Structural and Magnetic Properties of Cubic and Tetragonal Heusler Alloy Bilayers

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Abstract

Different cubic Co-based Heusler alloys — Co₂FeAl (CFA), Co₂FeSi (CFS), Co₂MnAl (CMA), and Co₂MnSi (CMS) — were grown epitaxially on tetragonal Heusler-like D₀₂₂-MnGa at different annealing temperatures (T_a) in the form of Heusler (20 nm)/MnGa (30 nm) bilayers. The CMA/MnGa bilayer showed the largest critical temperature (450°C), indicating good structural stability against interdiffusion. The experimental results showed an easier interdiffusion of the Fe and Si elements than of Mn and Al. An inverted hysteresis loop was observed in a polar magneto-optical Kerr effect (P-MOKE) measurement for the CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers. This is one piece of evidence for the existence of the antiferromagnetic (AFM) interfacial exchange coupling (J_{ex}). We evaluated J_{ex} for the Heusler/MnGa bilayers annealed at different temperatures. J_{ex} changed from ferromagnetic (FM) to AFM after annealing at 300°C for the CFA/MnGa bilayer. Other bilayers showed only AFM coupling. The strongest J_{ex} value of approximately -3.2 erg/cm² was obtained for the CMS/MnGa bilayer annealed at 400°C. It was found that the tendency of the Mn and Si elements to make AFM J_{ex} in the Heusler/MnGa bilayers interfaces is larger than that of Fe and Al.

Keywords: MnGa film, Heusler film, interfacial exchange coupling

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1. Introduction

In general, the requirements for high-density spin-transfer-torque magnetoresistance random access memory (STT-MRAM) are a tunnel magnetoresistance (TMR) ratio greater than 100 at room temperature, small critical current density, and a thermal stability factor greater than 60 for a lifespan of 10 years. Perpendicular magnetic anisotropy (PMA) materials with low saturation magnetization (M_s), low damping constant (α), and high spin polarization (P) are desired to meet the above-mentioned properties [1, 2]. So far, several types of PMA materials such as Co/(Pt, Pd, Ni) multilayers [3–8], (Fe, Co)Pt ordered alloys [9, 10], and TbFeCo and GdCoFe rare-earth alloys [11, 12] have been extensively studied. Although many types of materials have been investigated, the window is still open for researchers to fabricate new materials for STT-MRAM applications. Therefore, focusing on the structural and magnetic properties of new materials is essential in this area. The MnGa alloys show very unique properties, making them good candidates for STT-MRAM application. The structural, magnetic, and transport properties, as well as the spin polarization and magnetization dynamics, of MnGa with PMA have been investigated in previous studies [13–29]. However, this material shows a very low TMR ratio that is not comparable with the predicted TMR value (600%) reported by Kubota *et al.* for $D0_{22}$ -MnGa/MgO/ $D0_{22}$ -MnGa perpendicular magnetic tunnel junctions (p-MTJs) [30]. The TMR ratio is low because of the large lattice mismatch between the MnGa alloy and the MgO barrier layer and because of Mn diffusion at a high annealing temperature. One effective way to solve this problem is to insert an ultra-thin ferromagnetic metal/alloy interlayer such as Fe [27], Co [28], or FeCo [13] between the MnGa alloy and the MgO barrier. The key in this kind of research is the interfacial exchange coupling J_{ex} between the MnGa alloy and the insertion layer. Interestingly, the magnetic J_{ex} was found to be ferromagnetic (antiferromagnetic) for a Fe-rich (Co-rich) interlayer when FeCo-alloy interlayers were introduced [13]. In contrast to ferromagnetic (FM) coupling, the antiferromagnetic (AFM) coupling is very rare in magnetic films with PMA

and is technologically important for achieving synthetic ferrimagnetic structures for memory applications. In addition, through TMR enhancement with the insertion of an interlayer, we can control and tune the structural and magnetic properties depending on the desired application. Cubic Co-based Heusler alloys have potential for use as interlayers because the MnGa alloys have a structure derived from them. Moreover, the half-metal properties of Co-based Heusler alloys, such as a TMR ratio greater than 100% even at room temperature [31, 32] and a low damping constant, make these materials interesting in this area. However, there are no reports on such bilayers or the magnetic coupling of Co-based cubic Heusler alloys and MnGa, which is of fundamental importance to obtain the perpendicular magnetization of Co-based cubic Heusler alloys. We recently reported the epitaxy of bilayer films consisting of tetragonal Heusler-like $D0_{22}$ -MnGa and cubic Co-based Heusler alloys on (100) single-crystalline MgO substrates at different annealing temperatures [33, 34]. Therefore, in this work, the structural and magnetic properties of Heusler/MnGa bilayer films will be presented in more details for different Heusler alloys: Co_2FeAl (CFA), Co_2FeSi (CFS), Co_2MnAl (CMA), and Co_2MnSi (CMS).

