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

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L1<sub>0</sub>-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/cm<sup>3</sup> and saturation magnetization ( $M_s$ ) of 470 emu/cm<sup>3</sup> 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/cm<sup>3</sup>),  $M_s$  (= 302 emu/cm<sup>3</sup>) and  $R_a$  (= 1.45 nm) were decreased, respectively.

**Keywords:** L1<sub>0</sub>-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/cm<sup>3</sup>, a low  $\alpha$  $\leq$  0.01, a high  $K_{\rm u} \geq$  10 Merg/cm<sup>3</sup>, 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<sup>-3</sup> 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  $8.9 \times 10^{-7}$  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<sub>100-X</sub> (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<sub>x</sub>Ga<sub>100-x</sub> 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<sub>100-X</sub> 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  $\pm 70$  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 MnxGa<sub>100-X</sub> 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  $D_{022}$ -Mn<sub>3</sub>Ga 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<sub>x</sub>Ga<sub>100-X</sub> 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<sub>x</sub>Ga<sub>100-X</sub> 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/cm<sup>3</sup> were obtained. Highest  $M_{\rm s}$  of 470 emu/cm<sup>3</sup> and  $K_{\rm u}$  of 10.5 Merg/cm<sup>3</sup> 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  $Mn_{G2.0}Ga_{38.0}$  (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  $L1_0$ -MnGa phase were clearly observed for the  $t_{Mn}$ -Ga = 100 nm. The  $t_{Mn}$ -Ga = 20 nm film shows clear  $L1_0$ -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<sub>62.0</sub>Ga<sub>38.0</sub> (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<sub>62.0</sub>Ga<sub>38.0</sub> (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  $T_{\rm s} = 300$  °C and  $T_{\rm a} = 300$  °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<sub>62.0</sub>Ga<sub>38.0</sub> (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/cm<sup>3</sup>) and K<sub>u</sub> (10.0-6.01 Merg/cm<sup>3</sup>) 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<sub>62.0</sub>Ga<sub>38.0</sub> (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/cm<sup>3</sup> and  $K_{\rm u}$  of 10.5 Merg/cm<sup>3</sup> were obtained for  $L1_0$ -Mn-Ga highly oriented thin film. With decreasing the thickness (100-5 nm),  $M_{\rm s}$  (= 302 emu/cm<sup>3</sup>),  $K_{\rm u}$  (= 6.01 Merg/cm<sup>3</sup>) 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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