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Structural and Magnetic Properties of Co_2MnSi Heusler Alloy Films

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Recently half-metallic full-Heusler alloy films have attracted significant interests for spintronics devices. As these alloys have been known to have a high spin polarization, very large TMR ratio is expected in magnetic tunnel junctions. Among these alloys, Co_2MnSi full-Heusler alloy with a high spin polarization and a high Curie temperature is considered a good candidate as an electrode material for spintronic devices. In this study, the magnetic and structural properties of Co_2MnSi Heusler alloy films were investigated. TMR characteristics of magnetic tunnel junctions with a $Co_2MnSi/SiO_2/CoFe$ structure were studied. A maximum MR ratio of 39% with SiO_2 substrates and 27% with MgO(100) substrates were obtained. The lower MR ratio than expectation is considered due to off-stoichiometry and atomic disorder of Co_2MnSi electrode together with oxidation of the electrode layer.

Key words : Heusler alloy, half-metallic ferromagnet, atomic ordering,

1. Introduction

Half-metallic ferromagnets (HMFs), known to have only one occupied spin channel at Fermi level, have attracted significant interests for spintronics devices such as magnetic tunnel junctions (MTJs) and spin injection electrode. Since R. A. de Groot [1] has shown that NiMnSb Heusler alloy has the half-metallicity by simulation, many groups have studied Heusler alloys such as NiMnSb [2], Co_2MnSi [3, 4], $Co_2(Cr,Fe)Al$ [5-7], and Co_2MnAl [8].

Heusler alloys are based on ternary alloy system composed two groups; half-Heusler alloy (XYZ) and full-Heusler alloy (X_2YZ). X is a transition metal element such as Fe, Ru, Co, Rh, Ni, Pd, Pt, or Cu, Y is possessed by another transition metal element such as Ti, V, Cr, Mn, Nb, or Zr, and Z is an element from group III, IV, or V such as Al, Si, Ga, In, Ge, Sn, As, or Sb. Heusler alloys have the ordered structures, $C1_b$ (half-Heusler) structure and $L2_1$ (full-Heusler) structure, which can be briefly investigated X-ray diffraction (XRD).

Among these Heusler alloys, Co_2MnSi full-Heusler alloy can be suggested as one of the suitable candidates of electrode materials in spintronics, which has a high spin polarization and a high Curie temperature (~985 K) [9, 10]. In this study, structural and magnetic properties of Co_2MnSi full-Heusler alloys were investigated, and TMR characteristics of MTJs with Co_2MnSi electrodes were studied by micro-fabrication.

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2. Experiments

Co_2MnSi single layers were deposited by dc magnetron sputtering on thermally oxidized Si and MgO (100) substrates. Base pressure was below 1×10^{-8} torr and sputter rate is below 0.4 Å/sec. In order to obtain the $L2_1$ ordered structure of Co_2MnSi, the substrates were heated from 280°C to 470°C during deposition. Structural and magnetic properties of the films were investigated by XRD and vibrating sample magnetometer (VSM), respectively. Surface characteristics were measured by AFM. Full stack of MTJs is Co_2MnSi(20 nm)/Al-oxide(1.2 nm in Al thickness)/$Co_{75}Fe_{25}$(3 nm)/$Ir_{25}Mn_{75}$(10 nm)/Ta(2 nm) and oxidation of Al was done by plasma oxidation by changing oxidation time from 105 sec to 135 sec. MTJs were fabricated by photolithography with the junction size of 10×10 μm^2 and TMR characteristics were measured by a 4 point probe system. After micro-fabrication, MR mea-

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surements were performed at room temperature, then the junctions are annealed from 200°C to 350°C for 1 hour in the vacuum below 1×10^{-6} torr with the magnetic field of 2 kOe. After the annealing treatments MR was measured again at room temperature.

3. Results and Discussions

Fig. 1(a) shows XRD patterns of Co_2MnSi films deposited on heated SiO_2 substrates. Above 330°C, (111) superlattice peak as well as other diffraction peaks is shown, while only broad (220) peak is shown up to 300°C. This means that $L2_1$ ordered structure of Co_2MnSi is formed above 330°C [11]. Fig. 1(b) shows saturation magnetization and coercivity of Co_2MnSi films. Up to 300°C, M_s values are very small and increase very slowly and H_c values are very large and increase drastically with increasing the substrate temperature. On the other hand, above 330°C, M_s values increase up to about 950 emu/cc (4.58 μ_B/unit cell) and H_c shows very small values of around 10 Oe. This results that the film is mostly amorphous structure when the substrate temperature is lower than 330°C and when the film crystallizes it forms the superlattice although the degree of order is not studied in the present work. From these data, the substrate temperature of 330°C which is the lowest temperature showing (111) superlattice peak was chosen as the sputtering condition for Co_2MnSi films in order to reduce the surface roughness.

