REVIEW

Tunnel magnetoresistance effect and magnetic damping in half-metallic Heusler alloys

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Some full-Heusler alloys, such as Co2MnSi and Co2MnGe, are expected to be half-metallic ferromagnetic material, which has complete spin polarization. They are the most promising materials for realizing half-metallicity at room temperature owing to their high Curie temperature. We demonstrate a huge tunnel magnetoresistance effect in a magnetic tunnel junction using a Co2MnSi Heusler alloy electrode. This result proves high spin polarization of the Heusler alloy. We also demonstrate a small magnetic damping constant in Co2FeAl epitaxial film. The very high spin polarization and small magnetic constant of Heusler alloys will be a great advantage for future spintronic device applications.

Keywords: tunnel magnetoresistance; half-metal; Heusler alloy; magnetic damping

1. Heusler alloys

High spin polarization can be realized in half-metal ferromagnets (HMFs), which have a band gap at the Fermi energy level for one spin band. Numerous efforts have been made to realize half-metallicity of HMFs in magnetic tunnel junctions (MTJs). The most typical result was reported for an MTJ of La2/3Sr1/3MnO3(LSMO)/SrTiO3/LSMO, which indicated a tunnel magnetoresistance (TMR) ratio of 1800 per cent at 4.2K [1]. This experiment proved that an MTJ using an HMF material as the electrode can exhibit a large TMR effect. Nevertheless, the LSMO has a low Curie temperature (Tc): the TMR ratio at room temperature became nearly zero. Other potential oxides such as Fe3O4 [2] and CrO2 [3] with relatively high Tc have been investigated in an attempt to achieve a large TMR effect at room temperature. However, the expected high TMR ratio at room temperature has not been obtained to date.

The most interesting and attractive materials with half-metallicity are a series of Heusler alloys. The first successful study on fabrication of MTJs was that using half-Heusler NiMnSb with C1b crystal structure [4]. This type of crystal structure is unstable because of the existence of vacant sites

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in the unit cell. For this reason, full-Heusler alloys with the composition of $X_2YZ$ are the most promising materials. Electronic band structures of several compounds in these Heusler alloys with ordered $L2_1$ crystal structure, such as $Co_2Mn(Al, Si)$, $Co_2MnGe$ and $Co_2(Cr, Fe)Al$, have been calculated. Some of them are expected to have half-metallicity [5–7]. However, the spin polarization of $L2_1$-ordered full-Heusler alloys is very sensitive to site disorder because the upper and lower edge states of the minority band gap are derived from localized anti-bonding states on the Co site. Picozzi et al. [7] indicated by means of ab initio full potential linear augmented plane wave calculations that spin polarization of $Co_2MnSi$ and $Co_2MnGe$ largely decreases by Co antisites because of a defect-induced in-gap state for the minority spin band. Therefore, highly ordered Heusler alloy films are necessary in order to realize half-metallicity.

The magnetic damping constant of ferromagnetic materials is extremely important for achieving high-speed magnetization switching and reduction of critical current density for spin torque transfer switching [8,9]. Damping for various metals and alloys has been studied experimentally by several groups. It has been reported by these groups that very low damping constants can be exhibited in some Co-based Heusler alloys, half-Heusler alloys and iron–vanadium alloys [10–13]. In particular, Heusler alloys are very attractive materials because some have both small damping constants and very large spin polarization [14]. On the other hand, formal microscopic theories for the mechanism of damping have been presented theoretically, taking into consideration the electronic structures of some Heusler alloys [15]. However, the mechanism of intrinsic damping in Heusler alloys has not been clearly understood.

This review article presents our recent efforts in the fabrication of high-quality $Co_2MnSi$ Heusler alloy films as electrodes for MTJs with both amorphous Al–O and crystalline MgO barriers. We explain in detail the relationship between the structure of Heusler alloy films and the TMR effects. In particular, we emphasize that the control of the crystal structures and the interface structure are important for the realization of a large TMR ratio. In addition, we review the experimental results of magnetic damping in Heusler alloy films. Systematic results of atomic site ordering and composition dependences of magnetic damping constants in some Heusler alloys were obtained, and a very small damping constant was realized in an epitaxially grown $Co_2FeAl$ (CFA) film.

