Local Magnetism of Co$_2$MnSn Heusler Alloy Films and Magnetoresistance of Co$_2$MnSn-Based Magnetic Tunnel Junctions

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Abstract. Co$_2$MnSn Heusler alloy films were prepared by atomically controlled alternate deposition on MgO(001) substrates and the local magnetism was investigated using $^{119}$Sn Mössbauer spectroscopy. It turned out that the magnetic environments around the Sn sites in the films prepared by this method are uniform in comparison with those in bulk alloys prepared by arc melting. The local uniformity around the Sn sites depends on the growth temperature. The magnetoresistance of the magnetic tunnel junctions with a Co$_2$MnSn/MgO/Co$_2$MnSn/Co structure, where the Co$_2$MnSn layers were prepared by the same method, was also examined. Tunneling magnetoresistance effect has been observed for the samples where the local magnetism is relatively uniform, although the observed ratio is not so large at the present stage.

1. Introduction

Realization of large tunnel magnetoresistance (TMR) has been a key subject in the field of spintronics in this decade. One of the promising methods to achieve large TMR effects is the use of Heusler alloys, which are theoretically expected to be half metallic [1], for the ferromagnetic layers of magnetic tunnel junctions (MTJs) with a ferromagnetic-metal/nonmagnetic-insulator/ferromagnetic-metal structure. Several groups have investigated MTJs with Heusler alloys and realized large magnetoresistance ratio of a few hundred % at room temperature [2-4]. The magnitude of magnetoresistance is, however, not yet satisfactorily large as a system with half metals. Moreover, the TMR effects, which are large at low temperatures, reduce significantly around room temperature. In order to find a key to solve these problems, it seems important to prepare good quality Heusler alloy films with little site disorder and high interface uniformity, and examine the structure and magnetism with local experimental methods such as Mössbauer spectroscopy [5, 6] and NMR [7].

In this work, we fabricated Co$_2$MnSn Heusler alloy films by atomically controlled alternate deposition and investigated local magnetism using $^{119}$Sn Mössbauer spectroscopy. The influence of crystallographic disorder and interface effects on the local magnetism of the films can be examined through the magnetic hyperfine field induced at the Sn nuclear sites of the Heusler alloy films. MTJs with Co$_2$MnSn layers and an MgO barrier were also prepared, and the TMR effect was compared with the local magnetism.

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2. Experiments

Co$_2$MnSn films were prepared by depositing one atomic layer of Co, half an atomic layer of Mn and Sn alternately in a controlled manner on a Cr buffer layer grown on MgO(001) substrates using a vacuum vapour deposition system with e-gun heating. Sn was occasionally enriched with $^{119}$Sn to obtain $^{119}$Sn Mössbauer spectra effectively. Mössbauer spectra were measured by means of conversion electron Mössbauer spectroscopy. The spectra were fitted with magnetically split sextet with a distribution of magnetic hyperfine fields. Layered structures of Co$_2$MnSn(30.2 nm)/MgO(2 nm)/Co$_2$MnSn(10.2 nm)/Co(50 nm) were also prepared, with the Co$_2$MnSn layers made by the same method. The bottom Co$_2$MnSn layer was grown at the substrate temperature $T_{s1} = 400$ °C, MgO barrier at 30 °C, and the top Co$_2$MnSn and Co layers at $T_{s2} = 300$, 200, 100 and 30 °C. The Co layer deposited on the top Co$_2$MnSn layer is for making a difference in the coercive fields of the ferromagnetic layers at both sides of the MgO layer. A part of each layered structure was fabricated into MTJs of an ellipse-shaped pillar with the in-plane size of several μm using photolithography and Ar ion etching, and the rest of it was used for Mössbauer measurements. Thus, the correlation between the local magnetism and the magnetoresistance ratio was examined using the identical layered structure.

3. Results and Discussion

A typical absorption Mössbauer spectrum of bulk Co$_2$MnSn alloys prepared by arc melting, for comparison, is shown in figure 1(a). The spectrum can be fitted with a set of magnetically split sextets having two peaks in the hyperfine distribution curve and isomer shift of about 1.4 mm/s (relative to CaSnO$_3$), which agrees well with the spectra in the literatures [8-10]. In Co$_2$MnSn alloys with an ideal L$_{21}$ structure, Sn atoms occupy a single site with eight Co atoms in the nearest neighbours and six Mn atoms in the second nearest. The Mössbauer results indicate that the Co$_2$MnSn alloys are not perfectly L$_{21}$-ordered, and have two different environments as Sn sites, although the site causing the minor component with smaller magnetic hyperfine field has not been identified clearly.

