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Effect of substrate temperature on structure, magnetic and electrical transport properties of Mn\(_{1.60}\)Ga films

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Abstract

L\(_{10}\)-MnGa alloys have great application potential in the fields of permanent magnet and spintronics. L\(_{10}\)-Mn\(_{1.60}\)Ga thin films were prepared on MgO (100) substrates by magnetron sputtering under different substrate temperatures ranging from 200 to 600 °C, and the effect of the substrate temperature on the structure, magnetic and electrical transport properties of the films were observed. The results show that the films deposited at 300–600 °C substrate have good (001) orientation. The coercivity and effective anisotropy of film are enhanced at 400 °C substrate temperature, because the large tensile stress/strain induces a large coercivity and high effective anisotropy. When the substrate temperature increases to 500 °C, and the abnormal Hall resistivity of the film reaches the maximum value. The magnetic and electrical transport properties of the L\(_{10}\)-Mn\(_{1.60}\)Ga film are improved effectively with an appropriate substrate temperature.

1. Introduction

L\(_{10}\)-MnGa alloy is considered as one of the promising candidate for rare earth-free and noble metal-free permanent magnet due to its large coercivity (\(H_c\)), high magnetocrystalline anisotropy constant (\(K_u\)), high Curie temperature (\(T_c\)), and suitable saturation magnetization (\(M_s\)) [1–3]. Meanwhile, Mn-Ga alloy has a high spin polarization (\(P\)) and low Gilbert damping constant (\(\alpha\)). These properties make L\(_{10}\)-MnGa alloys have a prosperous application in spintronic device due to low power consumption magnetization switching [4]. For the tetragonal L\(_{10}\)-Mn\(_{1-x}\)Ga, it is ferromagnetic ordered alloy when 0.76 \(\leq x \leq 1.8\). According to the theoretical calculation results, it is concluded that the \(K_u\) of L\(_{10}\)-MnGa alloy can reach 26 Merg \(c^{-1}\). [5], \(M_s\) is 845 emu \(c^{-1}\). [6, 7], and the maximum magnetic energy product (\(BH\)\(_{max}\) = \((2\pi M_s)^2\)) is 28 MGOe [8]. P of Fermi level is 71% [5] and \(\alpha\) is 0.0003 [6]. L\(_{10}\)-MnGa alloys have great application potential in the fields of permanent magnet material and spintronic device. As previously reported, regulating annealing temperature [9, 10], thickness [11] and substrate [12, 13] could improve (001) preferred orientation growth, magnetic and electrical transport properties of L\(_{10}\)-MnGa film.

In our work, L\(_{10}\)-Mn\(_{1.60}\)Ga films with (001) orientation were deposited on (100) oriented MgO single crystal substrates under different substrate temperatures, the magnetic and electrical transport properties of L\(_{10}\)-Mn\(_{1.60}\)Ga films were studied. The lattice mismatch between L\(_{10}\)-MnGa alloy and MgO single crystal substrate is about 7.8%. The thermal expansion coefficient of MnGa is larger than that of MgO [14]. By changing substrate temperature, the lattice mismatch between the sample and the substrate can be adjusted, thus affecting the structure, magnetic and electrical transport properties of the thin film. We investigated systematically the effect of the substrate temperature on the structure, magnetic and electrical transport properties of L\(_{10}\)-Mn\(_{1.60}\)Ga films.

2. Experiments

Mn\(_{1.60}\)Ga films were deposited on MgO (100) substrates by direct current power on an ATC 1800-F magnetron sputtering system. The nominal thickness was 15 nm and the substrate temperature was 200–600 °C. The base
3. Results and discussion

Figure 1 (a) shows XRD patterns of Mn$_{1.60}$Ga films prepared at different substrate temperatures. There is no diffraction peak related to Mn-Ga phase in the film deposited on 200 °C substrate, which indicates that the film is amorphous at this temperature. When substrate temperature increases to 300 °C, $L1_0$-MnGa (001) and (002) diffraction peaks appear at $2\theta = 24.6^\circ$ and 50.3°. The relative intensities of (001) and (002) diffraction peaks increase with increasing substrate temperature. The unmarked peaks in the XRD patterns are from the MgO substrates. As the films deposited at temperature ranging from 300–600 °C, only $L1_0$-MnGa (00 l) diffraction peaks can be observed, showing highly epitaxial growth of the films on the MgO substrate. The variations of lattice constant c as functions of substrate temperature $T_s$ are shown in figure 1 (b). The c decreases firstly then increases with increasing $T_s$, but it is smaller than that of $L1_0$-MnGa bulk material, 3.690 Å (red line) [15]. This result means that the tensile stress/strain exists in the films. As substrate temperature increases to 400 °C, the c of Mn$_{1.60}$Ga film has the minimum value of 3.617 Å, which suggests that the tensile stress/strain is the largest in the film. To evaluate the crystal quality of Mn$_{1.60}$Ga films, we measure the XRD rocking curves and fit the full width at half maximum (FWHM) of the (002) diffraction peaks by Gaussian function. The variations of FWHM for $L1_0$-MnGa (002) diffraction peak with the substrate temperature are shown in figure 1 (c). FWHM decreases from 2.01° to 1.14°, indicating that increasing substrate temperature could improve the crystal quality of $L1_0$-Mn$_{1.60}$Ga film. The thermal expansion coefficient of MnGa is larger than that of MgO, so the lattice mismatch between $L1_0$-MnGa film and MgO single crystal substrate should be reduced in the film deposited at higher temperature, which could promote $L1_0$-MnGa film growth along MgO (001) crystal plane direction and lead to better crystal quality.