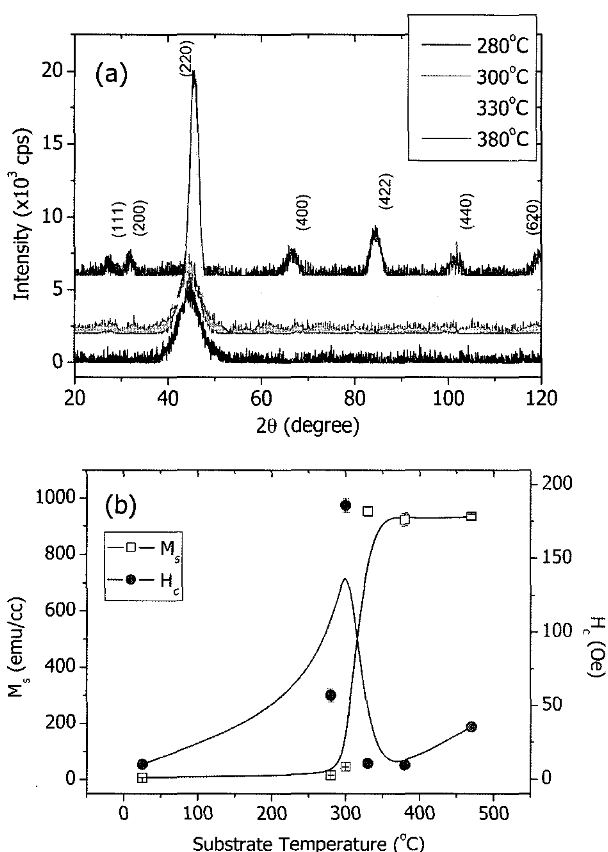

Fig. 1. (a) XRD pattern (α-2θ scan) and (b) saturation magnetization and coercivity of Co_2MnSi films on SiO_2 substrate heated at various temperatures.

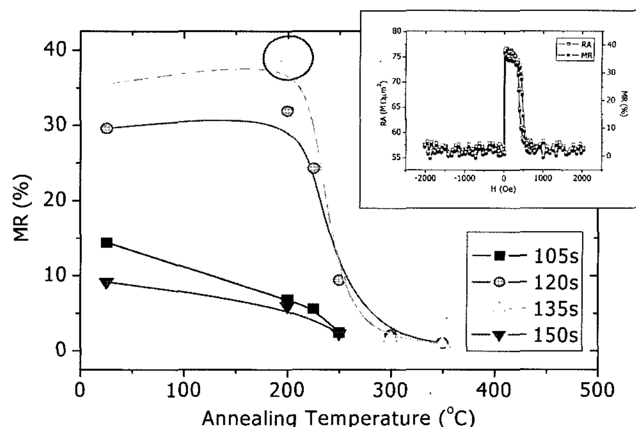

Fig. 2. Post-annealing temperature dependence of TMR ratio with various oxidation time; (inset) TMR curve of the condition which shows maximum MR value (red circle).

Fig. 2 shows the variation of MR ratio with varying oxidation times and post-annealing temperatures. A maximum MR ratio of 39% was obtained with Al oxidation time of 135 second and post-annealing at 200°C. The inset in Fig. 2 shows the TMR curve for the case of the maximum TMR value. Junction resistance was very high in the range of several tens of M$\Omega\mu m^2$. The reported AlOx junction resistance which shows an optimum TMR resistance in a CoFeB/AlOx/CoFeB MTJ with 1.1 nm thick Al was in several tenth k$\Omega/\mu m^2$ [12]. Therefore, the very high junction resistance must be due to the oxidation of manganese and silicon atoms of Co_2MnSi [13, 14]. More proper oxidation method should be developed improve the TMR ratio.

We have tried to the same process with MgO (100) substrates. As lattice misfit between MgO (100)<001> direction and Co_2MnSi (100)<011> direction is with 4.7%, Co_2MnSi films can grow on MgO (100) substrate epitaxially. In order to find the (111) superlattice peak, pole figures and image mappings of (111) peak is used. Fig. 3 shows the pole figures and mapping images of (111) peak by grazing incident XRD for the three different substrate temperatures. The x-ray pole figures show the Co_2MnSi films grow epitaxially on the MgO substrates when the substrate temperature is higher than 470°C. At 470°C, Co_2MnSi (111) peak starts to appear, while there is no (111) peak at 380°C. It is not clear from

