Magnetic Anisotropy and Reversal in Epitaxial FeGa/MgO(001) Films Deposited at Oblique Incidence

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We have epitaxially deposited FeGa films onto MgO(001) substrates at an oblique angle φ varying from 0° to 45° and systematically investigated their magnetization reversal. The square and two-step hysteresis loops were observed in the samples deposited at $\varphi = 0°$ and 15°. The reversed two-step and three-step hysteresis loops were observed in the samples deposited at $\varphi = 30°$ and 45°, in which a strong uniaxial magnetic anisotropy K_u was induced by the oblique deposition. A model based on domain wall (DW) nucleation and propagation was employed to quantitatively describe the angular-dependent behaviors of FeGa epitaxial films, which indicates that, for the samples deposited with increasing φ, the 180° magnetic transitions occurring in the magnetic field orientation between −45° and 45° gradually change from the two successive 90° DW nucleations to the 180° DW nucleation.

Index Terms—**Epitaxial film, magnetic anisotropy, magnetic reversal, oblique incidence.**

I. INTRODUCTION

FeGa ALLOYS exhibit low hysteresis, high tensile strength, and a moderate magnetostriction of 350 ppm under very low magnetic fields. The combination of these properties makes FeGa alloys excellent candidates for applications in actuators and sensors [1], [2]. Recently, with the development of magnetostrictive materials applied in multiferroic composites, spintronics, and microwave devices, FeGa alloys in thin-film form have attracted extensive attention [3]–[6]. Due to the average of crystallographic orientations and inherent stresses, polycrystalline FeGa thin films usually exhibit a reduced magnetostriction less than 100 ppm [7]. Parkes *et al.* [5] have epitaxially grown FeGa thin films on single-crystalline GaAs(001) substrates and achieved the magnetostriction as large as the value of bulk counterpart. In such epitaxial ferromagnetic films, the cubic magnetocrystalline anisotropy is a natural consequence of the fourfold lattice symmetry. In addition, a uniaxial magnetic anisotropy K_u is often found to be superimposed on the intrinsic fourfold anisotropy. The two kinds of magnetic anisotropies determine the magnetic switching processes of the epitaxial magnetic films and the area of their practical application [8], [9]. The oblique deposition is usually employed to control K_u, thus changes the magnetization reversal of magnetic films [10], [11]. This method, via a self-shadowing effect, results in the formation of grains in the plane of the film that are elongated perpendicular to the incident flux direction and with an aspect ratio increasing at a larger deposition angle [12], [13]. Consequently, an in-plane K_u with easy axis perpendicular to the incident flux direction is induced during the growth of magnetic films.

In this paper, the epitaxial FeGa films with various strength of K_u induced by the oblique deposition at different angles with respect to the surface normal were fabricated on single-crystalline MgO(001) substrates. The square, two-step, reversed two-step, and three-step loops were observed in FeGa films with various strength of K_u. A phenomenological model based on the domain wall (DW) nucleation and propagation was employed to quantitatively describe the angular-dependent behaviors of FeGa epitaxial films and explain that the 180° magnetic transitions occurring in the magnetic field orientation between −45° and 45° change from the two successive 90° DW nucleations to the 180° DW nucleation for the films deposited with increasing the oblique angles.

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II. EXPERIMENT

$Fe_{81}Ga_{19}$ films with a nominal thickness of 20 nm were epitaxially deposited on single-crystalline MgO(001) substrates in a magnetron sputtering system with a base pressure below 3×10^{-7} torr. The substrates were annealed at 700 °C for 1 h in a vacuum chamber and then held at 250 °C during deposition. FeGa layers were obliquely deposited at an angle φ varying from 0° to 45° with respect to the surface normal, as shown in the inset of Fig. 1(a). The projection of FeGa atom flux on the plane of the substrates was set perpendicular to the MgO[110] direction, which induces a uniaxial anisotropy perpendicular to the projection of FeGa atom flux, i.e., parallel to the MgO[110] direction. A reference FeGa film was also fabricated with rotating the substrate during the growth of FeGa layer. After deposition, a 5 nm protective Ta layer was deposited on the samples to avoid oxidation. The crystalline structure of FeGa films was characterized by X-ray diffraction (XRD) with Cu-$K\alpha$ radiation using Rigaku D/MAX-2500. The longitudinal (∥) and transverse (⊥) magneto-optical Kerr effect (MOKE) measurements were used to study the magnetic switching processes of epitaxial FeGa films.