HAADF-STEM image, cross-sectional HRTEM image and EDS mapping of Mn$_{1.60}$Ga film deposited at 500 °C are shown figure 2. From HAADF-STEM image, the film shows an island-like morphology. The total pressure of the vacuum chamber was below $8.0 \times 10^{-6}$ Torr and deposition pressure of high-purity Ar was held 3.0 mTorr. Mn and Mn$_{50}$Ga$_{50}$ targets with better than 99.95% were used co-sputtering to prepare the Mn$_{1.60}$Ga film. Before the film deposition, cleaned MgO substrates were heated to the desired substrate temperature for 5 min to avoid moisture contamination. The film was also in situ annealed at substrate temperature for 30 min after deposition. When the substrate was cooled to room temperature, the Ta layer of 8 nm was deposited on the surface of Mn$_{1.60}$Ga film to prevent oxidation.

The chemical component of the film was determined to be about Mn$_{1.60}$Ga using Rigaku ZSX Primus II x-ray fluorescence spectroscopy (XRF). Film thickness was measured by x-ray reflectivity (XRR). The structure of the films was observed by Rigaku D/max-2500 x-ray diffractometer (XRD) with Cu Kα radiation operating a voltage of 40 kV and current of 200 mA. The high-angle annular dark field-scanning transmission electron microscope (HAADF-STEM) image, high resolution transmission electron microscope (HRTEM) image, and energy dispersive x-ray spectroscopy (EDS) mapping of the cross-section of Mn$_{1.60}$Ga film were analysed by FEI Tecnai F20 transmission electron microscope (TEM). The magnetic properties were measured at room temperature on Lake Shore 7407 vibrating sample magnetometer (VSM) with magnetic field up to 20 kOe. The Hall effects were observed at room temperature by Lake Shore 775 HMS Matrix Hall effect measurement system (HMS) with magnetic field applied to vertical film plane.
thickness of the Mn$_{1.60}$Ga layer and the Ta layer is about 25.8 nm, and the thickness of Mn$_{1.60}$Ga layer is about 15.6 nm, which is close to experimental design value. The cross-sectional HRTEM image shows the Mn$_{1.60}$Ga film deposited at 500 °C has an epitaxial growth along MgO (001) orientation. Seen from the EDS mapping, the distribution of Mn and Ga elements is uniform, and the spaces between the islands are filled up with Ta element for Mn$_{1.60}$Ga film.

Figure 3 shows the out-of-plane, in-plane hysteresis loops and initial magnetization curves of the Mn$_{1.60}$Ga films. And table 1 summarizes corresponding out-of-plane coercivity $H_{c\perp}$, out-of-plane saturation magnetization $M_{s\perp}$, effective anisotropy constant $K_u^{\text{eff}}$ and maximum magnetic energy product $(BH)_{\text{max}}$. All films deposited at 300–600 °C show hard magnetic properties, the magnetic easy axis is perpendicular to the film surface. $H_{c\perp}$ and $K_u^{\text{eff}}$ increase firstly then decrease with substrate temperature increasing. $K_u^{\text{eff}} = H_AM_u/2$ [16],