2. Magnetoresistive devices using Heusler alloys

(a) Fundamental properties of Heusler alloy films

The (001)-oriented epitaxial $Co_2MnSi$ (CMS) Heusler alloy films were grown on a Cr-buffered MgO (001) substrate at ambient temperature by inductively coupled plasma (ICP)-assisted magnetron sputtering. The film was subsequently annealed at 450°C in order to reduce site disorder. In order to obtain the stoichiometric film composition, we used a composition adjusted Co–Mn–Si alloy sputtering target (Co: 43.7%, Mn: 27.95%, Si: 28.35%). The crystallographic structure and surface morphology of the CMS films were observed by X-ray diffraction (XRD) with CuK$_\alpha$ radiation and atomic force microscopy (AFM). The film composition was analysed by ICP spectroscopy.

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Figure 1. XRD pattern of MgO/Cr (40 nm)/Co$_2$MnSi (30 nm) film. Inset (a) shows $\phi$-scan
measurements of the (022) and (111) planes of Co$_2$MnSi. The surface morphology of Co$_2$MnSi
as observed by AFM is shown in inset (b).

The XRD profile for a CMS film is shown in figure 1. In addition to peaks
from MgO substrates, only the (002) Cr peak and (002) and (004) CMS peaks
were detected, indicating perfect (001)-preferred orientation. Since the full width
at half maxima of the rocking curve for the (002)-CMS peak was very small
(approx. 0.66°), the structural quality of the film in the perpendicular direction
to the plane was quite good. The XRD peaks with all odd (hkl) indexes (e.g.
111, 311) for a full-Heusler alloy originate in superlattice reflections in the $L_2^1$
structure. Therefore, we also measured the (111) $L_2^1$ peak and (022) peak using
$\phi$-scan measurements to obtain the degree of the $L_2^1$ order. As a result, clear (111)
and (022) peaks with fourfold symmetry were obtained as shown in inset (a)
of figure 1. The peak intensity ratio of $I(111)/I(022)$ is slightly smaller than the
simulated value for the $L_2^1$ structure. This result implies that a major fraction of
the film is $L_2^1$, but the film also includes a certain amount of $B2$ or $A2$ structure.
The surface morphology observed by AFM was extremely flat, as shown in inset
(b) of figure 1. An average surface roughness ($R_a$) of 0.2 nm was small enough to
form a thin tunnel barrier. In addition, the composition of CMS film analysed by
ICP spectroscopy was nearly stoichiometric (Co: 48.9%, Mn: 24.7%, Si: 26.4%).
Such a high-quality CMS Heusler alloy film is expected to realize half-metallicity
in an MTJ structure.

(b) Magnetic tunnel junctions with an Al–O tunnel barrier

The MTJ films, with a structure of MgO(100)-substrate/Cr(40)/Co$_2$MnSi(30)/
Al–O(1.3)/Co$_{75}$Fe$_{25}$(5)/Ir$_{22}$Mn$_{78}$(10)/Ta(3) (in nm), were prepared using an
ultrahigh vacuum magnetron sputtering system. The Al–O tunnel barrier was
formed by plasma oxidation of a pre-deposited 1.3 nm thick Al layer. The plasma
oxidation time $t_{ox}$ was varied from 10 to 240 s to examine the influence on interface
quality. The MTJ films were patterned into $10 \times 10$–$100 \times 100 \mu m^2$ elements by
photolithography combined with Ar ion milling. After micro-fabrication, MTJs
were annealed under a high vacuum and an external magnetic field of 300 Oe.
Magnetoresistance (MR) measurements were carried out using a standard DC
four-probe method.
Figure 2. (a) Dependence of the TMR ratio on the plasma oxidation time \( t_{\text{ox}} \) for MTJs prepared with an epitaxial CMS bottom electrode and a conventional Co\textsubscript{75}Fe\textsubscript{25} electrode. \((b,c)\) TMR curves corresponding to the MTJs with \( t_{\text{ox}} = 240 \text{s} \) (MTJ-A) and \( t_{\text{ox}} = 50 \text{s} \) (MTJ-B).