2. Experimental Procedure

Four series samples — CFA(20 nm)/ $D0_{22}$ -MnGa(30 nm), CFS(20 nm)/ $D0_{22}$ -MnGa(30 nm), CMA(20 nm)/ $D0_{22}$ -MnGa(30 nm), and CMS(20 nm)/ $D0_{22}$ -MnGa(30 nm) bilayer films — were epitaxially grown using an ultrahigh-vacuum magnetron sputtering system with a base pressure of $< 10^{-7}$ Pa onto a MgO (100) substrate buffered by a 10-nm-thick Cr buffer layer. All bilayer films were capped by a 5-nm-thick Cr protection layer. In-situ annealing was employed at 400°C after MnGa deposition. The influence of T_a on the structural and magnetic properties have been investigated for each Co-based Heusler alloy with ex-situ annealing using a rapid thermal annealing system. In addition, 30(20)-nm-thick Mn-Ga (Heusler) films with the same stacking structure, but without Heusler (MnGa) films, were fabricated as reference films. The film com-

Figure 1: XRD patterns of (a) CFA/MnGa, (b) CFS/MnGa, (c) CMA/MnGa, and (d) CMS/MnGa bilayers for unannealed (RT) samples and samples annealed at different temperatures (T_a). The data for the CFS/MnGa and CMS/MnGa bilayers were taken from Refs. [33, 34].

positions of MnGa, CFA, CFS, CMA, and CMS were $\text{Mn}_{70}\text{Ga}_{30}$, $\text{Co}_{46}\text{Fe}_{23}\text{Al}_{31}$, $\text{Co}_{47}\text{Fe}_{24}\text{Si}_{29}$, $\text{Co}_{46}\text{Mn}_{24}\text{Al}_{30}$, and $\text{Co}_{45}\text{Mn}_{25}\text{Si}_{30}$, respectively, as estimated using inductively coupled plasma spectroscopy. To characterize their structural properties, an X-ray diffractometer with Cu $K\alpha$ radiation was used. To examine the interfacial properties and element distribution of the CMS/MnGa structure, high-resolution transmission electron microscopy (HRTEM) and energy-dispersive X-ray spectroscopy (EDX) were used, respectively. To characterize the magnetic properties, a P-MOKE system operating at a laser wavelength of 400 nm and a vibrating sample magnetometer (VSM) with a 20-kOe applied magnetic field were used.

3. Experimental Results and Discussion

Figures 1(a), (b), (c), and (d) show the out-of-plane X-ray diffraction (XRD) patterns of the CFA/MnGa, CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers, respectively, for different T_a values. All unannealed samples show the (002) and (004) peaks of the tetragonal structure of $D0_{22}$ -MnGa. The (002) peaks of the cubic B2 structure of the Heusler alloys were observed for all the bilayers without post annealing. These observations indicate the good epitaxy of the Co-based Heusler-alloy layer on the $D0_{22}$ -MnGa film. In addition to these peaks, the (004) peaks of the Heusler films were observed. The (004) peaks overlap the Cr peak for the CFS and CMS films, whereas they can be separated in the XRD pattern for the CFA and CMA films. With annealing, different behaviors were observed for each Heusler film. By increasing T_a , the (004) peak position of $D0_{22}$ -MnGa did not change for CFA, CFS, CMA, and CMS when T_a was below 400, 350, 450, and 400°C, respectively. Here, we define T_{crit} as the temperature at which the interdiffusion effect arises. When the samples are annealed above T_{crit} , the Heusler elements diffuse into the MnGa film, forming the cubic MnGa–Heusler film. Clear evidence for the interdiffusion effect can be seen in Fig. 1(b). The CFS/MnGa bilayer film annealed at 400°C has a broad (004) peak for MnGa, and its position also changed. On increasing T_a further,

this peak disappears and another peak associated with the MnGa-CFS alloy
 90 appears very close to the Cr peak for the sample annealed at 450°C.