III. RESULTS AND DISCUSSION

In the θ–2θ XRD pattern [Fig. 1(a)], the (002) peak of FeGa indicates a good out-of-plane texture.

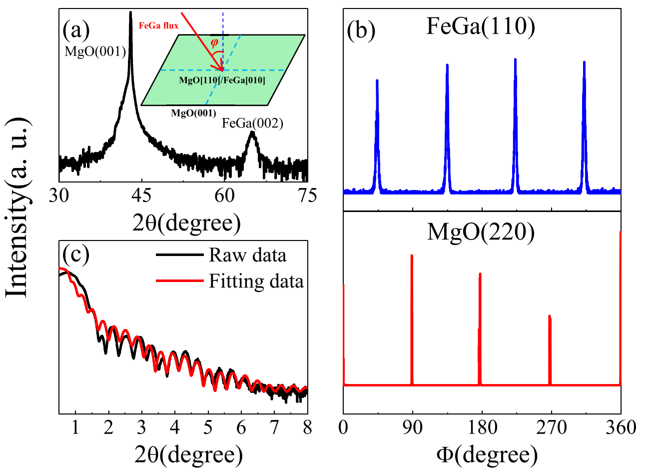

Fig. 1. XRD measurements. (a) θ–2θ scan and (b) in-plane Φ-scans for the reference sample. (c) X-ray reflectivity curves for the reference sample. Black curve: raw data. Red curve: fitting data. Inset: growth of FeGa films by oblique deposition.

The X-ray in-plane Φ-scans [Fig. 1(b)] display a fourfold symmetry, which confirm the epitaxial relationship of MgO(001)[100] ∥ FeGa(001)[110]. By fitting the X-ray reflectivity curves for the reference sample [Fig. 1(c)], the thicknesses of FeGa and Ta are obtained as $t_{\mathrm{FeGa}} = 19$ nm and $t_{\mathrm{Ta}} = 5$ nm, respectively. The root-mean-square roughnesses for both the lower interface between the FeGa layer and the MgO substrate and the upper interface between the FeGa layer and the Ta layer are 0.20 and 0.24 nm, respectively, which indicates the high quality of the epitaxial FeGa films.

Fig. 2(a) shows the geometries of the oblique deposition, the magnetic anisotropies, and the applied field used in this paper. A uniaxial magnetic anisotropy K_u perpendicular to the projection of FeGa flux is superimposed upon the magnetocrystalline anisotropy K_1 of FeGa. Both the longitudinal and the transverse hysteresis loops are obtained at different external magnetic field orientations ϕ in an interval of 5°.

The square and two-step loops, as typically presented in Fig. 2(b) and (c), respectively, are observed in the reference FeGa film deposited with rotating substrate and the samples deposited at $\varphi = 0°$ and 15°. For the square longitudinal loop, the corresponding transverse MOKE signal indicates the magnetization switches between the $[\bar{1}00]$ and [100] axes via the DW nucleation and propagation [Fig. 2(b)]. The corresponding spin orientations are given by the arrows enclosed in a square, as shown in Fig. 2. It is noted that the crystalline directions used in this paper refer to the FeGa lattice, unless indicated otherwise. For the two-step loop obtained at $\phi = 105°$ in the reference FeGa film, the switching route, which occurs for increasing field, is $[0\bar{1}0] \rightarrow [\bar{1}00] \rightarrow [010]$ [Fig. 2(c)]. Apart from the square loop, the reversed two-step and three-step loops, as typically shown in Fig. 2(d) and (e), respectively, are observed in the FeGa films deposited at $\varphi = 30°$ and 45°. The corresponding switching routes for the ascending branches of the loops, which are obtained at $\phi = 95°$ and 65° in the FeGa film deposited at $\varphi = 45°$, are $[0\bar{1}0] \rightarrow [100] \rightarrow [010]$ and $[0\bar{1}0] \rightarrow [\bar{1}00] \rightarrow [100] \rightarrow [010]$, respectively. The FeGa magnetization in the switching route for the reversed two-step loops rotates in the opposite direction when compared with the normal two-step loops observed in the same half quadrant of ϕ [see Fig. 2(c) and (d)].

