Influence of composition on structure and magnetic properties of epitaxial Mn-Ga films

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Abstract. In this study, influence of composition on structure and magnetic properties of epitaxial Mn-Ga films were investigated. The epitaxial Mn-Ga films with different composition were grown on metal (Pt and Cr) buffered MgO substrates by co-sputtering technique. By use of the Pt buffered MgO substrates, the D0 22 phases can be obtained in the Mn-Ga films with the composition ranging from Mn65Ga35 to Mn75Ga25. The crystalline quality and perpendicular magnetic anisotropic properties of the Mn-Ga films were deteriorated with increase of Mn content. The saturation magnetization decreased with the increase of Mn content, which is consistent with the reported results of bulk alloys.

1. Introduction

New materials with multiple properties, i.e. large perpendicular magnetic anisotropy (PMA), high spin polarization and low saturation magnetization ($M_s$), are desired to accelerate further progress of spintronics [1-2]. Mn-Ga alloy with D0 22 crystal structure has great possibility to be such material owing to its special crystalline and magnetic structure [3]-[6]. Recently, the D0 22-structured epitaxial Mn70Ga30 films have been reported to exhibit giant PMA ($K_{eff}=1.2\times10^7$ erg/cc) and low $M_s$ (250 emu/cc) simultaneously [6]. However, there is a strong influence of composition on the magnetic properties and electronic structure of the D0 22 Mn-Ga bulk alloy [4-5]. Investigation of the effects of the composition on the structure and magnetic properties of the Mn-Ga films is also very interesting for practical applications. In this work, the D0 22 epitaxial Mn-Ga films with wide composition range have been obtained by using suitable buffer, and the effects of the composition on its structural and magnetic properties were also studied.

2. Experimental details

Two different materials, namely, epitaxial Cr (40 nm) and Cr (40 nm) /Pt (10 nm) layers grown on MgO (001) single crystal substrates, were used as buffer layers for epitaxial growth of the Mn-Ga films. The experimental details for preparation of the Cr buffers have been described in literature [7]. The Pt layer was deposited at 350 °C on Cr buffered MgO substrate, and was confirmed to be epitaxially grown by pole figure measurement. The 30 nm thick Mn-Ga films were deposited at room temperature by co-sputtering technique in ultra-high vacuum magnetron sputtering system, the
composition was controlled from Mn$_{65}$Ga$_{35}$ to Mn$_{75}$Ga$_{25}$ by changing sputtering power of Mn$_{60}$Ga$_{40}$ and Mn target, respectively. Then, the films were in-situ annealed at 400°C. Finally, the samples were deposited with 5 nm thick Ta capping layers to protect oxidation of surface. The composition of the sample was analyzed by inductively coupled plasma mass spectrometry (ICP). The structural properties were measured by X-ray diffraction meter (XRD) with Cu $K\alpha$ radiation, and the magnetic properties were evaluated by superconducting quantum interference device (SQUID).

3. Results and Discussion

3.1 MgO/Cr/Mn-Ga

Figure 1 shows the XRD patterns of the Mn-Ga films grown on MgO/Cr with different composition. For the films with composition of Mn$_{65}$Ga$_{35}$ and Mn$_{68}$Ga$_{32}$, only (002) and (004) peaks from Mn-Ga can be observed, indicating that the Mn-Ga films are grown along $c$-axis. The films were confirmed to be epitaxially grown with the relationship as follows by pole figure measurements, MgO (001)<100>/Cr (001)<110>/Mn-Ga (001)<100>. The lattice constant of film (bulk) along $a$ and $c$ axis is 3.95 (3.91) Å and 7.04 (7.13) Å. The slight difference with the result of bulk alloy may be due to the existence of tensile strain existed in the Mn-Ga films, which results from lattice mismatch (about 5%) between Cr and Mn-Ga because the lattice constant of Cr and Mn-Ga is 0.408 nm and 0.391 nm, respectively. The full width of half maximum (FWHM) of Mn-Ga (004) peak is only 2.5° from rocking curve measurement. It should be noted that the Mn-Ga (004) peaks become broad and shift to higher angle with increase of Mn concentration to Mn$_{72}$Ga$_{28}$. This means that the Mn$_{72}$Ga$_{28}$ film does not form D0$_{22}$ phase. The difficulty might be due to meta-stability of the D0$_{22}$ structure when the composition is approaching to stoichiometric composition (Mn$_{75}$Ga$_{25}$) in bulk alloy, as described in [5].