The maximum TMR ratio achieved after annealing is plotted as a function of plasma oxidation time \( t_{\text{ox}} \) in figure 2a. High TMR ratios exceeding 140 per cent were observed at 2K, even for MTJs fabricated with the shortest oxidation time \( t_{\text{ox}} = 10 \text{s} \). The TMR ratio increased to 159 per cent with increasing \( t_{\text{ox}} \) up to 50 s. Interestingly, upon further oxidation \( (t_{\text{ox}} > 60 \text{s}) \), the TMR ratio then underwent a dramatic decrease to approximately 100 per cent. This result suggests that long plasma oxidation times, of over 60 s, led to the creation of a great deal of magnetic contamination at the CMS/Al–O interface owing to the large affinity of Mn and Si to oxygen, and resulted in the drastic reduction of the TMR ratio. Figure 2b,c shows the TMR curves corresponding to over-oxidized MTJ \( (t_{\text{ox}} = 240 \text{s}, \text{described as MTJ-A}) \) and optimum oxidized MTJ \( (t_{\text{ox}} = 50 \text{s}, \text{MTJ-B}) \). The shapes of two TMR curves show typical spin valve characteristics and an almost perfect anti-parallel magnetization configuration was observed at both room temperature (RT) and 2K. High-resolution transmission electron microscopy
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Figure 3. HR-TEM images of (a) over-oxidized ($t_{\text{ox}} = 240$ s, MTJ-A) and (b) optimally oxidized ($t_{\text{ox}} = 50$ s, MTJ-B) MTJs. Both MTJs were annealed at the temperature giving the highest TMR ratio.

(HR-TEM) images of the MTJ-A ($t_{\text{ox}} = 240$ s) and the MTJ-B ($t_{\text{ox}} = 50$ s) are shown in figure 3. Although extremely flat interfaces were observed near the Al–O barrier layer in both cases, a thin contaminant layer could be seen on the CMS surface in the over-oxidized case. The high-contrast layer between the CMS and the CoFe electrode in the MTJ-A was also considerably thicker (3 nm) than the pre-deposited Al layer (1.3 nm). This contaminant layer at the CMS surface and thickening of the barrier are strongly suggestive of the generation of an oxidized layer at the CMS interface, resulting in the observed reduction in the TMR ratio.

The stacking structure of the MTJ is MgO(001)-substrate/Cr(40)/Co$_2$MnSi(30)/Al(1.3)–O ($t_{\text{ox}} = 50$ s)/Co$_2$MnSi(10)/Ir$_{22}$Mn$_{78}$(10)/Ta(5). A plasma oxidation time $t_{\text{ox}}$ of 50 s was determined from the previous results for Co$_2$MnSi/Al–O/CoFe-MTJs. The upper Co$_2$MnSi electrode was grown on the Al–O barrier layer at ambient temperature, followed by annealing at 400°C prior to the deposition of the Ir$_{22}$Mn$_{78}$ anti-ferromagnetic pinning layer. This fabrication process suppresses the diffusion of Mn atoms in Ir$_{22}$Mn$_{78}$ towards the Al–O barrier during annealing, which generally degrades the interface and reduces the TMR ratio [16]. HR-TEM images of the CMS/Al–O/CMS MTJ are shown in figure 4. The MTJ has an excellent morphology, with an extremely smooth and flat interface (figure 4a). Both the upper and lower interfaces between the Al–O amorphous barrier and the Co$_2$MnSi electrode are very flat and sharp. Interestingly, a clear lattice image was obtained, even in the upper CMS layer (figure 4b), despite the non-epitaxial condition of growth on the Al–O amorphous barrier. Fourier transforms of the images of the local regions of the upper and lower CMS layers are shown in figure 4c,d, respectively. Although no in-plane orientation was observed in the upper CMS electrode, highly (001)-textured growth was confirmed in the upper CMS electrode, as well as in the lower epitaxial
Figure 4. (a,b) TEM images of an MTJ with a Co$_2$MnSi/Al–O/Co$_2$MnSi structure. The horizontal and vertical directions correspond to the bottom Co$_2$MnSi [110] (MgO substrate [100]) axis and the Co$_2$MnSi [001] (MgO substrate [001]) axis, respectively. (c,d) Fourier transforms of the images of the local regions of the upper and lower CMS layers, respectively.

