Highly (001) oriented MnAl thin film fabricated on CoGa buffer layer

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I. INTRODUCTION

Mn-Ga\textsuperscript{1-3} and Mn-Al\textsuperscript{4} alloys are quite attractive for applications to permanent magnet, magnetic recording media, and spin transfer torque magnetic random access memory (STT-MRAM), since they are made out of inexpensive and abundant elements as well as they have large uniaxial magnetic anisotropy, high spin polarization, low saturation magnetization and small Gilbert damping.\textsuperscript{5-9} Especially, Mn-Ga alloys such as L\textsubscript{1\textthinspace 0}-MnGa, are promising candidates as a ferromagnetic electrode for STT-MRAM because small Gilbert damping and large uniaxial anisotropy are required to achieve a sufficient thermal stability for data retention and a low writing current for low power operation simultaneously. L\textsubscript{1\textthinspace 0}-MnAl whose crystal structures similar to L\textsubscript{1\textthinspace 0}-MnGa, also has a large perpendicular anisotropy.\textsuperscript{10,11} L\textsubscript{1\textthinspace 0}-MnAl films were grown on MgO(001) substrates with Cr-based alloy buffer layers to obtain large perpendicular anisotropy.\textsuperscript{12-15} (001) oriented L\textsubscript{1\textthinspace 0}-MnAl films were known to grow epitaxially on the Cr-based alloys and reported to exhibit large perpendicular magnetic anisotropies of ~ 10\textsuperscript{7} erg/cc. However, magnetic properties of Mn-Al films on Cr-based alloy buffers with the thickness < 10 nm have not been reported, which is necessary to consider this material for the application to STT-MRAM. Reduction of the MnAl thickness will result in the degradation of its perpendicular magnetic anisotropy.

Recently, it was reported that MnGa films grown on the CoGa buffer layers exhibited a square hysteresis loop with perpendicular easy axis even in the MnGa thickness of 1 nm, and surprisingly, the thin MnGa films were grown at room temperature.\textsuperscript{16-18} Since both MnGa and MnAl have L\textsubscript{1\textthinspace 0} phase with similar lattice constants, the CoGa buffer will also be effective to realize highly oriented MnAl films with the thickness of several nm. In this study, we first report the growth of thin MnAl films on the CoGa buffer layers, where growth and post-annealing temperatures were varied to find the optimum growth condition.

II. EXPERIMENTAL METHOD

The samples were prepared on MgO(001) single crystal substrates by RF magnetron sputtering. The stack was Cr(2 nm)/MnAl(5 or 15 nm)/CoGa(0 or 30 nm)/Cr(20 nm)/MgO sub. The Ar gas pressure was 0.4 Pa for all the deposition. A Co\textsubscript{60}Ga\textsubscript{40} target was used for the deposition of the CoGa layer. The MnAl layer was fabricated by co-sputtering of Mn and Al targets and the nominal composition of the MnAl layer derived from the

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ABSTRACT

5 nm- and 15 nm-thick (001) oriented MnAl films were fabricated on CoGa buffer layers with various thermal treatments. The insertion of the CoGa layer was effective to obtain the square out-of-plane hysteresis loop even in the MnAl thickness of 5 nm. Highly (001) oriented MnAl film was obtained by depositing Mn and Al on CoGa at a substrate temperature of 200\textdegree C followed by annealing at 500\textdegree C. The perpendicular magnetic anisotropy was estimated to be 7.4±0.2 and 8.5±0.4 Merg/cc for 5 nm- and 15 nm-thick MnAl, respectively. Lower anisotropy in 5 nm-thick MnAl may be due to the interdiffusion between the MnAl and CoGa layers.

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sputtering rates of Mn and Al was 50:50 at\%.
Both Cr layer and CoGa layer were deposited at 400\°C and subsequently annealed at 600\°C for 30 min. The MnAl layer was deposited at \(T_s\), and some of the samples were annealed at \(T_a = 500\°C\) for 30 min after the deposition. Finally, the Cr layer was deposited as a protective layer. Magnetic properties were measured by an alternating gradient field magnetometer. Crystal structures were characterized by X-ray diffractometer (XRD) with Cu K\(\alpha\) radiation. Surface morphologies were observed by an atomic force microscope (AFM). A superconducting quantum interference device magnetometer combining vibrating sample magnetometer (SQUID-VSM) was used for measuring magnetization curves under high magnetic fields.

