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Interface magnetism of Co_2FeGe Heusler alloy layers and magnetoresistance of Co_2FeGe/MgO/Fe magnetic tunnel junctions

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The interface magnetism between Co_2FeGe Heusler alloy layers and MgO layers was investigated using ^{57}Fe Mössbauer spectroscopy. Interface-sensitive samples, where the ^{57}Fe isotope was used only for the interfacial atomic layer of the Co_2FeGe layer on the MgO layer, were prepared using atomically controlled alternate deposition. The ^{57}Fe Mössbauer spectra of the interface-sensitive samples at room temperature were found similar to those of the bulk-sensitive Co_2FeGe films in which the ^{57}Fe isotope was distributed throughout the films. On the other hand, the tunnel magnetoresistance effect of magnetic tunnel junctions with Co_2FeGe layers as the ferromagnetic electrodes showed strong reduction at room temperature. These results indicate that the strong temperature dependence of the tunneling magnetoresistance of magnetic tunnel junctions using Heusler alloy electrodes cannot be attributed simply to the reduction of the magnetization at the interfaces between the Heusler alloy and insulator layers.
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I. INTRODUCTION

Tunnel magnetoresistance (TMR) effect of magnetic tunnel junctions (MTJs) is a key technological element for spin electronic devices.[1,2] The performance improvement of the MTJs is required for further development of spin electronics. To increase the TMR effect, a current focus is much on highly spin-polarized materials as ferromagnetic electrodes of the MTJs. Some cobalt-based Heusler alloys are predicted to be highly spin-polarized materials[3–5] and have been used for ferromagnetic electrodes.[6–10] The high spin polarization of $Co_2FeGa_xGe_{1-x}$ Heusler alloy has been confirmed by point contact Andreev refection measurements[9] and TMR ratios of over 1000% at low temperature were reported for MTJs with $Co_2FeAl_{0.5}Si_{0.5}$/MgO/$Co_2FeAl_{0.5}Si_{0.5}$.[10] The TMR ratio of the MTJs with cobalt-based Heusler alloys, however, decreases drastically with increasing temperature. The strong temperature dependences of the TMR ratio are generally attributed to a spin-wave excitation at the interface between the Heusler alloy layers and insulator layers,[7,11–13] decrease in the magnetization near the interface[14,15] or inelastic tunneling due to dislocations in the insulator barrier.[16]

In this study, we investigated the interface magnetism between Heusler alloy Co_2FeGe layers and MgO layers. In order to investigate the interface magnetism, interface-sensitive samples were prepared for ^{57}Fe Mössbauer spectroscopy as previously reported for Fe/Pd multilayers.[17] In the samples for this study, the ^{57}Fe isotope was used for the interfacial atomic layers and the ^{56}Fe isotope was used for the other part in the Co_2FeGe layers. The Mössbauer effect only occurs at ^{57}Fe nuclei and therefore the interfacial magnetic properties can be examined by ^{57}Fe Mössbauer spectroscopy. We compared the magnetic hyperfine field of the Co_2FeGe/MgO interfaces with that of reference Co_2FeGe films with uniform distribution of ^{57}Fe at room temperature.

II. EXPERIMENTAL

Reference Co_2FeGe films were first prepared at the substrate temperatures T_{sub} of 200, 300, 400, and 500 °C on a Cr (5 nm) buffer layer grown on MgO(001) substrates using an electron beam deposition system. The base pressure of this system was about 8×10^{-7} Pa. Co_2FeGe films of 40 nm thick were grown by depositing one atomic layer of Co, and half an atomic layer of Fe and Ge, alternately in a controlled manner using a well-calibrated quartz thickness monitor. We have already reported that Co-based Heusler alloy films grown by this technique have more uniform magnetic environments than bulk alloys prepared by arc-melting.[18] An Fe metal ingot of the ^{57}Fe isotope with 20% enrichment was used as the Fe source in order to obtain sufficient signals in ^{57}Fe Mössbauer spectroscopic measurements. The deposition was started from an atomic layer of Co, so that the interface atoms of the Co_2FeGe layers on the Cr buffer layers were nominally designed to be Co. The compositions of the Co_2FeGe films were examined by an electron probe micro analyzer.