These results suggest that the upper crystalline Co$_2$MnSi is well formed, which is attributable to the flatness of the lower structure and the low surface energy of the amorphous Al–O layer. Although the degree of site order could not be estimated for the upper Co$_2$MnSi layer owing to experimental difficulties, the post-annealing temperature of 400°C is considered to be sufficiently high to obtain a highly ordered state. Figure 5 shows the temperature dependence of the TMR ratio for the Co$_2$MnSi/Al–O/Co$_2$MnSi MTJ (described as MTJ-C). The results for MTJ-A ($t_{ox} = 240s$), B($t_{ox} = 50s$) and conventional Co$_{75}$Fe$_{25}$/Al–O/Co$_{75}$F$_{25}$-MTJ are also shown for comparison. The applied bias voltage in all measurements was approximately 1 mV. The TMR ratio of 67 per cent observed at 300K for MTJ-C is similar to that observed for MTJ-B, yet increases dramatically to 570 per cent at 2K with decreasing temperature. This TMR ratio at low temperature is the largest reported for an MTJ with an Al–O barrier. The temperature dependence of the TMR ratio normalized at 2K is plotted in the inset of figure 5 for MTJ-A, B, C and D. Clearly, the temperature dependence of the TMR ratio is extremely large in Co$_2$MnSi-based MTJ, especially for MTJ-C, compared with conventional Co$_{75}$Fe$_{25}$/Al–O/Co$_{75}$F$_{25}$-MTJ, despite the approximately identical Curie temperature of Co$_{75}$Fe$_{25}$ and Co$_2$MnSi.

(c) Magnetic tunnel junctions with an MgO tunnel barrier

According to Julliere’s model [17], for MTJs with an amorphous tunnelling barrier, the TMR ratio depends only on the density of states (DOS) at the Fermi level of the ferromagnetic electrodes. On the other hand, in MTJs with a (001)-oriented crystalline MgO tunnelling barrier and body centred cubic structured ferromagnetic electrodes, $\Delta$-band selective coherent tunnelling occurs and enhances the TMR ratio drastically compared with an MTJ with an amorphous barrier [18]. Some groups have reported very large TMR ratios greater than 200 per cent at RT in the MTJs with Fe, CoFeB electrodes and
Figure 5. Temperature dependence of the TMR ratio for the Co$_2$MnSi/Al–O/Co$_2$MnSi-MTJ (MTJ-C). Data for the Co$_2$MnSi/Al–O/Co$_{75}$Fe$_{25}$ (MTJ-A, B) and Co$_{75}$Fe$_{25}$/Al–O/Co$_{75}$Fe$_{25}$ (MTJ-D) are shown for comparison. The inset shows the temperature dependence of the TMR ratio normalized at 2 K. MTJ-A (filled inverted triangles), Co$_2$MnSi/Al–O: $t_{\text{ox}} = 240\text{ s/Co}_{75}\text{Fe}_{25}$; MTJ-B (filled triangles), Co$_2$MnSi/Al–O: $t_{\text{ox}} = 50\text{ s/Co}_{75}\text{Fe}_{25}$; MTJ-C (filled circles), Co$_2$MnSi/Al–O: $t_{\text{ox}} = 50\text{ s/Co}_2\text{MnSi}$; MTJ-D (open diamonds), Co$_{75}$Fe$_{25}$/Al–O/Co$_{75}$Fe$_{25}$.

an MgO barrier [19,20]. Recently, some groups have attempted to use Heusler alloy electrodes and a crystalline MgO barrier together in the MTJs. Ishikawa et al. [21] observed a very large TMR ratio at low temperature and oscillation of the TMR ratio against the MgO barrier thickness; Tezuka et al. [22,23] observed a TMR ratio over 300 per cent at RT. These results are apparently related to the coherent tunnelling process through the crystalline MgO barrier. In addition, Miura et al. [24] suggested theoretically that coherent tunnelling can enhance the TMR ratio in an MTJ with Heusler alloy electrodes and an MgO barrier.

The epitaxially grown MTJs with the structure of MgO(001)-substrate/Cr(40)/Co$_2$MnSi(30)/MgO($t_{\text{MgO}}$)/CoFe(5)/IrMn(10)/Ta(5) (in nm) were prepared. The all-metal thin films were deposited using a direct magnetron sputtering system. The MgO barrier was formed using a direct magnetron sputtering and electron beam (EB) evaporation system. We describe the MTJs with a sputtered MgO barrier as SP-MTJ and the EB-evaporated MgO barrier as EB-MTJ. The Ar pressure was 0.1 Pa in the MgO sputtering and the deposition rate was about 0.008 nm s$^{-1}$. The pressure during evaporation was about $2 \times 10^{-6}$ Pa and the typical evaporation rate was 0.01 nm s$^{-1}$. The MTJ films were patterned into $8 \times 8 - 90 \times 90 \mu$m$^2$ elements using photolithography combined with Ar ion etching. After micro-fabrication, the MTJs were annealed under a high vacuum and an external magnetic field of 1 T.

