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Spin polarization of Co_2FeSi full-Heusler alloy and tunneling magnetoresistance of its magnetic tunneling junctions

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The authors report spin polarization (P) and tunneling magnetoresistance (TMR) of epitaxially grown Co_2FeSi thin films on a MgO (001) substrate. A Heusler-type $L2_1$ structure was observed in the samples sputter deposited at 473 K or above. The P value of the ordered film was measured as 0.49 ± 0.02 by the point contact Andreev reflection (PCAR) technique. The TMR values obtained from the magnetic tunneling junction (MTJ) using the Co_2FeSi electrode and Al-oxide barrier were 67.5% at 5 K and 43.6% at 298 K, respectively. The P value estimated from the TMR using Julliere's model matches the spin polarization measured by the PCAR very well, indicating that the TMR value from the MTJ is governed by the intrinsic value of P of the electrode material for incoherent tunneling. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2338025]

Ferromagnetic Heusler compounds have received much research interest for spintronics applications since the first prediction of their half-metallic behavior was made by de Groot *et al.*[1] Ideally, they show 100% spin polarization, due to a band gap for minority spin electrons at the Fermi level. Later, several other investigations[2–5] predicted half-metallic ferromagnetic properties of various X_2YZ compounds with the $L2_1$ structure, where X and Z are the transition metals and Z is a III_b or a IV_b group element. Since many Co-based Heusler alloys have high Curie temperature and a high magnetic moment,[6–8] they can be used as a ferromagnetic electrode of magnetic tunnel junctions (MTJs) for tunneling magnetoresistance[9,10] (TMR) or of spin valves for giant magnetoresistance. In MTJ structures, ferromagnetic half-metals sandwich a thin insulator layer, whose TMR value was predicted to follow Julliere's formula, $\text{TMR}=2P_1P_2/(1-P_1P_2)$, where P_1 and P_2 are the spin polarization of upper and lower ferromagnetic electrodes. The spin polarizations of ferromagnetic electrodes of MTJs are often deduced from TMR values using this formula, which involve interfacial effect. Thus, the P values deduced from Julliere formula may be different from the intrinsic spin polarization of the electrode materials. In fact, it is well known that this model is not valid for the spin-dependent tunneling with perfect crystalline barriers where the wave vector k is parallel to the interface and is conserved during the coherent tunneling.[11]

In previous studies, the magnetic moment of the Co_2FeSi compound was theoretically predicted to be $5.90\mu_B$ (Ref. 12) or $5.6\mu_B$ (Ref. 13) depending on the models used for calculations; however, these predictions were found to be systematically smaller than the experimentally obtained result of $6\mu_B$.[14] This contradiction was resolved by considering an on-site correlation in the local density approximation theory.[15] Due to the fact that Co_2FeSi shows integer magnetic moment, thus located at the Slater-Pauling curve of half-metallic Heusler alloys,[16] high spin polarization value was predicted. In this work, we measured the spin polarization (P) of an epitaxially grown Co_2FeSi thin film by the point contact Andreev reflection (PCAR) method[17] and investigated its structural characterizations. The TMR values of the MTJs using a Co_2FeSi electrode and the spin polarization estimated from Julliere's model are also reported for comparison.

Co_2FeSi films were grown on MgO (001) substrates using a stoichiometric composition target with a slow deposition rate of about 0.03 nm/s to obtain smooth interfaces. The substrate temperature (T_s) was controlled from room temperature (RT) to 773 K. The MTJs composed of Co_2FeSi (20 nm)/Al oxide (1.2 nm)/$Co_{75}Fe_{25}$ (3 nm)/IrMn (15 nm)/Ta (60 nm) for TMR measurements as well as Co_2FeSi (20 nm) films on MgO (001) for spin polarization measurements were prepared using an UHV sputtering machine with a base pressure below 2×10^{-7} Pa. The 1.2 nm thick Al-oxide layer for the tunneling barrier was fabricated by plasma oxidation inside the sputtering chamber. The crystal structures were characterized by x-ray diffraction using a Cu $K\alpha$ source. The TMR measurements were carried out on a 10×10 μm^2 junction area, created by electron beam lithography and Ar ion etching, using the conventional dc four-point technique. The spin polarization (P) was measured by the PCAR technique using Co_2FeSi/Nb contacts in liquid helium. Many contacts were mechanically made by positioning a sharp needle at various positions on the sample surfaces. The interface barriers can be described by introducing a dimensionless Z parameter into the Blonder-Tinkham-Klapwijk (BTK) theory,[18] which allows us to evaluate the *intrinsic* P value by extrapolating P values for various Z to $Z=0$.