In Fig. 2(a), the T_a dependence on the c lattice constant of MnGa is shown
 for each Heusler alloy. The c lattice constant of the reference MnGa film (the
 XRD pattern is not shown here) is about 7.00 Å, which is consistent with the
 value reported by Mizukami *et al.* [35]. For all the unannealed samples and the
 95 samples annealed at temperatures below T_{crit} , the c lattice constant of MnGa
 is constant and very close to the value of the reference MnGa film. For the
 samples annealed at temperatures above T_{crit} , the c lattice constant of MnGa
 decreased. Indeed, the tetragonal phase of MnGa changes to the cubic phase
 because of the formation of a MnGa-Heusler phase. In Fig. 2(a), the values of
 100 T_{crit} are marked for each Heusler film. As can be seen from this figure, the
 largest T_{crit} (450°C) was obtained for the CMA/MnGa bilayer, indicating good
 structural stability of the CMA/MnGa bilayer against the interdiffusion effect.
 The Heusler alloys used in this study are Co-based Heusler alloys. Therefore,
 the different behaviors of these films regarding annealing might have originated
 105 from different elements such as Al, Si, Fe, and Mn. The CFA film showed a
 smaller T_{crit} than the CMA film, which suggests that interdiffusion is easier for
 Fe than for Mn. A comparison between the CMS and CMA Heusler alloys also
 shows an easier interdiffusion of Si than Al.

The T_a dependence of the c lattice constant of the Heusler bilayer (Heusler/MnGa)
 110 and reference (Heusler/Cr) films are shown in Figs. 2(b) and (c), respectively.
 The bulk values of the c lattice constant of the Heusler alloys are marked for
 CFA, CFS, CMA, and CMS alloys in Fig. 2(c). The c lattice constant of
 Heusler/Cr did not change significantly with increasing T_a , and their values are
 close to the bulk values. However, the c lattice constants of the Heusler/MnGa
 115 bilayers show an enhancement over those of the Heusler/Cr reference films when
 the samples were annealed at a temperature below T_{crit} . In the high- T_a regime,
 the c lattice constants of the CFA and CMA alloys decrease, whereas for the CFS
 and CMS alloys, enhancements were observed. Such different behaviors might
 have originated from the different tendencies of the Heusler elements such as Al

Figure 2: Annealing-temperature dependence of the c lattice constant of (a) MnGa, (b) Heusler/MnGa, and (c) Heusler/Cr films. T_a dependence of FWHM of (d) MnGa, (e) Heusler/MnGa, and (f) Heusler/Cr films. The data of the c lattice constant of the CFA, CFS, and CMS films were taken from Refs. [33, 34].

and Si regarding diffusion. To estimate the mismatch and the a lattice constant of the MnGa and Heusler films, in-plane XRD measurements were performed for the unannealed CFA/MnGa and CMS/MnGa bilayers and the CFS/MnGa bilayer annealed at 200°C. The a values of the MnGa (Heusler) films for the CFA/MnGa, CFS/MnGa, and CMS/MnGa bilayers are 3.969 (5.620), 3.936 (5.557), and 3.937 (5.567) Å, respectively. The estimated mismatch value of the MnGa and Heusler films for the CFA/MnGa, CFS/MnGa, and CMS/MnGa bilayers are 0.42, 0.13, and 0.013%, respectively. However, the mismatch values for the MnGa and Heusler alloys estimated from the a lattice constant of the bulk values of MnGa [35] and Heusler alloys [36–39] are 3.62, 2.17, and 2.17%. This indicates that the lattices of the Heusler films are contracted to fit the MnGa lattice at the bottom in the Heusler/MnGa bilayer samples.

The T_a dependence of the full width at half maximum (FWHM) for the (004) peaks of MnGa and (002) peaks of Heusler/MnGa and Heusler/Cr are shown in Figs. 1(d), (e), and (f), respectively. The FWHM did not change for the MnGa

135 films when the samples were annealed at a temperature below T_{crit} . Then,
the FWHM drastically increased for samples annealed above T_{crit} because of
the interdiffusion effect. In general, the FWHM values of the Heusler films for
both the Heusler/MnGa bilayers and Heusler/Cr references films decrease with
increasing T_a . This indicates improvement of the film quality at high T_a due to
140 crystallization. However, the small FWHM values of the bilayers, especially for
the CFS and CMS films, compared with those of references films suggest that
the quality of these films on the MnGa layer is better than that of the Heusler
films on the Cr layer.

