Fabrication and magnetic properties of L10-MnGa highly oriented thin films

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L1₀-Mn-Ga highly oriented thin films were prepared on MgO (100) single crystalline substrates with a Cr buffer layer using an ultra-high-vacuum electron beam vapor deposition system. All growths are monitored in real-time using reflection high-energy electron diffraction (RHEED). The RHEED pattern shows clear oriented growth. In addition, XRD patterns for a fundamental (002) peak and (001) and (003) superlattice peaks were clearly observed. Large magnetic anisotropy (K_u) of 10.5 Merg/cm³ and saturation magnetization (M_s) of 470 emu/cm³ were observed for $L1_0$ -Mn-Ga film (100 nm) at $T_s = 300$ °C. When the thickness of $L1_0$ -Mn-Ga decreased from 100nm to 5nm, K_u (= 6.01 Merg/cm³), M_s (= 302 emu/cm³) and R_a (= 1.45 nm) were decreased, respectively.

Keywords: L1₀-Mn-Ga, magnetic anisotropy, saturation magnetization, electron beam evaporation method, thin film

1. Introduction

Mn-Ga alloy thin film is known to exhibit a saturation magnetization; $M_{\rm s} \sim 200\text{-}600 \text{ emu/cm}^3$ [1, 2, 5], a high magnetic anisotropy; $K_{\rm u} \sim 10{-}23.5 \text{ Merg/cm}^3$ [1], a high spin polarization; $P \sim 88$ % (it was theoretically predicted to be a half-metallic-like ferrimagnet) [3] and 58 % experimentally [4], and a low Gilbert damping constant; $\alpha \sim 0.008$ -0.015 [1]. It has been attractive attention as a new material for spin electronics device [5-12]. Recently, thin films of ordered Mn-Ga alloy is one of the most intensively studied materials for a magnetic tunnel junction (MTJ) for the super gigabit (Gbit) class magnetic random access memory operated by spin transfer torque (STT-MRAM) [14-20]. The primary issue to be addressed in MRAM applications is to reduce the critical current (I_c) required for STT-induced magnetization switching. Therefore, MTJ films should have a low $M_{\rm s} \sim 100$ emu/cm³, a low α \leq 0.01, a high $K_{\rm u} \geq$ 10 Merg/cm³, and a high $P \geq$ 70 %, Mn-Ga alloy thin film is very attractive to satisfy these required properties [13]. In addition, the thickness of the magnetic free layer in such STT device is required to be below 5nm in general [10]. At the moment, little has been reported on L10-MnGa thin film having a high perpendicular magnetic anisotropy (PMA) oriented perpendicular to the substrate by using an ultra high vacuum electron beam (UHV-EB) vapor deposition system.

In this paper, $L1_0$ -MnGa highly oriented thin films have been fabricated by using an UHV-EB vapor deposition system and their magnetic properties ware investigated.

2. Experimental procedure

Prior to film deposition, $Mn_{1.0}Ga$ target alloys were prepared from high purity manganese (99.999 %) and gallium (99.9999 %) by arc melting method in argon atmosphere. The base pressure of arc melting was less than 10⁻³ Pa. Mn-Ga thin films were prepared on MgO (100) single crystalline substrates with a Cr buffer layer using an ultra-high-vacuum electron beam evaporation system with a base pressure below 8.9×10^{-7} Pa. The stacking structure of sample was follows: MgO (100) substrate/ Cr (5 nm)/ Mn-Ga (100-5 nm)/ Cr (10 nm). The substrate was heated to $T_a = 300$ °C during deposition and annealed at 300 °C (3 h) for improve the quality of crystal. The compositions of the films were determined by an energy dispersive X-ray spectroscopy (EDX) and X = 77.8 (1st depo.), 71.5 (2nd depo.), 62.0 (3rd depo.), 69.9 (4th depo.), 59.3 (5th depo.) and 44.6 (6th depo.) for MnxGa_{100-X} (at. %) are confirmed. All growths are monitored in real-time using reflection high-energy electron diffraction (RHEED). The crystal structure of the samples was characterized by X-ray diffraction (XRD) with the Cu Ka radiation line



Fig. 1. RHEED patterns for the substrate, buffer and Mn_xGa_{100-x} thin films at $T_s = 300$ °C. (a) MgO (100) sub., (b) Cr buffer, (c) 77.8, (d) 71.5, (e) 69.9, (f) 62.0, (g) 59.3, (h) 44.6 with the electron beam azimuth [10] and [11] of MgO (100) substrate.



Fig. 2. XRD patterns for MnGa thin films prepared on MgO (100) substrate at $T_s = 300$ °C. The Mn content X for MnxGa_{100-X} films (100 nm) are (a) 77.8, (b) 71.5, (c) 69.9, (d) 62.0, (e) 59.3 and (f) 44.6 (at. %).

