Magnetoresistance Effect in Current-Perpendicular-to-Plane Magnetoresistive Devices using Co₂Fe_xMn_{1-x}Si Heusler Alloy

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1. Introduction

Current-perpendicular-to-plane giant magnetoresistive (CPP-GMR) devices have been extensively studied. The CPP-GMR head is expected to replace the tunneling magnetoresistance (TMR) head in future high-density hard disk drive (HDD) because of the small resistance area product (*RA*) that is suitable for achieving the high-speed reading. However, the MR ratio in the typical CPP-GMR devices using 3d-ferromagnetic electrodes is too small to apply them to high-density HDD read heads. Therefore, it is an important subject to improve MR ratio in CPP-GMR devices.

Using high spin polarization materials as ferromagnetic electrodes of CPP-GMR devices has been considered to be promising for achieving a large MR ratio. Ideal high spin polarization materials are half metallic ferromagnets (HMFs), which have a band gap at the Fermi level for one spin band. Although some HMFs are expected theoretically, full-Heusler alloys are interesting and attractive HMFs. Several Heusler alloy compounds such as Co₂Mn(Al, Si), Co_2MnGe , $Co_2(Cr, Fe)Al$ are expected to be HMFs theoretically [1, 2, 3], if they have ordered $L2_1$ structure. In addition, some Co-based full-Heusler alloys have a high Curie temperature. Magnetic tunnel junctions (MTJs) with Co-based Heusler alloy electrodes have been investigated by several group, and a very large TMR effect has already been demonstrated [4, 5, 6]. Recently, a relatively large magnetoresistance ratio around 30-40% has been observed in CPP-GMR devices with structure of Co-based Heusler alloy electrodes separated by a nonmagnetic spacer [7, 8]. However, the observed MR ratio in CPP-GMR devices is much lower than TMR ratio in MTJs and is not enough to be used as the magnetic read heads for high-density HDD.

Among the many Co-based full Heusler alloys, $Co_2Fe_xMn_{1-x}Si$ alloys are promising because they have a half metallic electronic structure and a very high Curie temperature. In addition, an $L2_1$ -ordered structure can be easily obtained [9]. In our previous work, MTJs with $Co_2Fe_xMn_{1-x}Si$ (x = 0.4-0.6) electrodes and an Al-oxide barrier exhibited a large TMR effect [10]. This result proves that $Co_2Fe_xMn_{1-x}Si$ (x = 0.4-0.6) alloys have a very high spin polarization. In this work, we investigated the MR properties in CPP-GMR devices with $Co_2Fe_xMn_{1-x}Si$ Heusler alloy electrodes and Ag spacer layers in order to observe a very large MR effect.

2. Experimental method

The GMR structure of MgO(001)-sub./Cr(20)/Ag(50)/ $Co_2Fe_xMn_{1-x}Si(20)/Ag(5)/Co_2Fe_xMn_{1-x}Si(3 \text{ or } 5)/Ag(3)/Ru$ (3) (in nm, x = 0.0, 0.4, 1.0) were prepared with an ultra-high vacuum (UHV) magnetron sputtering system. The MgO substrate was annealed at 600°C to remove surface contaminates. The Cr/Ag buffer layer and the bottom Co₂Fe_xMn_{1-x}Si layer were deposited at room temperature (RT). After the deposition of the $Co_2Fe_xMn_{1-x}Si$ layer, the film was annealed at $T_a = 300-500$ °C to improve the chemical ordering and surface flatness. The Ag spacer layer and the upper Co₂Fe_xMn_{1-x}Si layer were also deposited at RT, and the films were annealed at 450° C after the deposition of the upper Co₂Fe_xMn_{1-x}Si layer. The capping layer of Ag/Ru was prepared in order to prevent oxidation. The crystal structure of prepared films was characterized by X-ray diffraction (XRD) and dark-field scanning transmission electron microscopy (STEM). STEM measurement was carried out by Toray Research Center (TRC). The deposited films were patterned into pillars for CPP-type four terminal device structures using electron beam (EB) lithography and Ar ion milling. The size of the pillars in the substrate was varied from 0.0145 to 0.132 μ m². The pillars were insulated using SiO₂ insulating layers. The MR characteristics were measured at RT by using the DC four-terminal method by applying a magnetic field along the magnetic easy axis of both Co₂Fe_xMn_{1-x}Si layers.

