



## Research articles

Magnetic and structural properties of  $L1_0$  Mn-Ga epitaxially grown islands

Siqian Zhao<sup>a,b,\*</sup>, Yoshitomo Tanaka<sup>c</sup>, Takuya Sato<sup>a,c</sup>, Keisuke Kamiya<sup>a,c</sup>, Gary Mankey<sup>a,d</sup>, Takao Suzuki<sup>a,b,e</sup>

<sup>a</sup> Center for Materials for Information Technology, The University of Alabama, Tuscaloosa, AL 35487, USA

<sup>b</sup> Department of Metallurgical and Materials Engineering, The University of Alabama, Tuscaloosa, AL 35487, USA

<sup>c</sup> Materials Development Center, Technology & IP HQ, TDK Corporation, Ichikawa, Japan

<sup>d</sup> Department of Physics and Astronomy, The University of Alabama, Tuscaloosa, AL 35487, USA

<sup>e</sup> Department of Electrical and Computer Engineering, The University of Alabama, Tuscaloosa, AL 35487, USA



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## ABSTRACT

Magnetic and structural properties of  $L1_0$   $\text{Mn}_{53}\text{Ga}_{47}$  islands epitaxially grown onto  $\text{MgO}(1\ 0\ 0)$  substrates are discussed. The samples were fabricated by sputter-deposition at a substrate temperature  $T_s$  during deposition. The samples deposited at  $T_s$  above  $500^\circ\text{C}$  were found to form the island structure of Volmer-Weber type. The average size of the islands fabricated at  $T_s = 600^\circ\text{C}$  is around  $200\text{ nm}$  in width and  $60\text{ nm}$  in height. The XRD and TEM analyses indicate that those islands are of the  $L1_0$  phase with the  $\langle 001 \rangle$  axis perpendicular to the substrate surface. The lattice constants  $a$  and  $c$  are  $3.91 \pm 0.01\text{ \AA}$  and  $3.65 \pm 0.02\text{ \AA}$ , respectively, which are larger by  $0.8\%$ , and smaller by  $1.5\%$  respectively, as compared to the bulk values of the same composition. The order parameter of these islands is found to increase from  $0.7$  to  $0.8$  for  $T_s = 450\text{--}500^\circ\text{C}$ , and then remains nearly constant ( $\approx 0.8$ ) for higher  $T_s$ . The M-H hysteresis loops exhibit a high squareness for all the samples for  $T_s = 450\text{--}600^\circ\text{C}$ . The saturation magnetization  $M_s$  is about  $220$  and  $350\text{ emu/cm}^3$  for  $T_s = 450$  and  $600^\circ\text{C}$ , respectively. The out-of-plane coercivity  $H_c$  is  $5$  and  $13\text{ kOe}$  for  $T_s = 450$  and  $600^\circ\text{C}$ , respectively. The temperature dependence of  $M_s$  for the sample made at  $T_s = 600^\circ\text{C}$  was measured and fitted to an empirical relation in order to estimate  $T_c$ . The estimated  $T_c$  around  $600\text{ K}$  is in reasonable agreement with the reported value of bulk  $L1_0$   $\text{Mn}_{53}\text{Ga}_{47}$ . The uniaxial perpendicular magnetic anisotropy constant  $K_u$  estimated by the out-of-plane torque curves is around  $1.1 \times 10^7$  and  $8.3 \times 10^6\text{ erg/cm}^3$  at  $5$  and  $300\text{ K}$ , respectively. The initial magnetization curves of these islands exhibit a typical pinning mode for the coercivity mechanism. A phenomenological discussion of the coercivity is presented on the basis of Kronmuller's formula. It is found that the coefficient  $\alpha$  and effective demagnetizing factor  $N_{\text{eff}}$  in the formula by Kronmuller are approximately  $0.38$  and  $12$ . For the magnetic anisotropy mechanism, the power law relation between  $K_u(T)$  and  $M_s(T)$  is examined. It is found that the exponent  $n$  for  $K_u(T) \propto M_s(T)^n$  varies from  $2.3$  to  $3.5$  for  $T$  from  $350$  to  $5\text{ K}$ . However, the  $n$  in a whole temperature range from  $5$  to  $350\text{ K}$  is  $2.6$ . The  $n = 2.6$  suggests a deviation from the single-ion mechanism, and is consistent with the theoretical predictions of Ga contribution.

## 1. Introduction

Recently the manganese gallium (Mn-Ga) alloys have attracted much attention because of their versatile and attractive magnetic properties for various applications including rare-earth free permanent magnet, sensors and spintronics devices. The bulk phase diagram for this binary alloy system is complicated and has not been well established even now. In the case of thin films, the situation is even more complicated since the stability of the structure is much dependent on fabrication method and type of substrates. Furthermore, the origin of the magnetic properties such as coercivity and magnetic anisotropy

constants of a thin film which is often not of a single phase but of multi-phases is difficult to discuss, unless the sample is epitaxially grown.