(wavelength equal to 0.15418 nm). The surface roughness of the film was investigated by atomic force microscopy (AFM). The magnetic properties were measured by using a superconducting quantum interference device (SQUID) magnetometer in the field up to ± 70 kOe, and $M_{\rm s}$ and $K_{\rm u}$ for each thin film were evaluated from magnetization curves.

3. Results and discussion

RHEED patterns of Cr buffer and the growth of $Mn_XGa_{100\cdot X}$ films (100 nm) with various Mn content prepared on MgO (100) substrate are shown in Fig.1. The Mn content was varied as follows: X = (a) 77.8, (b) 71.5, (c) 69.9, (d) 62.0, (e) 59.3 and (f) 44.6 (at. %). The $Mn_XGa_{100\cdot X}$ films were fabricated from only $Mn_{1.0}Ga$ target alloy. The RHEED pattern shows clear oriented growth in c-plane, and the surface reconstruction structure in Mn-Ga layer was clearly observed. It should be noted that this oriented thin film exhibits a flat surface at the atomic level.

Since the composition is different by number of deposition by vapor pressure difference, Mn-Ga films were confirmed that it is a L_{10} structure by using XRD. XRD patterns for Mn_XGa_{100-X} films with various Mn content prepared on MgO (100) substrate are shown in Fig. 2. The Mn content was varied as follows: X = (a) 77.8, (b) 71.5, (c) 69.9, (d) 62.0, (e) 59.3 and (f) 44.6 (at. %). The intense peak from Cr buffer layer and MgO substrate ware clearly observed for all sample. In addition, a fundamental (002) peak, (001) and (003) superlattice peaks of the L_{10} -MnGa phase were clearly observed for X = 62.0 (at. %) (d) and X = 59.3 (at. %) (e). Furthermore, both fundamental (004) and superlattice (002) and (006) peaks of the D_{022} -Mn₃Ga phase were confirmed at the X = 71.5 (at. %) (b). The chemical order



Fig. 3. Magnetization curves for MnGa thin films prepared on MgO (100) substrate at $T_s = 300$ °C. The Mn content X for Mn_xGa_{100-X} films (100 nm) are (a) 77.8, (b) 71.5, (c) 69.9, (d) 62.0, (e) 59.3 and (f) 44.6 (at. %).

parameter S of X = 62.0 (at. %) (d) and X = 59.3 (at. %) (e) were shown S = 0.79 and 0.86. However, Mn-Ga thin film of X = 59.3 (at. %) (e) show decrease of (003) superlattice peak of the $L1_0$ -MnGa phase. Therefore, the Mn-Ga thin film of X = 62.0 (at. %) (d) shows best preferred orientation of $L1_0$ structure.

Magnetization curves for the Mn_xGa_{100-X} films prepared on MgO (100) substrate are shown in Fig. 3. All measurements were preformed at room temperature. A magnetic field was applied perpendicular to the film plane direction for the curves indicated by \perp , and it was applied along the in-plane direction for those indicated by //. Magnetization curve for X = 71.5 (at. %) (b) shows the curve of the case of a typical $D0_{22}$ structure [2,12]. Moreover, Mn-Ga films of X = 62.0 (at. %) (d) and X =59.3 (at. %) (e) had relatively high M_s and low H_c . To evaluate the PMA properties quantitatively, the K_u was



Fig. 4. RHEED patterns for the substrate, buffer and $Mn_{62.0}Ga_{38.0}$ (at %) thin films of different thickness (t_{Mn} -Ga nm) at $T_s = 300$ °C. (a) MgO (100) sub., (b) Cr buffer, (c) 100, (d)20, (e)10, (f)5 with the electron beam azimuth [10] and [11] of MgO (100) substrate.



Fig. 5. XRD patterns for $Mn_{62.0}Ga_{38.0}$ (at %) thin films of different thickness (t_{Mn} -Ga nm) prepared on MgO (100) substrate. t_{Mn} -Ga of (a) 100, (b) 20, (c) 10 and (d) 5 at $T_s = 300$ °C and $T_a = 300$ °C (3 h).

estimated using the relations $K_{\rm u} = M_{\rm s} \times H_{\rm k}^{\rm eff} / 2 + 2\pi M_{\rm s}^{2}$. Here, the effective anisotropy field $(H_{\rm k}^{\rm eff})$ was defined as the extrapolated intersection of the in-plane *M*·*H* curves with the saturation magnetization value of out-of-plane *M*·*H* curves. In Mn-Ga films of X = 71.5 (at. %) (b), X =62.0 (at. %) (d) and X = 59.3 (at. %) (e), Mn-Ga films with high $K_{\rm u} \ge 10$ Merg/cm³ were obtained. Highest $M_{\rm s}$ of 470 emu/cm³ and $K_{\rm u}$ of 10.5 Merg/cm³ were confirmed by epitaxial Mn-Ga film of X = 62.0 (at. %) (d).

