The effect of inserting thin Co$_2$MnAl layer into the Co$_2$MnSi/MgO interface on tunnel magnetoresistance effect

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Abstract. We fabricated an epitaxially grown B2-ordered Co$_2$MnAl Heusler alloy film by optimizing fabrication conditions, such as composition of the films and annealing temperature. Tunnel magnetoresistance (TMR) effect was investigated in Co$_2$MnSi/Co$_2$MnAl(0–1.0 nm)/MgO/CoFe magnetic tunnel junctions (MTJs). TMR ratio was enhanced by inserting a thin Co$_2$MnAl layer at the Co$_2$MnSi/MgO interface. The MTJ with Co$_2$MnAl thickness of 0.5 nm exhibited the highest TMR ratio at 310 K and 10 K.

1. Introduction

Development of ferromagnetic materials with high spin polarization is the most important subject in spintronics field. Ideal high spin polarization materials are half-metallic ferromagnets with a band gap at the Fermi energy level for one spin band [1]. Experimentally, we have observed a large tunnel magnetoresistance (TMR) ratio of up to 159% at 2 K in magnetic tunnel junctions (MTJs) with Co$_2$MnSi/Al–O/CoFe structure, thus proving the halfmetallic property of Co$_2$MnSi [2].

Similarly, MTJs using a crystalline MgO tunnel barrier producing coherent tunneling through the $\Delta_1$ band of ferromagnetic electrodes have attracting much interest for some spintronics devices, such as magnetic random access memory (MRAM) and read heads for hard disk drives (HDDs). Ikeda et al. showed a huge TMR ratio of up to 600% at RT in a CoFeB/MgO/CoFeB MTJ [3]. However, the TMR ratio using a CoFeB electrode and an MgO barrier is coming close to the theoretical limit [4]. Miura et al. showed theoretically that coherent tunneling through the $\Delta_1$ band of Co$_2$MnSi Heusler alloy could also occur in a Co$_2$MnSi/MgO/Co$_2$MnSi MTJ [5]. We have demonstrated a giant TMR ratio of 753% at 2 K in a MTJ with Co$_2$MnSi/MgO/CoFe structure, in which the MgO barrier was deposited by electron beam (EB) evaporation system. This result indicates that Heusler alloys as electrodes of MgO-based MTJs have great potential to achieve giant TMR ratio by both half-metallicity and coherent tunnelling [6]. However, the TMR ratio decreased rapidly with increasing of temperature in the Co$_2$MnSi/MgO/CoFe-MTJ. We considered that the large temperature dependence of the TMR ratio was originated from the inelastic tunneling at the antiparallel magnetization configuration. Y. Miura et al. suggested by their theory that Co$_2$MnSi/MgO interfacial states were created in the half metallic gap due to the non-bonding states [7]. Such interfacial states enhanced the inelastic tunneling [8]. They also suggested that a MTJ having an ultrathin Co$_2$MnAl layer between a Co$_2$MnSi electrode and a MgO barrier does not have the interfacial state and show the halfmetallic character at the MgO barrier interface [7]. In this study, we fabricated high-quality Co$_2$MnSi/Co$_2$MnAl/MgO/CoFe-MTJs and investigated the TMR effect.
2. Fundamental properties of Co$_2$MnAl thin films

The Co$_2$MnAl thin films were prepared by means of magnetron sputtering in order to characterize their crystal structure, surface morphology and magnetic properties. The MgO-buffer(20 nm)/Co$_2$MnAl(30 nm) films were deposited on single crystalline MgO (001) substrates at ambient temperature and subsequently annealed at 400-600 ºC in order to reduce site-disorder. In order to obtain the stoichiometric film composition, we used a composition-adjusted Co-Mn-Al alloy sputtering target (Co: 43.7%, Mn: 27.95%, Al: 28.35%). Crystal structure of the prepared Co$_2$MnAl thin films were verified by x-ray diffraction (XRD) with out-of plane geometries and surface roughness was verified by atomic force microscopy (AFM). We measured magnetization curves by means of a vibrating sample magnetometer (VSM). Film compositions were examined by means of inductively coupled plasma (ICP) analysis.

The XRD profiles for Co$_2$MnAl thin film with $T_a = \text{as depo.-600ºC}$ are shown in Figure 1(a). In addition to peaks from MgO substrates, only the (002) and (004) Co$_2$MnAl peaks were detected, indicating perfect (001)-preferred orientation. The full width at half maximum (FWHM) of the rocking curve for the (004) Co$_2$MnAl peak is very small (under 0.5º). Therefore, the structural quality of the film in the direction perpendicular to the plane was quite good. In addition, ideal epitaxial growth was confirmed with the epitaxial relation [110]Co$_2$MnAl||[100]MgO||[100]MgO from the φ-scan result shown in the Figure 1(b). Clear (220) Co$_2$MnAl peaks are observed in the φ-scan profile for all the prepared films, indicating that a highly $B2$-order structure can be obtained. For all films, the (111) peak of $L2_1$ structure is undetectable from the (111) pole-figure measurements. These results suggest that prepared epitaxial Co$_2$MnAl films are identified as having not $L2_1$ structure but $B2$ structure. In addition, the composition of Co$_2$MnAl film analyzed by ICP spectroscopy was nearly stoichiometric (Co: 49.3%, Mn: 24.2%, Al: 26.5%).

