Epitaxial $Mn_{2.5}Ga$ thin films with giant perpendicular magnetic anisotropy for spintronic devices

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We report on epitaxial growth and magnetic properties of $Mn_{2.5}Ga$ thin films, which were deposited on Cr/MgO single crystal substrates by magnetron sputtering. X-ray diffraction results revealed the epitaxial relationships as $Mn_{2.5}Ga(001)[100]\|Cr(001)[110]\|MgO(001)[100]$. The presence of (002) and (011) superlattice peaks indicates that the films were crystallized into DO_{22} ordered structures. The perpendicular magnetic anisotropy (PMA) properties were found to be related to the extent of DO_{22} chemical ordering. A giant PMA ($K_u^{\text{eff}}=1.2\times10^7$ erg/cm^3) and low saturation magnetization ($M_s=250$ emu/cm^3) can be obtained for the film with highest chemical ordering parameter ($S=0.8$). © *2009 American Institute of Physics*. [DOI: 10.1063/1.3108085]

Thin films with perpendicular magnetic anisotropy (PMA) are one type materials featured with easy magnetization axes perpendicular to the film surface. Since the discovery of PMA property in a Gd–Co amorphous film,[1] a variety of PMA films have been developed to overcome the superparamagnetic limit and enhance the thermal stability of ultrahigh-density magnetic storage devices.[2–6] Recently, research interest in applications of PMA films in the field of spintronics has increased remarkably. Spin valves[7–9] and magnetic tunneling junctions[10–13] with perpendicular anisotropy have been demonstrated to allow the realization of low-dimensional and highly reliable spintronic devices, e.g., spin-transfer switching magnetoresistive random access memory. Also, several physical phenomena including efficient electron spin injection into semiconductor from ferromagnetic contact in remanence[14,15] enhanced output signals in a spin-torque oscillator[16] and giant spin Hall effects[17] have been observed in spintronic devices made of PMA films.

For applications in spintronic devices, materials possessing large spin polarization (P) and low saturation magnetization (M_s) are strongly desired.[7,18] However, none of the present used PMA materials can satisfy this requirement. Mn_xGa ($x=2-3$) with DO_{22} crystal structure has the potential to possess such properties simultaneously according to previous theoretical and experimental results for polycrystalline alloys.[19–23] In this structure, an atomic layer of Mn and a layer containing both Mn and Ga atoms are arranged periodically along the c-axis [Figure. 1(a)]. A potential high PMA property has been proposed by approximating the M-H curves of polycrystalline alloy.[19] Classic neutron scattering experiments showed that this material had a ferrimagnetic structure and the magnetic moments of Mn atoms at sites I and II [Fig. 1(a)] were $2.8\pm0.3\mu_B$ and $1.6\pm0.2\mu_B$, respectively. Consequently, a low M_s can be observed at room temperature although the Curie temperature is higher than 770 K.[20] The theoretically calculated spin polarization value as high as 88% at the Fermi level originates from the strong difference in conductivity for minority and majority electrons,[21] which is a typical feature in type-III half metals.[24] To better comprehend the intrinsic properties of this material and realize its practical applications in spintronics, epitaxial $Mn_{2-3}Ga$ single crystal films are preferable, although they are still unavailable.

In this work, 100 nm thick $Mn_{2.5}Ga$ films were prepared on Cr-buffered MgO (001) single crystal substrates by a magnetron sputtering system with a base pressure of 10^{-6} Pa. Detailed experimental procedure for the epitaxial Cr buffer layer can be found in the literature.[25] For $Mn_{2.5}Ga$ films, Ar pressure and sputtering power were set as 0.1 Pa and 25 W, respectively. The substrate temperature (T_s) was varied from 250 to 500 °C. After deposition, the films were covered with a Ta layer of 2 nm for oxidation protection. The composition of the films was determined by an inductively coupled plasma mass spectrometer method. X-ray diffraction (XRD) patterns were measured by a high-resolution x-ray diffractometer (Bruker D8 Discover). The magnetization curves were measured at room temperature by a superconducting quantum interference device.