| $T_s$ (°C) | $H_{c\perp}$ (kOe) | $M_{s\perp}$ (emu/c.c.) | $K_u^{\text{eff}}$ (erg/c.c.) | $(BH)_{\text{max}}$ (MGOe) |
|---|---|---|---|---|
| 300 | 7.38 | 229.13 | $3.68 \times 10^6$ | 0.95 |
| 400 | 7.59 | 307.78 | $9.52 \times 10^6$ | 1.54 |
| 500 | 6.39 | 347.33 | $9.26 \times 10^6$ | 2.55 |
| 600 | 4.88 | 157.63 | $1.69 \times 10^6$ | 0.40 |
where $H_A$ represents the anisotropy field which could be obtained by concluding the in-plane and out-of-plane hysteresis loops. When the substrate temperature is 400 °C, $H_{c\perp}$ and $K_{\text{eff}}$ reach the maxium value, 7.59 kOe and $9.52 \times 10^6$ erg cm$^{-1}$c., respectively. The large tensile stress/strain could induce large coercivity and high effective anisotropy for the film. $M_{s\perp}$ and $(BH)_{\text{max}}$ increase firstly then decrease. For the film deposited at 500 °C, $M_{s\perp}$ and $(BH)_{\text{max}}$ reach the maxium value, 347.33 emu cm$^{-1}$c. and 2.55 MGOe, respectively. The latter is larger than

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**Figure 3.** Out-of-plane, in-plane hysteresis loops and initial magnetization curves of the Mn$_{1.60}$Ga films.

**Figure 4.** (a) Hall resistivity versus magnetic field curves and (b) variation of Hall resistivity as function of substrate temperature for the Mn$_{1.60}$Ga films.
the maximum magnetic energy product of 2.1 MGOe for the reported \( L_{10}\)-Mn\(_{1.60}\)Ga strip samples [17]. \( L_{10}\)-Mn\(_{1.60}\)Ga thin film with large magnetic energy product is an excellent candidate for new rare earth-free and noble metal-free economic permanent magnets.

We investigated the Hall resistivity of the Mn\(_{1.60}\)Ga films versus applied magnetic field at different substrate temperatures, the results are shown in figure 4(a). For ferromagnetic materials, the total Hall resistivity \( \rho_{H} \) is expressed with \( \rho_{H} = R_{H} + 4\pi R_{M} \), where the \( R_{H} \) is ordinary Hall effect coefficient associated with the Lorentz force acting on the moving charge in the magnetic field, \( H \) represents applied magnetic field, and anomalous Hall effect coefficient \( R_{M} \) is related to the irregularity of the left and right carrier scattering during their spin–orbit interactions in magnetic materials [18]. In the film deposited at 200 °C, there is no anomalous Hall effect, which is because the film does not form \( L_{10} \)-ordered MnGa ferromagnetic phase at this substrate temperature. As the substrate temperature increases from 300 to 600 °C, the films show an anomalous Hall effect. Figure 4(b) shows the variation of Hall resistivity \( \rho_{H} \) as function of substrate temperature \( T_{s} \) for the Mn\(_{1.60}\)Ga films. The anomalous Hall resistivity increases firstly then decreases with substrate temperature increasing. When \( T_{s} = 500 \) °C, \( \rho_{H} \) can reach the maximum value of 1.57 μΩ. cm, which is larger than that of \( L_{10}\)-FePt ferromagnetic thin film (\( \rho_{H} = 0.88 \) μΩ. cm) [19]. In the \( L_{10}\)-Mn\(_{1.60}\)Ga film, \( R_{H} \) is much larger than \( R_{H} \), thus the Hall resistivity can be described by \( \rho_{H} = 4\pi R_{M} \), the \( R_{M} \) values for the \( L_{10}\)-Mn\(_{1.60}\)Ga film deposited at 500 °C is 2.86 \( \times \) \( 10^{-11} \) Ωcm/G. These results mean that \( L_{10}\)-Mn\(_{1.60}\)Ga film has much more magnetic storage and logic devices applications compared to \( L_{10}\)-FePt ferromagnetic thin film [20].

4. Conclusion

We investigated the structure, magnetic and electrical transport properties of Mn\(_{1.60}\)Ga films prepared on single crystal MgO (001) substrates with different substrate temperatures. All films deposited at ranging from 300 to 600 °C have good (001) orientation and crystal quality. The crystal quality could be enhanced with increasing substrate temperature. When the substrate temperature is 400 °C, \( H_{c1} \) and \( K_{ef} \) of the sample can reach the maximum value of 7.59 kOe and 9.52 \( \times \) \( 10^{6} \) erg cm\(^{-3}\), respectively. The large tensile stress/strain induces large coercivity and high effective anisotropy. As the substrate temperature increases to 500 °C, \( M_{s} \) and (\( BH_{\text{max}} \) can obtain the maximum value of 347.33 emu cm\(^{-1}\) and 2.55 MGOe, respectively. And \( \rho_{H} \) reaches 1.57 μΩ. cm, \( R_{H} \) is equal to 2.86 \( \times \) \( 10^{-11} \) Ωcm/G. \( L_{10}\)-Mn\(_{1.60}\)Ga thin films deposited at an appropriate temperature have broad application prospects in the field of rare earth-free and noble metal-free permanent magnet materials and spintronics.

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