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Figure 6. HR-TEM images of (a) SP-MTJ with $T_a = 375^\circ$C and (b) EB-MTJ with $T_a = 475^\circ$C.

Figure 6a,b depicts cross-sectional HR-TEM images along the [110] direction of the CMS film for the SP-MTJ annealed at 375°C and the EB-MTJ annealed at 475°C, respectively. Both TEM images reveal epitaxial growth from the bottom Cr/CMS electrode to the top CoFe electrode and the crystallinity of the MgO barrier is almost the same. However, for the SP-MTJ, a disarray structure was observed at the CMS/MgO interface. On the other hand, for the EB-MTJ, extremely smooth and abrupt interfaces were formed, as shown in the TEM image.

Figure 7 portrays the annealing temperature ($T_a$) dependence of the TMR ratio at RT and the MR curves on both the MTJs. The TMR ratio increases with increasing annealing temperature. Previous investigations have indicated that the improvement of the TMR ratio by the annealing process come from improvements of both crystallinity and (001)-orientation of the MgO barrier at the electrode/barrier interface [25]. Furthermore, the degradation of TMR by high-temperature annealing was caused by atomic diffusion, especially of the Mn atom in the pinning layer [26]. The TMR ratio of the as-deposited EB-MTJ is almost the same as that of the as-deposited SP-MTJ, even though the anti-parallel magnetic configuration in the EB-MTJ is poor. This means that the quality of the MgO layer in the EB-MTJ should be better than that of the SP-MTJ. For the SP-MTJ, the TMR ratio showed a maximum of 80 per cent at $T_a = 375^\circ$C. This TMR ratio was rather smaller than those of the MTJs with CoFeB electrodes and an MgO barrier [25]. In addition, the resistance–area product (RA) value is almost $10^8 \Omega \mu$m$^2$ and larger than the EB-MTJ (approx. $10^5 \Omega \mu$m$^2$) and the previous CoFeB/MgO/CoFeB-MTJ (approx. $10^5 \Omega \mu$m$^2$), although the MgO barrier thickness was the same. Both the small TMR ratio and the large RA value for the SP-MTJ are considered to result from tunnelling electron scattering at the MgO barrier interface. On the other hand, the EB-MTJ showed a maximum TMR ratio of 217 per cent at $T_a = 475^\circ$C. This TMR ratio is much larger than that of the SP-MTJ; it is comparable to that of the CoFeB/MgO/CoFeB-MTJ. Both good crystallinity and
Figure 7. (a) Annealing temperature dependence of the TMR ratio. The MR curves are samples of various annealing temperatures for the (b) SP-MTJs and the (c) EB-MTJs.

high (001)-orientation of the MgO barrier can promote the coherent tunnelling process through the Δ1 band of the CMS. We note that the reduction in tunnel resistance caused by the coherent tunnelling process enhances the TMR effect at RT, compared with the SP-MTJ with the worse MgO interface.

Figure 8 shows that the temperature dependence of the TMR ratio, both for the SP-MTJ and for the EB-MTJ, exhibited a maximum TMR ratio at RT. As a reference, data for both the CoFeB/MgO/CoFeB prepared using EB evaporation are also shown in figure 8. The TMR curves measured at various temperatures on the SP-MTJ and the EB-MTJ are shown in figure 8. The TMR ratio drastically increased with decreasing temperature for both MTJs. We observed a large TMR ratio of 330 per cent for the SP-MTJ and a very large TMR ratio of 753 per cent for the EB-MTJ at 2 K. This result reveals that the half-metallicity of the CMS can be realized not only in the MTJ with an amorphous Al–O barrier, but also in the MTJ with a crystalline MgO barrier. However, contrary to our expectation, the temperature dependence of the TMR ratio is strong, even in the MTJ with the MgO barrier. The origins of the large temperature dependence of the TMR are still under consideration: small energy separation between the conduction band and the Fermi level [27]; non-quasi-particle state [28]; and inelastic tunnelling process owing to the magnon excitation [29]. In theoretical calculations, the exchange interaction energy of the Co atomic layer at the Co2MnSi/MgO interface is smaller than that of the Co2MnSi bulk, when the Co layer terminates with the MgO barrier layer [30]. Such weak exchange energy at the interface causes surface magnon excitation by increasing temperature. Therefore, the control of
the atomic termination at the Heusler/barrier interface is very important to improve the temperature dependence of the TMR effect. We attempt to improve the temperature dependence of the TMR by inserting a very thin layer into the Heusler/barrier interface [31].