Figure 2 shows the average roughness ($R_a$) of the prepared Co$_2$MnAl thin films as function of $T_a$. Roughness of the films decreases with increasing $T_a$ above $T_a = 300$ ºC. Inset in Figure 2 shows the AFM image for the MgO(001)-sub./MgO-buffer(20 nm)/Co$_2$MnAl(30 nm) ($T_a = 400$ ºC) film.
shows the AFM image for the films with $T_a = 400 \degree C$, where $R_a$ is about 0.2 nm. The $R_a$ of 0.2 nm was small enough to be formed a thin tunnel barrier.

Saturation magnetization $M_s$ estimated from $M$–$H$ curves is depicted in Figures 3 as functions of $T_a$. The maximum value of $M_s$ is about 20% smaller than the bulk $M_s$.

3. TMR in MTJs using Heusler alloy thin films

We prepared MTJs with two different structures of bottom electrodes. The stacking structure of MTJs is bottom-electrode/MgO(2.3 nm)/Co$_{50}$Fe$_{50}$(5 nm)/IrMn (10 nm)/Ta(5 nm)/cap layer. The structures of bottom electrodes are as follows.

Sample A: MgO(001)-sub./MgO-buffer(20 nm)/epitaxial Co$_2$MnSi(50 nm) ($T_a = 600 \degree C$)/Co$_2$MnAl(0.0-1.0 nm) ($T_a = 400-600 \degree C$).

Sample B: MgO(001)-sub./MgO-buffer(20 nm)/epitaxial Co$_2$MnAl(30 nm) ($T_a = 400-600 \degree C$).

All the MTJs were patterned into four-terminal structure by using photo-lithography and Ar ion milling. The TMR effect was measured at 310 and 10 K. To improve the crystallinity of the MgO barrier, patterned MTJs were annealed at the 250-525 °C for 1 hour in high vacuum by applying a magnetic field of 1 T.

TMR ratio increased with increasing annealing temperature because of crystallization of the MgO barrier, and shows a maximum at 450-475 °C. TMR ratio showed maximum for the Co$_2$MnSi/Co$_2$MnAl($T_a = 400 \degree C$)/MgO/Co$_{50}$Fe$_{50}$ MTJs and for the Co$_2$MnAl($T_a = 400 \degree C$)/MgO/Co$_{50}$Fe$_{50}$ MTJs. Figure 4(a) shows maximum TMR ratio measured at 310 K and 10 K in each inserted Co$_2$MnAl thickness. The MTJ with Co$_2$MnAl thickness of 0.5 nm shows the highest TMR ratio of 180% at 310 K and 600% at 10 K. These TMR ratios are larger than those of both Co$_2$MnSi/MgO/CoFe-MTJ and Co$_2$MnAl/MgO/CoFe-MTJ. Figure 4(b) shows the ratio of the TMR ratios measured at 310 K and 10 K (TMR$_{310K}$/TMR$_{10K}$) for each inserted Co$_2$MnAl thickness. TMR$_{310K}$/TMR$_{10K}$ ratio tends to saturate beyond a thickness of 1.0 nm. This result indicates that the temperature dependence of TMR ratio is slightly improved by insertion of Co$_2$MnAl.
Some origins of the large temperature dependence of TMR in the MTJs with Heusler alloy electrodes are considered both experimentally and theoretically: e.g., small energy separation between the conduction band and the Fermi level [9], non-quasiparticle state [10], and the weak exchange interaction energy of the Co layer at the Co₂MnSi/MgO interface [11] and interfacial states created in the half-metallic gap as mentioned above [7]. It is not clear which origins of the large temperature dependence were improved by insertion of the thin Co₂MnAl layer. In order to clarify the role of Co₂MnAl insertion layer, detailed investigations on the tunneling conductance properties are necessary.

4. Conclusion
We have fabricated MTJs with B2-ordered (001)-epitaxial Co₂MnAl bottom electrodes. Co₂MnAl thin films had good surface flatness (about 0.2 nm) and showed a highly-ordered B2 structure for $T_a \geq 400$ °C. We observed very high TMR ratios of 180% at 310 K and 600% at 10 K and improved the temperature dependence of TMR ratio by insertion of very thin Co₂MnAl films into the Co₂MnSi/MgO interface.

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