Figures 3(a) and (b) show the cross-sectional HRTEM image and nanobeam
145 electron diffraction of the CMS/MnGa bilayer annealed at 400°C, respectively.
As discussed in the previous paper [34], the HRTEM image clearly shows that
all the layers of the CMS/MnGa bilayer were grown epitaxially and single crys-
talline. The in-plane lattice of CMS follows that of MnGa, and there is no
dislocation at the interface of the MnGa and CMS films, which originates from
150 a very small mismatch (0.013%) between the MnGa film and CMS layer. Fur-
thermore, nanobeam electron diffraction patterns with a beam diameter of 1
nm for the CMS film show a (111) superlattice spot, indicating a $L2_1$ ordered
structure. However, to estimate the ordering level, both the (002) and (004)
peaks of the CMS film in the XRD pattern are needed. However, as can be seen
155 from the XRD pattern, the (004) peak of CMS is superimposed with the (002)
peak of Cr. Therefore, we are not able to distinguish these peaks. The EDX
analysis of the CMS/MnGa bilayer, annealed at 400°C, is shown in Fig. 3(c).
The MnGa and CMS layers were observed separately. The intensity of the Mn
and Ga elements for the MnGa layer are flat, implying that the distributions
160 of these elements are homogeneous. The intensity of the Mn element for CMS
is smaller than that for the MnGa layer and shows a small enhancement at the
interfaces of the MnGa film and the CMS layer. This figure also shows that the
Co atoms of the CMS layer diffused slightly into the MnGa layer. However, very
small interdiffusion was observed in the EDX analysis. As confirmed by XRD
165 and HRTEM measurements, the MnGa and CMS layers survived. Therefore,

Figure 3: (a) Cross-sectional HRTEM image, (b) nanobeam electron diffraction pattern with spot size = 1 nm (taken from Ref. [34]), and (c) EDX analysis of a CMS/MnGa bilayer film annealed at 400°C.

interdiffusion is negligible for this sample.

Typical out-of-plane and in-plane M - H loops measured using VSM for the unannealed CFA/MnGa, CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers are shown in Figs. 4(a), (b), (c), and (d), respectively. As can be seen from these figures, different M - H loops were observed for the different bilayers. To explain how the magnetization of the MnGa and Heusler films change with the application of an magnetic field, the magnetization process of the CMA/MnGa bilayer film will be explained as an example. The discussion below of the CMA/MnGa bilayer is similar to the case of the CMS/MnGa bilayer, reported in a previous study [34]. The 30-nm-thick MnGa and 20-nm-thick CMA films show hard

Figure 4: Typical VSM M - H loops of (a) CFA/MnGa, (b) CFS/MnGa, (c) CMA/MnGa, and (d) CMS/MnGa bilayers for unannealed samples. (e) Magnetization configurations of CMA and MnGa films corresponding to numbers 1, 2, and 3. The data of the CMS was taken from Ref. [34].

and soft magnetic properties, respectively (the data are not shown here). The magnetization of the CMA/MnGa bilayer film appears to be a combination of the magnetizations of MnGa and CMS films with the exchange coupling at the interface of the MnGa and CMA films taken into account. For clarity, certain points are marked with the numbers 1, 2, and 3 in Fig. 4(d) to show how the magnetization changes. We also marked the three magnetic fields with H_+ , H_- , and H_{sw} . H_+ and H_- are magnetic fields needed to align the magnetization of the CMA and MnGa films in parallel ($H_{\uparrow\uparrow}$) and antiparallel ($H_{\uparrow\downarrow}$) states, respectively. The H_{sw} denotes a switching field. In addition, the magnetization configurations of the CMA and MnGa films are shown in Fig. 4(e). The magnetization of MnGa and CMA are parallel for $H > H_+$ in step 1, as shown in Fig. 4(e). With decreasing magnetic field, the magnetization of CMA starts to rotate away from the parallel state to become antiparallel by step 2. Then, the magnetization of MnGa switches from the up to the down direction with

Figure 5: Polar Kerr hysteresis loops of (a) CFA/MnGa, (b) CFS/MnGa, (c) CMA/MnGa, and (d) CMS/MnGa bilayers for unannealed samples.

190 a further increase in the absolute value of the magnetic field in step 3. As the
 magnetic field is swept from -20 to 20 kOe, the magnetization of CMA starts to
 rotate from the parallel to the antiparallel state. The magnetization of MnGa
 finally switches from downward to upward with a further increase of the mag-
 netic field. If there is no exchange coupling at the interfaces of MnGa and CMA,
 195 then the H_+ and H_- fields have the same absolute value. Taking the AFM ex-
 change coupling at the interface into account, we would expect different values
 of H_+ and H_- . By increasing the coupling strength, H_+ increases, whereas the
 absolute value of H_- decreases. This indicates that larger and smaller magnetic
 fields are needed for the alignment of the total magnetization to the parallel and
 200 antiparallel states, respectively.