RHEED patterns of Cr buffer and the growth of $Mn_{62.0}Ga_{38.0}$ (at %) films of different thickness (t_{Mn} -Ga nm) on MgO (100) substrate are shown in Fig.4, t_{Mn} -Ga is (c) 100, (b) 20, (c) 10 and (d) 5. The observed RHEED patterns remain somehow bright and streaky. This indicates that the films are highly crystalline and have rough surfaces except surfaces of t_{Mn} -Ga = 5 nm.

XRD patterns for $Mn_{62.0}Ga_{38.0}$ (at %) film of different thickness (t_{Mn} -Ga nm) on MgO (100) substrate are shown in Fig.5. A fundamental peak, and superlattice peaks of the L10-MnGa phase were clearly observed for the t_{Mn} -Ga = 100 nm. The t_{Mn} -Ga = 20 nm film shows clear L10-MnGa (001), (002), (110) and (112) peaks (not shown (003)). The t_{Mn} -Ga = 10 and 5 nm films show relatively small Mn-Ga (002), (110) and (112) peaks (not shown (001)). With decreasing thickness (100-5 nm), the Mn-Ga films show shift of diffraction angle (superlattice peaks are small). In addition, the chemical order parameter S of the t_{Mn} -Ga = 20 nm film was shown S =0.57, it shows a significant decrease in the S compared to the t_{Mn} -Ga = 100 nm (S = 0.78).

Magnetization curves for $Mn_{62.0}Ga_{38.0}$ (at %) thin films of different thickness (t_{Mn} -Ga nm) prepared on MgO (100) substrate are shown in Fig. 6. All measurements were preformed at room temperature. The magnetic field was



Fig. 6. Magnetization curves for Mn_{62.0}Ga_{38.0} (at %) thin films of different thickness (t_{Mn} ·Ga nm) prepared on MgO (100) substrate. t_{Mn} ·Ga of (a) 100, (b) 20, (c) 10 and (d) 5 at $T_s = 300$ °C and $T_a = 300$ °C (3 h).



Fig. 7. AFM images for Mn_{62.0}Ga_{38.0} (at %) thin films of different thickness (t_{Mn-Ga} nm) prepared on MgO (100) substrate. t_{Mn-Ga} of (a) 100, (b) 20, (c) 10 and (d) 5 at $T_s = 300$ °C and $T_a = 300$ °C (3 h).



Fig. 8. $M_{\rm s}$, $K_{\rm u}$ and $R_{\rm a}$ as function of $t_{\rm Mn}$ -Ga (nm) for Mn_{62.0}Ga_{38.0} (at %) thin films prepared on MgO (100) substrate at $T_{\rm s} = 300$ °C, $T_{\rm a} = 300$ °C (3 h).

applied in the perpendicular (\perp) and in-plane (//) directions to the film. The easy magnetization axis is aligned perpendicular to the film plane for all the samples. With decreasing thickness (100-5 nm), $M_{\rm s}$ (= 520-302 emu/cm³) and K_u (10.0-6.01 Merg/cm³) were decreased. It can be considered that decreasing $M_{\rm s}$ and $K_{\rm u}$ originated primary from the decreased of chemical order parameter S. Furthermore, AFM images for Mn_{62.0}Ga_{38.0} (at %) thin films of different thickness $(t_{Mn-Ga} nm)$ prepared on MgO (100) substrate are shown in Fig. 7. The average roughness (R_a) for the Mn-Ga surfaces ($t_{Mn-Ga} = 100, 20, 10$ and 5 nm) was found to be $R_{\rm a}$ = 5.25, 4.74, 2.52 and 1.33 nm, respectively. With decreasing thickness (100-5 nm), Ra (= 5.25-1.33 nm) was decreased. The $M_{\rm s}$, $K_{\rm u}$ and $R_{\rm a}$ as function of $t_{\rm Mn-Ga}$ (nm) for Mn-Ga thin films are summarized in Fig.8. Considering that the growth temperature of PMA film should be as low possible for practical applications in spintronic devices, the L10-Mn-Ga highly oriented thin film is very promising because high PMA can be obtained at relative low growth temperature at $T_{\rm s}$ = 300 °C and $T_a = 300$ °C in this study.

4. Summary

Mn-Ga thin films have been fabricated by using UHV-EB vapor deposition and their magnetic properties were investigated. Variation of Mn composition has been confirmed by number of deposition. The clear oriented growth of Mn-Ga films has been confirmed on MgO (001) substrate by using RHEED in real time. Large $M_{\rm s}$ of 470 emu/cm³ and $K_{\rm u}$ of 10.5 Merg/cm³ were obtained for $L1_0$ -Mn-Ga highly oriented thin film. With decreasing the thickness (100-5 nm), $M_{\rm s}$ (= 302 emu/cm³), $K_{\rm u}$ (= 6.01 Merg/cm³) and $R_{\rm a}$ (= 1.45 nm) were decreased. The $L1_0$ -Mn-Ga highly oriented thin film is considered to be promising because relative high $K_{\rm u}$ and low $M_{\rm s}$ can be obtained at relatively low growth temperature at $T_{\rm s} = T_{\rm a} = 300$ °C.

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