The crystal structures were characterized by reflection high energy electron diffraction (RHEED) and X-ray diffraction (XRD) with Cu $K\alpha$ radiation. Magnetic hysteresis curves were measured using a superconducting quantum interference device magnetometer. To obtain information on local magnetism and structures, ^{57}Fe Mössbauer spectra were measured by means of conversion electron Mössbauer spectroscopy. The spectra were fitted with magnetically split sextets with a distribution of magnetic hyperfine fields.

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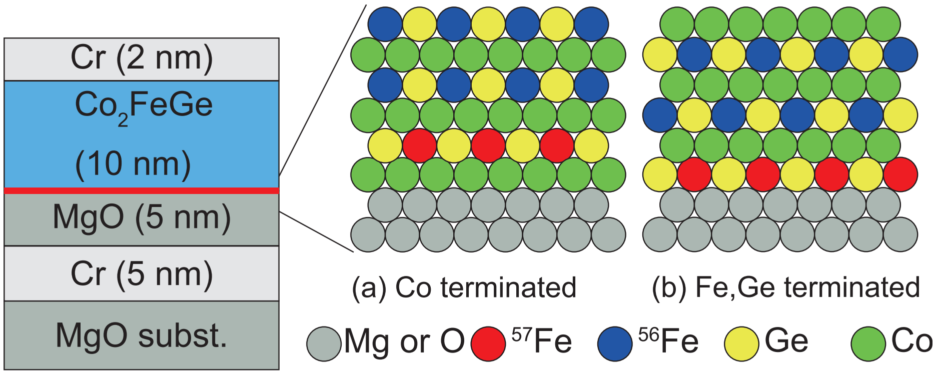

FIG. 1. Schematic structure of interface-sensitive Co_2FeGe/MgO samples for Mössbauer spectroscopic measurements. The interface atoms at the Co_2FeGe layers right on the MgO layers were nominally designed to be either (a) Co or (b) Fe/Ge.

The interface-sensitive Co_2FeGe/MgO samples, on the other hand, are designed as shown in Fig. 1. The Co_2FeGe layers were prepared by atomically controlled alternate deposition at the substrate temperature of 300 °C. The ^{57}Fe isotope was used for the interfacial atomic layer on the MgO layer and the ^{56}Fe isotope was used for the other part in the Co_2FeGe layer. The isotope enrichment in the ^{57}Fe and ^{56}Fe sources were 95.10% and 99.94%, respectively. Four interface-sensitive samples with different deposition orders were prepared as listed in Table I. Control of the interface atoms of the Co_2FeGe layers on the MgO barriers was attempted by changing the deposition order.

MgO (5 nm)/Cr (30 nm)/Fe (20 nm)/MgO (2.4 nm)/Co_2FeGe (10 nm)/Co (25 nm)/Cr (2 nm) layered structures were prepared on MgO (001) substrates for fabrication of MTJs. The Heusler alloy layers were used for one side of the ferromagnetic electrodes of the MTJs. The bottom ferromagnetic Fe layer was deposited at room temperature and annealed at 350 °C to improve the crystallographic quality. After deposition of the MgO barrier layer at room temperature, the top ferromagnetic Co_2FeGe layer was prepared by the atomically controlled alternate deposition. Transmission electron microscope images showed that the fully epitaxial Fe/MgO/Heusler alloy MTJ structure with an atomically flat MgO barrier can be realized for the systems prepared by this deposition method.[19] On the basis of the experiments for the reference Co_2FeGe films, the Co_2FeGe layers were deposited at the temperature of 300 °C. The interface atoms of the Co_2FeGe layer on the MgO barrier were designed to be either Co or Fe/Ge. The RHEED patterns right after the deposition of the Co_2FeGe layers showed that these layers were grown epitaxially with the cubic (001) orientation on the MgO (001) substrates.