3. Magnetic damping in Heusler alloys

(a) Magnetic damping

As mentioned earlier, manipulation of fast magnetization dynamics, in particular magnetic damping, in ferromagnetic materials is crucial for advanced applications in spintronics. Fast magnetization dynamics is well described by the Landau–Lifshitz–Gilbert (LLG) equation, which is expressed as

\[
\frac{d\mathbf{m}(t)}{dt} = -\gamma \mathbf{m}(t) \times \mathbf{H}_{\text{eff}}(t) + \alpha \mathbf{m}(t) \times \frac{d\mathbf{m}(t)}{dt}.
\]

Here, \( \mathbf{m} \) is a unit vector in the direction of macroscopic magnetization, \( \gamma \) is the gyromagnetic ratio (\( \gamma > 0 \)) and \( \mathbf{H}_{\text{eff}} \) is the effective magnetic field vector. \( \alpha \) is the so-called Gilbert magnetic damping constant and viscous friction coefficient for magnetization motion. The LLG equation is widely used for analysis of magnetization dynamics in the spintronics field, such as micro-magnetic simulation. On the right-hand side of equation (3.1), the first term corresponds to the torque driving precessional magnetization motion around the effective field, and the second term expresses the friction torque. Spin-transfer torque, first predicted by Slonczewski [9], is approximately parallel to the friction torque,
so that the current density required in devices based on the spin-transfer effect is proportional to the $\alpha$ value, in principle. Studies on magnetic damping in conventional ferromagnetic metals such as iron, cobalt, nickel and their alloys, have been carried out by various groups [32,33]. Past studies have revealed that Gilbert magnetic damping is an intrinsic property in ferromagnetic metals, as well as magnetic moment or resistivity if one excludes any other extrinsic influences, e.g. magnetic inhomogeneities and two-magnon scattering. Kambersky gave the first theoretical explanation in accordance with experiments [34]. This theory is based on the itinerant 3d-band model taking into account exchange splitting as the mean field, and it shows the relation

$$\alpha = \frac{(1/\gamma M_s)\mu_B^2 D(E_F)(g-2)^2}{\tau}. \quad (3.2)$$

Here, $M_s$ and $\mu_B$ are saturation magnetization and Bohr magneton number, respectively. $D(E_F)$ is the total DOS for both spin bands, $g$ is Lande’s $g$ factor and $\tau$ is the electron scattering time. This relation is too simplified to discuss magnetic damping quantitatively [35]. Nowadays, magnetic damping is predicted from Kambersky’s torque formula combined with first-principle band structure calculation [36]. Nevertheless, one can gain insights into engineering magnetic damping from equation (3.2). The deviation of the $g$ value from 2, $g-2$, is roughly proportional to the spin-orbit parameter in the 3d-band, and is also related to the orbital magnetic moment, so that magnetic damping tends to decrease with decreasing orbital magnetic moment or average atomic number of elements included in ferromagnetic metals. Such discussions are given, together with experimental low damping constants observed in the NiMnSb half-Heusler alloy [10] and Fe–V binary alloys [12]. From observation of equation (3.2), magnetic damping can also vary with DOS. The DOS for $B2$- or $L2_1$-ordered Co$_2$YZ full-Heusler alloys can be varied systematically with the total valence electron number, which is also changed with composition of Y and Z [37]. Thus, the Co$_2$YZ full-Heusler alloy enables us to investigate systematically the correlation between magnetic damping and DOS if Kambersky’s idea is applicable to such half-metals. In addition, the Co$_2$YZ full-Heusler alloy can exhibit a low damping constant because these alloys show soft magnetic properties, probably owing to low orbital magnetic moment [13,38].