Figures 5(a), (b), (c), and (d) show the typical polar Kerr hysteresis loop
 measured using P-MOKE for unannealed CFA/MnGa, CFS/MnGa, CMA/MnGa,
 and CMS/MnGa bilayers, respectively. The P-MOKE measurement result is
 consistent with the VSM measurement. H_+ , H_- , and H_{SW} are the same as

205 those estimated from the out-of-plane M - H loop. However, the shape of the
 hysteresis curve measured by the P-MOKE system is different from the VSM
 M - H loop. This might stem from the reflectance-phase difference of the light
 at the bilayer interface in the P-MOKE measurement. Owing to the optical
 properties of the materials, the light penetration is different in each material.
 210 Therefore, light cannot completely penetrate the bilayers. Indeed, the P-MOKE
 measurement picks up the signal of the up layer (Heusler films) and the up part
 of the down layer (MnGa film). Therefore, the polar Kerr hysteresis loop shows
 more information about the interfaces of the MnGa film and Heusler layer. The
 P-MOKE hysteresis loops of the CFS/MnGa, CMA/MnGa, and CMS/MnGa
 215 bilayers are different from those of CFA/MnGa. A normal hysteresis loop was
 observed for the CFA/MnGa bilayers, whereas the hysteresis loop was inverted
 for the other bilayers. This inverted hysteresis loop is evidence for the exist-
 ence of AFM J_{ex} at the interfaces of CFS, CMA, and CMS Heusler films and a
 MnGa layer in the CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers. Such
 220 hysteresis has been observed for inhomogeneous films and antiferromagnetically
 coupled bilayer films [40–42]. As the magnetic field decreases from a positive to
 a negative value, the Kerr signal is negative for the CFS/MnGa, CMA/MnGa,
 and CMS/MnGa bilayers when the magnetic field is zero. This indicates that
 the magnetic moments of the CFS, CMA, and CMS films coupled antiferromag-
 225 netically to the magnetic moments of the MnGa film at their interfaces.

The T_a dependences of the saturation magnetization of CFA, CFS, CMA,
 and CMS films for Heusler/MnGa bilayers and Heusler/Cr reference films are
 shown in Figs. 6(a), (b), (c), and (d), respectively. To estimate the saturation
 magnetization of the Heusler films, the out-of-plane VSM M - H loops were used.
 230 As can be seen from these figures, the saturation magnetization of the Heusler
 films for all the bilayers increases slightly with increasing T_a and then becomes
 maximum at the T_{crit} for each Heusler alloy. All the samples show a reduc-
 tion in saturation magnetization when T_a values are greater than T_{crit} . These
 results could be interpreted by taking the XRD analysis into account. The ex-
 235 planation is that the quality of the films was improved when the samples were

annealed at a temperature below T_{crit} . However, the structure of the bilayers changes in the high- T_a regime because of the interdiffusion effect. Therefore, the enhancement and reduction of the saturation magnetization originated from a better crystallization and interdiffusion effect below and above T_{crit} , respectively. Interestingly, the saturation magnetizations of all the bilayers films, except the CFS/MnGa bilayer, are larger than the values for the reference films (Heusler/Cr). This indicates that the quality of the Heusler films on a MnGa layer is better than the quality of the Heusler films on a Cr layer. A remarkable difference was observed for the CMS Heusler alloy, which might be attributed to the relatively small mismatch between the MnGa film and the CMS layer. We also showed that the saturation magnetization of the Heusler alloys from the bulk value reported in some articles [37, 38, 43, 44] for comparison. In all the figures these values only show the saturation magnetization for the optimized condition and do not contribute for different T_a . The saturation magnetizations of the Heusler films on MnGa are comparable with the bulk values. However, a small difference from the bulk value was observed, which corresponds to different c lattice constants of the bilayers.

The T_a dependences of the remnant magnetization ($M \times t_{(0kOe)}$) and H_{SW} are shown in Figs. 7(a) and (b), respectively, for different Heusler/MnGa bilayers. The values of $M \times t_{(0kOe)}$ and H_{SW} for a 30-nm-thick MnGa film are marked for comparison. Suppose there is no exchange coupling at the interfaces of the MnGa and Heusler films. Then, the expectation values for H_{SW} and $M \times t_{(0kOe)}$ are the same as those of a 30-nm-thick MnGa film. Indeed, J_{ex} plays an important role when a hard magnetic film combines with a soft magnetic film. Here, the 30-nm-thick MnGa film is a hard magnetic film, whereas the 20-nm-thick Heusler films are soft magnetic films. In general, when the exchange coupling between the hard and soft magnetic films is AFM, smaller $M \times t_{(0kOe)}$ and larger H_{SW} values are expected [45]. However, for an FM exchange coupling, opposite changes could be observed [45]. As can be seen from Fig. 7 (a), the unannealed CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers have smaller $M \times t_{(0kOe)}$ values than the remnant magnetization of a 30-nm-thick

Figure 6: Annealing-temperature dependence of the saturation magnetization for (a) CFA, (b) CFS, (c) CMA, and (d) CMS films on MnGa and Cr layers. The CMS data were taken from Ref. [34].