The Co layer (non-epitaxial) was deposited on the Co_2FeGe layer to reinforce the coercive field to realize an antiparallel configuration of magnetization between the lower Fe and upper Co_2FeGe layers. The layered structures were fabricated into MTJs comprising ellipse-shaped pillars with an in-plane size of several μm using photolithography and Ar ion etching. Magnetoresistance measurements were carried out using a standard dc four-probe method.

TABLE I. Deposition order of the interface-sensitive samples.

Label	Deposition order
Co terminated 1	MgO → Co → ^{57}Fe → Ge ···
Co terminated 2	MgO → Co → Ge → ^{57}Fe ···
Fe/Ge terminated 1	MgO → ^{57}Fe → Ge → Co ···
Fe/Ge terminated 2	MgO → Ge → ^{57}Fe → Co ···

III. RESULTS AND DISCUSSION

Figure 2 shows θ–2θ XRD patterns of the reference Co_2FeGe films. The peaks at about 31° and 65° in each XRD pattern can be attributed to the Co_2FeGe (002) and Co_2FeGe (004) reflections. Therefore, the $L2_1$ or B2 order is well established in these films. The lattice parameters of the Co_2FeGe films deposited at the substrate temperature of more than 300 °C were about 5.97 Å. From the intensity ratios of the Co_2FeGe (111) and (202) diffraction peaks in polar plots (not shown here), the degrees of $L2_1$ order of the Co_2FeGe films grown at 200, 300, 400, and 500 °C are estimated to be about 0%, 13%, 17%, and 19%, respectively. Note that the disordered B2 structure does not disturb the half metallicity of Co-based Heusler alloy materials.[20] Saturation magnetizations of 4.86 μ_B/unit cell, 5.63 μ_B/unit cell, 5.33 μ_B/unit cell, and 4.14 μ_B/unit cell were obtained from magnetic hysteresis loops for the Co_2FeGe films grown at substrate temperatures of 200, 300, 400, and 500 °C, respectively. Judging from the hysteresis loops, the reference Co_2FeGe films grown at substrate temperatures of 300 °C has similar magnetism to the bulk Co_2FeGe alloy fabricated by an arc-melting method.[23]

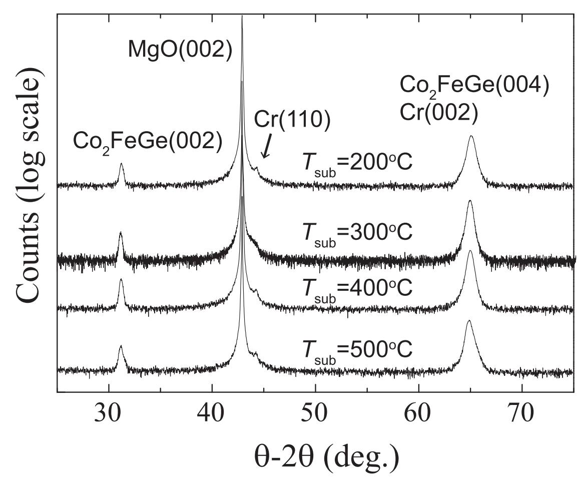

FIG. 2. X-ray diffraction patterns for the reference Co_2FeGe films grown at various substrate temperatures T_{sub}.

