Bias-voltage-dependence of magneto-resistance for epitaxial Fe/MgO/Co$_2$MnSn tunnel junctions

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Abstract. Tunnel magnetoresistance (TMR) was investigated for fully epitaxial Fe/MgO/Co$_2$MnSn magnetic tunnel junctions (MTJs) fabricated on MgO (001) substrates. The bias-voltage-dependence of the TMR ratio showed that the sign of TMR ratio at 300 K becomes negative for the bias voltage between -800 mV and +200 mV, which may be caused by the impurity state or other imperfections at the interface. The sign change in TMR effect was also observed with changing the temperature at low fixed bias voltage, implying that the effect is not simply due to a bias-dependent change of the density of state at the Fermi level.

1. Introduction
The tunnel magnetoresistance (TMR) of magnetic tunnel junctions (MTJs) made of two ferromagnetic electrodes separated by an insulating layer has attracted much interest for its use in new magnetic devices [1-2]. Within the Jullière model, the magnitude of TMR is determined by the spin polarization of the density of states at the Fermi energy of the two ferromagnetic electrodes [3]. Half metallic materials, which exhibit perfect spin polarization at the Fermi energy, are expected to be ideal materials to achieve large TMR ratio. Recently, TMR ratio of above 300 % at RT [4] was achieved in MTJs using Co-based Heusler alloys [4-7] which are theoretically predicted as half metallic materials [8-9]. On the other hand, we have reported that Co$_2$MnSn Heusler alloy films with relatively uniform local magnetism can be prepared when the films are grown by atomically controlled alternate deposition at 400 °C [10].

In this paper, we report on bias-voltage-dependence of the magneto-resistance for fully epitaxial Fe/MgO/Co$_2$MnSn MTJs. The negative TMR effect was observed between -800 mV and +200 mV at 300 K. Moreover, it was found that, at low bias voltage, the sign of TMR changed as a function of temperature.

2. Experiments
Layered structures of MgO (5 nm)/Cr (30 nm)/Fe (20 nm)/MgO (2.4 nm)/Co$_2$MnSn (26.1 nm)/Co (10 nm)/Cr (5 nm) were prepared on MgO (001) substrates using an electron beam deposition system. The bottom ferromagnetic Fe layer was deposited at RT and annealed at 350 °C in order to improve crystallographic quality. After the deposition of the MgO barrier layer, the top ferromagnetic Co$_2$MnSn layer was prepared by depositing one atomic layer of Co, half an atomic layer of Mn and Sn alternately in a controlled manner at the substrate temperature of 400 °C. The interface atoms of the
Co$_2$MnSn layer on the MgO barrier were designed to be Mn and Sn [10]. The crystal structures of the layered structures were investigated using x-ray diffraction (XRD) with $\theta$ - 2$\theta$ and polar plots. MTJs were patterned into ellipse-shaped pillars of 9.0 $\times$ 4.5 $\mu$m$^2$ or 6.0 $\times$ 3.0 $\mu$m$^2$ using a conventional photolithography and Ar ion milling process. Magnetoresistance measurements were carried out using a standard dc four-probe method. The positive current is defined as the current with the electrons flowing from the top Co$_2$MnSn layer to the bottom Fe layer.

3. Results

Figure 1(a) shows the $\theta$ - 2$\theta$ X-ray diffraction pattern of an MgO/Cr/Fe/MgO/Co$_2$MnSn/Co/Cr layered structure. The Co$_2$MnSn (002), Co$_2$MnSn (004) and Fe (002) peaks were observed, which indicates that the Co$_2$MnSn and Fe electrodes were grown with the (001) orientation epitaxially on the MgO (001) substrate. The polar plot for the (111) peaks of the Co$_2$MnSn layer is shown in figure 1(b). The pattern indicates that the Co$_2$MnSn layer has an L2$_1$ structure. The degree of L2$_1$ order of Co$_2$MnSn estimated from the intensity ratio of (111) and (002) was about 56%.

Figure 1. X-ray diffraction patterns of the MgO/Cr/Fe/MgO/Co$_2$MnSn/Co/Cr layered structure. (a) $\theta$ - 2$\theta$ plot with the scattering vector normal to the film plane, and (b) polar plot for the (111) peaks of the Co$_2$MnSn layer.

