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Tunnel Magnetoresistance Effect in CoFeB/MgO/Co_2FeSi and Co_2MnSi Tunnel Junctions

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We have fabricated MgO-based magnetic tunnel junctions (MTJs) with the CoFeB bottom electrode and top electrodes of poly crystalline Co_2FeSi and Co_2MnSi Heusler alloys. We have measured temperature dependence of the TMR ratio and TMR-V characteristics at 6 K. We have achieved a high TMR ratio of 90% at RT for the MTJ with Co_2FeSi electrode after annealing at 325°C. The MTJ with Co_2MnSi electrode showed a significant annealing temperature dependence of the TMR ratio. The increase of TMR ratio by annealing is due to the crystallization of the Co_2MnSi Heusler layer. Furthermore, the strong temperature dependence of TMR ratio and the anomalous TMR-V characteristics have been observed in the MTJ with Co_2MnSi.

***Index Terms*—CoFeB, Heusler alloys, MgO, MTJs.**

I. Introduction

MAGNETIC TUNNEL JUNCTIONS (MTJs) can be applied to the most promising spin electronics devices of magnetic random access memory (MRAM) and the high density magnetic reading head. Recently, several groups have reported giant tunnel magnetoresistance (TMR) ratio in MTJs using the MgO barrier [1], [2]. The observed TMR ratio in the MgO-based MTJ was much larger than those of conventional MTJs with amorphous aluminum oxide barriers [3], [4]. The origin of the giant TMR is due to coherent transport of Δ_1 electrons in the MgO insulating barrier, as predicted by theories [5], [6]. Moreover, employing the CoFeB as the ferromagnetic electrodes, larger TMR ratio has been observed [7], [8]. According to their report, highly textured MgO (001) barrier is one of the most important factors to obtain a large TMR ratio. On the other hand, recently a series of Heusler alloys has been extensively studied. NiMnSb was first reported by de Groot [9] showing half-metallic material. The minority band of this material shows an energy gap behaving like an insulator and the opposite spin band crosses the Fermi level that behaves like a metal. The predicted spin polarization of this material is expected to be 100%. Using the Heusler alloys as a spin injector, a perfect spin polarized current can be provided. Moreover especially Co-based half-metallic Heusler alloys have the high Curie temperature in the range between 400–1200 K. Thus this material is suitable for use in low power and high output spin-electronic devices. Some groups have attempted to use Co_2 MnSi and $Co_2Cr_2Fe_2$Al as a bottom magnetic layer of MTJs [10], [11]. Recently, we have succeeded in showing that half-metallic Co_2MnSi and the estimated spin polarization was about 89%, as reported previously [12]. In this study, we investigated the TMR effects in CoFeB/MgO/Heusler alloys (Co_2FeSi and Co_2MnSi). Co_2FeSi and Co_2MnSi have been theoretically predicted to have high Curie temperature of 1200 and 985 K [13], respectively. The lattice mismatch between the Heusler alloys and MgO is relatively small of about 5.3%, which indicates that the highly textured Heusler alloys can be grown on the tunneling barrier of MgO. Therefore, it is expected that the large TMR ratio can be observed in CoFeB/MgO/Heusler alloys.

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II. Experimental Procedure

Sub (thermal oxidized Si)/Ta(10)/Py(2)/IrMn(10)/$Co_{75}Fe_{25}$ (2)/Ru(0.85)/$Co_{40}Fe_{40}B_{20}$(5)/MgO(2.5)/Heusler alloys (Co_2FeSi and Co_2MnSi)/Ta(7)/Ru(7) were deposited at RT using magnetron sputtering. The thickness values indicated in parenthesis are nanometers. The MgO insulating layer was grown using the high density sintered MgO target. Conventional photolithography and the Ar ion milling technique were used to form the MTJ structure with a junction area of between 3×3–100×100 μm^2. The prepared MTJs were annealed at 275–400°C for 1 hour under vacuum with the magnetic field of 350 Oe to fix easy axis of ferromagnetic layers. The crystal structure of the Co_2FeSi and Co_2MnSi Heusler alloys deposited on the MgO insulating barrier were characterized by XRD measurement. The TMR $-V$ was measured at 6 K.