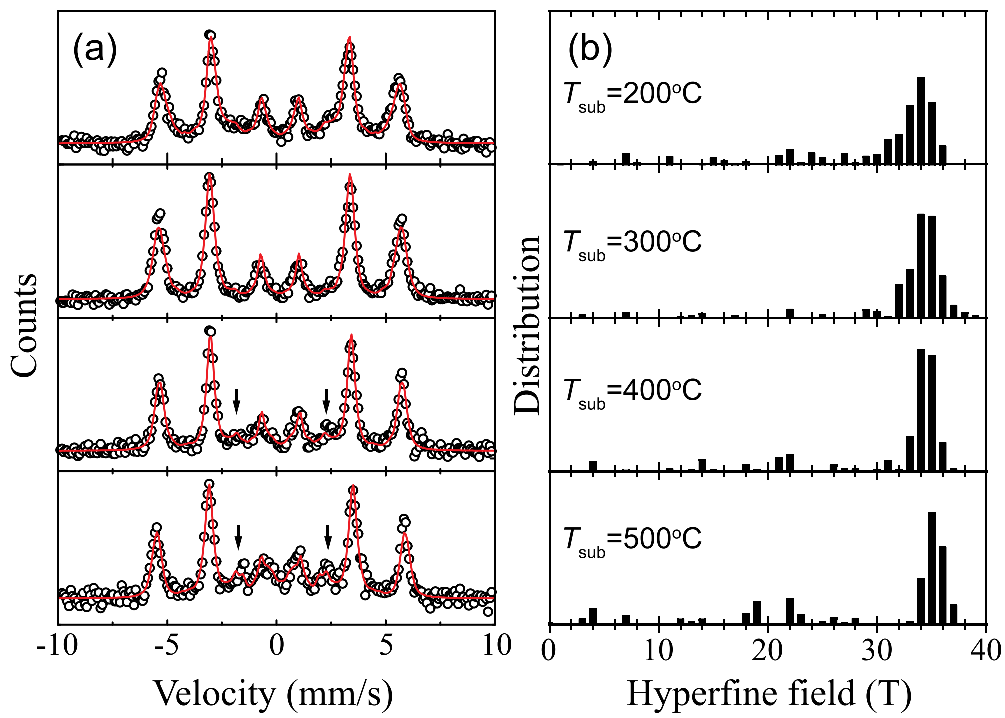

FIG. 3. (a) ^{57}Fe conversion electron Mössbauer spectra and (b) hyperfine-field distributions at the ^{57}Fe sites of the Co_2FeGe films grown at various substrate temperatures T_{sub}.

Figure 3 shows the ^{57}Fe conversion electron Mössbauer spectra and the hyperfine-field distributions at the ^{57}Fe sites for the reference Co_2FeGe films. The peak at around 35 T, which approximately agrees with the hyperfine field observed in bulk Co_2FeGe alloys prepared using a melt-spun method,[21] was observed in each hyperfine-field distribution. This peak becomes sharper with increasing substrate temperatures, which can be interpreted as the increase in the $L2_1$ order of the Co_2FeGe. There are faint additional peaks indicated by the arrows in the Mössbauer spectra of the films grown at $T_{sub} \geq 400\,°C$. These peaks correspond to the peaks at around 20 T in the hyperfine-field distributions. These peaks may be due to the substitution between Fe and Co atoms[22] or the interdiffusion between Co_2FeGe alloy layers and Cr layers. Thus Co_2FeGe films with sharp and single-peak hyperfine-field distribution can be obtained when the films are grown at the substrate temperature of 300 °C.

The XRD patterns for the interface-sensitive Co_2FeGe/MgO samples are shown in Fig. 4. The Co_2FeGe (002) and Co_2FeGe (004) reflections were observed in the XRD pattern of each film. Figure 5 shows the ^{57}Fe conversion electron Mössbauer spectra and hyperfine-field distributions at the ^{57}Fe sites of the interface-sensitive Co_2FeGe/MgO samples at room temperature. A sharp peak was observed at around 34–35 T in each hyperfine-field distribution. The value of hyperfine field for this peak is almost the same as that of the reference Co_2FeGe films in Fig. 3, and no drastic hyperfine-field reduction is observed. The results clarified that the magnetization at the interfaces between the Heusler alloy and insulator layers has no strong reduction at room temperature. The Mössbauer spectra also show that there are no oxidized iron atoms at the interfaces. These results provide experimental evidence that the strong temperature dependence of the TMR effect of the MTJs using Heusler alloy electrodes cannot be attributed simply to the reduction in the magnetization at the interfaces between Heusler alloy and insulator layers. There was no significant difference between the hyperfine-field distributions of the Co terminated samples and those of Fe/Ge terminated samples, which implies that the interface atoms to the MgO layer were eventually the same, regardless of the designed deposition order.