Among many phases of the Mn-Ga binary alloys, the  $L1_0$  phase ( $P4/mmm$ ) has been reported to be stable over a composition range from about  $54$  to  $64\text{ at\%}$  in a temperature range from ambient to about  $600^\circ\text{C}$  [1–4]. The  $L1_0$  MnGa thin films were reported to exhibit high perpendicular uniaxial magnetic anisotropy constant  $K_u$  in the order of  $10^7\text{ erg/cm}^3$ , accordingly high coercivity ( $> 10\text{ kOe}$ ) at room temperature. It is of interest to note that Mn and Ga do not have a large spin orbit coupling coefficient, but they do exhibit high magnetic anisotropy constant. In order to understand the origin of such large magnetic

\* Corresponding author at: Department of Metallurgical and Materials Engineering, The University of Alabama, Tuscaloosa, AL 35487, USA.  
E-mail address: [szhao6@crimson.ua.edu](mailto:szhao6@crimson.ua.edu) (S. Zhao).

anisotropy, Kota and Sakuma [5] employed the tight-binding linear muffin tin orbital (TB-LMTO) method to calculate the electronic structure in the atomic sphere approximation. They concluded that the large magneto-crystalline anisotropy results from the selection rule of spin orbit interaction (SOI) even though SOI for the individual atoms is weak. Also, Al-Aqtash and Sabirianov discussed on the basis of self-consistent electronic structure calculations using the density functional theory about the effect of strain on magnetic anisotropy constant of  $L1_0$  MnGa alloys, and concluded that the magnetic anisotropy energy is sensitive to a change in lattice constant  $c$  [6].

In order to clarify experimentally about the magnetic anisotropy mechanism, the temperature dependence of  $K_u$  was discussed in terms of a power law with  $M_s$ . This way, one would differentiate the single-ion model from the two-ion model. For example, in the  $L1_0$  FePt case  $n = 2$  was found for the power law of  $K(T) \approx c M_s(T)^n$  [7,8]. This result suggested that the two-ion model was responsible for the magnetic anisotropy mechanism. However, there has been little report found in literature regarding  $L1_0$  MnGa alloy thin films, and its mechanism is still open for discussion.

Coercivity  $H_c$ , is one of the extrinsic quantities sensitive to film morphology. The observed  $H_c$  is much smaller than the intrinsic magnetic anisotropy field,  $H_k$ . The reduction from  $H_k$  is often discussed in conjunction with the demagnetizing effect and the coupling between grains. Kronmüller [9] discussed extensively the role of pinning sites and the demagnetizing effect associated with the defect and impurities in this reduction. Suzuki et al. [10] discussed the coercivity mechanism in Co/Pt multilayers based on the Kronmüller's formula. Kronmüller et al also discussed it for NdFeB magnets [9]. It is of great interest to study the coercivity mechanism of  $L1_0$  MnGa thin films with the high perpendicular magnetic anisotropy, and to correlate it with the film morphology.

The present paper describes both the coercivity mechanism and also the magnetic anisotropy mechanism of  $L1_0$  MnGa epitaxially grown single crystalline thin films.

## 2. Experimental

Multilayer thin films of  $Mn_{50}Ga_{50}$  and Mn with various thicknesses were deposited onto MgO(1 0 0) single crystal substrates by using DC magnetron sputtering at substrate-deposition temperature  $T_s = 450$ – $600$  °C. The base pressure was better than  $10^{-8}$  Torr. The fabricated thin films were then capped by a 5 nm thick Ru layer at ambient temperature after the system cooled down. The  $Mn_{50}Ga_{50}$  alloy targets and Mn targets (purity 99.999%) were used. The deposition rates of MnGa and Mn were 0.03 and 0.01 nm/s, respectively, calibrated by X-ray reflectivity.

Based on preliminary experiments [11], it was found that the optimum layer-thicknesses for MnGa and Mn were 2 nm and 0.5 nm, respectively, and the total repetition was 25.