Figure 1(b) shows the XRD θ-2θ patterns of the $Mn_{2.5}Ga$ films prepared at different T_s. In addition to peaks from MgO and Cr, only $Mn_{2.5}Ga$ (002) and (004) peaks can be observed, indicating that the $Mn_{2.5}Ga$ films were grown with the tetragonal c axis along the normal direction. The crystalline quality of the films was evaluated by full width at half maximum (FWHM) value of the (004) peak [Fig. 1(c)]. With T_s increasing from 250 to 400 °C, the FWHM decreases from 0.70° to 0.42°, suggesting the crystalline qualities of the $Mn_{2.5}Ga$ films be improved. However, the FWHM increases suddenly to 0.70° again when T_s changes to 450 °C and (004) peak shifts toward high angle in θ-2θ pattern. We speculated this deterioration of crystalline quality originated from the diffusion of Cr buffer into the $Mn_{2.5}Ga$ films, although further study should be done to clarify this speculation. Since the XRD patterns of the DO_{22} and $L1_0$ crystal structure are quite similar,[19,23] it is impossible to differentiate between them using only θ-2θ measure-

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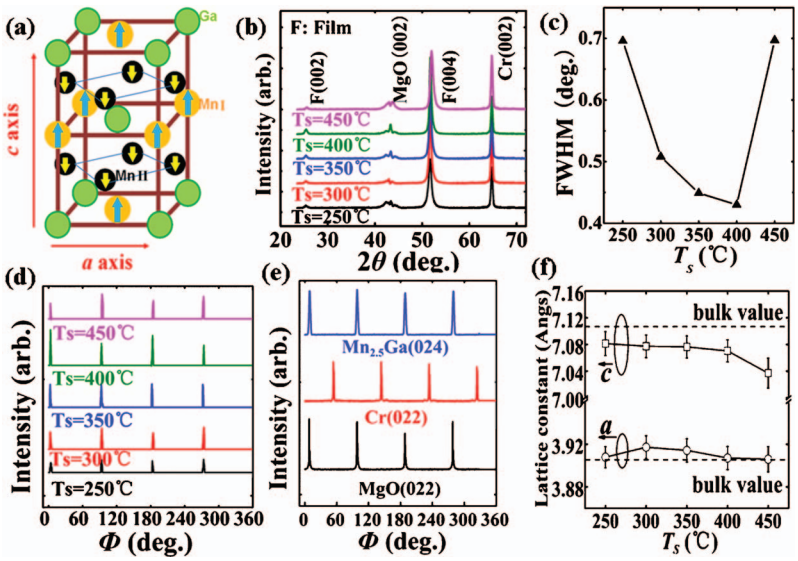

FIG. 1. (Color) (a) Schematic illustration of DO_{22} tetragonal crystal structure. The arrows in the Mn sites denote the direction of magnetization. (b) XRD θ-2θ patterns and (c) FWHM of 004 peaks of films prepared at different T_s. (d) Pole figures of the (011) peaks for the $Mn_{2.5}Ga$ films prepared at different T_s. (e) Pole figures of the $Mn_{2.5}Ga/Cr/MgO$ heteroepitaxial structure in which the $Mn_{2.5}Ga$ film was prepared at 400 °C. (f) The relationships between lattice constants and T_s.

ments for our films. Hence, pole figure measurements (Φ-scan) were employed to confirm the crystal structure of the $Mn_{2.5}Ga$ films. The presence of fourfold (011) peaks in all $Mn_{2.5}Ga$ films clearly indicates that the films have crystallized into the DO_{22} structure because the (011) peak is a forbidden diffraction peak in the $L1_0$ crystal structure [Fig. 1(d)]. To determine the epitaxial relationships between the $Mn_{2.5}Ga$ films and Cr/MgO substrates, we measured the Φ-scans of the MgO (022), Cr (022), and $Mn_{2.5}Ga$ (024) peaks. A typical result for the film prepared at 400 °C is displayed in Fig. 1(e). The epitaxial relationship can be determined as $Mn_{2.5}Ga(001)[100]\|Cr(001)[110]\|MgO(001)\times[100]$, which also existed in the films prepared at other substrate temperatures. Lattice constants c and a for epitaxial $Mn_{2.5}Ga$ films were plotted as a function of T_s in Fig. 1(f). The value of a is approximately equal to the bulk value while the value of c was found to be almost constant in the temperature region from 250 to 400 °C and smaller than the corresponding value for polycrystalline alloy.[22] Such deviation implies that tensile strain exists in the $Mn_{2.5}Ga$ epitaxial films because of lattice mismatch, which results from the different lattice spacing in $Mn_{2.5}Ga$ [100] (3.909 Å) and Cr [110] (2.036 Å). When one lattice of $Mn_{2.5}Ga$ matches two lattices of the Cr buffer, the lattice of $Mn_{2.5}Ga$ would contract along the c axis.