Figure 6(a) shows magnetoresistance curves for the Fe/MgO/Co_2FeGe MTJ with the bias voltage of 5 mV at 5 K, 100 K, and 300 K. The interface atoms at the Co_2FeGe layer were designed to be Co. TMR ratios of 67%, 50%, and 25% were observed at 5 K, 100 K, and 300 K, respectively. The temperature dependence of the TMR effect for the MTJs

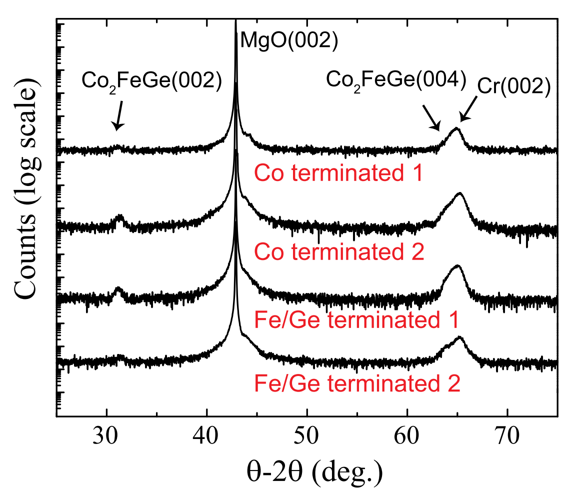

FIG. 4. X-ray diffraction patterns for the interface-sensitive Co_2FeGe/MgO samples.

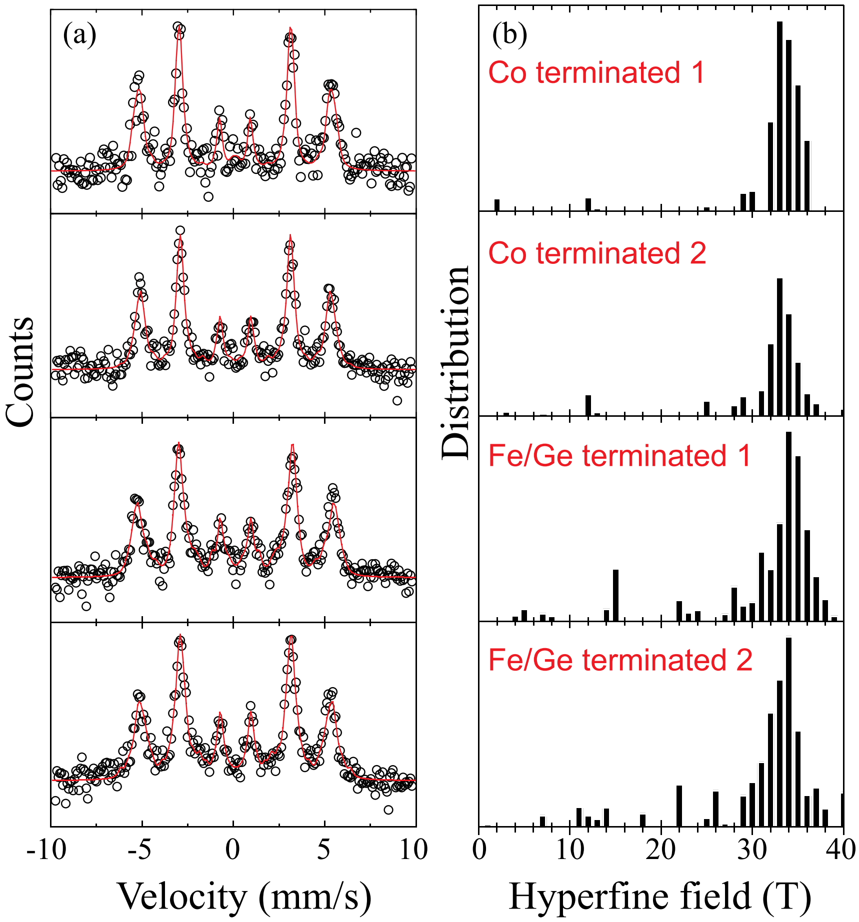

FIG. 5. (a) ^{57}Fe conversion electron Mössbauer spectra and (b) hyperfine-field distributions at the ^{57}Fe sites of the interface-sensitive Co_2FeGe/MgO samples at room temperature.