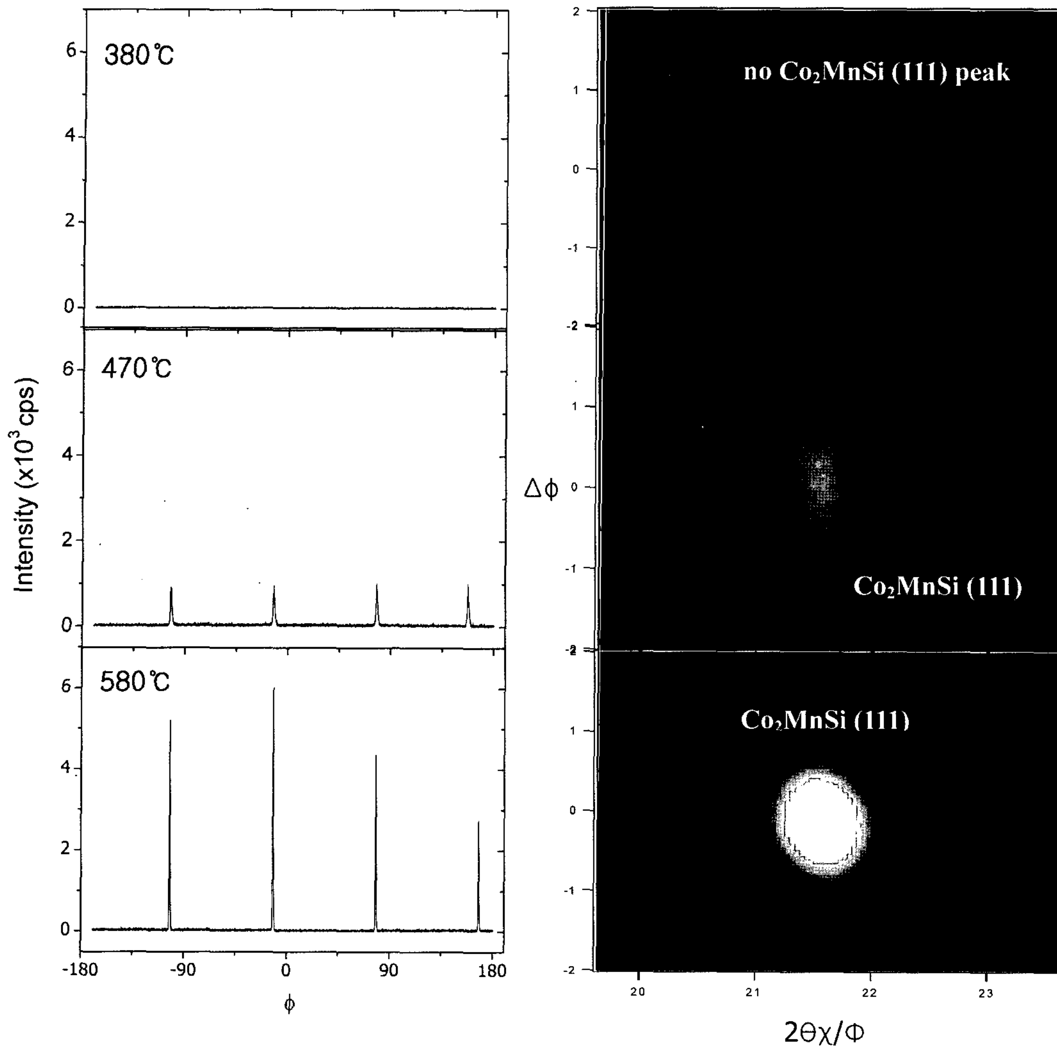

Fig. 3. X-ray pole figures and mapping image of Co_2MnSi (111) peak on MgO(100) substrate.

x-ray diffraction peak analysis that if the film deposited on 380°C heated MgO substrate is mainly amorphous phase or nano-crystalline phase. However, the high M_s of 380°C case in Fig. 4 strongly suggests the film is composed of nano-crystalline phase. It may be the same case for 330°C heated substrate. The reason (111) superlattice peak was not observed in Fig. 3 may be due to very weak diffraction. If this analysis is true, there is no big dependency of the substrate materials on crystallization behavior. Magnetic properties of the film deposited on MgO substrate in Fig. 4 show some difference in M_s and H_c compared with those on SiO_2 substrate. It shows higher saturation magnetization over 1100 emu/cc even at 330°C. As the substrate temperature increases, M_s is around 1120 emu/cc, which corresponds to 5.43 μ_B/(unit cell). This value is higher than 5 μ_B/(unit cell) predicted by the theoretical calculation. The reason of the discrepancy is not clearly known. H_c values increase very