III. Results and Discussion

We have examined the crystallization of Co_2FeSi and Co_2MnSi thin films by using X-ray diffraction. Fig. 1 shows the XRD patterns for sub./Ta(10 nm)/$Co_{40}Fe_{40}B_{20}$ (5 nm)/MgO(2.5 nm)/Co_2FeSi(20 nm) and Co_2MnSi(30 nm) annealed at various temperatures in the range between R.T. and 350°C. The MgO(002) peak is observed at 43°, which indicate that the fabricated MgO film is highly oriented along the (002) direction. We can conclude that highly textured MgO(002) have been obtained. Annealing at sufficiently high temperature over 325°C, Co_2FeSi resulted in the appearance of the $A2$(004) peaks showing improvement in crystallization. It is confirmed that the Co_2FeSi has grown perfectly (001) oriented in the direction perpendicular to the film plane. Co_2FeSi have exhibited Co_2FeSi $B2$(002) peaks at around 32°, however, the intensity

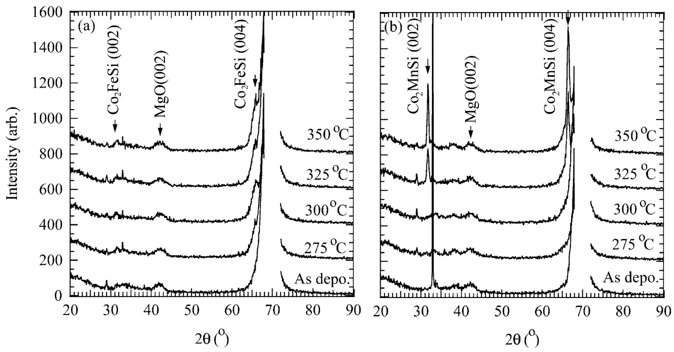

Fig. 1. XRD patterns for (a) sub./Ta(10 nm)/$Co_{40}Fe_{40}B_{20}$ (5 nm)/MgO(2.5 nm)/Co_2FeSi(20 nm) and (b)sub./Ta(10 nm)/$Co_{40}Fe_{40}B_{20}$ (5 nm)/MgO(2.5 nm)/Co_2MnSi(30 nm) annealed at various temperatures with the range of R.T. to 350°C.

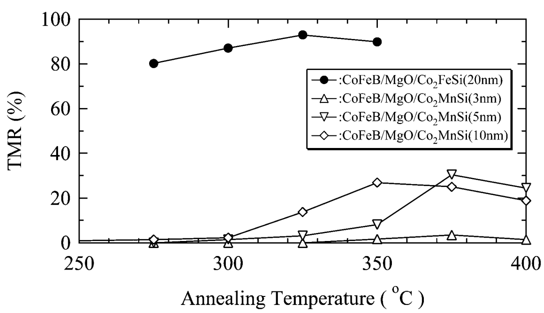

Fig. 2. Annealing temperature dependence of the TMR ratio at RT for the MTJs with various thickness of Heusler alloy layers.

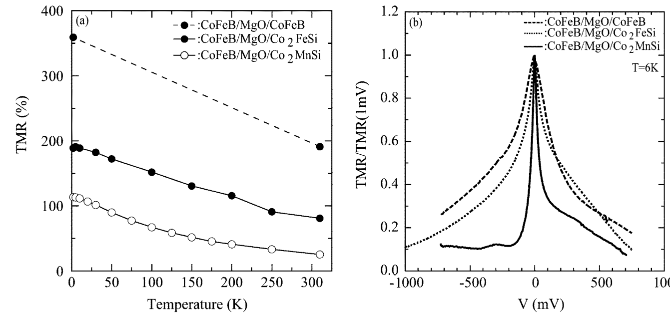

Fig. 3. (a) Temperature dependence of the TMR ratio at a bias voltage of 1 mV for $Co_{40}Fe_{40}B_{20}$/MgO/$Co_{40}Fe_{40}B_{20}$, $Co_{40}Fe_{40}B_{20}$/MgO/Co_2FeSi(20 nm) and $Co_{40}Fe_{40}B_{20}$/MgO/Co_2MnSi(10 nm) MTJs. (b) Bias voltage dependence of the normalized TMR ratio at 6 K.