Measurements of magnetic properties were performed by using an alternating gradient magnetometer (AGM) in fields  $H$  up to 18 kOe at room temperature, and a vibrating sample magnetometer (VSM) and a torque magnetometer in  $H$  up to 90 kOe over a temperature range from 5 to 400 K. The structural properties were characterized by X-ray diffraction (XRD) with Cu  $K\alpha$  radiation and scanning/transmission electron microscopy (S/TEM) together with energy dispersive spectroscopy (EDS). The cross-sectional sample was prepared by focused ion beam (FIB). The lattice constants  $a$  and  $c$  were estimated by X-ray diffraction at  $\psi$  angle 0 and  $43.5^\circ$ , from  $L1_0$  (0 0 2) and (2 0 2) peaks, where the  $\psi$  is the angle between the film normal and the bisector of the incident and diffracted beam. The angle  $43.5^\circ$  is the angle between  $L1_0$  MnGa (2 0 2) and (0 0 2) planes.

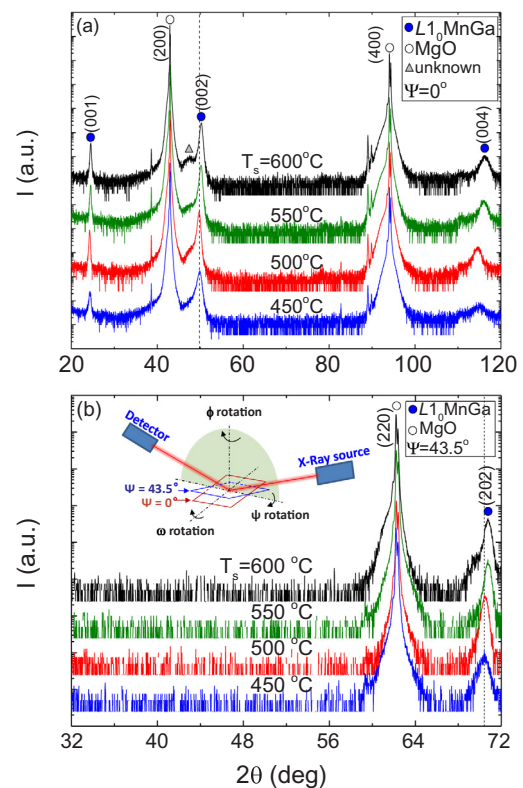


Fig. 1. The  $2\theta$ - $\omega$  scan XRD patterns measured at (a)  $\psi = 0$  and (b)  $43.5^\circ$  for the samples deposited at the various  $T_s$ . The dotted lines show a guideline for observation of peak shift. The inset figure in (b) shows the illustration of the measurement.

## 3. Results and discussions

### 3.1. Structure

Fig. 1 shows the  $2\theta$ - $\omega$  scan XRD patterns measured at (a)  $\psi = 0$  and (b)  $43.5^\circ$  for the samples with various  $T_s$ . The dotted lines for (2 0 0) and (2 0 2) are merely the guide for eyes for the peak shift with  $T_s$ . As shown in Fig. 1(a) for  $\psi = 0^\circ$ , the diffraction peaks from the {0 0 1}, {0 0 2} and {0 0 4} planes of the  $L1_0$  phase are observed. It is thus concluded that the samples are epitaxially grown with the  $\langle 001 \rangle$  axis along film normal. The unknown peaks at around  $47.6^\circ$  are observed, whose intensity increases with  $T_s$ . One possibility is the unknown peak belongs to the (4 2  $\bar{1}$ ) peak of  $Mn_5O_8$  with monoclinic  $c2/m$  structure, since the sample at  $T_s = 600$  °C was oxidized with 15 at% O. On the other hand, the  $2\theta$ - $\omega$  scan at  $\psi = 43.5^\circ$  shows  $L1_0$  (2 0 2) peaks. The peak intensities exhibit low fluctuation with various  $T_s$  except the sample with  $T_s = 450$  °C shows lower intensity. The inset figure in Fig. 1(b) shows the illustration of the measurement at  $\psi = 0$  and  $43.5^\circ$ , as well as the in plane rotation angle  $\phi$ .

Fig. 2 shows the  $\phi$  scan XRD patterns for  $L1_0$  {2 0 2} peaks at  $\psi = 43.5^\circ$  and MgO {2 2 0} peaks at  $\psi = 45^\circ$  for a range of  $360^\circ$ , where the angle  $\phi$  is the rotation angle whose axis is the thin film normal, as shown in Fig. 1(b). As can be seen, the  $L1_0$  {2 0 2} peaks show a four-fold symmetry, demonstrating a tetragonal structure. On the other hand, the MgO {2 2 0} peaks show similar four-fold symmetry. No offset was observed for the  $L1_0$  {2 0 2} and MgO {2 2 0} peaks. Therefore, it can be determined that the  $L1_0$  MnGa is grown on the MgO lattice epitaxially and well ordered.

The order parameter  $S$  was estimated from the superlattice (0 0 1) and fundamental (0 0 2) peak intensities. Considering that the theoretical integrated intensity  $I$  of x-ray diffraction peaks can be written as

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