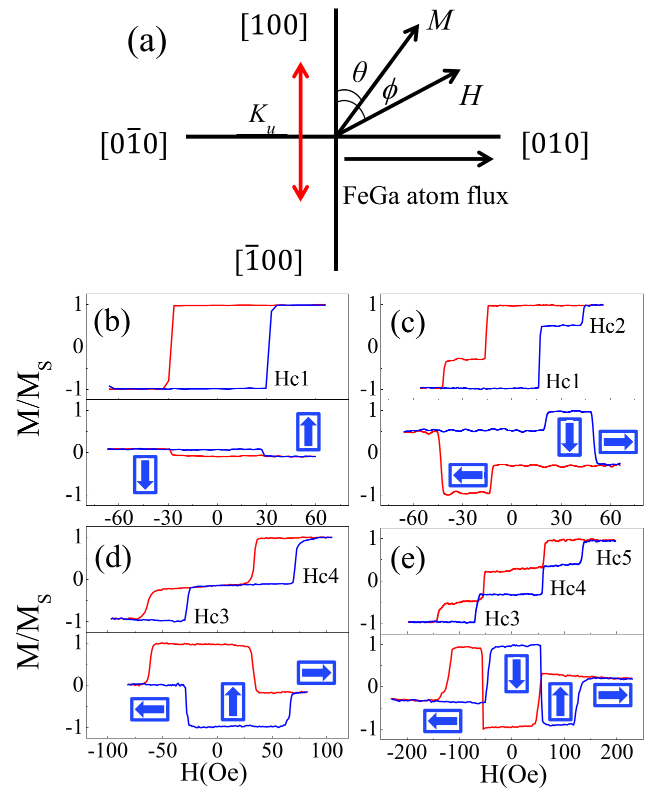

Fig. 2. (a) Geometries of the oblique deposition, the magnetic anisotropies, and the applied field for the FeGa films. A uniaxial anisotropy K_u perpendicular to direction of the FeGa atom flux is induced by the oblique deposition and superimposed on the FeGa cubic anisotropy. Typical longitudinal (∥) and transverse (⊥) MOKE loops measured at various field orientations. (b) Square loop and (c) two-step loop are obtained at $\phi = 0°$ and 105°, respectively, for the reference sample. (d) Reversed two-step loop and (e) three-step loop are obtained at $\phi = 95°$ and 65°, respectively, for the FeGa film deposited at $\varphi = 45°$. Red curve: descending branches of the hysteresis loops. Blue curve: ascending branches of the hysteresis loops. Arrows enclosed by a square: orientation of FeGa magnetization. The switching fields based on the magnetic switching routes are presented as well.

Fig. 3 shows the angular dependence of the switching fields for the reference sample and the obliquely deposited samples. For the reference FeGa film, the square loops with the switching field (H_{c1}) occur in $-15° < \phi < 15°$ and the two-step switching events with the switching fields (H_{c1}, H_{c2}) occur in $-35° < \phi < -20°$, $20° < \phi < 35°$, and $60° < \phi < 125°$ [Fig. 3(a)]. In the range of $-45° < \phi < 45°$, the angular dependence of H_{c1} reveals a peak around $\phi = 0°$ [see Fig. 3(d) for a detailed view]. The FeGa film deposited at $\varphi = 0°$ displays the similar angular-dependent features as the reference sample. For the film deposited at $\varphi = 15°$, only square loops are observed in $-45° < \phi < 45°$ and the two-step loops are obtained in $45° < \phi < 135°$. The corresponding angular dependence of H_{c1} also reveals a peak at $\phi = 0°$, as shown in Fig. 3(b). For the FeGa film deposited at $\varphi = 45°$, the reversed two-step switching processes with the coercive fields (H_{c3} and H_{c4}) are observed at $75° < \phi < 100°$, and the three-step loops with the switching fields (H_{c3}, H_{c4}, and H_{c5}) are obtained at $55° < \phi < 70°$ and $105° < \phi < 125°$, as shown in Fig. 3(c). Only the square loops with the switching field (H_c) are observed in the range of $-45° < \phi < 45°$, and the angular dependence of H_c shows a minimum at $\phi = 0°$ [see Fig. 3(e) for a detailed view]. The FeGa film deposited at $\varphi = 30°$ displays the similar