The in-plane and out-of-plane M-H curves of the film prepared at 400 °C are given in Fig. 2(a). A well-squared loop is observed for the out-of-plane M-H curve. However, the in-plane M-H curve exhibits almost zero remnant magnetization and cannot be saturated even under a magnetic field of 50 kOe. The results indicate that the easy axis of the magnetization is perpendicular to film plane. Also, this feature holds for the films prepared at other temperatures. The dependence of M_s on T_s for epitaxial $Mn_{2.5}Ga$ films is shown in Fig. 2(b), where the value of M_s was obtained from the out-of-plane M-H curve. From 250 to 300 °C, a sharp increase in M_s is observed. Further increase in T_s has a little effect on M_s and M_s stays at a nearly invariant value of 250 emu/cm^3, which is larger or smaller than the bulk value.[22,23] For polycrystalline alloys, the M-H curves cannot

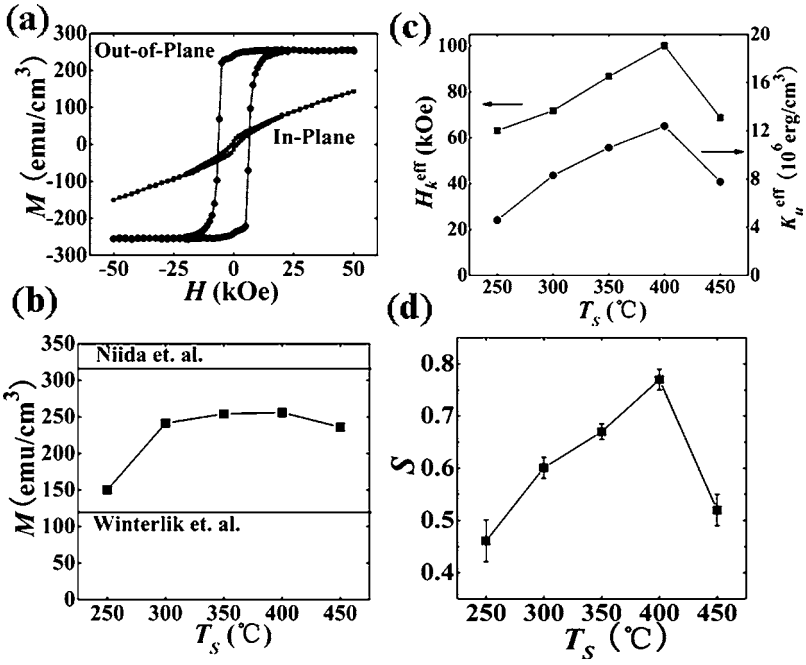

FIG. 2. (a) In-plane and out-of-plane M-H curves of the epitaxial $Mn_{2.5}Ga$ film prepared at 400 °C. (b) Saturation magnetization (M_s) of films prepared at different T_s. (c) Dependence of effective anisotropic field (H_k^{eff}) and anisotropic energy (K_u^{eff}) on T_s. (d) The chemical ordering parameter (S) at different T_s.

be saturated fully under a high magnetic field of 150 kOe, thus, such a difference between our films and those of polycrystalline alloys is not unusual. As mentioned above, obtaining a PMA material with low M_s is crucial for applications in spintronic devices. In this respect, our films are superior to the $L1_0$-ordered PMA films, like FePt (Refs. 4 and 9) and CoPt,[13] and can be comparable with the well-known ferrimagnetic PMA films, e.g., TbFeCo.[10]