Figure 2. (a) Magnetoresistance curves for the epitaxial Fe/MgO/Co$_2$MnSn MTJ under various bias voltages at 300 K. (b) Bias-voltage-dependence of TMR ratio and RA products for anti-parallel (AP) and parallel (P) configurations at 300 K.
The magnetoresistance curves under various bias voltages at 300 K are shown in figure 2(a). At bias voltages of +500 mV and -200 mV, positive and negative TMR with the ratios of +6.3% and -10.9% were observed, respectively. Figure 2(b) displays the bias-voltage-dependence of the TMR ratio and resistance area products (RA products) for anti-parallel (AP) and parallel (P) configurations. The bias-voltage-dependence of TMR exhibits an asymmetric character regarding the polarity of the voltage, and negative TMR ratios are realized in the bias voltage range between -800 mV and +200 mV. The RA products exhibit resistance drops at -200 mV and +400 mV for AP and P configurations, which correspond to the maximum and minimum TMR ratio, respectively. Figure 3 shows the magnetoresistance curves at a bias voltage of +100 mV at 5 K, 100 K and 300 K. A negative TMR effect was observed at 300 K, whereas a positive TMR effect appeared at 5 K. The sign change of the TMR effect with temperature was observed at bias voltage between -100 mV and +150 mV.

4. Discussion

The change in the sign of the TMR effect as a function of bias voltage at 300 K similar to that in figure 2(a) has been reported for some systems. Sharma et al. performed magnetoresistance measurements for Permalloy/Al₂O₃/Ti₂O₅/Permalloy MTJs and observed positive and negative TMR effects depending on the bias voltages [11]. They concluded that the phenomenon was caused by the difference of the spin polarization of two barrier/electrode interfaces and by the sign change of the spin polarization with the bias voltage in one side. The sign change of TMR effect with changing the bias voltages can also occur by the impurity states near the Fermi surface [12], Co anti-sites [13] and non-bonding states at the interface [14]. For comparison with the Fe/MgO/Co₂MnSn MTJs, Fe (30 nm)/MgO (2.4 nm)/CoFe (10 nm) MTJs were prepared. The magnetoresistance of the Fe/MgO/CoFe MTJs displayed large TMR ratio of about 150% at a bias voltage of +100 mV and the TMR ratio simply decreased with increasing the bias voltage at 300 K, which means that the TMR behaviour of the Fe/MgO/Co₂MnSn MTJs may be due to the impurity states or other imperfections at the Co₂MnSn/MgO interface.

The temperature-dependent magnetoresistance curves in figure 3 indicate that the sign change of TMR ratio is not simply due to the difference of spin-dependent density of states (DOS) because the DOS-based spin polarization in barrier/electrode interfaces would be almost the same in the range between 5 K and 300 K. In other words, the temperature independent electron transport coexists with temperature dependent one in the TMR effect for the Fe/MgO/Co₂MnSn MTJs. The strong
temperature dependence of the TMR ratio at low bias voltage may be due to the magnon-assisted inelastic tunnelling. Marukame et al. reported that the TMR ratio for the Co$_2$MnGe/MgO/CoFe MTJs showed strong dependence on the temperature at a low bias voltage [15]. The TMR curves in their paper showed that the temperature-dependent sign change occurs in the negative bias voltage range between -300 mV and -200 mV, which is larger voltage than the present case. The result in the present paper implies that the mechanism is not simply due to the DOS-based spin-dependent tunnelling for the Fe/MgO/Co$_2$MnSn MTJs.

5. Conclusion
We have investigated the tunnel magnetoresistance effect for fully epitaxial Fe/MgO/Co$_2$MnSn MTJs with the Co$_2$MnSn layer prepared using atomically controlled alternate deposition. The TMR measurements at various bias voltages showed a negative TMR effect at the bias voltage between -800 mV and +200 mV at 300 K. At the low bias voltage, positive and negative TMR effects were observed at 5 K and 300 K, which may be caused by the increase of magnon scattering with increasing temperature.

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