The conversion electron Mössbauer spectrum and distribution of magnetic hyperfine fields for the single Co$_2$MnSn layer with 40.2 nm in thickness, which is prepared by atomically controlled alternate deposition with a substrate temperature of 400 °C, are shown in figure 1(b). The peak with smaller hyperfine field is not significant and the width of the peak at around 10 T is relatively narrow.

![Figure 1](image-url)

**Figure 1.** (a) $^{119}$Sn absorption Mössbauer spectrum and distribution of magnetic hyperfine field for a bulk Co$_2$MnSn alloy prepared by arc melting. (b) $^{119}$Sn conversion electron Mössbauer spectrum and distribution of magnetic hyperfine field for a Co$_2$MnSn(40.2 nm) film grown by atomically controlled alternate deposition at 400 °C.
suggesting that the magnetic environments around the Sn sites are uniform in comparison with those in the bulk alloys. The distribution was sharpest when the film was deposited at 400 - 500 ºC, so that we have chosen the substrate temperature of 400 ºC to prepare the bottom Co$_2$MnSn layer in the MTJ structure. The sharp distribution in hyperfine fields also makes it possible to examine interface-originating local magnetism of Co$_2$MnSn of magnetic tunnel junctions.

The spectra for Co$_2$MnSn/MgO/Co$_2$MnSn/Co layered structures, where the Co$_2$MnSn layer on top of the MgO barrier was grown with different temperature ($T_{s2}$), are shown in the left of figure 2. The line width of the magnetically split sextet is broader than that for the single Co$_2$MnSn layer in figure 1(b). Noticeable intensity exists at the center of the spectra (around 1.5 mm/s), especially for the samples grown at 300ºC and 30ºC. The distribution of hyperfine field is shown in the right of figure 2. The peak at around 10 T in the histogram is broader and the portion in the lower field increases in comparison with the distribution for the single layer shown in figure 1(b). Since the preparation condition of the bottom Co$_2$MnSn layer is the same as that of the single layer, the origin of the distribution of the hyperfine field can be attributed to the interface of the bottom Co$_2$MnSn layer and/or to the top Co$_2$MnSn layer. The distribution around 0 - 3 T is larger for the samples prepared at 300ºC and 30ºC, indicating that a part of the Co$_2$MnSn layers in these samples is paramagnetic or with very small magnetization. If the Co$_2$MnSn layers react with MgO to form a nonmagnetic oxide, a peak would appear at 0 mm/s, which results in an asymmetric shape of spectra. Such asymmetry is not so significant, but exists in the spectrum for the sample grown at 300 ºC. In this way the local magnetic uniformity appears better in the sample grown at 200ºC and 100ºC. Since the preparation condition is the same up to the MgO barrier, the upper Co$_2$MnSn layers, including the interface, are thought to have slightly different local magnetism. Note that the magnetization value is a little smaller than 5 $\mu_B$ per formula unit, which is an expected value as a half metal, and does not depend significantly on the growth temperatures.

The TMR curve for the samples grown at 200ºC and 100ºC are shown in figure 3. Small but clear magnetoresistance effect due to the parallel and antiparallel magnetic configurations between the two Co$_2$MnSn layers is observed. On the other hand, magnetoresistance change was not detected at all for the sample grown at 300ºC and 30ºC. As mentioned above, the local magnetism is less uniform in the

Figure 2. $^{119}$Sn Mössbauer spectra and distribution of magnetic hyperfine field for Co$_2$MnSn(30.2 nm)/MgO(2.0 nm)/Co$_2$MnSn(10.2 nm)/Co(50.0 nm) layered structures grown at $T_{s1}$ = 400 ºC for the bottom Co$_2$MnSn and at $T_{s2}$ = 300, 200, 100, and 30 ºC for the top Co$_2$MnSn.
latter samples. The magnetoresistance ratio has correlation with the local magnetic uniformity of the upper Co$_2$MnSn layers.

4. Summary

MTJs with high quality Co$_2$MnSn were prepared by atomically controlled alternate deposition on MgO(001) substrates and the local magnetism was investigated using $^{119}$Sn Mössbauer spectroscopy. It turned out that the magnetic environments around the Sn sites in the samples prepared by this method are uniform in comparison with those in bulk alloys prepared by arc melting. The local uniformity around the Sn sites depends on the growth temperature. The magnetoresistance effect is observed for the MTJs with a Co$_2$MnSn/MgO/Co$_2$MnSn/Co structure when the local magnetism is relatively uniform, although the magnitude of the magnetoresistance ratio is not so large at the present stage. Further improvement in local magnetic uniformity would be important to achieve larger TMR ratio.

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