(b) Magnetic damping for Co$_2$FeAl Heusler alloys

The DOS at the Fermi level for CFA is lowest in those for Co$_2$YZ full-Heusler alloy [37], so we have investigated magnetic damping in CFA to clarify the correlation between magnetic damping and DOS at the Fermi level [11]. The 50 nm thick CFA films were deposited on MgO(001) single crystalline substrates by co-sputtering with Co, Fe and Al targets. The capping layer was thin aluminum oxide. Ex situ post-annealing was performed at temperatures ($T_a$) of 400–600°C in high vacuum. Structural characterization of the films was performed by XRD, and it is confirmed that all films had the (001)-oriented $B2$ structures and the epitaxial relationship was CFA (001)[100]//MgO(001)[110]. Moreover, the degree of $B2$ order and mosaicity improved with increasing $T_a$. The lattice constant for the films was close to the reported value and was almost independent of $T_a$. The surface morphology for the films was measured by AFM, and the average
surface roughness $R_a$ was around 0.4 nm and did not change with annealing. Magnetic moments measured by a vibrating sample magnetometer for the films were also independent of $T_a$, and the value for the film annealed at 600°C was $4.6 \mu_B$ formula units$^{-1}$, which was also consistent with the reported value [39]. Magnetization curves were rectangular for any films and showed soft magnetic properties. Ferromagnetic resonance (FMR) was carried out for characterization of fast magnetization dynamics. Microwave absorption in films was recorded as a function of magnetic field with different frequencies of 2–20 GHz. The FMR linewidth $\Delta H$ was evaluated from a microwave absorption curve fitted to a linear combination of dispersion and absorption curves. Figure 9a shows $\Delta H$ as a function of frequency in CFA films with different annealing temperatures. In an as-deposited film, the $\Delta H$ versus $f$ data exhibit a linear relationship with isotropic behaviour. This is expected from the linear relation of $\Delta H = 4\pi a f / \gamma$ derived from equation (3.1), so that the slope of the $\Delta H$ versus $f$ data is proportional to magnetic damping. With increasing annealing temperature, the slopes of $\Delta H$ versus $f$ decrease further, especially in the [110] direction, and simultaneously, the nonlinearity and anisotropy of $\Delta H$ against $f$ is enhanced. This nonlinear and anisotropic behaviour of $\Delta H$ is observed widely in the epitaxial films, which originates from two-magnon scattering in the existence of a misfit dislocation network [40]. In order to determine magnetic damping precisely, the analysis was
Figure 10. (a) Experimental relaxation frequency $G$ and the calculated total DOS at Fermi level $D(E_F)$ in CFA films as a function of $B_2$ ordering parameter $S$. (b) Calculated spin-dependent DOS for $A_2\,(S = 0)$ and $B_2\,(S = 1)$ CFA as a function of energy.

performed using the model taking account of line broadening owing to anisotropic two-magnon scattering [11]. Figure 9b shows $\alpha$ and $G$ for the films as a function of $T_a$, respectively, where $G$ is the relaxation frequency of the Gilbert damping, defined as $G = \alpha \gamma M$. As shown in figure 9b, $\alpha$ and $G$ for the film without annealing are rather large and are comparable to the values for permalloy, while they decrease remarkably with increasing $T_a$. The $\alpha$ and $G$ values for the film annealed at 600°C were approximately 0.001 and 20 Mrad s$^{-1}$. The lowest values of $\alpha$ and $G$ of elemental ferromagnetic metals or binary alloys were reported to be 0.0021 and 35 Mrad s$^{-1}$, respectively, in the epitaxial Fe–V alloy film [12]. The values of $\alpha$ and $G$ found in our study are even lower than in the Fe–V film.

Figure 10a displays $G$ values as a function of long-range order parameter for $B_2$ structure $S$ in CFA epitaxial films. The $S$ value was estimated from the integrated intensity ratio of the (002) to (004) peak observed in $\theta$–$2\theta$ XRD patterns for the films annealed at different temperatures, taking into account Lorentz polarization absorption corrections. Fully disordered $A_2$ or ordered $B_2$ structures correspond to $S = 0$ and 1, respectively. Magnetic damping seems to be affected by this ordering because the other properties, such as magnetic moment and coercivity, did not change much. Theoretically, atomic ordering can influence the DOS significantly in Co-based full-Heusler alloys, as well as the total valence electron number. Figure 10b shows the spin-dependent DOS versus energy profile in $A_2$- and $B_2$-CFA calculated from first principles with a coherent potential

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approximation (Y. Miura 2009, private communication). The DOS for the minority spin band for $A_2$-CFA is several times larger than in $B_2$-CFA, which indicates that the ordering dependence of the $G$ values is due to modification of the DOS. The DOS profiles in CFA with intermediate $S$ values were calculated, and the calculated $D(E_F)$ values are also plotted as a function of $S$ in figure 10b [11]. The trend of $D(E_F)$ versus $S$ is quite similar to that of $G$ versus $S$, and $G$ seems to be roughly proportional to $D(E_F)$, as seen in equation (3.2). Thus, the very low damping observed in CFA can be attributed to low DOS values at the Fermi level owing to its special band structure close to half-metals. It should be noted that a similar idea was also given theoretically by the group in Alabama University [15]. To understand further the underlying physics of magnetic damping in half-metals, it is necessary to investigate systematically the correlation between magnetic damping and DOS in $Co_2YZ$ with different compositions of $Y$ and $Z$.