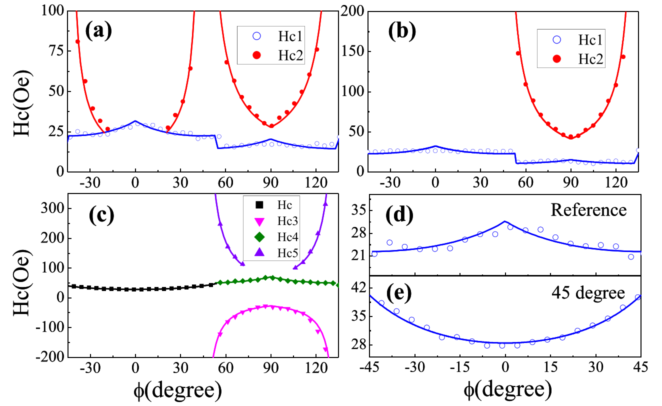

Fig. 3. Typical field orientation dependence of the experimentally observed switching fields (symbols) and the corresponding theoretical fitting results (curves) for (a) reference sample. The films are obliquely deposited at (b) $\varphi = 15°$ and (c) $\varphi = 45°$. The detailed view for the angular dependence of the switching fields of (d) H_{c1} for the reference sample and (e) H_c for the film deposited at $\varphi = 45°$.

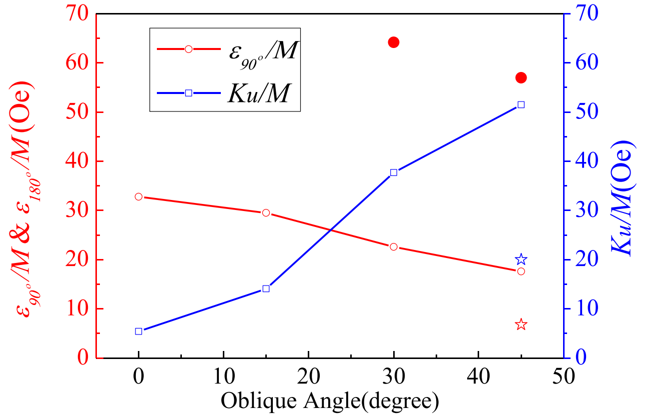

Fig. 4. Parameters $\varepsilon_{90°}/M$, $\varepsilon_{180°}/M$, and K_u/M are given by the fitting for the angular dependence of switching fields for all the FeGa films with different oblique angles. Red dots: values of $\varepsilon_{180°}/M$ for the films deposited at $\varphi = 30°$ and 45°. The red and blue stars represent the values of K_u/M and $\varepsilon_{90°}/M$, respectively, for the reference sample.

angular-dependent features as the one deposited at $\varphi = 45°$, and therefore are not shown here.

IV. Magnetization Reversal Mechanism

The complicated angular dependence of magnetic switching processes of FeGa films with different geometries of anisotropies can be described using a model based on the DW nucleation and propagation [14], [15]. The total free energy for an arbitrary single-domain spin orientation can be written as

$$E = (K_1/4)\sin^2 2\theta + K_u \sin^2\theta - MH\cos(\phi - \theta)$$

where θ is the angle between the magnetization M and the direction of K_u.