of $B2$(002) peak is much smaller than that of $A2$(004) superlattice. These results indicate that the grown Co_2FeSi films had $A2$ poly-crystalline structure. Magnetization saturation of $A2$- Co_2FeSi was around $4(\mu_B$/f.u.) at 2 K which is about 67% of the theoretically predicted value for bulk $L2_1$-Co_2FeSi [14]. And also, in the X-ray diffraction pattern of Co_2MnSi annealed at over 325°C, sharp (002) peaks reflecting $B2$-type superlattice were observed. Therefore we conclude that the Co_2MnSi was identified as a highly ordered $B2$ structure. The obtained magnetization saturation of $B2$-Co_2FeSi was around 4 (μ_B/f.u.) at 2 K which is about 80% of the value for bulk $L2_1$-Co_2 MnSi.

Fig. 2 shows the annealing temperature dependence of TMR ratio measured at RT. Although Co_2FeSi have $A2$ structure, the MTJ with Co_2FeSi electrode showed a high TMR ratio of 80% annealed at 275°C, and the TMR ratio increased up to 90% after annealing at 325°C. MTJs with various thickness of Co_2MnSi showed a significant annealing temperature dependence of the TMR ratio. The TMR ratio was only a few % in the MTJs with 5 and 10 nm thick Co_2MnSi annealed at 275°C, but drastically increases with increasing annealing temperature up to 30% for 5 nm and 27% for 10 nm Co_2MnSi thickness, respectively. The increase in TMR ratio probably corresponds to the improvement of crystallization of Co_2MnSi films with annealing temperature as observed in the XRD results. On the other hand, the MTJ with 3 nm thick Co_2MnSi showed low TMR even after annealing at 400°C. It is thought that the thickness of 3nm is too thin to crystallize. Thus the increase in the TMR ratio with annealing temperature was not observed for the MTJ with 3 nm thick Co_2MnSi.

Fig. 3(a) shows the temperature dependence of the TMR ratio for $Co_{40}Fe_{40}B_{20}$/MgO/$Co_{40}Fe_{40}B_{20}$,$Co_{40}Fe_{40}B_{20}$/MgO/Co_2 FeSi(20 nm) and $Co_{40}Fe_{40}B_{20}$/MgO/Co_2MnSi(10 nm) MTJs annealed at 350°C. The TMR ratio at 2 K is 189% for $Co_{40}Fe_{40}B_{20}$/MgO/Co_2FeSi (20 nm) and 113% for CoFeB/MgO/Co_2MnSi (10 nm), respectively. TMR ratios for both MTJs were almost independent of the temperature between 2–20 K and then decreased drastically over 50 K. Sub./Ta(10 nm)/Py(2 nm)/IrMn(10 nm)/$Co_{75}Fe_{25}$ (2 nm)/Ru(0.85 nm)/$Co_{40}Fe_{40}B_{20}$ (5 nm)/MgO(2.5 nm)/$Co_{40}Fe_{40}B_{20}$ (3 nm)/Ta(7 nm)/Ru(7 nm) junctions showed the TMR ratio of 191% at RT and 359% at 2 K. Compared with $Co_{40}Fe_{40}B_{20}$/ MgO/$Co_{40}Fe_{40}B_{20}$, $Co_{40}Fe_{40}B_{20}$/MgO/Co_2FeSi(20 nm) and $Co_{40}Fe_{40}B_{20}$/ MgO/Co_2MnSi(10 nm) showed strong temperature dependence on the TMR ratio. The TMR ratio strongly depends on the quality of tunneling barrier. If the tunneling barrier contains the disorder site such as vacancy or various oriented crystal structures inside MgO, the electrons will be scattered or pass through two step tunneling path, consequently reduction in the TMR ratio with the bias voltage would be observed. However, the deposited MgO thin films in our study have high quality (001) oriented crystal structure, therefore the drastic decrease of TMR was rather due to the reduction of the interfaces spin polarization of Heusler alloys. The spin polarization of CoFeB in our sample estimated from Julliere's equation are $P(\mathrm{Co_{40}Fe_{40}B_{20}}) = 70\%$ at RT and 80% at 2 K, respectively. Using these values, the spin polarization P of Heusler alloys were estimated as $P(\mathrm{Co_2FeSi}) = 41\%$ and $P(\mathrm{Co_2MnSi}) = 16\%$ at RT and $P(\mathrm{Co_2FeSi}) = 61\%$ and $P(\mathrm{Co_2MnSi}) = 45\%$ at 2 K. The spin polarization of Co_2FeSi and Co_2MnSi increases about 1.5 times and 2.8 times by changing the temperature from 310 to 2 K. The significant temperature dependence of the TMR is reported also in a $L2_1$-Co_2MnSi/AlO/CoFe MTJ [12], which may be the intrinsic feature of MTJs using Heusler alloys.