Figure 7: Annealing-temperature dependence of (a) the remnant magnetization and (b) the switching field for different Heusler/MnGa bilayer films. The data of the CMS was taken from Ref. [34].

MnGa film. As T_a increases below T_{crit} , a linear reduction of the $M \times t_{(0k0e)}$ values is observed. However, in the high- T_a regime, the reduction of $M \times t_{(0k0e)}$ values is non-linear, which is attributed to structural changes due to the interdiffusion effect. Here, we should mention that the $M \times t_{(0k0e)}$ value of the CFS/MnGa bilayer annealed at 400°C is larger than that of the 30-nm-thick MnGa film. The reason is the change of the magnetic anisotropy direction from out-of-plane to in-plane. For the CFA/MnGa bilayers, a linear reduction is observed. However, their $M \times t_{(0k0e)}$ values are larger than the values for the 30-nm-thick MnGa film. The T_a dependence of H_{SW} exhibits larger values for unannealed CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers than those for the 30-nm-thick MnGa film. With increasing T_a , a linear enhancement is observed. Finally, the CFS/MnGa (CMA/MnGa) bilayer annealed at 450°C (550°C) shows a very small H_{SW} due to a change in the magnetic anisotropy from the perpendicular to the in-plane direction. A linear enhancement was also observed for the CFA/MnGa bilayers, in which the values of H_{SW} are smaller than the value in the 30-nm-thick MnGa film when T_a is less than 350°C. With a further increase in T_a , the values of H_{SW} became larger than that for the 30-nm-thick MnGa film. The small $M \times t_{(0k0e)}$ and large H_{SW} for CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers, compared with those for the 30-nm-thick MnGa film, suggest an AFM exchange coupling at the interfaces of the MnGa and Heusler films. On the other hand, the large $M \times t_{(0k0e)}$ and small H_{SW} for the CFA/MnGa bilayer, compared with those for the 30-nm-thick MnGa film, indicate that the FM exchange coupling is dominant when T_a is less than 350°C.

The T_a dependences of H_+ and H_- for the CFA, CFS, CMA, and CMS films on MnGa and Cr layers are shown in Figs. 8(a), (b), (c), and (d), respectively. To obtain H_+ and H_- , the following equations, in which energy is minimized, were used:

$$H_+ = 4\pi M_{\text{eff}}^H - (J_{\text{ex}}/M_S^H d_H), \quad (1)$$

$$H_- = -4\pi M_{\text{eff}}^H - (J_{\text{ex}}/M_S^H d_H), \quad (2)$$

where M_S^H and d_H are the saturation magnetization and thickness of the Heusler film, respectively. By combining equations 1 and 2, the constant J_{ex} can be estimated as follows:

$$J_{ex} = -((H_+ + H_-)/2)M_S^H d_H. \quad (3)$$

Three conditions were considered in this study. An AFM exchange coupling exists at the interface between the Heusler film and the MnGa layer if H_+ is larger than $-H_-$, whereas an FM exchange coupling exists when H_+ is smaller than $-H_-$. In the absence of coupling, H_+ is equal to $-H_-$. To evaluate J_{ex} , the thickness of the Heusler film was fixed at 20 nm. To estimate H_+ and H_- , an P-MOKE measurement was employed, consistent with reported values estimated from VSM hysteresis in previous studies [33, 34]. Although both method yield the same result, it is easier to estimate H_+ and H_- from a P-MOKE hysteresis loop than a VSM hysteresis loop. In addition, the P-MOKE measurement give us more insight about the behavior of the magnetic moments at the interfaces of the Heusler and MnGa films. The saturation magnetization of the Heusler film was estimated also from the out-of-plane VSM hysteresis loop. As can be deduced from Fig. 8, the H_+ and H_- for the Heusler films on Cr layers are symmetric with respect to the horizontal axis, as expected for the no-coupling case. However, an asymmetry was observed for H_+ and H_- of Heusler films on MnGa layers as a result of the exchange coupling between the MnGa and Heusler films. A remarkable asymmetry of H_+ and H_- was observed for the CMS/MnGa bilayer. In addition, H_+ is larger than $-H_-$, which indicates an AFM exchange coupling at the interface of the CMS film and the MnGa layer.