**III. RESULTS AND DISCUSSIONS**

Figure 1 shows (a), (c), and (e) AFM images and (b), (d), and (f) \(M-H\) curves of 5 nm-thick MnAl films grown on the CoGa layer with \(T_s = [(a) and (b)] 200, (c) and (d)] 300, and (e) and (f)] 400\°C\). These samples were not annealed after the deposition of MnAl layer. Step signals observed in the \(M-H\) curves near zero magnetic field originate from the CoGa layer which exhibits a small magnetization depending on the annealing condition. The average roughness \(R_a\) of MnAl, which is obtained by calculating an average of the vertical deviation of each point from the mean height in the AFM image, increased from 0.52 to 2.2 nm with increasing \(T_s\), and the island growth of MnAl was confirmed for \(T_s \geq 300\°C\). The hysteresis loop of MnAl was also sensitive to \(T_s\). The coercivity of MnAl increased with increasing \(T_s\), and the abrupt change of magnetization near coercivity was not observed for \(T_s = 400\°C\). This suggests that the island structures act as pinning centers of the domain walls and prevents the smooth propagation of domain walls in the samples.

Figure 2 shows out-of-plane XRD profiles of the 5 nm-thick MnAl films grown at \(T_s = 200, 300, 400\°C\) on the CoGa buffer layer. Dotted lines indicate the 001 and 002 peak positions of bulk L\(_{10}\)-MnAl. 001 and 002 peaks of CoGa and MnAl, where 002 peak of Cr overlaps to 002 CoGa, were observed for all the films, indicating all layers are grown on the MgO substrate with (001) orientation. The appearance of the 001 peak of MnAl means the existence of ordered L\(_{10}\)-MnAl phase. The positions of 001 and 002 peaks largely deviate from those of bulk L\(_{10}\)-MnAl. One may note that MnAl 002 peak shifts toward lower angle with increasing \(T_s\), whereas 001 peak does not shift as much as the 002 peak. The reason is considered as follows. According to the previous report on the growth of MnGa on CoGa,\(^1\) MnGa first grows pseudomorphically on the CoGa layer up to a certain thickness, and then the growth mode changes to Stranski–Krastanov (SK) mode.\(^2\) In this case,
there exist two types of MnGa layers through the thickness, i.e., the pseudomorphically- and 3-dimensionally-grown MnGa layers. Similar growth mode will be observed in the present MnAl films. One may note that there are two peaks at $2\theta \sim 53$ and $57^\circ$ for the sample with $T_s = 200$ and 400°C, which may correspond to 002 peaks of the two layers: pseudomorphically- and 3-dimensionally-grown MnAl. If we assume L1$_0$ phase is existed only in the 3-dimensionally-grown layer, 001 peak is considered to come only from the 3-dimensionally-grown layer in which the strain is gradually relaxed, and the peak position will be close to that of bulk L1$_0$-MnAl. From the peak position of MnAl 001 of the 3-dimensionally-grown layer, it is considered that 002 peak from the 3-dimensionally-grown layer appears at lower angle side and that from the pseudomorphically-grown layer appears at higher angle side. Increase of $T_s$ changed the ratio of the two peaks, indicating the change of the volume fraction between pseudomorphically- and 3-dimensionally-grown layers.

Figure 3 shows (a), (c), and (e) AFM images and (b), (d), and (f) $M$-$H$ curves of 5 nm-thick MnAl film grown (a) and (b) on the Cr layer and (c)-(f) on the CoGa layer with $T_s = 200$ °C. No post-annealing was performed on the samples shown in Fig. 3 (a)–(d), whereas the sample shown in Fig. 3(e), (f) was annealed at $T_a = 500$ °C after the MnAl deposition. All the films were confirmed to exhibit perpendicular magnetic anisotropy. For the MnAl film grown on the Cr layer, recesses on surface were observed in the AFM image, and small magnetization and large coercivity compared to the film grown on CoGa were confirmed. On the other hand, the post-annealed MnAl film grown on the CoGa exhibited square-shape hysteresis with large remanence $M_r = 420$ emu/cc ($M_r$ was almost the same as the saturation magnetization of MnAl if we neglect the contribution from the CoGa layer near zero magnetic field) and smaller coercivity than that of the film without post-annealing. The surface flatness of the MnAl film was also improved from $R_a = 0.52$ to 0.27 nm by the post-annealing.