The x-ray diffraction (XRD) patterns taken in conventional θ-2θ geometry (out of plane) from the Co_2FeSi thin films on MgO (001) substrates at various deposition temperatures are shown in Fig. 1(a). The absence of the (200)

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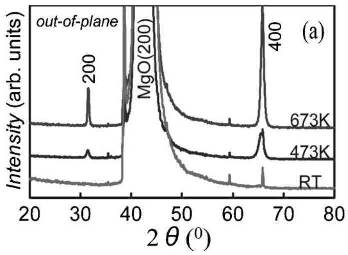

FIG. 1. Out of plane XRD pattern (a) and pole figure of the $L2_1$ (111) peak (b) of the Co_2FeSi film deposited at various temperatures on MgO (001) substrate.

superlattice reflection expected from the $B2$ structure indicates that the sample sputtered at T_s=RT has a disordered $A2$ structure. However, the (200) peak is visible in the thin films prepared at T_s=473 and 673 K, suggesting the presence of ordered $B2$ phase. The φ scan with an incident angle of 0.5° taken from the (111) superlattice reflection of the ordered $L2_1$ phase is shown in Fig. 1(b). The higher substrate temperature results in a higher peak intensity indicating a higher degree of ordering. Furthermore, the fact that the (111) peaks have a fourfold symmetry is an evidence for the single-crystalline Co_2FeSi film. A detailed study of the effect of substrate temperature and postannealing on structural ordering in the Co_2FeSi alloy is presented elsewhere.[19,20] These previous results suggest a lower spin polarization value; however, a theoretical calculation on the effect of disordering in the Co_2FeSi compound has not been reported yet.

The selected area electron diffraction (SAED) pattern of the [110] zone axis of the Co_2FeSi single crystal including the MgO substrate is shown in Fig. 2(a). Arrows indicate the fundamental reflections corresponding to the $A2+B2+L2_1$ phases, while dotted circles mark the reflections of both $B2+L2_1$ phases. The evidence for the presence of the $L2_1$ ordered structure is emphasized using solid circles. Furthermore, we can conclude that Co_2FeSi grows epitaxially with the [110] zone parallel to the [100] zone of the MgO substrate (a=4.23 Å), which is a consequence of the smallest lattice mismatch (~5%) between the corresponding planes in the two structures. Figure 2(b) shows a dark field transmission electron microscopy (TEM) image excited with the (111) spot of the Co_2FeSi film grown on a MgO (001) substrate at T_s=473 K. The fully enlightened image also confirms the presence of the $L2_1$ phase sample widely. The lattice parameter of Co_2FeSi thin film deduced from XRD was 5.67 Å, slightly larger than it was recently reported elsewhere.[21] Figure 3. shows the cross-sectional high-resolution TEM (HRTEM) image of the MTJ. The interfaces of Co_2FeSi/Al-oxide/$Co_{75}Fe_{25}$ layers prepared at 473 K are all atomically sharp. The Al–O is an amorphous oxide with a uniform thickness (2 nm) with no defects.

Maximum TMR values of MTJs with a Co_2FeSi (100) electrode were 67.5% at 5 K [Fig. 4(a)] and 43.6% at RT [Fig. 4(b)] after deposition at 473 K and followed by post-annealing at 573 K. From the TMR values at 5 K, the spin

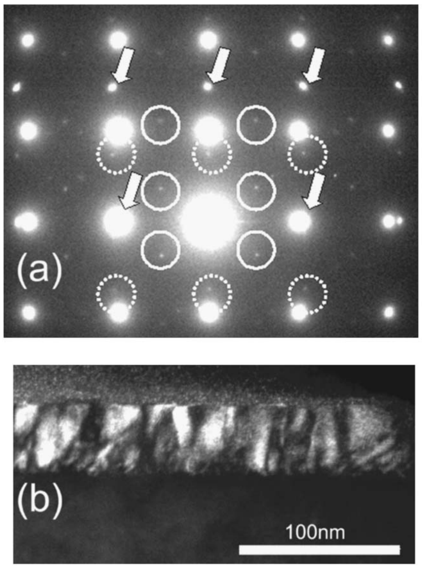

FIG. 2. (a) SAED pattern taken from the interface area of the Co_2FeSi [110] thin film and the MgO substrate. Arrows denote the fundamental reflections of the $A2+B2+L2_1$ phases and dotted circles indicate the reflections of both $B2+L2_1$ phases. The evidence for the presence of the $L2_1$ ordered structure is marked by solid circles. (b) Dark field TEM image including the $L2_1$ super-reflection.

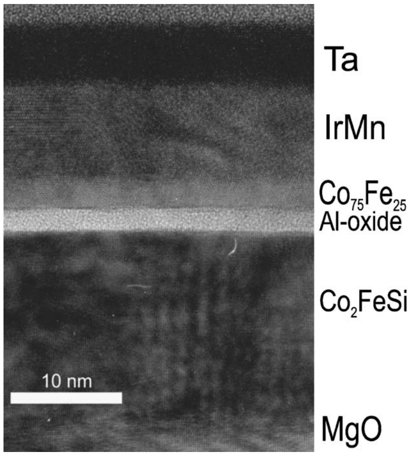

FIG. 3. HRTEM cross-sectional image of the MgO/Co_2FeSi (20 nm)/Al-oxide (2 nm)/$Co_{75}Fe_{25}$ (3 nm)/IrMn (10 nm)/Ta (7 nm) magnetic tunnel junction.