Fig. 3(b) shows the bias voltage dependence of the TMR ratio normalized by the value at 1 mV measured at 6 K. The TMR ratio decreases with increasing bias voltage for each

MTJs. The curves were obtained from $dI/dV - V$ curves at both anti-parallel and parallel magnetization configurations. The shape of TMR-V curve for the MTJ with Co_2MnSi showed unusual voltage dependence of the TMR ratio. The $V_{\rm half}$ value at which the TMR ratio reduced by halves were about -30 mV and 39 mV for each side of the bias curve. These values are much smaller than that of $Co_{40}Fe_{40}B_{20}$/MgO/$Co_{40}Fe_{40}B_{20}$ MTJs. Since the TMR ratio strongly depends on the shape of density of states for ferromagnetic electrodes, we suggest that the strong bias voltage dependence observed in the junctions was due to the half-metal narrow energy gap of Co_2MnSi near the Fermi level. The first principle calculation by *Kandpal* [15] shows that the shape of the density of state and the position of the Fermi level for Co_2MnSi is changed with lattice parameter. The predicted maximum width of energy gap is 0.9 eV at the most. Moreover, the half—metallicity can not be preserved by anticite disorder. The disorder states that were induced inside the energy gap and spin polarization were strongly reduced [16]. Furthermore, it should be noted that spin-fluctuation at finite temperature can reduce spin-polarization of Heusler alloys even though the ideal MgO/Heusler alloys interface is formed. Therefore, the further optimization of fabrication process to improve MgO/Heusler alloys interface state is necessary to obtain the MTJs with a larger TMR ratio with small temperature and voltage dependences.

IV. Conclusion

MTJs of sub./Ta(10 nm)/Py(2 nm)/IrMn(10 nm)/$Co_{75}Fe_{25}$(2 nm)/Ru(0.85 nm)/$Co_{40}Fe_{40}B_{20}$(5 nm)/MgO(2.5 nm)/polycrystalline Heusler alloys (Co_2 FeSi and Co_2MnSi)/Ta(7 nm)/Ru(7 nm) have been fabricated. The MTJ with Co_2FeSi showed a high TMR ratio of 90% at RT and a strong temperature dependence of TMR ratio compared to the $Co_{40}Fe_{40}B_{20}$/MgO/$Co_{40}Fe_{40}B_{20}$ MTJs were observed. The MTJ with Co_2MnSi have shown the obvious correlation between the increase in the TMR ratio and crystallization of the Co_2MnSi Heusler layer by annealing. Moreover, the significant temperature dependence of the TMR ratio has been observed. This result implies that half-metallicity of Co_2MnSi was observed in MgO-based MTJs. However, the estimated spin polarization of Co_2MnSi was rather low if compared with expected value, even at 2 K. The further optimization of fabrication process to improve the MgO/Heusler alloys interface state is necessary to obtain a larger TMR ratio.

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