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will be also related to low magnetization and squareness in the \(M-H\) curve of MnAl on Cr (see Fig. 3 (b)). The post-annealing of the MnAl film grown on the CoGa layer enhances the (001) orientation of the MnAl film, which is the reason of the improvement of the magnetic property shown in Fig. 3 (f).

Figure 5 shows [(a) and (c)] AFM images and [(b) and (d)] \(M-H\) curves of 15 nm-thick MnAl films grown on [(a) and (b)] CoGa and [(c) and (d)] Cr layers at \(T_a = 200^\circ\text{C}\) followed by annealing at \(T_a = 500^\circ\text{C}\). The insertion of the CoGa layer was also effective to improve the magnetic property of the thick MnAl film. However, the magnetization reversal of 15 nm-thick MnAl on CoGa was not smooth compared to the case of 5 nm-thick MnAl. As mentioned above, we consider two layers: pseudomorphically- and 3-dimensionally-grown layers, exist in the MnAl film, and thicker film will have thicker 3-dimensionally-grown layer. The 3-dimensionally-grown layer will have defects, which will prevent the smooth propagation of domain walls.

Figure 5 shows [(a) and (c)] AFM images and [(b) and (d)] \(M-H\) curves of 15 nm-thick MnAl films grown on [(a) and (b)] CoGa and [(c) and (d)] Cr layers at \(T_a = 200^\circ\text{C}\) followed by annealing at \(T_a = 500^\circ\text{C}\). The insertion of the CoGa layer was also effective to improve the magnetic property of the thick MnAl film. However, the magnetization reversal of 15 nm-thick MnAl on CoGa was not smooth compared to the case of 5 nm-thick MnAl. As mentioned above, we consider two layers: pseudomorphically- and 3-dimensionally-grown layers, exist in the MnAl film, and thicker film will have thicker 3-dimensionally-grown layer. The 3-dimensionally-grown layer will have defects, which will prevent the smooth propagation of domain walls.

Figure 6 shows (a) out-of-plane and [(b) and (c)] in-plane XRD profiles of 5 nm- and 15 nm-thick MnAl films grown on the CoGa layer at \(T_a = 200^\circ\text{C}\) followed by annealing at \(T_a = 500^\circ\text{C}\). Dotted lines indicate the peak positions of a bulk \(\text{Li}_0\text{MnAl}\). In out-of-plane XRD profiles, MnAl 001 and 002 peaks were clearly observed and the lattice parameters \(a\) were estimated to be 0.328 and 0.346 nm for 5 nm- and 15 nm-thick MnAl, respectively. MnAl 200 and 220 peaks were confirmed as shoulders at high angle side of Cr, CoGa 110 and 200, respectively, in in-plane XRD profiles. For 5 nm-thick MnAl, these peaks were not visible as shown in Fig. 6 (b) and (c). One of the reasons is the overlap of these peaks with the Cr, CoGa 110 and 200 peaks. If we assume the overlap of the peaks, the lattice parameters \(a\) of 5 nm- and 15 nm-thick MnAl are estimated to be \(a = 0.408 \pm 0.04\) and \(0.400 \pm 0.03\) nm, respectively. Using these values, the unit cell volumes of MnAl for both cases are calculated to be \(a^2c \sim 0.055\) nm\(^3\), which agrees well with the unit cell volume \(a^2c = 0.0552\) nm\(^3\) of a bulk \(\text{Li}_0\text{MnAl}\). This suggests that the MnAl film grown on CoGa is significantly strained due to the lattice mismatch between MnAl and CoGa, while conserving the unit cell volume. Similar dependence of the lattice parameters and unit cell volume on the thickness are also reported in the previous paper for MnGa on CoGa.\(^\text{18}\) The long-range order parameter \(S\), which describes a degree of ordering of the atomic arrangement, was estimated from the integral intensity ratio of MnAl 001 and 002 peaks. The parameters \(S\) were \(\sim 0.85\) and 1.0 for 5 nm- and 15 nm-thick MnAl, respectively.

