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Transmission electron microscopy of $Co_2(Cr_{1-x}Fe_x)Al$ sputtered films and their magnetic tunneling junctions

Y.K. Takahashi[a,*], T. Ohkubo[a], K. Hono[a], S. Okamura[b], N. Tezuka[b], K. Inomata[b]

[a]National Institute for Materials Science (NIMS), 1-2-1 Sengen, Tsukuba 305-0047, Japan
[b]Department of Materials Science, Graduate School of Engineering, Tohoku University, Sendai 980-8579, Japan

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Abstract

The microstructures of Co_2FeAl and $Co_2(Cr_{0.4}Fe_{0.6})Al$ sputtered films and of their magnetic tunnel junctions (MTJs) have been investigated to discuss the possible reasons for an unexpectedly low tunneling magnetoresistance (TMR). The structure of the Co_2FeAl film changed from B2 to $L2_1$ with increasing substrate temperature, while that of the $Co_2(Cr_{0.4}Fe_{0.6})Al$ film remained B2 up to 500 °C. The thermodynamically predicted phase separation was not observed in the films. The low TMR values obtained from the MTJs using the Co_2FeAl and $Co_2(Cr_{0.4}Fe_{0.6})Al$ films are attributed to the low-spin polarization expected from the low degree of order in these films. The TMR values depend sensitively on the interfacial structure of the tunnel junctions when the degree of order of the film is low.

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Keywords: $Co_2(Cr_{1-x}Fe_x)Al$ film; Microstructure; Magnetic tunnel junctions

1. Introduction

A magnetic tunnel junction (MTJ) using a half-metallic ferromagnetic material whose spin polarization is 100% at the Fermi energy is expected to exhibit an infinitely large tunnel magnetoresistance (TMR) according to Jullière's equation. A large giant magnetoresistance is also expected to appear in the current that is perpendicular to the plane spin valves using half-metallic electrodes. These magnetoresistance devices are required for developing a high-density magnetic random access memory and read heads for hard disk drive systems. Among many theoretically predicted half-metallic materials, Co-based full Heusler alloys with the $L2_1$-ordered structure is thought to be as promising as the MTJ electrode, since they have Curie temperatures high enough for their magnetoresistive devices to operate at room temperature (RT) [1–3]. However, the TMR values obtained from the MTJs using Heusler alloys are relatively small compared with that of the Fe/MgO/Fe junctions [4,5], in which the coherent MgO/Fe interface is believed to act as a spin filter. The experimentally low-TMR values using the Co-based full Heusler alloys have been thought to be caused by an atomic disorder [6], a roughness of barrier, lack of half metallicity, and a difference of electronic state between bulk and interface; however, its real reason has not been identified because of the lack of direct structural observations of MTJs.

Full Heusler alloys whose chemical form is X_2YZ with the $L2_1$ structure consists of four BCC unit cells having Y and Z atoms in the body-centered positions avoiding them in the nearest or second nearest positions in the $L2_1$ unit cell. Since, $L2_1$ is a highly ordered structure, it transforms into less-ordered structures, B2 $X_2(Y,Z)$, and then to A2 (X,Y,Z), with an increasing temperature. Miura et al. [1] predicted that the spin polarization of $Co_2(Cr_{1-x}Fe_x)Al$ alloys does not show strong dependence on the site disorder from $L2_1$ to B2 by ab initio calculation. Following this prediction, Okamura et al. [7] fabricated Co_2FeAl/Al–O/$Co_{75}Fe_{25}$ MTJs and observed a TMR value of 47% at RT despite the A2 type atomic disorder in the Co_2FeAl

*Corresponding author. Tel.: +81 298 59 2719; fax: +81 298 59 2701.
E-mail address: takahashi.yukiko@nims.go.jp (Y.K. Takahashi).

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electrode. They also reported almost the same TMR value from the MTJ with the B2 type Co_2FeAl electrode. This work suggests that the change in the MR values in these MTJs cannot be explained from the change in the spin polarization of the electrode materials. Although a phase separation was reported in bulk $Co_2(Cr_{1-x}Fe_x)Al$ alloys by Kobayashi et al. [8], it has not been confirmed in sputtered thin films. Although TMR values are thought to depend strongly on the structure and spin polarization of electrode materials as well as the interfacial structure of MTJs, little work has been done on the direct observation of the microstructure of the electrode thin films and their MTJs. In this study, we have investigated the microstructures of Co_2FeAl and $Co_2(Cr_{0.4}Fe_{0.6})Al$-sputtered films and those of the MTJs to discuss the TMR values obtained from the MTJs that were composed of Co_2FeAl and $Co_2(Cr_{0.4}Fe_{0.6})Al$ alloy electrodes.