The switching fields can be achieved by calculating the energy gain between the local minima at the initial and final easy axes involved in a magnetic transition [14]. Consequently, in the range of $-45° < \phi < 45°$, the theoretical switching fields can be obtained as

$$H_c = \frac{\varepsilon_{180°}}{2M|\cos\phi|}, \quad H_{c1} = \frac{\varepsilon_{90°} + K_u}{M(\mp\sin\phi + \cos\phi)}$$
$$H_{c2} = \frac{\varepsilon_{90°} - K_u}{M(\pm\sin\phi + \cos\phi)}$$

where $\varepsilon_{180°}$ and $\varepsilon_{90°}$ are the 180° and 90° DW nucleation energies, respectively.

In $45° < \phi < 135°$, one can obtain

$$H_{c1} = \frac{\varepsilon_{90°} - K_u}{M(\sin\phi \pm \cos\phi)}, \quad H_{c2} = \frac{\varepsilon_{90°} + K_u}{M(\sin\phi \mp \cos\phi)}$$
$$H_{c3} = \frac{\varepsilon_{90°} - K_u}{M(\sin\phi \mp \cos\phi)}$$
$$H_{c4} = \frac{\varepsilon_{90°} + K_u}{M(\sin\phi \pm \cos\phi)}, \quad H_{c5} = \frac{\varepsilon_{90°} + K_u}{M(\sin\phi \mp \cos\phi)}.$$

The detailed derivation for the equations can be seen elsewhere [14].

For the reference sample and the FeGa films deposited at $\varphi = 0°$ and 15°, the angular-dependent switching fields in the whole range of ϕ can be fitted by the theoretical equations of H_{c1} and H_{c2}. The fitting parameters K_u/M and $\varepsilon_{90°}/M$ are shown in Fig. 4. It should be noted that the switching fields for the square loops, which correspond to the 180° magnetic transition between $[\bar{1}00]$ and [100], can be fitted by the equation of H_{c1}, but not H_c. In the model of DW nucleations, when $-45° < \phi < 45°$, the equation of H_c, which theoretically shows a minimum at $\phi = 0°$, is obtained by considering the energy gain between the $[\bar{1}00]$ and [100] axes. However, H_{c1}, which predicts a maximum at $\phi = 0°$, is obtained using the energy gains between $[\bar{1}00]$ and $[0\bar{1}0]$ for $-45° < \phi < 0°$ and between $[\bar{1}00]$ and [010] for $0° < \phi < 45°$. The nicely fitting for the peak of angular-dependent switching fields for the square loops by the equation of H_{c1} indicates that the 180° magnetic transitions are mediated via the intermediate axis of $[0\bar{1}0]$ or [010] following the two successive 90° DW nucleations rather than the 180° DW nucleation [16], [17]. The critical angle ϕ_1 separating the appearance of the square and two-step loops is determined by the comparison between H_{c1} and H_{c2}. The relations of $H_{c1} > H_{c2}$ and $H_{c1} < H_{c2}$ correspond to the occurrence of two successive and two separate 90° DW nucleations, respectively. Consequently, one can obtain $\phi_1 = \arctan(K_u/\varepsilon_{90°})$. Using the fitting parameters, the critical angle ϕ_1 can be predicted at 18.8° and 9.3° for the reference sample and the FeGa film deposited at $\varphi = 0°$, respectively, which are in good agreement with the experimental observations. For the FeGa film obliquely deposited at $\varphi = 15°$, a relatively large critical angle $\phi_1 = 25.7°$ is obtained. However, we have not experimentally observed the two-step loops in $-45° < \phi < 45°$. This is because when ϕ is close to the hard axis, i.e., 45°, a large external magnetic field is need to saturate the magnetization, but in the experiment, we did not apply such a large field.