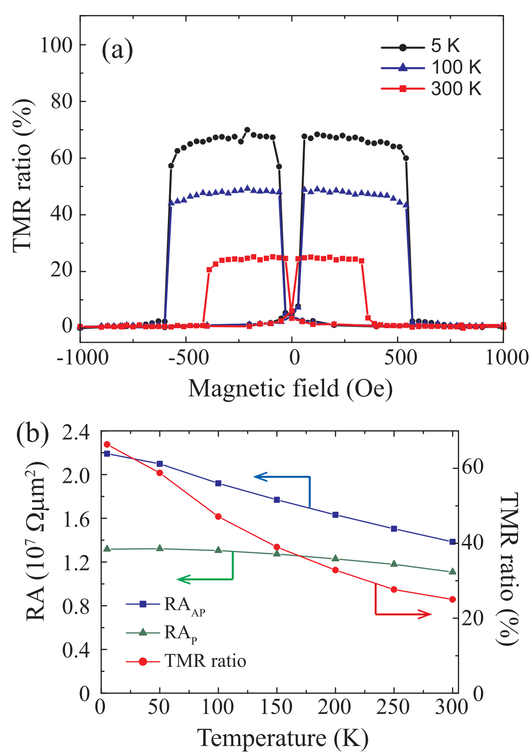

FIG. 6. (a) Magnetoresistance curves at 5 K, 100 K, and 300 K for the Fe/MgO/Co_2FeGe magnetic tunnel junction. The interface atoms at the Co_2FeGe layer on the MgO barrier were designed to be Co. (b) Temperature dependence of TMR ratio and RA products of antiparallel (RA_{AP}) and parallel (RA_P) magnetization configurations.

with the Co_2FeGe layer whose interface atoms on the MgO barrier were designed to be Fe/Ge showed a similar behavior. In spite of the expected half metallicity of the Co_2FeGe alloy,[5] the TMR ratio was not satisfactorily large even at low temperature, which may arise from the spin depolarization in Fermi levels of the Co_2FeGe alloy due to the disorder of the crystal structure at the interfacial region. The strong temperature dependence of the TMR effect is similar to that in a previous report on the TMR effect for the MTJs with Heusler alloy layers as one side of the ferromagnetic electrodes.[7] Figure 6(b) shows the temperature dependence of TMR ratio and resistance area (RA) products of antiparallel and parallel magnetization configurations. The situation is not the same for the previous report,[16] where the strong temperature dependence is attributed to inelastic tunneling due to the dislocations within the MgO barrier. In any case, it can be concluded that the strong temperature dependence of the TMR effect for the MTJs with Heusler alloy is not caused by the reduction of the interface magnetization in Heusler alloy on insulator layers.

IV. CONCLUSION

In this study, we investigated the interface magnetism between Co_2FeGe layers and MgO layers. The XRD patterns and Mössbauer spectra showed that reference Co_2FeGe films with sharp single peak in hyperfine-field distribution can be grown at substrate temperatures of 300 °C. The ^{57}Fe Mössbauer spectra at room temperature of the interface-sensitive samples were similar to those of Co_2FeGe films in which the ^{57}Fe isotope was distributed throughout the films, though the Fe/MgO/Co_2FeGe MTJ showed strong temperature dependence of the TMR effect. These results indicate that the strong temperature dependence of the tunneling magnetoresistance effect in magnetic tunnel junctions using Heusler alloy electrodes cannot be attributed simply to the reduction of the interface magnetization in Heusler alloy on insulator layers.

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