3. Experimental results and discussion

Figure 1 (a) shows the XRD patterns (2θ -scan) for the Cr(20)/Ag(50)/Co₂Fe_xMn_{1-x}Si(20)/Ru(3) layer (x = 0.0, 0.4, 1.0) deposited on the MgO(001) substrate. The film was deposited at RT and annealed at 500°C after the deposition of the Co₂Fe_xMn_{1-x}Si layer. The peaks of Cr(002), Ag(002), Co₂Fe_xMn_{1-x}Si(002), and Co₂Fe_xMn_{1-x}Si(004) were observed except for the peaks from the MgO substrate. This result indicates that all the layers of Cr, Ag, and Co₂Fe_xMn_{1-x}Si had a (001)-orientation. Figure 1(b) shows the ϕ -scan profile of the (111) peak, which is a superlattice peak for the *L*2₁ ordering. We clearly observed four-fold peaks of (111) and found that the fabricated films were epitaxially grown and had an *L*2₁ ordered structure.

Figure 2 shows a cross-sectional STEM image of $Co_2Fe_{0.4}Mn_{0.6}Si$ layer in the MgO-sub./Cr(20)/ Ag(50)/Co_2Fe_{0.4}Mn_{0.6}Si(20)/Ru(3) film annealed at 500°C. The $Co_2Fe_{0.4}Mn_{0.6}Si$ layer shows (001)-orientation. In



Figure 1 XRD patterns of (a)2 θ -scan and (b) ϕ -scan of (111) peak for the MgO(001)-sub./Cr(20)/Ag (50)/Co₂Fe_xMn_{1-x}Si (20)/Ru(3) films (x = 0.0, 0.4, 1.0).



Figure 2 Cross-sectional STEM image of the MgO(001)sub./Cr(20)/Ag(50)/Co₂Fe_{0.4}Mn_{0.6}Si (20)/Ru(3) film.

addition, we can easily distinguish the Co, Fe and Mn from the Si atoms, since the image contrast is roughly proportional to the square of the atomic number [11]. The STEM image indicates that the $Co_2Fe_{0.4}Mn_{0.6}Si$ layer has an $L2_1$ ordered structure. This result is consistent with the XRD results.

We investigated the annealing temperature dependence of MR ratio in the CPP-GMR devices with 5-nm-thick $Co_2Fe_xMn_{1-x}Si (x = 0.0, 0.4, 1.0)$ upper electrodes. Maximum MR ratios for each $Co_2Fe_xMn_{1-x}Si$ compositions were 31.6%(x = 0.0), 37.1%(x = 0.4) and 36.2%(x = 1.0). The behavior that the large MR was observed for $Co_2Fe_{0.4}Mn_{0.6}Si$ electrodes is similar to that in MTJs [10]. Figure 3 shows the MR curve for the CPP-GMR device with a 3-nm-thick $Co_2Fe_{0.4}Mn_{0.6}Si$ upper electrode. The maximum MR ratio was 50.1%. This is the highest value to date for CPP-GMR devices. The observed large MR ratio is originated from the large ΔRA value of $55.8 \text{ m}\Omega\mu\text{m}^2$. The large ΔRA may be due to the large spin asymmetry in the bulk $Co_2Fe_{0.4}Mn_{0.6}Si (\beta)$ and at the $Co_2Fe_{0.4}Mn_{0.6}Si/Ag$



Figure 3 MR curve for the $Co_2Fe_{0.4}Mn_{0.6}Si(20)/Ag(S)$ $Co_2Fe_{0.4}Mn_{0.6}Si(3)$ CPP-GMR device.

interface (γ). In addition, *RA* value of 111.4 m $\Omega\mu$ m² is suitable for the high-density HDD head application.

4. Conclusions

CPP-GMR devices with $Co_2Fe_xMn_{1-x}Si/Ag/Co_2Fe_x$ - $Mn_{1-x}Si$ structure were fabricated. The $Co_2Fe_{0.4}Mn_{0.6}Si/Ag/Co_2Fe_{0.4}Mn_{0.6}Si$ CPP-GMR device showed a very large MR ratio of 50.1%. This MR ratio is the largest to date in CPP-GMR devices. The CPP-GMR device with $Co_2Fe_{0.4}Mn_{0.6}Si$ electrodes would be useful for the high-density HDD read heads.

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