(c) Composition dependence of magnetic damping in Heusler alloys

We systematically investigated magnetic damping in series of $Co_2Fe_xMn_{1-x}Si$ (CFMS) and $Co_2MnAl_xSi_{1-x}$ (CMAS) Heusler alloys. The films with structures of MgO(001)-substrate/Cr(40)/CFMS or CFMS(30)/Ta(2) (in nm) were prepared by magnetron sputtering system. All films were annealed at 450°C after deposition. Crystalline structures and magnetic properties were evaluated, respectively, using XRD and a superconducting quantum device. The magnetic damping constant $\alpha$ was measured using FMR measurements with an X-band microwave source ($f = 9.4$ GHz) and a TE$_{011}$ cavity. The samples were fixed on a quartz rod. Subsequently, a goniometer was used to measure the out-of-plane angular dependence of the resonance field and line width of the FMR spectra. The magnetic damping constant was determined by analysing the angular dependence of the line width of FMR spectra.

The results shown for XRD measurements confirmed the $Co_2MnAl_xSi_{1-x}$ had $B_2$-ordered structure and $Co_2Fe_xMn_{1-x}Si$ had $L_2_1$-ordered structure for all concentrations $x$. Saturation magnetizations of $Co_2MnAl$, $Co_2MnSi$ and $Co_2FeSi$ were, respectively, $780 \times 10^3$, $800 \times 10^3$ and $1015 \times 10^3$ A m$^{-3}$ cm$^{-3}$. These values were about 20 per cent lower than reported values for bulk samples [41], although magnetization varied systematically according to the Slater rule [5].

Obtained values of $\alpha$ depending on valence electron number are shown in figure 11 (solid line). The $\alpha$ showed a minimum value of 0.003 at $x = 0.4$ in $Co_2Fe_xMn_{1-x}Si$ and increased rapidly for $x > 0.8$. The Gilbert damping constant is considered to be proportional to the square of the spin-orbit coupling parameter $\xi$ and total DOS at Fermi energy $D(E_F)$, as mentioned above. Therefore, Gilbert damping in half-metallic Heusler alloys is expected to be small. Electronic structures for $B_2$-ordered $Co_2MnAl_xSi_{1-x}$ and $L_2_1$-ordered $Co_2Fe_xMn_{1-x}Si$ were calculated using first principles. The calculated DOS values at the Fermi energy are plotted in figure 11 (dashed line). As seen in the results for the CFA Heusler alloy, the magnetic damping constant seems to be roughly proportional to $D(E_F)$. The small magnetic damping constants of $Co_2MnAl_xSi_{1-x}$ and $Co_2Fe_xMn_{1-x}Si$ ($x < 0.6$) can be attributed to the half-metallic electronic band structure. Within this region, the DOS in the majority spin channel at the Fermi level decreases concomitantly with increasing electron number. On the other hand, a rapid
increase of damping for Co$_2$Fe$_x$Mn$_{1-x}$Si ($x > 0.8$) can be explained by degradation of the half-metallicity owing to the increasing Fe content. The magnetic damping increases rapidly because of the rapid increase of localized d-states in the minority spin channel. Such descriptions are consistent with results of the TMR effect in MTJs using Co$_2$Fe$_x$Mn$_{1-x}$Si electrodes [14].

4. Applications of Heusler alloys in future spintronics

Heusler alloys are promising materials for the application of spintronic devices based on the MR effect and spin-transfer torque, owing to their high-spin polarization and low magnetic damping constants. It is expected that Heusler alloys are suitable for memory layers for MTJ cells in so-called spin transfer torque magnetic random accesses memory. A magnetoresistive reading head in a hard disk drive is also one of the candidates for Heusler alloy applications, as already reported by several groups. On the other hand, nano-scaled microwave oscillators made of MTJs with Heusler electrodes are considered to be advanced spintronic devices for inter-chip wireless communications since the device performance, e.g. noise and operating power, can be improved by decreasing the magnetic damping. Further enhancements of spin polarization and reducing magnetic damping in Heusler alloys will enable us to realize these innovative devices in the near future.

References


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