Finally, perpendicular magnetic anisotropy constants of the (001) oriented MnAl were estimated by using SQUID-VSM measurements. Figure 7 shows \(M-H\) curves of (a) 5 nm- and (b) 15 nm-thick MnAl films grown on the CoGa layers at \(T_a = 200^\circ\text{C}\) followed by annealing at \(T_a = 500^\circ\text{C}\). The hysteresis seen in the in-plane \(M-H\) curve shown in Fig. 7(a) is due to the sample tilting during the measurement. The effective anisotropy field \(H_{\text{eff}}\) estimated from the in-plane curve was \(H_{\text{eff}} = 30.3 \pm 0.6\) kOe for 5 nm-thick MnAl and \(H_{\text{eff}} = 27.5 \pm 1.5\) kOe for 15 nm-thick MnAl. The perpendicular anisotropy constant \(K_u\) was calculated as \(K_u = M_iH_{\text{eff}}/2 + 2\pi M_i^2\), and it was \(K_u = 7.4 \pm 0.2 \times 10^6\) erg/cc for 5 nm-thick MnAl and \(K_u = 8.5 \pm 0.4 \times 10^6\) erg/cc for 15 nm-thick MnAl. Those values are slightly lower than the bulk value of \(\sim 10^7\) erg/cc. Theoretical calculations\(^\text{18}\) are reported that the order parameter and...
The c/a ratio affects the magnetic moment of Mn atoms and $K_u$. When c/a ratio decreases from 0.865 (15 nm) to 0.804 (5 nm), the Mn moment decreases from 2.38$\mu_B$ to 2.25$\mu_B$ while $K_u$ increases from 1.9 to 2.4 $\times 10^7$ erg/cc. Based on the theoretical calculations, pseudomorphically-grown layer with small c/a ratio will have large perpendicular anisotropy compared to 3-dimensionally-grown layer with large c/a ratio. However, 15 nm-thick MnAl, which may have thicker 3-dimensionally grown layer, exhibited large $K_u$. The difference in the order parameter between 5 nm- and 15 nm-thick MnAl should be taken into account, but it was 1.0 for 15 nm-thick MnAl and 0.85 for 5 nm-thick MnAl, which will reduces $K_u$ by 10%. Therefore, the difference in $M_s$ and $K_u$ between the 5 nm- and 15 nm-thick MnAl films can not be explained only by the variation of the order parameter and c/a ratio (growth mode). One possible mechanism to explain the decrease in $K_u$ and $M_s$ is the interdiffusion between the MnAl and CoGa layers which was not considered in theoretical calculations. In the previous study, the substitution of Al atoms by Ga or Co atoms was reported to modifies $K_u$ of MnAl films, and the substitution by Ga increased $K_u$, while the substitution by Co decreased $K_u$. Therefore, the diffusion of Co atoms of the CoGa layer into the MnAl layer during the deposition or post-annealing will explain the reduction of $K_u$ in 5 nm-thick MnAl. Further optimization of the thermal treatment may improve the magnetic property of thin MnAl grown on CoGa layers.

IV. CONCLUSION

5 nm- and 15 nm-thick MnAl films were fabricated by using CoGa buffer layers and highly (001) oriented films were obtained by optimizing the condition of thermal treatments. 5 nm-thick MnAl film grown at a substrate temperature of 200°C on the CoGa layers exhibited a square-shape hysteresis loop with perpendicular easy axis. Subsequent annealing of the film at 500°C improved (001) orientation and magnetic properties of the MnAl film. For 15 nm-thick MnAl films, the CoGa buffer layer was also effective to obtain highly (001) oriented MnAl with L1$_0$ ordered structure. The perpendicular magnetic anisotropy constants were estimated to be $7.4\pm0.2 \times 10^6$ for 5 nm- and $8.5\pm0.4 \times 10^6$ for 15 nm-thick MnAl. The decrease in the perpendicular anisotropy with decreasing the thickness of the MnAl film may be due to the interdiffusion between the MnAl and CoGa layers, and further improvement of the perpendicular magnetic anisotropy in thin MnAl will be expected by suppressing the interdiffusion.

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