2. Experimental

Twenty nanometer thick $Co_2(Cr_{1-x}Fe_x)Al$ ($x = 0.6,1$) films were prepared by the magnetron sputtering method using alloy targets. MgO(1 0 0) single crystalline substrates were used and their temperature (T_{sub}) varied from RT to 500 °C. Before the film deposition, the MgO substrate was cleaned by annealing at 600 °C for 1 h in the ultrahigh vacuum-sputtering chamber. The MTJs with stacking structure of Ta (2 nm)/$Ir_{22}Mn_{78}$ (10 nm)/$Co_{75}Fe_{25}$ (3 nm)/Al (1.2 nm)–O/$Co_2(Cr_{1-x}Fe_x)Al$ (20 nm)/MgO(0 0 1) were also fabricated by the magnetron sputtering method. The tunnel barriers were fabricated from the Al layer by inductively coupled plasma (ICP) oxidation. The MTJs were annealed with the magnetic field of 2 kOe at 200–350 °C for 1 h. By the standard photolithography and Ar ion etching technique, $10 \times 10\,\mu m$ MTJs were patterned. Magnetoresistance of the MTJs was measured by the four-point probe technique. Microstructures were characterized by transmission electron microscopy (TEM) using a Philips CM200 TEM and a high resolution TEM, JEOL JEM-4000EX. The composition of the film was analyzed by ICP optical emission spectroscopy.

3. Results

3.1. Microstructure of $Co_2(Cr_{1-x}Fe_x)Al$ ($x = 0.6$, 1) films

Fig. 1(a) shows a cross-sectional bright-field (BF) TEM image and (b) a corresponding selected area electron diffraction (SAED) pattern of the Co_2FeAl film deposited on a MgO(0 0 1) substrate at RT. The morphology of the Co_2FeAl film is continuous, and no grain boundaries are observed. The SAED pattern is a superposition of those for MgO and Co_2FeAl, indicating that the Co_2FeAl film was epitaxially grown on the MgO substrate with the orientation relationship of $(0\,0\,1)_{Co2FeAl}//(0\,0\,1)_{MgO}$ and $(1\,1\,0)_{Co2FeAl}//(1\,0\,0)_{MgO}$. The SAED pattern also indicates that the Co_2FeAl film is ordered to the B2 structure, but its

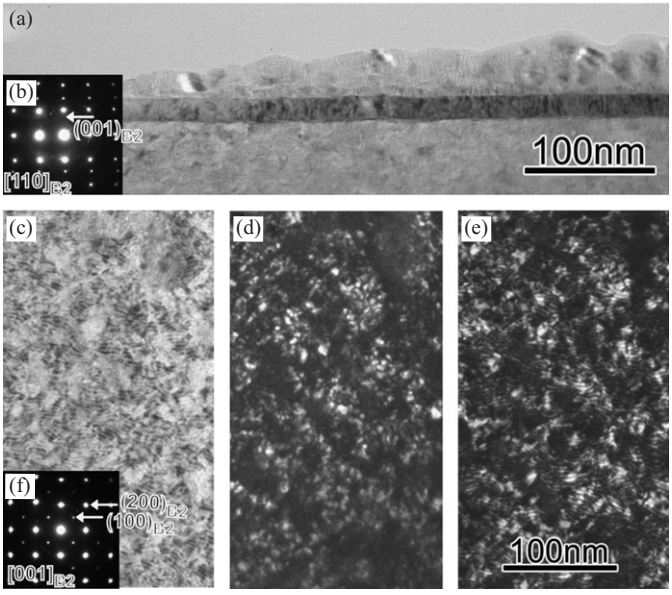

Fig. 1. (a) Cross-sectional BF TEM image and (b) the corresponding SAED pattern of the Co_2FeAl film deposited on a MgO (0 0 1) substrate at RT. (c) In-plane BF TEM image, (d) corresponding DF images excited with the $(1\,0\,0)_{B2}$ superlattice spot and (e) the one excited with the $(2\,0\,0)_{B2}$ fundamental spot and (f) the in-plane SAED pattern.

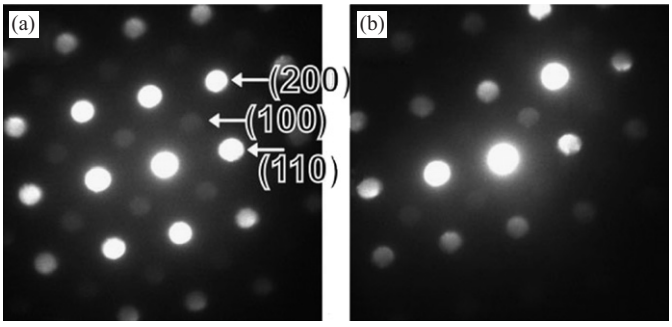

Fig. 2. Nanobeam diffraction patterns taken from the brightly (a) and darkly (b) imaging regions in the DF image in Fig. 1(d).

degree of order appears to be low since the intensity of the (0 0 1) superlattice spot of B2 is very weak. The dark-field (DF) images excited with the $(1\,0\,0)_{B2}$ superlattice spot (Fig. 1(d)) and the one excited with the $(2\,0\,0)_{B2}$ fundamental spot that is overlapped with $(2\,0\,0)_{A2}$ and MgO (Fig. 1(e)) show that almost the same region is excited brightly. This means that the contrast observed in the BF and DF images are simply because of the diffraction contrast arising from the local strains within the film. The contrast does not reflect local change in the degree of order. This can be further confirmed by Fig. 2, which are the nanobeam diffraction patterns taken from the bright imaging region (a) and from the dark imaging region (b) in the DF image of Fig. 1(d). Since the superlattice spots are clearly observed from both the regions with a slight difference in orientation, the contrasts observed in Fig. 1(c)–(e) are concluded to be due to the slight local orientation difference within the film. Note that the rings in the SAED patterns are due