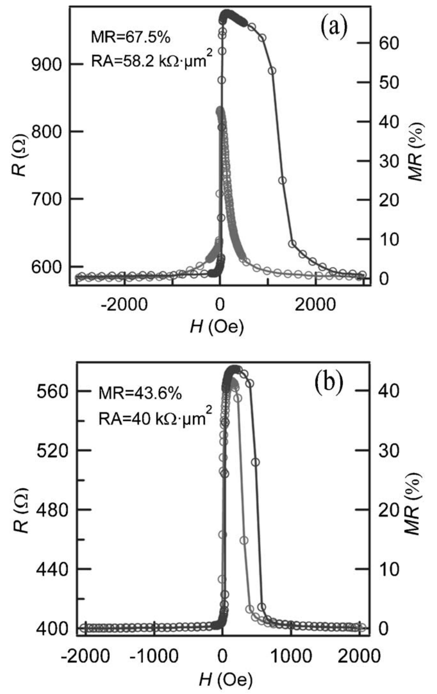

FIG. 4. Magnetoresistance curves of the Co_2FeSi junction measured at 5 K (a) and RT (b).

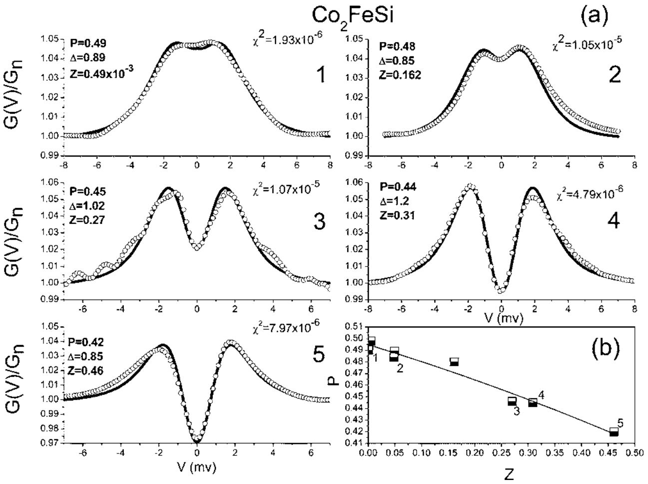

FIG. 5. Representative conductance vs voltage curves of the Co_2FeSi/Nb contacts for various Z interface strengths (a). Open circles are experimental data and solid lines are the fits using the modified BTK model. Extrapolation to $Z=0$ results in the inner spin polarization value (b).

polarization of the Co_2FeSi electrode material is deduced to be $P \sim 0.5$ assuming that the spin polarization value for the $Fe_{75}Co_{25}$ electrode is 0.5.

Figure 5(a) shows the normalized conductance $[G(V)/G_n]$ curves that were obtained by the PCAR measurement using the Co_2FeSi/Nb contacts. The following parameters are evaluated during the fitting process using the BTK theory: spin polarization (P), superconducting gap (Δ), and dimensionless parameter Z to describe the strength of the interfacial scattering barrier. According to the modified BTK theory,[22] the P value depends on the Z barrier strength and contacts with a higher Z result in a lower spin polarization value. Only the P value with a transparent interface ($Z=0$) corresponds to the intrinsic spin polarization. This scattering free state can only be measured directly in a few cases; however, extrapolation to $Z=0$ using the (Z,P) data pairs obtained from the different contacts results in the intrinsic P value [see Fig. 5(b)]. Thus, the spin polarization value of the ordered Co_2FeSi was found to be $P=0.49\pm0.02$. The P value derived from the TMR value of the MTJ using Julliere's model[23] was $P \sim 0.50$, which is in excellent agreement with the PCAR measurement result and indicates that the TMR value of the defect-free high quality MTJ (Fig. 3) can be expressed with Julliere's model. The experimentally determined P value is smaller than the value predicted from the density of state (DOS) calculations, because the DOS calculations were performed for the fully $L2_1$ ordered state.

In summary, we have measured the spin polarization of epitaxially grown Co_2FeSi thin film as $P=0.49\pm0.02$ using the PCAR method. This value is in excellent agreement with the value that was deduced from the TMR value of the Co_2FeSi/Al-oxide/$Co_{75}Fe_{25}$ MTJ using Jullier's formula. This means that the TMR value that was obtained from the MTJ is governed by the intrinsic value of P without much influence from the interface in case that the interfacial structure of the junctions is smooth as confirmed by the TEM observation and that TMR appears with the incoherent tunneling. The experimentally determined P value is smaller than it was predicted by the DOS calculations, which may be attributed to the site disorder in the Co_2FeSi films. TEM observations do not confirm perfectly ordered $L2_1$ structure in the samples. Therefore, further optimization of thin film preparation conditions to achieve a higher degree of order in the Co_2FeSi electrode should improve the TMR effect even for the incoherent MTJ.

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