vibrating sample magnetometer (VSM), respectively.
fabrication of thin tunneling barrier layer on it, and the obtained D0₃-ordered films can be thus used as bottom electrodes of MTJ devices.

Figure 4 (a) and (b) show magnetization curves measured along [100] direction of FeAlSi films with various temperature for Samples-A and B, respectively. In Fig. 4 (a), Sample-A showed a small coercivity at $T_a = 400^\circ$C due to the D0₃-ordering, but the coercivity increased at $T_a = 500^\circ$C as summarized in Fig. 4 (c), even though the films had a D0₃-ordered structure. We infer that the small lattice distortion at $T_a = 500^\circ$C observed as the slight peak shift of XRD in Fig. 1 (a) contributes increase in the coercivity. On the other hand, for Sample-B, the as-deposited film showed a slightly large coercivity, but the coercivity decreased as the annealing temperature increased because of the improvement of D0₃-ordering by the annealing process. A minimum coercivity was 1.8 Oe for Sample-A with $T_a = 400^\circ$C and for Sample-B with $T_a = 500^\circ$C. The observed small $H_c$ is comparable to NiFe and CoFeSiB free layers in the MTJ sensor devices. We also measured the magnetization curve for Sample-B with $T_a = 500^\circ$C measured along [110] direction (not shown) and the magnetic anisotropy field $H_K$ was ca. 13 Oe and the evaluated crystalline anisotropy constant $K_1$ was $5.7 \times 10^8$ erg/cc. These values are larger than those for bulk FeAlSi and slightly larger than induced magnetic anisotropy for polycrystalline NiFe films and amorphous CoFeSiB films. Further improvement of the quality for the FeAlSi films is needed by optimization of the film composition and deposition conditions to decrease magnetic anisotropy.

**SUMMARY**

We fabricated FeAlSi films with nm-order-thickness and investigated their structural and magnetic properties. The D0₃-ordered epitaxial FeAlSi thin films with (001)-orientation were successfully fabricated on single crystalline MgO(100) substrates by the post-annealing process. We found that the B2- and D0₃-ordering temperature and the morphology for the FeAlSi films depend sensitively on the film composition. The D0₃-ordered FeAlSi films showed a small surface roughness of $R_a \approx 0.3$ nm even after the post-annealing process. We also achieved a low coercivity of 1.8 Oe in the D0₃-ordered FeAlSi films. The obtained coercivity is comparable to those for free layers in MTJ based sensor devices such as NiFe and CoFeSiB. Although the further reduction in the magnetic anisotropy is needed, the developed D0₃-ordered FeAlSi thin films with (001)-orientation, smooth surface, and soft magnetic property is a promising candidate for the free layer of MTJ based sensors.

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