Figure 9 shows the T_a dependence of J_{ex} for CFA/MnGa, CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers. Some samples annealed at high temperature are neglected for the evaluation of J_{ex} because of the interdiffusion effect. Both the FM and AFM exchange coupling are observed in the CFA/MnGa bilayer, depending on T_a . The J_{ex} value for the unannealed CFA/MnGa bilayer is approximately 0.8 erg/cm²; it then decreases monotonically with increasing T_a . The sign of J_{ex} for the CFA/MnGa bilayer changes from positive to negative

Figure 8: Annealing-temperature dependence of the saturation fields (H_+ and H_-) for (a) CFA, (b) CFS, (c) CMA, and (d) CMS (taken from Ref. [34]) films on MnGa and Cr layers.

at $T_a = 300^\circ\text{C}$, and then the J_{ex} value continuously decreases to approximately -1.3 erg/cm^2 at $T_a = 400^\circ\text{C}$. On the other hand, only the AFM exchange coupling is observed for the CFS/MnGa, CMA/MnGa, and CMS/MnGa bilayers. Indeed, the sign of J_{ex} is negative at all annealing temperatures for these bilayers. The CMS/MnGa bilayer film shows the strongest AFM exchange coupling. The J_{ex} value for the unannealed CMS/MnGa bilayer film is -2.5 erg/cm^2 ; it becomes stronger with increasing T_a , reaching -3.2 erg/cm^2 in the sample annealed at 400°C .

Why is the exchange coupling different for each Heusler alloy? In order to answer this question only unannealed bilayer films were considered to neglect the structure change and interdiffusion effect due to the annealing temperature. All the Heusler alloys in this study are Co-based alloys, therefore, the different properties for each bilayers may have originated from the Fe, Mn, Al, and Si elements. To understand the dependence on the elements in this study, CFA

was compared with the CFS alloys and also CMA with the CMS alloys. The
 340 bilayers of the CFS and CMS Heusler alloys show a stronger AFM coupling than
 with the CFA and CMA bilayers, respectively. This indicates the tendency of Si
 to make the AFM exchange coupling larger than for Al. On the other hand, we
 can compare the CFA and CFS alloys with CMA and CMS, respectively. The
 CMA and CMS bilayers show a stronger AFM coupling than the CFA and CFS
 345 bilayers, respectively. Indeed, the tendency of Mn to make an AFM exchange
 coupling is larger than for Fe. This is consistent with the previous result, in
 which the FM coupling is dominant in the Fe/MnGa bilayer film [27].

In addition, the different c lattice constants of the Heusler films in Heusler/MnGa
 bilayers may be the reason for the existence of the different exchange couplings
 350 in this study. The XRD results show a smaller c lattice constant of the CMS
 film than for CFA. This implies that the atoms of the CMS film are closer to
 the atoms of the MnGa film. Thus, the magnetic moments of the CMS atoms
 tend to be antiparallel to their nearest magnetic atoms (MnGa) according to
 the Bethe-Slater curve [46]. A comparison between CFS and CFA is also consis-
 355 tent with this interpretation. Although the c lattice constant of CFA is smaller
 than that of the CMA film, an AFM J_{ex} was observed in the CMA/MnGa
 bilayer. The reason might stem from the Mn and Fe atoms in the CMA and
 CFA Heusler alloys, respectively. As we discussed, the AFM J_{ex} is dominant for
 Heusler alloys that contain Mn atoms, whereas FM J_{ex} is dominant for Heusler
 360 alloys with Fe atoms at the interface of the Heusler/MnGa bilayers.

The T_a dependence of J_{ex} in the Heusler/MnGa bilayers could be interpreted
 as interface-structure changes, which is apparent from the XRD results. The
 interpretation can be found as follows:

First, at low T_a , atomic diffusion is not significant, implying that the ex-
 365 change coupling in the Heusler/MnGa bilayers may stem from the intrinsic
 electronic structure at the interface, as reported in previous studies concern-
 ing Co/MnAl [47] and Co/MnGa [28] bilayers. The electronic structure of
 B2-ordered CFA and CFS are quite similar, though they have an important
 difference: the position of the Fermi-level. The Fermi level of CFA lies in the

370 middle of the minority-spin band gap, while for CFS it resides at the edge of the
 minority conduction band [48]. Thus, the spin polarizations of the spin band
 density of states are opposite for CFA and CFS, which could be related to the
 sign of the interfacial coupling for the unannealed bilayer films. This is similar
 to the interpretation of the physical mechanism of the Fe-Co/MnGa bilayer [13].
 375 The B2-ordered structures of CMA and CMS are similar to the structure of the
 CFA Heusler alloy [49]. Therefore, an FM exchange coupling is expected at the
 interfaces of the CMA/MnGa and CMS/MnGa bilayers. However, only an AFM
 exchange coupling was observed in the experimental results. The reason may
 be related to Mn. The CMA and CMS Heusler alloys have Mn atoms, which
 380 make the coupling exchanges more complicated when these films are coupled
 with the MnGa alloy. Indeed, the interaction between the Mn atoms of the
 Heusler films and the nearest Mn atoms of the MnGa layer might be unique to
 the AFM exchange coupling in the CMA/MnGa and CMS/MnGa bilayers.