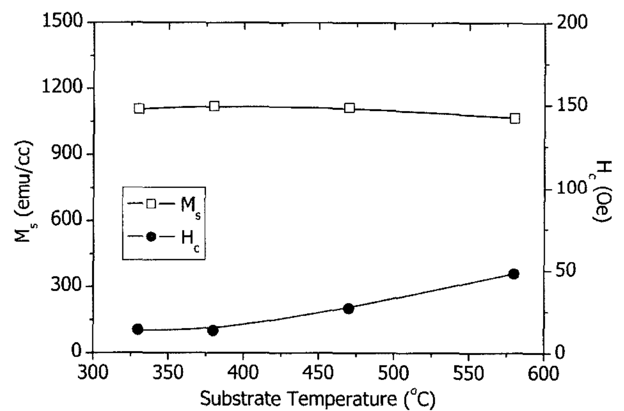

Fig. 4. Saturation magnetization and coercivity of Co_2MnSi films on MgO (100) substrate heated at various temperatures.

slowly with increasing substrate temperature. For the MTJ junction fabrication, the substrate temperature of 470°C was selected for the Co_2MnSi deposition. This

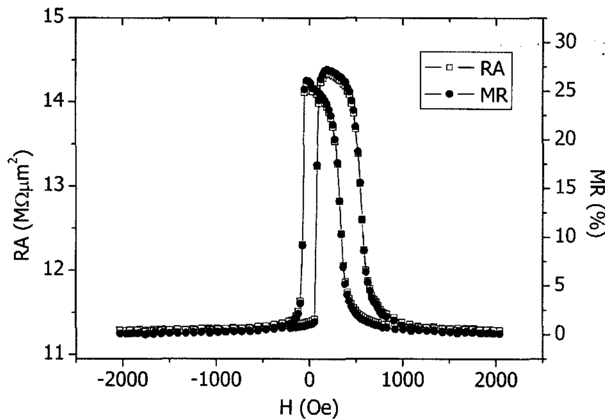

Fig. 5. TMR curve of MTJ with 120 sec oxidation after annealing at 200 °C.

temperature is the lowest temperature that shows clear (111) peak.

Fig. 5 shows the TMR curve of the MTJ with Co_2MnSi electrode deposited at 470°C which shows the maximum MR value. In this case, the optimum oxidation time was 120 seconds and post-annealing temperature was 200°C. The highest TMR ratio of 27% with MgO substrate was lower than that of 39% with SiO_2 substrate. The reason of lower TMR ratio of the MTJ on MgO substrate than that on SiO_2 substrate is not clearly understood. The junction resistance of the MTJ stack on MgO substrate was lower than that of SiO_2 substrate although they are in the same order of magnitude. This suggests that the lower TMR ratio is not due to over-oxidation difference. In order to study surface roughness contribution, roughness of the Co_2MnSi films deposited at 470°C on MgO substrate and deposited at 330°C on SiO_2 substrate was compared. It was 2.3 nm and 7 nm in R_{ms}, respectively, although the bare substrate surface roughness of MgO showed much higher roughness. Therefore the roughness effect can be excluded.

The MTJ stacks on MgO (100) substrates as well as SiO_2 substrates did not show higher MR ratio, or higher spin polarization than theoretically expected. The first reason may be off-stoichiometric problem. The nominal composition of Co_2MnSi films in this study was $Co_{49.8}Mn_{17.7}Si_{32.5}$ by EDS analysis. This is Mn-poor alloy. The second reason may be atomic disorder of Co_2MnSi films, which is already known [15, 16]. Theoretically $L2_1$ ordered structure expected Heusler alloy can show 100% of spin polarization and atomic disorder reduces spin polarization of half-metallic Heusler alloy. However, in real experiments, it is very difficult to control the degree of order of the films. The third reason may be due to the oxidation of Co_2MnSi electrode during Al layer oxidation in the present work. These might be the main reasons of the low MR ratio of the magnetic tunnel junctions with Co_2MnSi half-metallic Heusler alloy films.

4. Conclusions

Co_2MnSi half-metallic full Heusler alloy was investigated as a possible candidate of electrode materials in magnetic tunnel junction. In this study, we found the possibility of Co_2MnSi electrode with TMR ratio of 39% with SiO_2 substrate and 27% with MgO substrate. These values are much lower than theoretical prediction. Off-stoichiometry and atomic disordered structure of Co_2MnSi films, and oxidation of the Co_2MnSi free layer during the barrier oxidation are considered the main reasons of the low MR ratio. Further studies are needed to enhance TMR ratio by optimizing stoichiometry of the composition, oxidation conditions of barrier materials and degree of atomic order.

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