To evaluate the PMA properties quantitatively, the effective magnetic anisotropy energy (K_u^{eff}) was estimated using the relations $K_u^{\text{eff}}=M_s H_k^{\text{eff}}/2$. Here, the effective anisotropy field (H_k^{eff}) was defined as the extrapolated intersection of the in-plane *M-H* curve with the saturation magnetization value of the out-of-plane *M-H* curve. Figure 2(c) displays H_k^{eff} and K_u^{eff} versus T_s. With increasing T_s, both H_k^{eff} and K_u^{eff} increase monotonically and reach maximum values at 400 °C and then decrease with further increase in T_s. The highest K_u^{eff} value (1.2×10^7 erg/cm^3) is comparable to that of other high PMA films, e.g., PtFe and CoFe.[4,9,13] Considering that the growth temperature of PMA films should be as low as possible for practical applications in spintronic devices, the epitaxial $Mn_{2.5}Ga$ film is very promising because high PMA can be obtained at relative low growth temperature.

The degree of chemical ordering as a parameter reflecting the probability of correct site occupation in the crystal structure often has significant influence on the final PMA property of the $L1_0$-ordered alloy film.[4,13] We investigated the degree of chemical ordering in the epitaxial $Mn_{2.5}Ga$ films to understand why PMA properties are enhanced with increasing T_s. In the DO_{22} structure with a chemical formula of X_3Y, the atom positions are (0,0,0) and (1/2,1/2,1/2) for Y atoms, (1/2,1/2,0) and (0,0,1/2) for X(I) atoms, and (0,1/2,1/4), (1/2,0,1/4), (0,1/2,3/4), and (1/2,0,3/4) for X(II) atoms. Reflection conditions for the DO_{22} structure permit only refection from the (hkl)-plane with the relationship of $h+k+l=2n$ (h, k, and l: Miller indices; n: integer). Fundamental and superlattice peaks can be determined from the structure factor F using a scattering factor for the atoms occupying each of sites f_Y, $f_{X(I)}$, and $f_{X(II)}$ as described below. When l is even and h, k, and $l/2$ are all even or all odd, i.e., (004) and (024), the structural factor for the fundamental peaks is $F=2|f_{X(I)}+2f_{X(II)}+f_Y|$ and the intensities of these peaks are independent of the degree of order. On the other hand, when l is even and h, k, and $l/2$ are mixtures of even and odd, i.e., (002), the structural factor for superlattice peaks is $F=2|f_{X(I)}+f_Y-2f_{X(II)}|$, which is affected by the disorder between the Mn and Mn–Ga planes. When l is odd, i.e., (011), the structural factor for another superlattice peaks is $F=2|f_{X(I)}-f_Y|$, which decreases only by the preferential disorder between Y and X(I) atoms in the Mn–Ga planes. Based on the above guidelines, the (011) and (024) peaks were chosen as the superlattice and fundamental peak, respectively, to evaluate the ordering parameter (S) using the equation of $S=\sqrt{(I_{011}/I_{024})_{\text{meas}}/(I_{011}/I_{024})_{\text{calc}}}$. Here, I_{011} and I_{024} are the integrated intensities of the (011) superlattice and (024) fundamental peak and $(I_{011}/I_{024})_{\text{meas}}$ and $(I_{011}/I_{024})_{\text{calc}}$ are the measured and calculated diffraction intensity ratios, respectively. We took into account the Lorentz-polarization factor, temperature factor, absorption factor, multiplicity factor, etc. in the calculation of the ordering parameter S. As shown in Fig. 2(d), the change tendency of chemical ordering parameter (S) with T_s is nearly the same as that of K_u^{eff} versus T_s. Thus, it can be considered that the enhancement of K_u^{eff} originates primarily from the improved chemical ordering in the DO_{22} crystal structure.

In summary, we investigated the effects of substrate temperature on structural and magnetic properties of epitaxial $Mn_{2.5}Ga$ films. At optimum growth condition, the film exhibits giant PMA ($K_u^{\text{eff}}=1.2\times10^7$ erg/cm^3) and low saturation magnetization ($M_s=250$ emu/cm^3). Further experiments are being carried out to testify its high spin polarization.

