Local magnetism of Co$_2$MnSn Heusler alloy films prepared by atomically controlled alternate deposition

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Abstract. Co$_2$MnSn Heusler alloy films were prepared by atomically controlled alternate deposition on MgO(001) substrates. The local magnetism was investigated through the distribution of magnetic hyperfine fields obtained from $^{119}$Sn Mössbauer spectroscopic measurements. It turned out that the magnetic environments around the Sn sites in the films prepared by this method with appropriate growth temperatures are relatively uniform in comparison with those in bulk alloys prepared by arc melting. The Mössbauer spectroscopy was applied also to Co$_2$MnSn/MgO/Co$_2$MnSn/Co layered structures and Co$_2$MnSn/Ag multilayers to examine crystallographic uniformity of the Co$_2$MnSn layers and possibility of magnetic anomaly at the interface regions.

1. Introduction

Half-metallic materials, where the conduction electrons are 100% spin-polarized, have recently been studied intensively from theoretical and experimental viewpoints in the field of spintronics. Such materials are promising candidates for ferromagnetic layers of magnetic tunnel junctions (MTJs) with a ferromagnetic-metal/nonmagnetic-insulator/ferromagnetic-metal structure for the realization of quite large tunnel magnetoresistance (TMR) effects. One series of materials which are expected to be half-metallic are X$_2$YZ Heusler alloys with an L2$_1$ structure [1]. Several groups have investigated MTJs with Heusler alloys and realized large magnetoresistance ratio of a few hundred % at room temperature [2-4]. However, the magnetoresistance change is not yet satisfactorily large as expected for systems of 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 using 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. The Mössbauer measurements were performed also for MTJs composed of Co$_2$MnSn layers and an MgO barrier to investigate the correlation between the local magnetism and the TMR effects. For more interface-sensitive measurements, multilayers of thinner Co$_2$MnSn layers and nonmagnetic Ag layers were prepared instead of Co$_2$MnSn/MgO multilayers, since it is easier to grow thin Co$_2$MnSn and Ag layers repeatedly to form multilayers with good crystallographic quality.
2. Experiments

Co$_2$MnSn films were prepared using a vacuum vapour deposition system with e-gun heating. One atomic layer of Co, half an atomic layer of Mn and Sn, correspondingly to the L$_2$$_1$ structure, were deposited alternately in a controlled manner on a Cr buffer layer grown on MgO(001) substrates. Sn was moderately enriched with $^{119}$Sn to obtain $^{119}$Sn Mössbauer spectra effectively. 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. A part of each sample was micro-fabricated into MTJs and the TMR effect was examined using the identical layered structure. Multilayers of Co$_2$MnSn(2.2 nm) and Ag(3 nm) with the bilayer repetition of 10 times were also prepared for more interface sensitive measurements. For all the layered structures, the interfaces of the Co$_2$MnSn layers were designed to be terminated with an atomic layer of Co, not with Mn and Sn. Mössbauer spectra were measured at room temperature by means of conversion electron Mössbauer spectroscopy using a Ca$^{119m}$SnO$_3$ source and a He+1%(CH$_3$)CH gas flow counter. The peak position of CaSnO$_3$ was used as a standard of zero velocity.

3. Results and Discussion

In Co$_2$MnSn alloys with an ideal L$_2$$_1$ structure, Sn atoms occupy a single crystallographic site with eight Co atoms as the nearest neighbours and six Mn atoms as the second nearest. However, the Mössbauer results on bulk Co$_2$MnSn alloys published so far indicate that two different environments with different hyperfine fields coexist as Sn sites, although the site causing the minor component with smaller hyperfine field has not been identified clearly [8-11]. A typical absorption Mössbauer spectrum of bulk Co$_2$MnSn alloy prepared by arc melting is shown in figure 1(a). The spectrum can be fitted with (i) two magnetic components with Voigt-type absorption lines (the solid lines in the left figure), i.e., with two Gaussian distributions of hyperfine fields (the solid lines in the right figure) or (ii) a set of magnetically split sextets having two peaks in the hyperfine distribution (the histogram in the right figure), with the isomer shift of about 1.4 mm/s.

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 was prepared by atomically controlled alternate deposition with a substrate temperature of 500 °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, suggesting that the magnetic environments around the Sn sites are uniform in comparison with those in the bulk alloys. The distribution changes as a function of deposition temperatures as shown in figure 2, and becomes sharp when the film is deposited at 400 °C or above. The sharp distribution in

![Figure 1](image-url)
hyperfine fields makes it possible to examine interface-originating local magnetism of Co$_2$MnSn-containing layered structures.

A typical example of the Mössbauer spectrum and the distribution of magnetic hyperfine field for Co$_2$MnSn/MgO/Co$_2$MnSn/Co layered structures is shown in figure 3, for the sample where the bottom Co$_2$MnSn layer was deposited at the substrate temperature of 400 °C and the top Co$_2$MnSn layer on the MgO barrier was grown at 100 °C. The peak at around 10 T in the histogram is broader and the distribution in the lower field range is larger in comparison with the histogram for the single layer deposited at 400 - 500 °C, which reflects the local crystallographic disorder in the top Co$_2$MnSn layer and possibly the interface magnetism of the bottom and top Co$_2$MnSn layers. For this sample, a small but clear magnetoresistance effect was observed associated with the parallel and antiparallel magnetic configurations between the two Co$_2$MnSn layers [12]. 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 the spectra. Such asymmetry is not significant in this sample. Actually, the distribution histogram changes dependent on the preparation conditions, and TMR effect is not observed when the component with smaller hyperfine fields is more significant [12].

A typical Mössbauer result for Co$_2$MnSn/Ag multilayers prepared at 400 °C for more interface sensitive experiment is shown in figure 4. In this sample, the nominal structure is designed in such a
way that 2/5 of Sn atoms are contacting the interfacial Co atoms and 3/5 is located inside the Co$_2$MnSn layers. The spectrum resembles that of the thicker single Co$_2$MnSn layer and no strong interface effect is observed in the histogram. The tiny peak around 2 mm/s may be due to a minor nonmagnetic Sn-rich site, which would not be intrinsic to the magnetic interface effect. Note that a preliminary result shows that there is also no strong interface effect even when the interfaces of the Co$_2$MnSn layers are designed to be terminated with atomic layers of Mn and Sn.

4. Summary
High quality Co$_2$MnSn 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 samples prepared by this method are relatively uniform in comparison with those in bulk alloys prepared by arc melting. The local uniformity around the Sn sites depends on the growth temperatures. The TMR effect was observed for the MTJs when the local magnetism of the Co$_2$MnSn/MgO/Co$_2$MnSn/Co layered structure was relatively uniform. No strong magnetic anomaly at the interface region of Co$_2$MnSn layers has been detected at the present stage. Further investigation is required to clarify the interface magnetism of Co$_2$MnSn films.

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