In the high- T_a regime, two conditions should be considered. First, when
 385 samples were annealed at temperatures below T_{crit} , the crystallization of the
 Heusler films is one of the reasons for a stronger exchange coupling at the in-
 terfaces of the Heusler films and the MnGa layers. In addition, the c lattice
 constant of the Heusler films decreases slightly with increasing T_a for all the
 Heusler/MnGa bilayers. This suggests an increasing AFM exchange coupling
 390 at the interfaces of the Heusler and MnGa films with increasing T_a according
 to the Bethe-Slater curve. Another reason might be related to the existence of
 more Co atoms at the interface of the Heusler/MnGa bilayers. As reported in
 previous studies, the AFM exchange coupling is dominant in the Co/MnGa bi-
 layer [28, 29]. Second, when samples were annealed at temperatures above T_{crit} ,
 395 the interdiffusion plays an important role and makes the exchange coupling in-
 creasingly complicated. As an example, structural changes due to interdiffusion
 are the reason for the decrease in the absolute J_{ex} value when the CFS/MnGa
 bilayer was annealed at a temperature above T_{crit} . For a more comprehensive
 understanding, further studies on the origin of these behaviors are required.

Figure 9: Annealing-temperature dependence of the J_{ex} constant for different Heusler/MnGa bilayer films.

4. Summary

We have investigated the annealing-temperature dependence of the structural and magnetic properties of cubic Co-based Heusler alloys — CFA, CFS, CMA, and CMS — on $D0_{22}$ -MnGa films as Heusler/MnGa bilayer films. All the bilayer films were successfully grown epitaxially, even in samples without post annealing. The XRD measurement results show that the Heusler films on MnGa experienced better epitaxy than Cr did. Good structural stability against interdiffusion were observed for the CFA, CMA, and CMS Heusler alloys. The largest structural stability against interdiffusion (T_{crit} : 450°C) was obtained for the CMA/MnGa bilayer. Experimental results showed easier interdiffusion of the Fe and Si elements than of the Mn and Al. A normal P-MOKE hysteresis loop was observed for the CFA/MnGa bilayer, whereas the hysteresis loop was inverted for other bilayers. This inverted loop is one piece of evidence for the existence of AFM J_{ex} at the interfaces of the Heusler/MnGa bilayers. The J_{ex} constant was evaluated from VSM and P-MOKE hysteresis loops for bilayers at different annealing temperatures. Both FM and AFM exchange couplings were observed for the CFA/MnGa bilayer, depending on T_a . For the CFS/MnGa,

CMA/MnGa, and CMS/MnGa bilayers, only an AFM J_{ex} was observed. A very strong J_{ex} of approximately -3.2 erg/cm^2 , which is comparable with that for Fe-Co/MnGa, was found in the CMS/MnGa bilayer. A comparison between
420 different Heusler/MnGa bilayers suggests that the smaller c lattice constant of the Heusler films may be one of the reason for the stronger AFM exchange coupling according to the Bethe-Slater curve. Finally, the experimental results show that the magnetic moments of the Heusler films with Mn and Si elements tend to make a stronger AFM exchange coupling with the magnetic moment of
425 the MnGa film than with Heusler films with Fe and Al elements.

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Introduction

Hopeful materials for perpendicular magnetic tunnel junction device

Results & Conclusions

Heusler alloys	Interdiffusion temperature (T_{crit} (°C))	Magnetization at T_{crit} (M_s (emu/cm ³))	Exchange coupling ($-J_{\text{ex}}$ (erg/cm ²))
Co ₂ FeAl	400	1275	-0.8-1.5
Co ₂ FeSi	350	1045	0.4-1
Co ₂ MnAl	450	520	0.75-1.15
Co ₂ MnSi	400	800	2.5-3.2

Best properties

Comparison between Heusler alloys	Element tendency for interdiffusion	Element tendency to make AFM coupling
Co ₂ FeAl & Co ₂ FeSi	Si > Al	Si > Al
Co ₂ MnAl & Co ₂ MnSi	Si > Al	Si > Al
Co ₂ FeAl & Co ₂ MnAl	Fe > Mn	Mn > Fe
Co ₂ FeSi & Co ₂ MnSi	Fe > Mn	Mn > Fe