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[1] P. Chaudhari, J. J. Cuomo, and R. J. Gambino, IBM J. Res. Dev. **17**, 66 (1973).
[2] S. Iwasaki and K. Ouchi, IEEE Trans. Magn. **14**, 849 (1978).
[3] P. F. Carcia, J. Appl. Phys. **63**, 5066 (1988).
[4] T. Seki, T. Shima, K. Takanashi, Y. Takanashi, E. Matsubara, and K. Hono, Appl. Phys. Lett. **82**, 2461 (2003).
[5] J. Sayama, K. Mizutani, T. Asahi, and T. Osaka, Appl. Phys. Lett. **85**, 5640 (2004).
[6] S. N. Piramanayagam, J. Appl. Phys. **102**, 011301 (2007).
[7] S. Mangin, D. Ravelosona, J. A. Katine, M. J. Carey, B. D. Terris, and E. E. Fullerton, Nature Mater. **5**, 210 (2006).
[8] H. Meng and J. P. Wang, Appl. Phys. Lett. **88**, 172506 (2006).
[9] T. Seki, S. Mitani, K. Yakushiji, and K. Takanashi, Appl. Phys. Lett. **88**, 172504 (2006).
[10] N. Nishimura, T. Hirai, A. Koganei, T. Ikeda, K. Okano, Y. Sekiguchi, and Y. Osada, J. Appl. Phys. **91**, 5246 (2002).
[11] J. H. Park, C. Park, T. Jeong, M. T. Moneck, N. T. Nufer, and J. G. Zhu, J. Appl. Phys. **103**, 07A917 (2008).
[12] M. Nakayama, T. Kai, N. Shimomura, M. Amano, E. Kitagawa, T. Nagase, M. Yoshikawa, T. Kishi, S. Ikegawa, and H. Yoda, J. Appl. Phys. **103**, 07A710 (2008).
[13] G. Kim, Y. Sakuraba, M. Oogane, Y. Ando, and T. Miyazaki, Appl. Phys. Lett. **92**, 172502 (2008).
[14] N. C. Gerhardt, S. Hovel, C. Brenner, M. R. Hofmann, F. Y. Lo, D. Reuter, A. D. Wieck, E. Schuster, W. Keune, and K. Westerholt, Appl. Phys. Lett. **87**, 032502 (2005).
[15] A. Sinsarp, T. Manago, F. Takano, and H. Akinaga, Jpn. J. Appl. Phys., Part 2 **46**, L4 (2007).
[16] D. Houssameddine, U. Ebels, B. Delaet, B. Rodmacq, I. Firastrau, F. Ponthenier, M. Brunet, C. Thirion, J. P. Michel, L. Prejbeanu-Buda, M. C. Cyrille, O. Redon, and B. Dieny, Nature Mater. **6**, 447 (2007).
[17] T. Seki, Y. Hasegawa, S. Mitani, S. Takahashi, H. Imamura, S. Maekawa, J. Nitta, and K. Takanashi, Nature Mater. **7**, 125 (2008).
[18] K. Yagami, A. A. Tulapurkar, A. Fukushima, and Y. Suzuki, Appl. Phys. Lett. **85**, 5634 (2004).
[19] H. Masumoto, K. Watanabe, and M. Mitera, J. Jpn. Inst. Met. **42**, 474 (1978).
[20] E. Krén and G. Kádár, Solid State Commun. **8**, 1653 (1970).
[21] B. Balke, G. H. Fecher, J. Winterlik, and C. Felser, Appl. Phys. Lett. **90**, 152504 (2007).
[22] J. Winterlik, B. Balke, G. H. Fecher, and C. Felser, Phys. Rev. B **77**, 054406 (2008).
[23] H. Niida, T. Hori, H. Onodera, Y. Yamaguchi, and Y. Nakagawa, J. Appl. Phys. **79**, 5946 (1996).
[24] J. M. D. Coey, M. Venkatesan, and M. A. Bari, in *Lecture Notes in Physics*, edited by C. Berthier, L. P. Levy, and G. Martinez (Springer, New York, 2002), Vol. 595, pp. 377–396.
[25] Y. Sakuraba, J. Nakata, M. Oogane, H. Kubota, Y. Ando, A. Sakuma, and T. Miyazaki, Jpn. J. Appl. Phys. **44**, 6535 (2005).