For the obliquely deposited films at $\varphi = 30°$ and 45°, the angular-dependent switching fields for the square loops obtained in the range of $-45° < \phi < 45°$ reveal a minimum at $\phi = 0°$, and can be fitted by the equation of H_c based on the 180° DW nucleation, which suggests that the 180° magnetic transition in $-45° < \phi < 45°$ is no long the two successive 90° DW nucleation but changes to the 180° DW nucleation. For the reversed two-step and three-step loops shown in $45° < \phi < 135°$, the switching fields of H_{c3} and H_{c5} for the 90° magnetic transitions can be nicely fitted by the corresponding theoretical equations. It is interesting that the switching field for the 180° magnetic transition between

$[\bar{1}00]$ and [100] in the three-step loops can be described by the equation of H_{c4} based on the 90° DW nucleation, which indicates that this 180° magnetic transition is still mediated via the two successive 90° DW nucleations. Thus, the magnetic switching route for the ascending branch of the three-step loop actually is $[0\bar{1}0] \rightarrow [\bar{1}00] \rightarrow [010] \rightarrow [100] \rightarrow [010]$. The switching field H_{c4} for the magnetic transition from $[\bar{1}00]$ to [010] is larger than the switching field for the magnetic transition from [010] to [100], which gives rise to the occurrence of the two successive 90° DW nucleations in the middle step of three-step loops. Previous studies have demonstrated that the three-step magnetic switching process appears only when the relation $45° < \phi \leq \arctan(K_u/\varepsilon_{90°})$ is satisfied [15]. For the sample deposited at $\varphi = 45°$, the fitting parameter of $K_u/M = 51.5$ Oe is larger than $\varepsilon_{90°}/M = 17.6$ Oe, which ensures the appearance of three-step loops in such epitaxial ferromagnetic films with the combined fourfold magnetocrystalline anisotropy and uniaxial magnetic anisotropy. Consequently, the critical angle, which separates the three-step and reversed two-step loops, can be predicted at 71.2°, which agree well with the experimental results.

The fitting for the angular-dependent behaviors of the FeGa films grown with different oblique angles indicates that K_u/M increases from 5.4 to 51.5 Oe with increasing φ from 0° to 45°, whereas $\varepsilon_{90°}/M$ and $\varepsilon_{180°}/M$ decrease from 32.8 to 17.6 Oe and 64.2 to 57 Oe, respectively. The increase of K_u is obviously due to the oblique deposition. The decrease of $\varepsilon_{90°}$ and $\varepsilon_{180°}$ results from the thickness reduction of FeGa layers when increasing φ. Previous research has shown that $\varepsilon_{180°} \approx 2\varepsilon_{90°}$ in Fe films [14], while in FeGa films, our result is $\varepsilon_{180°} > 2\varepsilon_{90°}$. The enhanced K_u leads to the occurrence of the reversed two-step loops and the three-step loops. The critical angle of the oblique deposition for the onset of the two kinds of loops in FeGa epitaxial films can be estimated at $\varphi = 22.8°$ via the intersection of the curves of K_u/M and $\varepsilon_{90°}/M$ shown in Fig. 4. In addition, the increase of K_u caused by the oblique deposition gives rise to an evolution in the magnetization reversal mechanism from the two successive 90° DW nucleations to the 180° DW nucleation for the magnetic switching from the $[\bar{1}00]$ to [100] axes in $-45° < \phi < 45°$, as shown in Fig. 3(d) and (e).

V. Conclusion

In conclusion, we have epitaxially grown FeGa films on MgO(001) substrates by oblique deposition at an incidence angle varying between 0° to 45°. The square and two-step loops were observed in the reference sample and the FeGa films deposited at $\varphi = 0°$ and 15°. Besides the square loop, the reversed two-step and three-step loops were observed in the samples deposited at $\varphi = 30°$ and 45°, in which a strong K_u was induced by the oblique deposition. A model based on the DW nucleation and propagation was employed to quantitatively describe the evolution of angular-dependent behaviors of epitaxial FeGa films. In $-45° < \phi < 45°$, the angular dependence of H_{c1} for the reference sample and the obliquely deposited samples with $\varphi = 0°$ and 15° reveals a peak around $\varphi = 0°$, which can be described with the magnetization reversal mechanism of two successive 90° DW nucleations. However, for the samples deposited at $\varphi = 30°$ and 45°, the angular dependence of H_c shows a minimum around $\phi = 0°$, which can be explained by the 180° DW nucleation. The change in the magnetization reversal mechanism for the 180° magnetic transitions can be ascribe to the enhanced K_u caused by the oblique deposition.

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