Figure 2 shows the M-H curves of the epitaxial Mn-Ga films with composition of Mn$_{65}$Ga$_{35}$ and Mn$_{68}$Ga$_{32}$. Both of them exhibit perpendicular magnetic anisotropy (PMA) property because the magnetization easy axis is perpendicular to film plane. The saturation magnetization and effective anisotropy energy ($K_{u}^{eff}$) is estimated to be around 350 emu/cc and 8×10$^6$ erg/cc. However, there is step in M-H loop of the film with composition of Mn$_{68}$Ga$_{32}$ in perpendicular direction, indicating other phases may be co-existed with the D0$_{22}$ phase although it cannot be detected by X-ray diffraction meter.
It can be concluded that preparation of D0\(_2\) structured Mn\(_{75}\)Ga\(_{25}\) films on MgO/Cr substrates is very difficult. There are at least two reasons, one is the relative large lattice mismatch between Cr and Mn-Ga, the other is the meta-stability of the stoichiometric D0\(_2\) Mn-Ga.

![Figure 2. M-H curves of Mn-Ga films with different composition grown on MgO/Cr substrates, (a) Mn\(_{65}\)Ga\(_{35}\), and (b) Mn\(_{68}\)Ga\(_{32}\). Perpendicular means the applied magnetic field is perpendicular to the films’ plane, and the In-plane means the field is parallel to the films’ plane.](image)

3.2 MgO/Cr/Pt/Mn-Ga

The reason of using Pt as buffer is that the lattice mismatch between Pt and Mn-Ga is almost zero because the lattice constant along a-axis of Mn-Ga and Pt is 3.91 Å and 3.93 Å, respectively.

Only (002) and (004) peaks from Mn-Ga can be observed for the films with composition ranging from Mn\(_{65}\)Ga\(_{35}\) to Mn\(_{75}\)Ga\(_{25}\), and peaks’ position does not change with composition, as shown in Figure 3. The FWHM is only around 1° from rocking curve measurements of Mn-Ga (004) peaks, this value is much smaller than that of the film grown on MgO/Cr substrates, indicating that the crystalline quality was much improved. The epitaxial relationship is MgO (001)<100>// Cr (001)<110>// Pt (001)<100>// Mn-Ga (001)<100> according to pole figure results. The lattice constant along a and c axis is 3.91 Å and 7.20 Å, respectively, which is independent on composition. This independence is quite different from that of bulk alloy, and the reason is unclear.

![Figure 3. XRD patterns of the Mn-Ga films with different composition grown on MgO/Cr/Pt substrates](image)

Figure 4 gives M-H curves of the films with different composition grown on MgO/Cr/Pt substrates. The perpendicular magnetic anisotropy can be observed in all the samples. However, the squareness of loop in perpendicular direction becomes poor with increase of Mn content, suggesting that the quality
of PMA properties was deteriorated. Figure 5 shows intensity of Mn-Ga (004) peak as function of composition. As can be seen, the intensity of (004) peak decreases with increase of Mn content. That means the crystallization quality decrease with increasing Mn content. Therefore, the deteriorated crystalline quality leads to the poor perpendicular magnetic anisotropy property. Further work is needed to improve the quality of the epitaxial Mn-Ga films.

**Figure 4.** M-H curves of the Mn-Ga films with different composition grown on MgO/Cr/Pt substrates, (a) Mn$_{65}$Ga$_{35}$, (b) Mn$_{68}$Ga$_{32}$, (c) Mn$_{72}$Ga$_{28}$, and (d) Mn$_{75}$Ga$_{25}$. Perpendicular means the applied magnetic field is perpendicular to the films’ plane, and the In-plane means the field is parallel to the films’ plane.

**Figure 5.** The intensity of (004) peak as function of Mn content.

**Figure 6.** The dependence of saturation magnetization on Mn content.
Figure 6 summaries saturation magnetization of the films with different composition. The saturation magnetization decreases continuously from 270 emu/cc to 50 emu/cc when the composition approach to Mn$_{75}$Ga$_{25}$, which is roughly in agreement with the reports results for Mn-Ga bulk alloy including in figure 6 [4].

**Conclusion**

The D0$_{22}$ structured epitaxial Mn-Ga films with wide composition range can be prepared on Pt buffer. And the effects of composition on structure and magnetic properties were investigated. However, the perpendicular magnetic anisotropy property becomes poor with composition approaching to stoichiometry. Further studies are needed to overcome this difficulty.

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